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# Advances in control assembly materials for water reactors

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#### FOREWORD

The safe, reliable and economic operation of nuclear reactors depends to a large extent upon the reliable operation of control rods. In water reactors, these consist of neutron absorbing materials, clad in stainless steel or zirconium based alloys. Current designs have worked extremely well in normal conditions, but occasionally reported failures, associated with the irradiation degradation of the neutron absorbing materials themselves, or wear from contact between the cladding and guide cards or guide tubes, underline the fact that improvements could be made if the underlying mechanisms were better understood.

Neutron absorbing materials currently in use include the ceramic boron carbide, the high melting point metal hafnium and the low melting point complex alloy Ag-In-Cd. Other promising neutron absorbing materials, such as dysprosium titanate, are being evaluated in Russia. These control materials exhibit widely differing mechanical, physical and chemical properties, which must be understood in order to be able to predict the behaviour of control rod assemblies.

Identification of existing failure mechanisms, end-of-life criteria and the implications of the gradual introduction of extended burnup, mixed oxide (MOX) fuels and more complex fuel cycles constitutes the first step in a search for improved materials and designs. In the IAEA's programmes in the past, the subject of control rods and control assemblies has been treated only as a part of more extensive safety studies. It became clear that treatment of the subject area from the point of view of materials science was long overdue.

To obtain an overall picture of the current usage of control materials, research under way on new materials and to identify areas where new materials are necessary to improve the safety, reliability and/or economics of water reactors, the IAEA convened a Technical Committee Meeting on Advances in Control Materials for Water Reactors. This meeting was recommended by the International Working Group on Fuel Performance and Technology and was held in Vienna from 29 November to 2 December 1993. Twenty-seven participants from twelve different countries attended the meeting and twelve papers were presented and are reproduced in these proceedings together with a summary of the meeting.

The IAEA wishes to thank all the participants for their contributions to this publication, which summarizes experience to date with control materials and ongoing research in the field in the participating countries. The officer of the IAEA responsible for the organization of the meeting and for the compilation of this TECDOC was I.G. Ritchie of the Nuclear Materials and Fuel Cycle Technology Section.

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### SUMMARY OF THE TECHNICAL COMMITTEE MEETING

### INTRODUCTION

For the purposes of this meeting, "Control Materials" included all of the solid materials used in the construction of control assemblies for the regulation or shut down of water-cooled, power reactors, i.e. neutron absorber(s), cladding, guide tubes and guide cards, and other structural components of the assembly. Liquid neutron absorbers and poisons were not considered and, for the most part, the deliberations dealt with the behaviour of control materials in normal operating and shut down conditions.

The meeting was ably chaired by L. Hallstadius of ABB Atom, Sweden. Twenty-seven participants representing twelve countries attended the twelve formal presentations of papers and the subsequent discussions. The absence of representation from the United States of America was noted and unanimously regretted by the participants.

The twelve papers presented covered the control materials currently used in PWRs, BWRs, WWERs and CANDUs as well as improved materials under investigation for their replacement or for advanced water reactor designs. The technical discussions focused on the performance of absorber materials, cladding, guide-tubes and guide-cards in reactor and, more particularly, on issues affecting and limiting the lifetimes, and hence the economic viability, of control assemblies.

Current designs of control assemblies together with the stringent criteria followed by the utilities for their safe operation, inspection and replacement has led to an outstanding safety record. However, mechanical mechanisms (fretting and wear), corrosion mechanisms (oxidation and hydriding), radiation damage mechanisms (enhanced creep and growth), and mechanisms such as irradiation assisted stress corrosion cracking (IASCC), involving synergism between two or more fundamental mechanisms, all detract from the ideal behaviour of control materials. More importantly, this lessthan-ideal behaviour limits the lifetimes of control materials, imposing an economic penalty which acts as a strong incentive to produce improved materials and designs that are more reliable and approach closer to the ideal lifetime of a control assembly, which should depend solely on the neutronics of the chosen absorber. Improved understanding of the damage mechanisms mentioned above is leading to improved behaviour of control assemblies through design changes, wear-resistant coatings and surface treatments, and better control over microstructures and compositions of the materials at the fabrication stage.

Experience with control materials in water reactors is spread over many different types of reactors in many different countries. Similarly, research, development and design of the different types of control assemblies is carried out by several different vendors. Presentations at the meeting dealt with all of the currently used control materials in most of the water-cooled, power reactor types operated at present. In addition, some of the presentations and discussions were devoted to alternative, advanced absorber materials and developments aimed at increasing the lifetimes of control assemblies.

International Conferences and Symposia on the materials science aspects of control assemblies have been relatively few and far between. Although the IAEA has held many meetings on the safety aspects of control rods and control systems, in both normal operating conditions and severe accident conditions, to our knowledge this is the first meeting to focus on the materials aspects of reactor control materials. Consequently, in bringing together experts in the materials science and materials engineering of control materials, we believe that this meeting and its published proceedings has helped to fill an important gap in the information exchange opportunities in this branch of nuclear research.

Following the formal presentations of the papers and some preliminary discussions, the meeting split into two groups to facilitate discussions and prepare a summary, recommendations for further work in the field and conclusions of the meeting. The first group considered PWRs and WWERs,

while the second group considered BWRs and CANDUs. The results of these discussions are presented in the following sections.

### **PWRs AND WWERs**

This group was led by I. Kennett of BNFL, UK and the initial decision was taken to structure the discussions under the headings of plant experience, detection of damage, management criteria, and improvements and recommendations for further research and development. As is common practice among PWR operators, the control assemblies for both PWRs and WWERs are referred to as RCCAs for simplicity in the following. This acronym stands for Rod Cluster Control Assembly.

### PLANT EXPERIENCE

For WWER-1000 control rods there is no indication to the present that they are significantly affected by wear. Unlike PWRs where the use of guide cards leads to localised fretting and wear, WWERs use a continuous guide tube which tends to spread any wear over a larger area. The life-limiting factor in WWER-1000 is absorber-cladding interaction.

In most PWRs the lifetime of RCCAs is limited by wear or by absorber-clad interaction and is less than expected. Wear is highly dependent on the guide tube design and the dynamic forces generated by the hydraulic flow conditions. For example, the so-called T-hot and T-cold conditions (hot upper-head plants with reduced upper-head by-pass flow, versus cold upper-head plants with increased upper-head by-pass flow) define an important characteristic of the plant's hydraulic flow leading to wear of different severity. Although wear is a generic problem, the spread of the results from plant to plant or from RCCA to RCCA inside a given plant is very large. It is possible to obtain some statistical correlation of wear amplitudes with design characteristics, but very accurate prediction of wear depth with location is still difficult at present, due to the non-linear behaviour of the flowinduced vibration phenomena.

The main causes of swelling are the neutron absorber materials themselves (Ag-In-Cd or  $B_4C$ ) and the neutron fluence accumulated, which depends strongly on the type of RCCA bank (shutdown or regulation) in question and on the plant operating conditions (base versus load-follow).

### **DETECTION OF DAMAGE**

On-site inspection techniques must be able to detect wear, cracks and swelling with good accuracy in an acceptable time. The most commonly used techniques are eddy current testing, ultrasonic testing and mechanical profilometry. To detect leaking rods, the sipping test technique is complementary to the above, while the monitoring of the chemical composition and activity of the coolant can also yield useful information. The cost of surveillance was identified as an important issue.

#### **RCCA MANAGEMENT CRITERIA**

The criteria are first and foremost based on a careful consideration of the consequences of the damage in question (wear or swelling). For safety reasons, damage that is extensive enough to prevent an RCCA from dropping cannot be tolerated under any circumstances. Consequently, rodlet ruptures must be avoided at all cost. The loss of RCCA efficiency must also be taken into account: for example, absorber depletion, and dissolution of  $B_4C$ , but these phenomena may be less critical from a safety point of view. Replacement of RCCAs with leaking Ag-In-Cd absorber is always beneficial in reducing the primary circuit contamination man-rem exposure. Cracks at the bottom end of an RCCA in a PWR lead to much less contamination than under-card, worn-through cladding. Possible damage to the fuel assembly guide tubes in the case of swelling must be taken into account also.

The other concern identified was the possibility of the control rod jamming in the dashpot (due to rod swelling) in the French design.

The criteria also include the kinetics of the damage process, which have to be assessed from previous measurements using a statistical approach.

Clearly the criteria must consider the limitations of the monitoring and inspection measurement techniques, specifically their efficiency and accuracy.

The management strategy takes into account the above criteria, the frequency of inspection, and any axial shifting or shuffling of the RCCAs, which depends upon the normal operating practices of the individual plants.

#### **IMPROVEMENTS AND RECOMMENDATIONS - PWRs**

French designers of new PWR plants have included a new guide tube design for RCCAs which reduces vibrations of the rods.

Some vendors of RCCAs have introduced wear-resistant coatings of chromium or chromium carbide for the cladding of control rods. Others have adopted wear-resistant surface hardening treatments such as ion nitriding for control rod cladding. Up to now, the in-reactor experience with coated cladding of all three types has justified their use. Significant wear reduction has already been measured in plants where previously the uncoated RCCAs were considerably worn. The question of whether or not wear resistant claddings had caused additional wear on the untreated guide cards/guide tubes and whether or not such wear was of concern was not answered satisfactorily.

The efficiency of introducing a gap increase between the absorber material and the cladding at the bottom of the RCCAs is obvious, but it is difficult to evaluate the lifetime gain because it greatly depends on the plant operating conditions.

Further improvements might be brought about by material changes, for example, the use of Hf instead of Ag-In-Cd, but more research and development effort is still required in order to demonstrate acceptable wear resistance, corrosion behaviour and hydrogen pick-up rates.

For future reactors (e.g. reactors designed specifically to burn MOX fuels) that might need more efficient control materials, compounds based on enriched boron and hafnium show considerable promise. Again, more research and development is recommended in this area.

#### **IMPROVEMENTS AND RECOMMENDATIONS - WWERs**

For WWERs it is possible either to change the clad material in the zone of highest fluence to a material less prone to swelling, or to adopt a new absorber such as dysprosium titanate  $(Dy_2O_3.TiO_2)$ or Hf, i.e., replace the n, a-absorber B<sub>4</sub>C in which He-induced swelling occurs with an n, g-absorber. Further, detailed evaluations of  $Dy_2O_3.TiO_2$ , already begun in Russia, are recommended.

### **BWRs AND CANDUs**

This group was led by E.G. Price of AECL CANDU, Canada and the initial decision was made to structure the deliberations around the headings issues (arising from plant experience), inspection and end-of-life criteria, and recommendations for research and development. In contrast to the conditions experienced by control materials in BWRs, the conditions experienced in CANDUs are rather benign. Consequently, little discussion about control materials in CANDU took place.

### **BWR CONTROL RODS: ISSUES**

The main issue discussed was the limited life time of BWR control rods due to the swelling of the absorber material. Boron carbide  $(B_4C)$  is the reference design material in many countries. Its swelling by helium formation leads to absorber-clad interaction and this together with the degradation of the mechanical properties of the cladding material (sensitisation of the stainless steel) under irradiation leads in turn to cracking of the cladding. The cause of failure has often been identified as irradiation assisted stress corrosion cracking (IASCC), but the precise mechanisms of this complex process are not yet clearly understood.

It was emphasised that operating conditions (residence time and integrated flux) vary greatly between cell control rods (CCRs) and shutdown rods (SRs). Consequently, the issues for these two types of control rods are quite different.

For CCRs the major issue is swelling of the  $B_4C$  and sensitisation of the stainless steel with fluence leading to IASCC. Replacement of boron carbide with hafnium in some Japanese BWRs has shown considerable promise. The hafnium is in a flow of coolant water rather than tightly clad, and appears to be self-protecting from hydrogen pick-up under these conditions. However, the well known similarities between the behaviour of zirconium and hafnium suggest that further work on both hydrogen pick-up and irradiation induced growth in hafnium, particularly at high fluences, would be prudent.

For SRs the major issue is swelling of the  $B_4C$  and cracking of the cladding. Added to this is the degradation of the mechanical properties of the structural sections, which can come under stress during handling procedures.

### **INSPECTION AND END-OF-LIFE CRITERIA**

For safety reasons, in the countries represented in the meeting, it is not allowed to operate a reactor with a control rod which has deteriorated to the extent that it could lead to a significant decrease in function or efficiency. End-of-life criteria for control assemblies have been set differently in different countries.

In Japan, for instance, the criterion is based on the integrated fluence and the threshold fixed by results reported in the General Electric database. The limit has been set in order to have a very low probability of failure. Therefore, inspections are not considered to be needed on these standard control rods.

In Scandinavian countries the strategy is based on examination of CCRs after a certain integrated fluence has been achieved. It has been demonstrated that once a failure has been detected, the control rod can sustain an additional irradiation cycle without losing a significant amount of absorber material by wash out. In the future, based on these observations, a threshold will be determined which will take into account any improvements in technology. In any case, the end of life criterion will be based on the integrated fluence.

On-site detection of failed rods is performed by means of visual examination and further information is obtained from neutron radiography.

Another technique for the detection of failed control rods, based on the detection of tritium increase in the primary circuit, is under development by some utilities.

It was noted that little work has been done on disposal of control rods. The assemblies are dismantled, the highly active parts are stored at the reactor site and the remainder is sent to medium level waste storage. In Sweden it is planned to use the repository (CLAB) for both spent fuel and control rods.

### **RECOMMENDATIONS FOR FURTHER R&D**

The main developments necessary have been identified as follows:-

### Swelling

- Evaluate high density  $B_4C$ .
- Look at design solutions to counter swelling.
- Evaluate alternatives such as Dy<sub>2</sub>O<sub>3</sub>.TiO<sub>2</sub> or HfB<sub>2</sub>.
- Evaluate the consequences of reactions between  $B_4C$  and  $H_2O$ .

### Hafnium

- Investigate the possible onset of accelerated growth (very unlikely to be life limiting).
- Establish the optimum, as-fabricated microstructure, texture and composition to reduce growth (or at least accommodate growth in the design).
- Evaluate possible accelerated hydrogen pick-up rates adjacent to joints with stainless steel.

### Cladding and structural material

- The move from 304 to 304L to 316L was approved by the participants as well as the evaluation of other grades of stainless steel found to be resistant to IASCC.
- Investigations to improve our understanding of all of the fundamental mechanisms contributing to IASCC are needed.
- More complete post irradiation examination (PIE) of failed control rods is strongly recommended.

### CANDU

 Since there is some question about the long-term commercial availability of cadmium, which is the neutron absorber used in control and shutdown assemblies of CANDU reactors, further work on other absorbers such as boron loaded stainless steels is recommended.

### **OVERALL RECOMMENDATIONS**

- The participants agreed that the opportunities for information exchange in the field of control materials are far too limited, but that future meetings of this type must include US participation to be able to reflect the status and trends of the industry more accurately.
- It is recommended that a technical meeting should be organised jointly with EPRI on iradiation assisted mechanical degradation and cracking of control rod cladding in water reactors.
- Vendors of CCRs are recommended to pursue more vigorously the development of absorber materials less prone to swelling.
- A database on experience with wear-resistant coated and surface hardened control rod claddings should be established and maintained.

### **OVERALL CONCLUSIONS**

- Although the current designs of control assemblies have an outstanding safety record, their reliability and economic viability can be enhanced by improvements in design and materials behaviour that allow increased lifetimes in the core.
- Wear-resistant coatings and surface treatments of RCCAs in PWRs show great promise, but for new plants improved guide tube designs will reduce fretting and wear.

- It appears that "naked" hafnium, which gained a poor reputation in the clad form because of swelling due to hydrogen pick-up, has excellent properties provided that it resides in the coolant flow where its oxide coating protects it from hydrogen pick-up. This has been demonstrated by research activities in Japan and in a research reactor in France.
- More efficient absorber materials based on enriched boron and hafnium will probably have to be developed for advanced reactor types, for example, reactors designed to burn MOX fuels.

# ADVANCES IN CONTROL ASSEMBLY MATERIALS FOR PWRs AND WWERs

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### DESIGN OF CONTROL RODS FOR PRESSURIZED WATER REACTORS WITH SPECIAL CONSIDERATION OF ABSORBER SWELLING AND CLAD CREEPDOWN

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Abstract

One important mechanism which can influence the life of rod cluster control assemblies is the absorber-clad interaction due to swelling of the absorber material and creepdown of the surrounding cladding. The possible residence time of a control rod is limited by the time it takes for the swelling induced cladding strain to reach the permissible strain limit. Therefore, knowledge of the dimensional behaviour of the absorber and cladding material as a function of neutron fluence is an essential basis for control rod design and life time prediction. Various specimens of absorber material have been irradiated in extended material test programs during the last 15 years under different conditions (restrained, unrestrained by the surrounding clad). Resulting measurement data have been evaluated and compared to results of other published measurements. For design purposes, a conservative swelling curve has been derived. In the same material test program, diameter changes of the cladding enclosing the absorber specimens were also measured and evaluated. These data serve as a basis for the design curve describing the in-reactor creepdown of steel cladding tubes. Meanwhile, a large number of irradiated control rods accumulated fluences near or even higher than the conservatively calculated fluence limit. The experience feedback, based on the measurements of those highly irradiated control rods, allows a verification of the currently used design method and its conservatism on the one hand. On the other hand, the experience feedback enables recommendations for an extension of irradiation periods beyond the once established fluence limits, which had been determined conservatively on the basis of the above mentioned material test programs.

# 1. Introduction

The life of rod cluster control assemblies (RCCAs) is of particular interest to the operator of a reactor plant. Therefore, mechanisms which possibly influence RCCA life have to be analyzed and must be taken into account in the RCCA design.

As demonstrated in Table 1, RCCA design can be considered as an iterating process or a quality circle: starting points are general requests on RCCAs (e. g. shut down capability), anticipated loads during operation and irradiation, material behaviour as known from experiments and literature, and a design methodology, prescribed by guide lines etc..

Then, the material for the absorber and cladding is selected and the design analysis is performed. In parallel to engineering and fabrication of control rods, a test program with specimens and test rods is started.

 Table 1

 Advances in control materials for water reactors: design as iterative process



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Figure 1 PWR RCCA and Control Rod for SIEMENS Convoy Plants

After irradiation and inspection, the evaluation of the operational experience is a basis for design verification or review, leading – if necessary – to improvements of the material and the design process, to enlarged test programs and improved inspection techniques, resulting in an advanced design. In the following, examples of single steps are considered, which constitute this still ongoing design process.

# 2. Description of RCCAs

A typical RCCA design (Figure 1) features a spider assembly with compression spring, bolt and spring cup, and control rods which are fastened onto the spider assembly. The spider assembly arms are machined from a forging using milling and electrical discharge machining; this fabrication method avoids welded or brazed connections and results in an optimum mechanical strength.

The individual control rods are fastened by a lock-welded nut onto the spider assembly. Each control rod comprises of an absorber which is enclosed gas-tight in a stabilized austenitic steel cladding tube with welded end plugs. The alloy Ag80In15Cd5 is used as absorber material. The remaining volume in the control rod is filled with Helium at atmospheric pressure.

Other types of RCCAs are characterized as follows:

absorber materials:	AgInCd, Hf, B <sub>4</sub> C, and combinations ("hybrid")
cladding materials:	stainless steel, Inconel
length of control rods:	full length, part length

The main difference between the types described above is the use of other absorber materials or combinations of absorber and cladding materials.

# 3. Life–limiting Mechanisms

The life of PWR RCCAs is limited on the one hand by the absorber/clad interaction due to absorber swelling and creepdown of the cladding surrounding the absorber and, on the other hand, by possible wall thinning due to wear. The neutronic life, i.e. the time up to a reduction in RCCA worth of 10 %, is not considered here since it is considerably higher than 30 years [1].

# 3.1 Wear

Wear of control rod cladding may occur as a result of axial movement of the RCCAs and vibration of the control rods against the inner walls of the tubes or plates (sometimes called guidance zones and guidance cards) of the guide assemblies in the upper core structure, vibrations of the guide assemblies themselves, and/or vibrations against the inner walls of the guide thimbles of the fuel assemblies. Wear results in wall thinning, resulting in an increase of the stresses in the cladding, or even in clad wearthrough.

# 3.2 Absorber / Clad Interaction

The alloy AgInCd is a widely used absorber material. As a result of neutron capture, different isotopic conversion processes take place in the absorber, e.g. conversion of In —> Sn and Ag —> Cd as a result of the resonance capture of epithermal neutrons (0.6 eV < E < 0.8 MeV), the sum of which result in an increasing volume of the absorber as a function of neutron fluence [2]. If boron carbide is used as absorber material, swelling takes place due to the formation of lithium and helium. The swelling rate of boron carbide is significantly higher compared to AgInCd. In case of hafnium as the absorber, hydriding leads to a uniform, nearly isotropic volume expansion [3]. The pressure inside the reactor vessel produces an external overpressure onto the cladding, resulting – in connection with neutron irradiation – in creepdown of the steel cladding. After gap closure all further absorber swelling leads to tensile stress in the clad. The allowable residence time is limited by the ductility of the cladding material.

# 4. Design

For German licensing conditions, the task of design is, according to [4], to assure that the control rods are fully capable of withstanding the mechanical loads occurring during normal operation and anticipated operational occurrences. The associated loading conditions are specified in [5].

In addition to the mechanisms described in Section 3, which will be considered in greater detail below, the design has also to fulfil a number of other criteria:

- The melting temperature of the absorber shall not be reached.
- No surface boiling shall occur in the gap between control rod and guide thimble.
- The cladding shall not collapse under the maximum pressure loading.
- The stress loading shall not result in impermissible cladding deformation.
- The cyclic stresses shall remain below the permissible limits.

Observance of these criteria is demonstrated in each case, but is not covered in any further detail here since none of the above-mentioned mechanisms has proven to have a life-limiting effect.

# 4.1 Wear

As described in Section 3.1, wear results in a reduction of the cross-sectional area of the cladding tube and thus in an increase in cladding stresses. The maximum permissible wall thinning is reached when – in accordance with the design criteria [4] – the required margin to control rod fracture is reached.

a) Types of Stress

The following stresses are considered:

- Stresses resulting from forces of inertia during RCCA stepping motion, i.e. compressive/tensile stresses (membrane stresses), distributed uniformly over the cross section. They are described by the equation:

 $\sigma = F/A$ 

where

- $\sigma$ : Membrane stress
- F : Force
- A : Cross-section
- Stresses resulting from bending moments.

Here the maximum rod bending and thus the maximum bending stress is limited by the gap between cladding tube and guide thimble.

# Cyclic stresses. Cyclic stresses occur as a result of bending vibration of the control rods.

b) Conservative Assumptions

The possible shapes of wear marks, connected with the manner in which the control rods are guided in the guide assemblies, are as shown in Figure 2. Considering the maximum bending stress, the postulated one-sided, crescent-shaped wall thinning is conservative for calculation purposes.



Figure 2 Possible shapes of wear marks of control rods

Further conservative assumptions are:

- Minimum cladding cross section
- Maximum bending moment calculated from the maximum guide thimble inside diameter and minimum rod diameter
- Maximum RCCA acceleration during stepping motion
- Force of inertia resulting from the entire control rod mass at the bottom end plug and thus at the location of maximum wall thinning.

# 4.2 Absorber / Clad Interaction

# 4.2.1 Absorber Swelling

Knowledge of the dimensional behaviour of the absorber as a function of neutron fluence is necessary in design analysis. Therefore, in 1979, a material test program has been started to study the swelling behaviour of various absorber materials under irradiation.

# 4.2.1.1 Unrestrained Swelling of AgInCd

Samples of 100 mm length were fabricated with gaps between 110 and 330  $\mu$ m, pre-characterized and then assembled to produce test rods. The gap width was selected in such a way, that the absorber material could easily be extracted from the surrounding cladding in a Hot Cell and measured. The samples have been irradiated for up to now 10 cycles in the guide thimbles of a PWR fuel assembly. To date the specimens have attained a maximum fluence of almost 9 x 10<sup>22</sup> cm<sup>-2</sup> (E > 0.6 eV).

After each cycle the samples were examined visually at pool side, and also non-destructive dimensional measurements were performed. After two, three and seven irradiation cycles samples were unloaded and shipped to the Karlstein hot cells for destructive examinations.

# 4.2.1.2 Results

Density, diameter and length measurements were performed on the AgInCd samples. Further measurements were performed on two control rods. The measuring accuracy is better than 0.1 % for those measurements. The measured data are shown in Figure 3.

After normalizing all swelling data to the same dimensions, an analysis of the measured data yields the following results:

- The swelling behaviour of AgInCd material is a linear function of fluence up to 6 x 10<sup>22</sup> cm<sup>-2</sup> (E > 0.6 eV).
- The swelling behaviour of AgInCd material is isotropic.
- The measured data agree well with results of measurements, which are published in [1, 6, 7].

For design purposes a swelling curve is used which covers the present measuring points with adequate statistical certainty.

diameter change



neutron fluence (E > 0.6 eV) [ $10^{21} \text{ cm}^{-2}$ ]



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diameter change
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Figure 4 Unrestrained and restrained swelling of AgInCd

# 4.2.1.3 Restrained Swelling of AgInCd

In an additional irradiation program, the deformability of austenitic stainless steels and nickel–base alloys was studied combining the influence of irradiation in a light water reactor core environment and high stresses and strains [8]. Tubular specimens filled with AgInCd–swelling mandrels were inserted into fuel assemblies and exposed to the high neutron flux and to the pressure of the coolant water in reactor cores. The gap of the specimens was less than 10  $\mu$ m. At hot condition, this small gap is closed according to the elastic strain by the coolant pressure and the greater thermal expansion of the absorber material compared to stainless steel. The samples are still under irradiation in their forth irradiation cycle. The accumulated fluence up to now is  $3.5 \times 10^{22}$  cm<sup>-2</sup> (E > 0.6 eV).

Under these conditions, the influence of the coolant pressure and of the stainless steel cladding on the swelling behaviour of AgInCd can be studied.

# 4.2.1.4 Results

Figure 4 shows the measured diameter changes, normalized to the dimensions of the unrestrained AgInCd samples, together with the unrestrained swelling values and their best-estimate and conservative swelling curve. As can be seen from Figure 4, the measured normalized diameter changes are well within the scattering band of the unrestrained swelling values and below the conservative swelling curve, derived from the unrestrained swelling values. From these results the following can be derived:

- The swelling rate of AgInCd remains nearly unchanged after gap closure.
- The coolant pressure and the stainless steel cladding have no influence to the swelling rate of AgInCd.

# 4.2.1.5 Influence of the Chemical Composition on the Swelling

A comparison of the chemical composition of the absorber AgInCd in both irradiation programs shows the following table:

Ag in %	In in %	Cd in %	comments
80	15	5	nominal
balance	14.8	5.1	unrestrained
balance	15.5	4.9	restrained

As can be seen from Figure 4, slight changes of the absorber composition have no measurable influence on the swelling behaviour. Therefore, no correction term, related to the chemical composition of the absorber, is necessary.

# 4.2.2 Cladding Creepdown

In the material test program mentioned in Section 4.2.1.1, changes in the diameter of the stainless steel cladding enclosing the absorber specimens were also measured.

The individual segments of the material test rods were visually inspected, measured and evaluated after each irradiation cycle. These data serve as the basis for a design curve which describes the in-reactor creepdown of stainless steel cladding tubes. Figure 5



Figure 5 Diameter changes of stainless steel cladding tubes

shows the measured values as a function of fluence. An analysis of the measured diameter changes yields the following results:

- There is no primary creep under PWR coolant conditions.
- The diameter change depends linearly on the tangential stress, induced by the coolant pressure, as a function of the cladding dimensions.
- The diameter change of stainless steel claddings under unrestrained conditions is a linear function of fluence up to  $1.5 \times 10^{22} \text{ cm}^{-2}$  (E > 1 MeV).

Therefore the diameter change  $\Delta d$  of stainless steel claddings under PWR conditions can be described by the product of the material dependent creep constant K, the tangential stress  $\sigma_{tg}$ , the neutron flux  $\varphi$ , and the irradiation time t:

$$\Delta d = K \cdot \sigma_{tg} \cdot \phi \cdot t$$

The design curve is specified such that it covers the measured points with adequate statistical certainty.

# 4.4 Permissible Control Rod Residence Times

### 4.4.1 Absorber / Clad Interaction

The possible residence time of a control rod is limited by the time it takes for the swelling induced cladding strain to reach the permissible equivalent strain.

For calculations, the fluence at gap closure is first determined using the design curves for absorber swelling and cladding creepdown and the fabrication data (e.g. filling gap). The permissible cladding strain is then calculated as follows:

After gap closure, the cladding tube is strained isotropically in the axial and tangential directions ( $\varepsilon_t = \varepsilon_a$ ) as a result of absorber swelling, and from the equation for the permissible equivalent strain

$$\varepsilon_{v} = \frac{2}{\sqrt{3}} \sqrt{\varepsilon_{t}^{2} + \varepsilon_{a}^{2} + \varepsilon_{t} \varepsilon_{a}}$$

one obtains  $\varepsilon_t = 0.5 \times \varepsilon_v$  for the permissible tangential cladding strain.

This is used to determine the permissible swelling expansion of the absorber and the associated permissible fluence.

Performing a conservative analysis (absorber swelling, clad creepdown and fabrication data) one obtains, for example, a permissible fluence (E > 0.6 eV) of approx. 5 x 10<sup>22</sup> cm<sup>-2</sup> for a control rod in a reactor loaded with 16x16 fuel assemblies.

For comparison, using best-estimate data results in a permissible fluence of approx. 8 x  $10^{22}$  cm<sup>-2</sup>.

The resultant permissible residence time or the permissible number of operating cycles cannot be fixed definitively. The reason is, that a specific fluence may be accumulated over different residence periods as a function of in-core position, operating mode and insertion depth of control rods, which is in turn determined by the bank allocation.

The following figures are therefore only reference values: Using a conservative calculation method the permissible residence time for a RCCA inserted in a control bank is approx. 14 years and in a shutdown bank approx. 22 years. In a best-estimate calculation the equivalent values are approx. 23 years and approx. 34 years, respectively.

# 4.4.2 Wear

Using the above-mentioned conservative assumptions, the maximum permissible cross section reduction due to wear is approx. 23 to 31 %, depending on control rod design (different mass of the control rods due to different control rod lengths). Here the stress category "membrane and bending stress" is limiting; the membrane stress still shows a large margin to the design limit.

A permissible residence time cannot be inferred directly. Knowing the permissible clad thinning, it is, however, possible to ensure reliable RCCA insertion by continuous RCCA monitoring in the form of eddy current testing/profilometry and/or visual inspection.

In case of sharp wear marks, a reduction in cross section below the design limit may already result in exposure of the absorber material. Continued operation of a control rod with exposed absorber is in principle permissible since the corrosion resistance of AgInCd is very satisfactory (tenacious protective oxide layers form in the area of the exposed absorber which means that annual local wall thinning of the exposed absorber is negligible).

# 5. Operational Experience

### 5.1 Overview

The operational experience with different types of RCCAs is described in detail in the literature, e. g. [9, 10, 11, 12]. As reported in [12], the maximum residence time of a RCCA to date was 21 cycles and the maximum cumulative fluence (E > 0.6 eV) has reached a value of 6.3 x 10<sup>22</sup> cm<sup>-2</sup>. The mechanical integrity of the spiders and the screwed connections to the control rods has been maintained in all cases.

In 1978 some evidence of accelerated rod wear on RCCAs was observed in a reactor in its seventh cycle [12]. This reactor is one of the four older Siemens plants that has experienced appreciable control rod wear. The same occurred in some plants of other vendors [1, 9, 10, 13]. The wear was discovered during routine visual inspections as part of a refueling outage. The Siemens RCCAs showing wear were identified and partly replaced with improved wear-resistant Siemens RCCAs (see Section 6) on which no further wear has since been observed.



Figure 6 Possible fretting wear on a fully withdrawn RCCA

The detected wall thinning is located on the control rods at the position of the tubes or plates of the guide assemblies and in some cases also at the top end of the fuel assembly guide thimbles. Figure 6 shows a schematic of these positions. The different possible shapes of wear marks on the control rods as shown in Section 4.1, were, in fact, observed during inspection, as demonstrated in Section 5.3. Similar shapes are reported in [9].

# 5.2 Measurement of Wall Thinning Using a Fixed Reference System

Behaviour with respect to crack formation as a result of absorber swelling and wear due to control rod movement and vibration is monitored as part of regular inspection using special examination techniques (eddy current testing, profilometry). Eddy current testing and profilometry are described in greater detail in [14, 15], an overview is given in [16]. An outline of a new development for improved measurement of wall thinning is offered in [12]:

The dual-coil system used up to now (cf. Figure 7) is replaced by a new improved fixed reference system. The advantage of this new method is that axial cross section



Figure 7 Eddy current testing of RCCAs

changes, which are relatively smooth and which cannot be detected with the dual-coil system or only with difficulty, can be reliably determined. This method has already been successfully introduced.

As before, the time required for measurement is very short; it takes only a few minutes to test one RCCA. This means that such inspections can be performed during routine inspection periods as before.

### 5.3 Wear Behaviour

Figure 8 shows the inspection results for control assembly wear of one plant with 16x16 fuel assemblies as an example. The results are typical for this type of plant.

Figure 9 shows the result of a profilometry of a wear mark found during an eddy current examination. The agreement with the shape of the mark shown in the top part of Figure 2 is readily apparent. The measured wall thinning at this position is approx. 19 % of the cladding cross section.

As stated in [12], the cumulative experience gained in all control rod inspections reveals a statistical average cross section reduction of approx. 0.5 % per cycle for 16x16 plants. The values attained for 15x15 plants are in part considerably higher.

### 5.4 Measurements of Absorber / Clad Interaction

Dimensional changes of the control rod tip caused by cladding creep down and absorber swelling, are monitored by diameter measurements. Such diameter measurements have been made at first sporadically in several plants in order to ensure that the control rods behave as expected. The scope of measuring was extended when the neutron fluence accumulated by the leading control rods was approaching the conservative design limit.

Typical examples of diameter traces obtained are shown in Figures 10 to 13: Free cladding creep—down without any firm contact between clad and absorber is assumed if the diameter decrease towards the control rod tip is following the neutron fluence profile, as shown in Figure 10. Beginning mechanical interaction between clad and absorber causes slight deviation from that diameter profile near the control rod tip, see Figure 11. In the next stage, a small hump is formed as a clear indication of interaction as in Figure 12. Further swelling of the absorber causes the hump to increase close to or even above the as fabricated diameter, see Figure 13.

Because actual data of the as-fabricated cladding diameter of those control rods are not available, evaluation of the diameter change was related to the average diameter at axial positions above 300 mm from the absorber tip where cladding creep-down is negligible because of low neutron fluence.

It is remarkable that all the different stages of absorber / clad-interaction, as shown in Figure 10 to 13, occur at the same point in time in one RCCA. To understand why individual control rods behave differently at the same fluence level, it should be considered that cladding creep-down as well as absorber swelling are rather slow processes and,



Wear (Reduction of Cladding Cross-Section)





Figure 9 Profilometry of control rods



Figure 10 Diameter measurements on control rods: no absorber / clad contact



Figure 11 Diameter measurements on control rods: beginning absorber / clad interaction



Figure 12 Diameter measurements on control rods: significant absorber / clad interaction



Figure 13 Diameter measurements on control rods: restrain of the clad above as fabricated diameter



Figure 14 Diameter changes of on control rods

hence, the dimensional changes are small and well within the range of manufacturing tolerances of cladding tubes and absorber rods. Most likely, differences in the as fabricated gap between absorber and cladding within the tolerance range have caused the different dimensional behaviour observed.

The measurements performed in several plants up to now provide a very consistent picture, as shown in Figure 14. Here, the diameter changes measured near the control rod tips are plotted vs. neutron fluence. Additionally, best estimate curves of cladding creep–down and absorber swelling based on nominal as fabricated dimensions are shown. In agreement with theory, the measured data indicate start of clad–absorber interaction at a fluence of about  $3 \times 10^{22}$  cm<sup>-2</sup> (E > 0.6 MeV). The rate of cladding restrain, determined from repeatedly measured control rods, is in good agreement with the expected AgInCd swelling rate for which, in all cases, the measured value is smaller than the conservative value, derived from unrestrained swelling measurements.

Based on these measurements, the remaining life time of control rods can be predicted with great confidence and the control rods can thus be operated beyond the conservative design limit.

In one Siemens plant, deformation of control rod cladding in excess of the conservative design limit and formation of isolated, longitudinal cracks have been detected in a few control rod cladding tubes. Due to early examinations the problem was recognized in time, and meanwhile the control rods of that plant have been replaced by new ones after 21 years service without any complication for reactor operation.

# 6. Coating for Improvement of Wear Resistance

An effective way for improved wear resistance in plants showing accelerated wear is the coating of control rods [12]. Coating requirements, selection of chromium carbide  $(Cr_3C_2)$  as coating material and the qualification of the coating by detonation gun process for reactor application is described in [15, 17, 18]. Other coating materials are in use, e. g. nitrided claddings [9].

The operating experience confirms the positive results obtained in the selection and qualification of the  $Cr_3C_2$  coating. In no case wear indication has been detected up to now (insertion time up to 14 years, more than 264 accumulated cycles) on any of the coated control rods during routine eddy current testing or visual inspection [12].

# 7. Optimization of RCCA Insertion

The objective of optimizing the RCCA insertion strategy is to minimize the total number of RCCAs used during the specified life of a PWR. In addition, the need for optimization arises also from the problem of disposal of control rods [19].

In plants with more pronounced wear, coated RCCAs showing no wear are used for reloads. Their life is only limited by absorber / clad interaction. Here, care should be taken only to ensure that, sufficiently before the fluence limit is reached, the RCCAs are shuffled from a control bank to a shut down bank where only a minimum increase in fluence occurs.

The insertion of existing uncoated RCCAs has to be optimized such that each RCCA reaches its end of life almost simultaneously as a result of absorber / clad interaction and wear, i.e. it should not be necessary to replace a RCCA because of wear without having attained a considerable fluence.

The extent to which a RCCA is approaching the design limit as a result of absorber swelling and thus the end of its life can be easily quantified:

The cumulative fluence can be calculated as a function of

- insertion depth
- bank allocation (control or shut down bank)
- in-core position
- operating mode

for the area of the tip of the absorber since this is the most highly-stressed part of the control rod.

For all PWR cores supplied by Siemens, accumulation of neutron fluences of the individual RCCAs can be continuously followed up with the aid of a computer code.

Determination in advance of the expected wear per cycle as a function of

- RCCA position in core
- control rod position in RCCA
- insertion depth

seems to be difficult as operating experience has shown to date. The reason for this is that the excitation of vibrations is strongly dependent on the prevailing conditions, e.g. coolant flow, part and location tolerances. (Wall thinning was, for example, found in a single control rod in a RCCA, while all the other 19 control rods of the same assembly showed no noticeable wall thinning.) Furthermore, the boundary conditions change from cycle to cycle due to fuel assembly shuffling, control assembly operating mode, etc..

For this reason the wear of RCCAs has to be determined by an adequate number of measurements using the eddy current method; it is common practice to test 25 to 33 % of the RCCAs in the core per cycle.

RCCA insertion strategy can be optimized on the basis of the known fluence, the wear of an individual RCCA and a conservative assumption of the maximum increase in wear per cycle (determined on the basis of eddy current measurements).

# 8. Summary

By taking appropriate account of life-limiting material effects (cladding creep and absorber swelling) and of the results of regular inspections using advanced techniques (detection of possible wall thinning due to wear), it is possible to operate RCCAs reliably for the duration of the permissible residence time and to optimize RCCA insertion strategy.

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### BEHAVIOUR OF PWR TYPE B<sub>4</sub>C IRRADIATED AT HIGH CAPTURE RATE

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### Abstract

In the French 1300 MW PWRs,  $B_4C$  is used in the upper part of the control rods and therefore undergoes only a low capture rate.

An irradiation experiment on  $B_4C$  at high capture rate (average burn-up:  $32.10^{20}$  capt/cm<sup>3</sup>, max :  $115.10^{20}$  capt/cm<sup>3</sup>) has been performed in the OSIRIS reactor in order to test the performance of this material.

The different phenomena which have been studied are : swelling, cracking, helium and tritium release of  $B_4C$  pellets. The main conclusion which has been drawn from this study concerns the influence of a heterogeneous swelling in the outer rim of the  $B_4C$  pellets, due to the effect of self-shielding in the thermal neutron flux, leading to a mechanical interaction with the clad. The mechanical consequences of the  $B_4C$  swelling are clearly described by mean of a finite elements three-dimensional computation whose results are in good agreement with the experimental observations.

### 1. INTRODUCTION

Boron carbide is presently one of the most used absorbing materials for the control rods of the nuclear power plants. The main advantages of this material are : high neutron worth, high melting point (2450°C), very low radioactive waste easy to reprocess, low cost in case of natural  $B_4C$ . However it presents some drawbacks, the main one being the swelling due to the formation of helium atoms. For this reason, in the French 1300 MW PWRs,  $B_4C$  is only used in the upper part of the control rods where the neutron flux is low enough to minimize the swelling rate.

If we look ahead for a wider use of boron carbide in the control rods it is necessary to improve our knowledge concerning the phenomena which limit the life time of this absorber. For that purpose we present in this paper the results of an experiment of stainless steel clad  $B_4C$  pellets irradiated at high capture rate in the OSIRIS reactor. The average burn-up achieved on  $B_4C$  after 122 days of irradiation in the OSIRIS reactor was  $32.10^{20}$  capt/cm<sup>3</sup>.
### TABLE I : CHARACTERISTICS OF THE EXPERIMENTAL ABSORBERS

ABSORBER RODLET	1	2	3	
Absorbing material				
Nature	B <sub>4</sub> C	B <sub>4</sub> C	B <sub>4</sub> C	
Fabrication – powder	magnesiothermal	magnesiothermal	carbothermal process	
– pellets	pressureless sintering	pressureless sintering	pressureless sintering	
Boron content w %	74	77	77	
Carbon content w %	25	23	23	
<sup>10</sup> B at %	19,8 %	19,8 %	19,8 %	
Density % TD	71	71	71	
Pellet grain size µm	ellet grain size µm 5		15	
Pellet diameter mm	ellet diameter mm 7,50		7,46	
Cladding material				
Nature	S.S. 304 L	S.S. 304 L	S.S. 304 L	
Diameter mm	7.72 - 9.72	7.72 - 9.72	7.72 - 9.72	
Filling gas	helium	helium	helium	

## 2. EXPERIMENTAL PROCEDURE

## 2.1. The absorbing materials

Two different types of  $B_4C$  pellets have been stacked into stainless steel rodlets sealed tightly, and whose characteristics are given in Table I. The two types of  $B_4C$  pelllets differ in their production process of powder.

The rodlets 1 and 2 contain  $B_4C$  pellets sintered with a powder produced from the magnesiothermal process according to the following reaction :

$$2 \text{ B}_2\text{O}_3 + 6\text{Mg} + \text{C} \rightarrow \text{B}_4\text{C} + 6\text{MgO}$$

This exothermal reaction gives directly a fine grain size powder (average size about 1 micron) ready for sintering after chemical elimination of magnesia. The average grain size of the pellets is 5 microns.

The rodlet 3 contains a column of  $B_4C$  pellets sintered with a powder produced from the carbothermal process according to the following reaction :

$$2 \text{ B}_2\text{O}_3 + 7\text{C} \rightarrow \text{B}_4\text{C} + \text{CO}^\uparrow$$

This endothermal reaction gives  $B_4C$  aggregates which have to be crushed, milled and sieved in order to obtain a fine powder. The average grain size of the pellets fabricated from this process is 15 microns.

The cladding material of the three rodlets is the 304 L stainless steel

#### 2.2. Irradiation conditions

The rodlets have been loaded in a capsule designed for the irradiation of materials at controlled temperature in the core of the OSIRIS reactor. It consists of two stainless steel containers, placed one inside the other. The gap between these two volumes corresponds to a gas jet whose conductivity can be varied continuously during irradiation, by adjusting the helium and nitrogen content.

The heating of the absorber rodlets arises from two sources : the  ${}^{10}B$  (n, $\alpha$ )<sup>7</sup>Li reaction on one hand, the gamma radiation generated from the structures of the capsule on the other hand. The regulation of temperature is obtained by adjustment of the content of a gas gap as mentioned above. The temperatures of the rodlets have

ABSORBER RODLI	ET	1	2	3
Irradiation time	days	122	122	122
<u>Average burn-up in</u> 10 <sup>20</sup>	capt/cm <sup>3</sup>			
CALCULATION		45	50	50
• DOSIMETRY	(1)	39	42	42
MASS SPECTROMETRY	(2)	22	32	32
Températures	°C			
• B <sub>4</sub> C Pellets	(3)	430-510	485-540	480-540
• CLAD	(3)	320-350	340-380	340-380

#### TABLE II : IRRADIATION CONDITIONS OF THE EXPERIMENT

(1) From AI-Co dosimeter inserted in the capsule

(2) From mass spectrometry measurements of the <sup>10</sup>B/<sup>11</sup>B ratio after irradiation

(3) From thermocouples measurements of the clad external temperature during irradiation.

been determined from thermocouples fixed on the external surface of the clads. The irradiation characteristics given in Table II indicate a wide range of variation for the temperature of cladding (320-380°C) and  $B_4C$  (430-540°C).

The values of burn-up achieved after 122 irradiation days are presented in Table II. The predicted values from calculation ( $50.10^{20}$  capt/cm<sup>3</sup>) appear to be overestimated compared to the experimental values obtained either from Al-Co dosimeters inserted in the capsule ( $40.10^{20}$  capt/cm<sup>3</sup>) or by direct mass spectrometry measurements of boron 10 concentration in the B<sub>4</sub>C pellets ( $32.10^{20}$  capt/cm<sup>3</sup>) after irradiation. The results given by the latter method were used to quantify the phenomena observed after irradiation.

## 3. RESULTS

The post-irradiation results presented in this section have been analyzed in relation to the average values of burn-up measured directly on the B<sub>4</sub>C pellets according to the method described in section 2. A computation of the capture rate profile along the pellet radius shows a heterogeneous distribution (Fig. 1) which is due to an effect of self-shielding in the thermal neutron flux. This leads to a steep capture rate gradient in the outer rim of the pellet, such that the local capture rate at the surface is nearly 6 times higher than the average capture rate value of the pellet at the beginning of the irradiation. At the end of the irradiation, for an average burn-up of  $32.10^{20}$  capt/cm<sup>3</sup> in the whole pellet, the calculation shows a maximum local burn-up of  $117.10^{20}$  capt/cm<sup>3</sup> at the surface of the pellet.

Non destructive examinations have been performed in order to measure the dimension changes of the clads (direct measurements) and of the  $B_4C$  columns (from X-ray radiography). The results (Table III) indicate a low but significant diameter increase of the rodlets 2 (0,25 %) and 3 (0,35 %) along the  $B_4C$  column, whereas no variation is observed along the plenum. As for the  $B_4C$  column, no variation of length has been detected from the measurements done on the radiographs. These results reveal clearly a mechanical interaction between the absorber and the clad induced by the  $B_4C$  swelling in the the rodlets.

It is interesting to notice the heterogeneous nature of the swelling which occurs only in the radial direction corresponding to the high flux dip in the pellets. The determination of the  $B_4C$  swelling deduced from the cladding diameter measurements (free swelling + stress swelling) leads to identical results for the different rodlets, as follows :

linear swelling	:	$\frac{\Delta \phi}{\phi} = 3,9 \%$
volumetric swelling	:	$\frac{\Delta V}{V} = \frac{2\Delta \phi}{\phi} = 7,8 \%$
linear swelling rate	:	1.2.10 <sup>-3</sup> /10 <sup>20</sup> capt/cm <sup>3</sup>
volumetric swelling rate	:	2.4.10 <sup>-3</sup> /10 <sup>20</sup> capt/cm <sup>3</sup>

It appears clearly that the swelling of  $B_4C$ , for these operating conditions, is independent of the fabrication process (magnesiothermal and carbothermal processes leading respectively to fine and large grain size pellets).



FIGURE 1 : RADIAL PROFILE OF THE CAPTURE RATE IN THE  $\mathsf{B}_4\mathsf{C}$  PELLETS

TABLE III : DIMENSIONAL VARIATION OF THE $B_4$	C RODLETS AFTER IRRADIATION
--	-----------------------------

	CLA	VARIATION OF		
RODLET	Β <sub>4</sub> C μm	Column %	Plenum	COLUMN LENGTH
1	-	_	-	-
2	25	0,25	_	—
3	32	0,35		_



FIGURE 2 : ASPECT OF B<sub>4</sub>C PELLET IRRADIATED UP TO A BURN-UP OF 32.10<sup>20</sup>/cm<sup>3</sup>

Another result must be focussed on : the absence of clad deformation along the plenum of the rodlets reveals that the pressure of helium released during irradiation is sufficiently low to avoid a high level of tensile stresses into the cladding.

On the micrographs of the rodlets 2 and 3 (Fig. 2) an extensive damage can be seen specially in the outer rim of the  $B_4C$  pellets where the gradient of burn-up is very steep. The external annular zone of the pellets is characterized by the presence of some typical circular and concentric cracks. Near the surface of the pellet, in the highest burn-up zone, where  $B_4C$  interacts with the cladding, the material is severely damaged (dark zone), and specially the rodlet 3 containing the large grain size  $B_4C$  pellets (carbothermal process).

The Figure 3 shows a detail of this zone which reveals the disintegration of  $B_4C$ . Consequently it seems obvious from these results that the carbothermal  $B_4C$  with large grain size presents a lower mechanical resistance to cracking compared to the magnesiothermal  $B_4C$  with fine grain size.

The results mentioned above have been analyzed and modelled by means of the finite elements CASTEM 2000 code [1] taking into account the three-dimensional effects, in order to describe the mechanical behaviour of the absorber. The main results obtained with this computation concern the different events occuring during the irradiation : swelling, cracking of  $B_4C$  and absorber cladding/mechanical interaction.



20 µm

#### FIGURE 3 : MICROGRAPH OF RODLET 3 SHOWING THE DESINTEGRATED ASPECT OF B<sub>4</sub>C AT THE PELLET SURFACE IN CONTACT WITH THE CLAD



relative density (%)







FIGURE 5 : EVOLUTION OF THE MAXIMUM PRINCIPAL STRESS IN THE B4C PELLET DURING IRRADIATION

The main data used in the model are the common thermal and mechanical properties of  $B_4C$  and 304 L stainless steel. The volumetric swelling has been taken equal to  $2.10^{-3}/10^{20}$  capt/cm<sup>3</sup>, whereas the rupture stress of the 70 % theoretical density  $B_4C$  has been deduced from values collected in the literature [2-7] as shown on Figure 4. In these conditions the main events described by the model are the following :

step 1 – 9 <sup>th</sup> day :	A first circular crack occurs at position 0,85 R (Radius)
step 2 – 85 <sup>th</sup> day :	The external ring of the B <sub>4</sub> C pellet is in contact with the
	clad.
step 3 – 90 <sup>th</sup> day :	A second circular crack occurs at position 0,72 R.
step 4 - 122 <sup>nd</sup> day :	End of irradiation, the clad diameter increase reaches
	0.6 % due to the mechanical $B_{A}C$ /cladding interaction.

The evolution of the maximum principal stress in the  $B_4C$  pellet during irradiation is shown on Figure 5.



FIGURE 6 : COMPUTED DRAWING SHOWING THE ELASTIC DEFORMATION AND THE STRESSES CONCENTRATION OF THE IRRADIATED B<sub>4</sub>C PELLETJUST BEFORE FISSURATION.THE MAXIMUM OF THE STRESS CONCENTRATION IS LOCATED INSIDE THE DARK AREAS FROM WHICH THE FIRST CIRCULAR CRACK WILL BE INITIATED.

RODLET	1	2	3
Burn up 10 <sup>20</sup> capt/cm <sup>3</sup>	22	32	32
HE PRODUCED (CALCULATED) CM <sup>3</sup>	128	727	774
HE RELEASED (MESURED) CM <sup>3</sup>	17,8	182,5	89,6
%	14	25	12
HE RELEASED (CALCULATED) % (1)	18	22	22
<sup>3</sup> H RETENTION MCI/G (MEASURED)	_	22±0,7	25±0,7
<sup>3</sup> H PRODUCTION MCI/G (CALCULATED)	_	32±6	32±6

## TABLE IV : HELIUM AND TRITIUM PRODUCED AND RELEASED IN THE IRRADIATED $${\rm B}_4{\rm C}$$ RODLETS

<sup>(1)</sup> From Homan relation [8]

The comparison with experimental results shows that the calculated position of the circular cracks is compatible with the micrographic observations (cracks are located in the range 0,65-0,82 R). On Figure 6 we present a computed drawing showing the elastic deformation of the pellet just before step 1. The maximum of the stress concentration is located in the outer radial zone towards the ends of the pellet where the first circular crack will be initiated. Concerning the cladding deformation, we observe that the computed results are slightly higher than the experimental results (0,6 % compared to 0,35 %). This discrepancy may be due to an effect of "rearrangement" of the outer rim of  $B_4C$  when the interaction occurs with the clad, phenomenon which has not yet been taken into account with the calculation (incompressibility of  $B_4C$  is assumed).

The helium release in the  $B_4C$  rodlets has been collected after irradiation by clad puncture and analyzed by mass spectrometry. The results presented in Table IV show that the percentage of helium release varies in the range 12-25 %. The following relation proposed by F.D. HOMAN [8], to calculate the percentage of helium release :

He % = Exp (6,69 - 1,85 D) exp -Q/RT

- with D = Fraction of theoretical density
  - R = Gas constant : 1.98 cal mole $^{-1}K^{-1}$
  - Q = Activation energy : 3600 cal mole<sup>-1</sup>
  - T = Temperature of  $B_4C$  pellet in K.

leads to values in good agreement with the experimental results, excepted for rodlet 3, as shown in Table IV. For the latter, the difference is probably due to the fact that the Homan's model does not take the grain size of the  $B_4C$  into account. It is interesting to notice that for such a low density  $B_4C$  (70 % TD) the porosity is totally open and consequently 30 % of the  $B_4C$  column volume could be used to store the helium released.

Another point of interest is the tritium retained in the  $B_4C$  pellets after irradiation. For this purpose measurements of tritium concentration have been performed on  $B_4C$  fragments. The extraction of tritium is obtained by heating the samples at 1000°C, then the gaseous tritium is converted into tritiated water which is analyzed by scintillation spectrometry. The experimental results given in Table IV compared to calculated production of tritium, reveal that this isotope is mainly retained in the  $B_4C$ .

#### 4. CONCLUSION

An irradiation experiment on  $B_4C$  at high burn-up (average :  $32.10^{20}$  capt/cm<sup>3</sup>, max. on the outer rim of the pellet :  $117.10^{20}$  capt/cm<sup>3</sup>) has been achieved after 122 EFPD in the OSIRIS reactor, in order to test the performanc of this absorbing material in view of a wider use of boron carbide in the PWRs control rods. For this purpose, two types of boron carbide pellets, differing in their fabrication process (carbothermal and magnesiothermal processes) have been tested in this experiment. The main conclusions which have been drawn from this study are the following :

- The volumetric swelling rate which has been determined reaches  $2.4.10^{-3/10^{20}}$  capt/cm<sup>3</sup>, whatever the B<sub>4</sub>C fabrication process. This swelling is responsible for a mechanical interaction leading to a low but significant diameter increase (0,35 %) of the cladding (304 L).

- Due to an effect of self shielding in the thermal neutron flux, a gradient of swelling occurs leading to circular cracks close to the outer rim.

– The mechanical behaviour of the absorber has been described by mean of a finite elements three-dimensional computation whose results are in good agreement with the experimental observations.

– The helium and tritium produced during irradiation is mainly retained in the  $B_4C$  pellets.

Finally this set of results allows to predict the behaviour of the  $B_4C$  used in the upper part of the French control rods up to a burn-up of  $30.10^{20}$  capt/cm<sup>3</sup>.

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# **BORON AND HAFNIUM BASE ABSORBERS FOR ADVANCED PWR CONTROL RODS**

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#### Abstract

For the future generation of PWRs, improvements in efficiency, life time and safety of the control rods are considered. An increase of reactivity worth will be particularly requested in case of reactors highly loaded with MOX fuel assemblies or operating with a limited boron content in water.

A preliminary research has been undertaken to improve the design of the absorber material. We present calculations and out of pile experiment results revealing the advantage of using a combination of boron and hafnium base materials. These results show that two types of absorbers should be studied and developped : boron 10 enriched  $B_4C$  pellets/hafnium cladding and boron 10 enriched HfB<sub>2</sub> pellets/hafnium cladding. Due to the neutron efficiency of the hafnium, a significant lowering of the irradiation damage of the pellets is obtained. The different aspects of the expected irradiation behaviour of such control rods are discussed.

## 1. INTRODUCTION

The next PWR generation will have to face with higher requirements in the fields of costs and safety. This is particularly true for the control rods to be used for the regulation and the shut-down of the future plants. As a matter of fact, important evolutions of the design of those reactors are to be considered, such as : high MOX content (> 50 %) in the fuel assemblies, total or partial suppression of the diluted boron in water, better behaviour to LOCA. This leads to the choice of different or improved materials and designs. In this frame, a material such as Silver-Indium-Cadmium (AIC) should be eliminated, for the following reasons : low mechanical properties, inducing creep and premature absorbercladding interaction ; weak melting temperature ( $\approx 800^{\circ}$ C), which means that AIC is the first core material which melts in the case of LOCA.

In order to satisfy the new requirements of safety, life time, efficiency, of the control rods materials, we can either consider new concepts or new materials. On an efficiency and cost point of view, boron carbide is very interesting but it undergoes an important swelling and an extended fracturation [1]. A significant part of those damages come from important capture gradients throughout the pellets radius, due to the high thermal neutron absorption of <sup>10</sup>B, leading to self-shielding effects. This could be reduced by the use of other cladding materials, with a high neutron absorption cross-section. Hafnium is such a material and different studies are in progress, aiming first to the elaboration of hafnium tubes [2], [3] and second to the estimation of the efficiency of control rods consisting of a hafnium

cladding containing  $B_4C$  pellets, which is related in section 4. Other materials, such as europium or other rares earths are very efficient neutron absorbers, but they are much too expensive and lead to too difficult waste management to be extensively used. We shall then focuse here only on the compounds containing boron and/or hafnium.

In this paper, we also present an evaluation of a 'new' absorbing material, hafnium diboride : the thermo-mechanical, chemical and neutronic properties of this material make it very interesting, potentially able to be used with or in place of  $B_4C$ . In section 4, are also presented estimations of the efficiency of control rods consisting of  $HfB_2$  pellets stacked in steel or hafnium tubes.

## 2. ABSORBING MATERIALS TO BE CONSIDERED

The most used absorbing materials are presently  $B_4C$  and the ternary alloy Silver-Indium-Cadmium (AIC 80-15-5). The main reasons for the choice of  $B_4C$  are : high and adjustable efficiency (according to <sup>10</sup>B enrichment and density of  $B_4C$ , the <sup>10</sup>B content can range from less than 1.5  $10^{22}$  to 1.1  $10^{23}$  at/cm<sup>3</sup>) ; high melting temperature (2450 °C) ; good chemical inertness (although some data concerning a possible reaction with water during irradiation are missing) ; very low radioactive wastes (He and Li) and low cost of the natural  $B_4C$ . Then, although its thermo-mechanical properties are quite low, this material will certainly be of large use in the future.

In the case of AIC, the situation is quite different since this material was designed in the seventies only in order to simulate an other 'rare' and dear material, hafnium. Taking into account the low melting temperature of this material, it would certainly no longer be used in the next reactors generations. But since hafnium is nowadays, as a by-product of the zirconium industry, neither dear nor rare, it would be very favourable to reconsider it. This material could then be used first as rods, like the present AIC rods (or even as un-cladded rods) and second as cladding tubes ; the last possibility is as component of new materials, such as HfB<sub>2</sub>.

We have reported on table 1 the main neutronic properties of AIC, boron, hafnium and two derived compounds :  $B_4C$  and  $HfB_2$ , for thermal neutrons. Hafnium appears less efficient than AIC, but since the main part of the efficiency of AIC comes from <sup>113</sup>Cd, which is rapidly burnt, this is not really a disadvantage. Moreover, the neutron absorption reactions by hafnium mainly lead to the formation of hafnium isotopes, first resulting in a much slower efficiency decrease and second to an easier management after use. At last, the absorption cross section of hafnium is better for epithermal neutrons, the flux of which is expected to be higher in the case of MOX fuels.

However, two difficulties remain for an extensive use of hafnium as control rods material. First, it can absorb high quantities of hydrogen, this resulting in an important swelling (up to 15 volumic % : [4]); but it has been shown that an oxydation treatment of the surface can completely inhibit this phenomenon [5] : rods of pure hafnium have been used in the CAP French experimental reactor for years without any operating problem. Second, it has low mechanical properties (hardness, wear resistance : [6]) and it will be possible to use

Element	Atomic	Isotopic	Absorption cross section (thermal neutrons)		absorption	descer	descendants	
Compound	mass	abundance	microscopic	macrosc.	resonances		abs. cross	
1			(barn)	(cm <sup>-1</sup> )	(barn)		section	
							(barn)	
В	10.82		$755 \pm 2$	104	280			
	10	0.198	3813			$7_{Li}$	0.033	
						<sup>4</sup> He		
B <sub>4</sub> C				81				
Ag	107.88		63 ± 1	3.63	700			
1	107	0.5135	$31 \pm 2$		80.6	<sup>108</sup> Ag		
						<sup>108</sup> Cd		
	109	0.4865	87 ± 7		1870	<sup>110</sup> Ag		
						<sup>110</sup> Cd	0.2	
Cd	112.41		$2450 \pm 50$	118				
	113	0.1226	$20000 \pm 300$			114Cd	1.2	
In	114.82		196 ± 5	7.3	2700			
1	113	0.0423	58 ± 13		891	<sup>114</sup> In		
						<sup>114</sup> Sn		
	115	0.9577	$207 \pm 21$		2294	<sup>116</sup> In		
J	ļ	ļ				116Sn	0.006	
AIC	109.15		202	9.9	965		Í	
80-15-5								
Hf	175.58		$105 \pm 5$	4.81	1860			
	174	0.0018	$1500 \pm 1000$			<sup>175</sup> Hf		
						<sup>175</sup> Lu		
	176	0.0515	$15 \pm 15$			177Hf	380 ± 30	
	177	0.1839	$380 \pm 30$		1	1/8Hf	$75 \pm 10$	
	178	0.2708	$75 \pm 10$			1/9Hf	$65 \pm 15$	
	179	0.1378	$65 \pm 15$		15.7		$14 \pm 5$	
	180	0.3544	14 ± 3		15.7	181T-	21 2 1 1	
HfB		<u> </u>		55		1a	$\frac{21.3 \pm 1}{1}$	
		1		22	[			

Table 1 : nuclear properties of elements and compounds considered as neutron absorbers

hafnium as cladding material only as far as those properties be improved : the elaboration of a 'Hafnaloy', which would be improved in the same extent than zirconium into 'Zircaloy', is required.

## 3. AN ADVANCED ABSORBER : HfB2

We present here a brief review of the main properties of this compound. Hafnium diboride is a very refractory material, its melting temperature is about 3380 °C (table 2) [7]. It is a ceramic which has then a brittle mechanic behaviour. High density materials are then obtained by sintering or hot-pressing at high temperature (around 2000 °C), in conditions similar to B<sub>4</sub>C densification. The complete ternary Hf-B-C diagram is well known [7]. It shows that HfB<sub>2</sub> reacts neither with graphite nor with B<sub>4</sub>C : it will then be possible, first, to

	Hf	B <sub>2</sub>	B <sub>4</sub> C	
Temperature (°C)	20	1000	20	1000
Density d (g/cm <sup>3</sup> )	11.22		2.52	
Thermal conductivity $\lambda$ (W/m.K)	83	74	26	12
Specific heat Cp (J/g.K)	0.298	0.418	1.11	2.21
Dilatation coefficient $\alpha$ (10 <sup>-6</sup> /K)	5.8	7.9	4.5	4.5
Vickers micro hardness (GPa)	25-30		30-35	
Young modulus E (GPa)	505	462	420	400
Bending stress $\sigma_r$ (3 points) (MPa)	340	450	250	230
Poisson coefficient	0.12	0.13	0.18	
Figures of merit :				
$R1 = \sigma_r / \alpha E (K)$	116	123	132	128
$R2 = \sigma_r k/\alpha E (W/cm)$	96	91	34	15

Table 2 : properties of hafnium diboride  $HfB_2$ , as compared with  $B_4C$ 

Table 3 : Swelling and helium release during irradiation (GETR reactor, irradiation temperature ≈ 350-550°C) of borides (from [14] and [15])

Material	Density (%)	Burnup (% <sup>10</sup> B)	(nα)/cm <sup>3</sup> /10 <sup>20</sup>	Released He (%)	Sweling (vol. %)	Sweling rate (vol. %/10 <sup>20</sup> /cm <sup>3</sup> )
BN	97	99		84	14	
B (93 % <sup>10</sup> B)	100	10	120	89	38	0.32
natural B	100	31	81	2.2	32	0.40
TiB <sub>2</sub>	97	67	98	1.1	40	0.41
ZrB <sub>2</sub>	85	83	91	3.6	25	0.27
VB <sub>2</sub>	90	50	81	7.9	19	0.21
HfB <sub>2</sub>	83	87	101	0.3	15	0.15
B <sub>4</sub> C	100	37	81	11	36	0.44
DyB <sub>4</sub>	86	44	55	39	broken	
YB <sub>4</sub>	95	62	95	4.9	23	0.24
YB <sub>6</sub>	92	42	67	14	12	0.18
SmB <sub>6</sub>	86	44	64	18	15	0.23
EuB <sub>6</sub>	87	37	57	19	7	0.12
DyB <sub>6</sub>	87	43	63	82	broken	

obtain high density pellets by hot-pressing in graphite dies, as  $B_4C$ , and second, to elaborate  $HfB_2/B_4C$  composites materials [8]. Its main thermo-mechanical properties are reported in table 2 [9], [10]. It can be seen that its thermal conductivity is much better than  $B_4C$ 's one, leading to a higher figure of merit. An important difference, when compared with  $B_4C$ , is the presence of dislocation networks [11] confering some plasticity to the material. This in turn could allow a better accomodation, without breaking, of high density of helium atoms or other irradiation defects, which is a very interesting property in the perspective of nuclear applications.

Hafnium diboride is also characterised by a good chemical inertness. Its oxydation under air becomes notable above 700 °C, leading to the formation of a passivating film of  $B_2O_3$ . Addition of SiC highly improved its resistance to oxydation in air, up to 2200 °C [12]. In water (T < 350 °C, p < 169 bar), HfB<sub>2</sub> is the boride which has the best resistance to corrosion [13]. The reactions of HfB<sub>2</sub> with the cladding materials (steel, hafnium) are not known. It can however be deduced from the equilibrium phases diagrams that the reactions should occur at high temperatures (HfB<sub>2</sub>-Fe eutectic at 1265 °C, HfB<sub>2</sub>-Hf eutectic at 1800 °C).

The main data concerning the behaviour of  $HfB_2$  under neutron irradiation have been obtained in Vallecitos atomic laboratories in the sixties [14], [15] and to a lower extent in U.S.S.R. in the seventies [16], [17]. In Vallecitos, different high density borides pellets (table 3) have been irradiated in the 350-500 °C temperature range in the GETR high flux reactor. The results show that, even though all the materials are extensively cracked, the diborides and particularly  $HfB_2$ , have a relatively low swelling rate and second have the lowest helium release rate during subsequent annealings (figure 1). These conclusions, i.e. the good behaviour of diborides under irradiation, are confirmed by [16] and [17].



Figure 1 : Helium release in selected borides during post-irradiation annealings, from [14] First points of the curves : released helium during irradiation



The crack has been initiated during a biaxial bending test. It propagates slowly and it is deviated by the pre-cracked HfB<sub>2</sub> aggregates



Top : microstructure of a B<sub>4</sub>C/HfB<sub>2</sub> (20 volumic % HfB<sub>2</sub>, 400 μm aggregates) composite Bottom : strain/stress curve of B<sub>4</sub>C/HfB<sub>2</sub> composite (biaxial bending test)

All those results show very interesting potentialities for the diborides, in the perspective of the next generation of nuclear reactors. In this frame, we have elaborated  $B_4C/HfB_2$  reinforced boron rich composites, made of pre-cracked HfB<sub>2</sub> spherical aggregates dispersed in an homogeneous  $B_4C$  matrix. The preliminary results we have obtained show that those materials have significantly improved thermo-mechanical properties as compared with pure  $B_4C$  (subcritical crack growth, pseudo-plastic strain/stress curves) [8] (figure 2) : the thermal gradients for crack initiation by thermal shocks (> 600°C/cm) are twice higher than those required for breaking pure  $B_4C$  ( $\approx 350°C/cm$ ); moreover, the samples do not break for higher thermal gradients. Their toughness is around 60 % higher than  $B_4C$ 's one and evidence for a R-curve behaviour (i.e.  $K_{IC}$  increase with cracks extension) has been observed. Consequently, such materials should then have a much better resistance to swelling and cracking than pure  $B_4C$ .

## 4. NEUTRONIC ASSESSMENT OF NEW MATERIALS AND CONCEPTS

We present here the main results of a neutronic assessment of the efficiency of new control rods concepts and materials, compared to the stainless steel cladded AIC rod as a reference [18]. A calculation has been performed with APOLLO 1 program [19], on the basis of a 900 MWe PWR reactor core loaded with 100 % MOX fuel, without diluted boron in water. The following configurations have been considered :

Claddings (outer diameter : 9.73 mm) :	Stainless steel S.S. 304, thickness 1 mm Hafnium, thicknesses 0.5, 1, 1.5 mm		
Cladding/absorber gap :	0.03 mm		
Absorber rod or pellets :	AIC (rod) Hf (rod) $B_4C$ HfB <sub>2</sub> $\begin{cases} natural boron or 90 \% {}^{10}B \\ 90 \% density pellets \end{cases}$		

The efficiencies of the materials are reported in table 4. Obviously, the efficiency of AIC and hafnium are much lower than those of  $B_4C$  or  $HfB_2$  (here containing only natural boron) : AIC and pure hafnium could then not be used in shut-down rods but only in regulation rods. Hafnium can of course also be used as cladding material. The absorber of the shut-down rods must then contain a high density of  $^{10}B$ . On the other hand, we can see that  $HfB_2$  is nearly as efficient as  $B_4C$ , this because of self-shielding effects. The influence of the thickness of a hafnium cladding can be checked on table 5 : even though a thicker cladding leads to a better protection of  $B_4C$  (and then a lower cracking), the efficiency of the rod is a decreasing function of hafnium thickness. In the following, the hafnium thickness is always 1 mm. On table 6, we have reported the efficiencies of the different rods (90 %  $^{10}B$  enriched boron) and their evolution from loading up to a fuel burnup of 45 GWj/t. It can be deduced that the use of a hafnium cladding improves the efficiency of the rods, even though the efficiency of the hafnium cladded rods decreases faster than the stainless steel ones.

## Table 4 : Efficiency of the tested materials (in pcm : $part/10^5$ of core reactivity)

- 90 % density B<sub>4</sub>C and HfB<sub>2</sub>
- natural boron  $({}^{10}B/({}^{10}B+{}^{11}B) = 0.198)$
- no evolution of the composition of the absorbers

	AIC	Hf	HfB <sub>2</sub>	B <sub>4</sub> C
0 GWj/t	6543	6876	9650	9700
30 GWj/t	8101	8425	11712	11972

Table 5 :  $B_4C$  absorber (90 % density)/Hf cladding : influence of the thickness of the hafnium cladding (efficiency in pcm)

Cladding	S.S. 304		Hafnium		
Thickness (mm)	0.53	1	0.53	1	1.5
natural B <sub>4</sub> C	9683	8369	10639	10142	9665
enriched (90% <sup>10</sup> B) B <sub>4</sub> C	14381	12401	15070	13829	12661

Table 6 : Evolution of the efficiency of different control rods -90% density B<sub>4</sub>C and HfB<sub>2</sub>

- cladding thickness = 1 mm

-  ${}^{10}\text{B}$  enrichment = 90 %

fuel burnup (GWj/t)	0	10	20	30	45
$B_4C/S.S.$	12513	12444	12307	12175	12018
B <sub>4</sub> C/Hf	13957	13711	13372	13029	12534
HfB <sub>2</sub> /S.S.	11360	11319	11210	11112	11014
HfB <sub>2</sub> /Hf	12870	12651	12341	12028	11590

The main effect of a hafnium cladding can be seen on figure 3 where the efficiency of isovolume rings in the absorber is reported as a function of the cladding material. It appears clearly that the hafnium cladding leads to a much smoother absorption profile in the absorber (the same result is obtained for a  $HfB_2$  absorber). This is a very favourable result, since an important part of boron carbide damage during irradiation arises from the high absorption gradients throughout the pellets radius and the subsequent differential swelling.





Figure 3 : Efficiency of iso-volume rings in  $B_4C/S.S.$  and  $B_4C/Hafnium$  control rods ring 1 : center of the absorber pellet ; ring 5 : cladding

## 5. CONCLUSION

Significant improvement of efficiency, safety and life duration of the control rods to be designed for the future PWR nuclear reactors can be obtained. We have briefly reviewed the properties of new materials, hafnium and hafnium diboride and assessed the efficiency of control rods consisting of a boron-rich absorber ( $B_4C$  or  $HfB_2$ ) cladded with hafnium tubes. The main result is that, provided a significant improvement of the mechanical properties of hafnium ('Hafnaloy') is obtained, such an absorber rod would be very interesting :

- introducing high boron contents (<sup>10</sup>B enriched boron) allows the regulation and shut-down of cores highly loaded with MOX fuels and without diluted boron in water ;

- on a safety point of view, the melting and reactions temperatures of those materials are much higher than AIC ones and in the same range than fuel-cladding reactions temperatures;

- using a hafnium cladding leads to a smoother absorption profile in the central absorber  $(B_4C \text{ or } HfB_2)$  and then a lower differential swelling and subsequent cracking;

- new composites materials, such as heterogeneous  $B_4C/HfB_2$  would have a still better resistance to cracking.

Those materials and the derived absorber rods concepts present much interest. Different studies are in progress, such as the elaboration of hafnium claddings, the high temperature interactions of  $HfB_2$  with stainless steel and  $B_4C$  with hafnium, the characterisation of  $HfB_2$  microstructure, or helium implantations in  $HfB_2$ . The mechanical properties of hafnium should then be improved, in order to obtain a 'Hafnaloy' as different from the pure metal as Zircaloy is different from zirconium. At last, experimental irradiations should be performed, in order to test the behaviour of the different materials (especially  $HfB_2$ , for which very few data are available), according to the new rods concepts we have described.

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#### RCCA'S LIFE LIMITING PHENOMENA: CAUSES AND REMEDIES

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#### Abstract

The main objective of a rod cluster control assembly (RCCA) supplier is to provide RCCA's with the longest possible lifetime, but also to offer surveillance services in order to ensure that their aging has not reached a level critical for their structural integrity.

By the end of 1993, the FRAMATOME surveillance equipment will have performed 115 campaigns, on about 50 PWR's plants of ELECTRICITE DE FRANCE as well as in other countries.

Based on this experience RCCA life limiting phenomena were investigated and the findings used to devise remedies for an increase of their lifetime. Two types of causes potentially damage RCCA's with Ag-In-Cd or with hybrid Ag-In-Cd and  $B_4C$  absorbers :

- cladding wear at the continuous and discontinuous guideways or at the bottom end, which may lead to perforation or failure of the rodlet,

- absorber swelling with clad cracking at the lower part of the rodlet, potentially inducing some release of the absorber material into the reactor coolant system, and jamming of the swollen RCCA in the fuel assembly guide thimble dashpot.

These phenomena and their evolution rate with time, were analyzed from hot cell examinations, out-of-pile experiments and calculations. On this basis, taking into account the inspection frequency and measurement accuracy, RCCA management criteria can be defined, providing the utility with a guaranty of the safe operation of its RCCA's in the interval between two inspections.

Furthermore, FRAMATOME developped a new RCCA design characterized by :

- ionitriding treatment of the cladding outer surface,
- reduction in absorber diameter at the bottom,
- helium backfilling of the rodlet,
- the use of AISI 316L for the cladding.

Ionitriding is a process for diffusing nitrogen at the surface of heated workpieces. This causes a reduction of wear, not only on the treated claddings, but also on the pieces in contact with them. Helium filling reduces the Ag-In-Cd temperature and creep, thus delaying the time of contact between the absorber and the cladding; the absorber diameter reduction further increases this delaying effect. The use of AISI 316L improves the cladding resistance to creep, and delays the cracking of the rodlet bottom section.

More than 300 new design RCCA's have been progressively loaded in PWR's since 1988. Today the manufacturing capacity is over 600 RCCA's per year.

#### **1. INTRODUCTION**

As RCCA supplier, FRAMATOME's concern is to design a product which will fulfil its function as long as possible, and also to provide the plant utility with measurement equipment and criteria allowing him to take the right decision on the reloadability of its RCCA's. It is well-known that the phenomena affecting PWR RCCA's during their in-reactor stay time can be classified into two categories: those of mechanical origin (wear) and those of thermal and irradiation origin (swelling and cracking). This paper is an outline of FRAMATOME's knowledge to date, built up mainly in conjunction with ELECTRICITE DE FRANCE.

The first step is to analyze the phenomena and their consequences, then attention is given to the ways of interpreting them (R and D, tests, examinations, etc). A prominent role is played by experience feedback, and the FRAMATOME measurement stands are described, illustrating how the parameters under evaluation (diameters, thickness) match the apparatus used. The next step is to define the applicable criteria and to state how they are calculated in terms of wear or swelling. Then a few indications are given on the RCCA management strategies.

Lastly, the knowledge base built up from RCCA in-core loadings is used to develop wear and swelling remedies, which are described in detail.

#### 2. DESCRIPTION OF THE PHENOMENA AND THEIR CONSEQUENCES

### <u>2.1 Wear</u>

Wear has been found at the following rodlet locations, from bottom to top (figure 1): - Rodlet tips : extending from the bottom end plug to the cladding over about 50 mm and exhibiting a "crescent" azimuthal shape.

- Continuous guidance zone : stepping-induced friction produces wear traces axially spread over a fairly large span. Concurrently, pointwise wear at the discontinuous areas (inlet and outlet) of the zone was found on low-mobility RCCA's. These wear traces mainly arise from rodlet hard contact towards the RCCA center and therefore assume a "2V" shape.

- Guide cards : this wear affects RCCA's not used for reactor control, which therefore accumulate wear over a narrow span. Depending on whether the rodlet bears against the card mostly on the spider side or on the opposite side, these wear traces take on a "2V" or a "crescent" shape.

In order of increasing importance, the consequences of wear are :

- occurrence of under-card perforation and release of absorber into the reactor coolant system,

- possible fatigue fracture of the rodlet in the event of excess wear if the number of steps per RCCA is high.

This latter risk is very slight in the case of base-load operated units since the RCCA's move very little. Preventive measures must nevertheless be taken since a broken rodlet could prevent the RCCA from dropping.

#### 2.2 Swelling and cracking

These phenomena appear generally in the lower 30 centimeters of the RCCA rodlet. Typical swelling is up to .2 mm of diameter increase, and typical crack length is a few centimeters. A combination of thermomechanical and irradiation-induced phenomena are involved :

- Slump and creep of the Ag-In-Cd absorber under the effect of compression by the rodlet spring; the effect of slump is to bridge the gap between the absorber and the cladding. The higher the absorber temperature is, the greater is the slump.

- Irradiation-induced swelling: neutron flux induces changes of absorber phase and transmutation of Indium into Tin, Silver into Cadmium, and creates microstructural defects.



FIGURE 1

## TYPICAL RECORDED WEAR FOR A LOW MOBILITY RCCA

The two phenomena act together to close the gap between absorber and cladding. When contact has occurred, the slump-induced stresses are not sufficient to cause cladding distortion. Only irradiation swelling therefore continues until the cladding ultimate uniform strength is reached. A longitudinal crack then appears in the cladding, releasing the built-up stresses, and swelling continues.

The consequences of swelling are, in the case of rod drop, an increase of the overpressure in the guide thimble dashpot system whose integrity must be checked. Excessive swelling could even theoretically cause the RCCA to become jammed at the bottom of the guide thimble.

Since the crack is always longitudinal, as demonstrated by stress calculations and rodlet observation in hot cells, failure of the lower part of the rodlet can be ruled out. At the very most, release of Silver into the reactor coolant is to be expected. However, even in this case, the quantities of Ag 110 released are well below those resulting from a clad perforation in case of wear with absorber friction against a guide card.

#### 3. ANALYSIS HARDWARE AND METHODS

#### 3.1 On-site measurements

As already stated, extensive experience feedback (115 in-reactor measurement campaigns by end 93, up to 20 campaigns per year) is a key source of knowledge, especially since the large variability of the loadings and wear phenomena found are clear proof that in-reactor spot checks cannot give a true indication of the possible full range of the phenomena and, likewise, a test program too narrow in scope cannot thoroughly predict all the potential behaviour patterns.

The measurement stand is described in detail in Appendix. It is used to characterize damages as follows:

- Using the Eddy Current (EC) technique :

- Cladding percentage cross-sectional wear is determined to within 5 % over the entire cladding height in 2 mm steps. The height accuracy is 2 mm at the bottom and about 15 mm at the top.

- Swelling in the lower section, with or without cracking. In the former case, crack elevation and length are determined to within 2 mm.

- Using the Ultrasonic (UT) technique :

The cladding outside radius, worn or swollen, is measured every  $3^{\circ}$  (i.e 120 points per cross-section) in vertical steps of 3 to 10 mm, chosen according to the extent of the defect. The UT measurement accuracy is .03 mm.

In addition to the measurement stand described below, of which two models exist, FRAMATOME has developed a "fine measurement" stand, used to inspect nitrided RCCA's. The wear measurement is taken with a high-frequency EC pancake coil, over an area of about  $2 \text{ mm}^2$ , with a depth range of 0 - 15 µm. The coil moves spirally along the rodlet, in 2 mm steps. Its aim is to detect any wearing of the nitrided layer. Compared with the two production stands, which have an "industrial" application, this rotary coil is only used for "R and D" purposes.

#### **<u>3.2 MAGALY test facility</u>**

Knowledge of rodlet vibration-induced wear was yielded mainly by the MAGALY hydraulic loop, located at the FRAMATOME Technical Center of Le Creusot.

A view of the loop flow chart is shown in <u>figure 2</u>. The section comprises, from bottom to top :

- the top end of a 17 x 17 fuel assembly (last span to top nozzle);

- a water box specially designed to adjust flow rates Q1 (main flow through assembly), Q2 (bypass flow through the guide thimbles), Q3 (crossflow at the top of the assembly if necessary);

- a guide tube with a water box for outlet flow;

- an upper plenum for injecting or removing flow Q4 (upper head bypass flow);

- a complete control rod drive mechanism (CRDM) with its driveline.

The comprehensive instrumentation package features absorber rod position indicators, for six rods (at every card location and in the fuel top nozzle), impact detectors and force measurements at the guide cards, pressure sensors at different guide tube locations, Pitot tubes in RCCA rodlets, translation force measurement on rodlets, displacement measurement of the spider, and flow visualisation. One of the difficulties that had to be surmounted in MAGALY is the instrumentation. Optical displacement sensors with an accuracy of .3  $\mu$ m and force sensors over the 0. - 50. N range (accuracy .03 N) have been developed, with the constraint







of not disturbing the phenomena. Software programs have also been devised to control the experiments, perform data acquisition and deduce wear power "on line" with an accuracy of about  $5 \times 10^4$  Watt.

The main MAGALY tests have been the following :

- Tests for understanding the phenomenon, during which it was shown that the major source of rodlet excitation is the main flow exiting the guide tube (Qout); flow Q4 leaving the top end of the guide tube, plays also a role : above a given threshold, it develops an instability of the CRDM. This is responsible of higher upper card wear in cold upper head plant operation condition.

- Tests for optimizing the guide tube. They were of major interest since they allowed to design a new guide tube called "M1" that equips all FRAMATOME new plants like Guang Dong or the french N4.

- Rodlet stress measurement tests, in which some RCCA rodlets were instrumented at the upper guide card with strain gauges providing tensile, compressive and bending stress data. The results showed that stresses due to flow-induced vibrations were relatively slight compared to the stepping loads.

Regarding wear power, the MAGALY tests demonstrated the major importance of the hydraulic conditions and of the rodlet hard contact conditions in the continuous guideway or against the guide cards. From this standpoint, the choice of running the tests with a real guide tube proved the only one capable of faithfully representing reality, including all the hydraulic features (pressure drops, flow distribution, type of turbulence). On the other hand, the tests made in a more simplified configuration (less guide cards, different geometry) sometimes showed that they were less representative. This is due to the highly nonlinear character of the involved phenomena.

All the trends observed in MAGALY have been found on site, particularly the relative importances of the different guide cards for wear, and the classification of the rodlets into different categories according to their position on the spider vanes: for example in the 17 x 17 geometry, single rodlets on vanes wear most, while those referred to as "external on double vane" wear the least, and the "internal" rodlets exhibit behaviour between the two.

#### 3.3 Other test facilities

Some loops of the Commissariat à l' Energie Atomique (CEA) have also been used for RCCA tests. These include HERMES and SUPERBEC. These facilities have pressure and temperature conditions representative of full-scale PWR's and comprise 1 to 4 assemblies.

In order to qualify a mechanical model to compute the stepping-induced loads on each part of the RCCA rodlet, a small test facility in air named MEXCIC was built by FRAMATOME, in which accelerations and forces on different parts - springs, bottom and top plugs, cladding, absorber - can be measured in detail. From this and from the complete hydraulic loops the dynamic behaviour of the RCCA is known, and the mechanical model allows to perform sensitivity studies.

In addition to stepping studies, endurance tests have also been run (several millions of CRDM steps). One conclusion arising from these tests and from reactor experience feedback is that, contrary to expectations at the start of the load follow studies about fifteen years ago, it is not the axial motion which most wears the RCCA's, but rather flow-induced vibrations; as a result, the RCCA's which remain immobile and do not participate in core control are the most heavily worn. The following calculation, although simplified, is a clear illustration of this phenomenon.

It is known that wear is, to within a "wear" coefficient (see next paragraph), proportional to the work of the friction forces, which is in turn proportional to the friction distance. h bd follow, an RCCA performs in one cycle hundreds of thousands of 16 mm steps. The friction

distance is therefore a maximum of 1.6 km for 100 000 steps. Now, for flow-induced vibrations a rodlet rubs against the guide card with a typical frequency of about 5 Hertz and amplitudes on the order of  $\pm$  .25 mm, which represents 5 mm per second or 160 km per year! It is clear from comparing 1.6 km with 160 km that stepping motion is not necessarily the worst-case configuration for wear, even if the friction forces involved are not comparable.

The endurance tests also showed the good behaviour of the spider and of the rodlet-tovane connections under such extreme number of steps. Furthermore MEXCIC is being equipped with electric resistances in order to run endurance tests on absorbers in hot conditions.

#### 3.4 Knowledge of the wear regime

Knowledge of friction forces, and of the upstream hydraulic conditions which cause these forces must be supplemented by study of the wear regimes in special-purpose machines. The wear mechanism is a combination of two types of behaviour:

- impacts with a relatively high normal force and little sliding.
- friction, with smaller normal forces but longer contact times.

According to Archard's law, the wear coefficient is the ratio of the worn volume to the work of the wear forces, which is the product of the tangential forces by the sliding speeds. This simple law does not allow for the wear mechanism (impact or friction), nor the normal component of the friction force, and more detailed models have to be used.

Metallographic examination of worn parts has shown that the wear facies depends on the wear mechanism: in a dominant impact regime, the parts are cold worked over a large thickness (several tens of microns), many cracks occur under the cold-worked layer, and cladding ovalization may even be caused in some tests (which was never observed in reactor). In a dominant friction regime, the appearance of the parts differs, the cold-worked layer is thin ( $\sim 5$  microns) and wear forms by abrasion of small-sized chips. It has been demonstrated that these examinations, using scanning electron microscopy and metallographic cross-sections, provided accurate characterization of the wear regime.

The MAGALY tests have shown that the rodlet motion was rather of the sliding type, but with a few impacts. Examinations of parts worn in the reactor, both in RCCA's and bottom mounted instrumentation (BMI) tubes, have confirmed this trend; thus, after using a variety of wear machines in PWR conditions (FROTTEAU, VIBREAU), FRAMATOME has developed in Le Creusot a machine called AURORE which exactly simulates this wear mechanism.

#### 3.5 Wear calculations

Software programs for predicting vibratory wear have been developed and benchmarked to the experimental base. A key predictive parameter is obviously the hydraulic excitation source, but calculations have also shown very large dependency on geometrical conditions: initial rodlet bowing, hard contact forces, rod-guide card gaps, off-centering, etc. It is a feature of these "non-linear" wear phenomena that a very slight variation in a parameter can have major consequences on the calculated wear powers.

This explains why RCCA's which are apparently identical wear in very different ways and confirms the benefits of our extensive field measurement database. It also shows the limits of using software tools, but this difficulty in prediction must not prevent us from going ahead with the development effort!

#### 3.6 Analysis of swelling and cracking

The increase in diameter at the bottom of the Ag-In-Cd stack has two causes:

- creep slumping under the effect of axial stresses, accelerated by temperature. It has been evaluated on site by stack length measurement statistics yielded by the EC technique. Out of pile tests on hot creep machines have added more accurate further data. In addition to the temperature effect, the effect of manufacturing conditions was also investigated.

- Irradiation swelling: RCCA hot cell examinations have been conducted with ELECTRICITE DE FRANCE. Studies have also been undertaken on the Ag-In-Cd phases. Lastly, RCCA swelling and fluence were correlated by means of neutronic calculations.

In the same way as for wear, the swelling kinetics is difficult to predict since it depends on the RCCA utilization which may be very variable under load follow conditions.

Irradiation swelling of boron carbide  $(B_4C)$  will not be addressed here. In French reactors whose RCCA's contain  $B_4C$ , the RCCA design and the core control mode are such that irradiation of  $B_4C$  pellets is small enough not to limit RCCA use.

#### 4. RCCA MANAGEMENT CRITERIA

The RCCA management criteria include the mechanical integrity criteria values and the phenomena kinetics to be considered for future cycles. They are closely linked to the RCCA management strategy (reshuffling, shifting) and to the planned inspection frequency. They are also related to the inspection techniques and the results achieved.

#### 4.1 Wear criteria

It is of course necessary to prevent rodlet fracture and so mechanical studies have to be carried out. Furthermore, it may also be advisable to avoid contact of the absorber with water, and this requires other studies.

Getting criteria on absorber rod mechanical integrity calls for knowledge of the phenomena against which precautions must be taken, of the different loadings sustained by the cladding, and of the shapes and locations of axial and azimuthal wear. It is then possible, either on an analytical basis or by means of finite element models, to set threshold values above which rodlet mechanical integrity can no longer be guaranteed.

#### 4.1.1 Maximum allowable wear

Worn rodlet studies consist in finite element calculations of the worn area, with the minimum cladding cross section, applying the most penalizing loads. These are :

- for the RCCA stepping, axial force in traction and compression, combined with bending moments,

- for the hydraulic vibrations, bending moments,

- of course, the primary pressure - not considered if the rodlet is worn through.

The other loads - like RCCA drop during a scram - are bounded by the previous ones. All these loads are obtained from tests and calculations. The result of these studies is the maximum allowable wear the RCCA's must not exceed, in order to be reloadable for one or more cycles, depending on the specific utility operating conditions. This wear is expressed in terms of cladding cross-section or in local wear depth, since both kinds of values are available from the on-site measurement stands.

#### 4.1.2 Wear kinetics

The wear kinetics is wear rate expressed as the cladding depth worn in one operating cycle.

Mainly for the upper cards, kinetics has a tendency to slow down as the wear keeps growing up to the cladding perforation. This is due to the fact that at the beginning few rodlets are in contact with the card, and when a certain level of wear is reached more rodlets come into contact with the card, thus reducing the contact load per rodlet. This trend of wear slow down has actually been measured on a number of units.

However, when a new RCCA (non surface treated) replaces a rejected one, wear kinetics often increases significantly.

#### 4.1.3 Shifting

Axial shifting of the RCCA's is a good way to extend their life. Usable positions are for instance, between 222 and 231 steps for 900 MWe reactor or between 260 and 268 for 1300 MWe reactors. Lower positions would impact the power level of the unit. It is recommended to use an operating procedure that reduces the positioning uncertainty, by raising the RCCA's up to the uppermost groove of the drive shaft, on plant startup and after every scram.

#### 4.1.4 Rejection criteria

When maximum allowable wear and wear kinetics are known, it is possible, according to the control frequency and possible shiftings, to define rejection criteria. Compared to these criteria, measured rods can be rejected, reloaded for one or more additional operating cycles, or shifted.

#### 4.2 Swelling rejection criteria

#### 4.2.1 Interference with the guide thimble tube dashpot

To avoid jamming the RCCA in the assembly, the gap between the guide thimble tube dashpot and the swollen rodlet is calculated. The worst case situation is encountered under hot conditions, due to Zircaloy and Ag-In-Cd expansions. The criterion relates to the largest increase in diameter measured at the elevation where swelling is largest, at pool temperature.

#### 4.2.2 Overpressure in the dashpot

The second criterion relates to the maximum permissible overpressure at the bottom of the guide thimbles. This overpressure is obtained intentionally, by fuel assembly guide tube design, to decelerate RCCA drop. When swollen rodlets drop, the amount of water flowing between the guide thimble and the rodlet decreases and the overpressure increases; it is important at this stage to verify the mechanical strength of the guide thimbles.

The calculations are made with values bounding the rod drop velocities at the dashpot entry. These values are deduced from the RCCA drop recordings made during periodical tests, or from a computer code.

Sensitivity studies are made on crack elevation and length. It is shown that for a given swelling, the lower crack elevation is, the greater the overpressure is. As far as length is concerned, longer cracks also lead to greater overpressure. The overpressure also increases with the swelling (see <u>figure 3</u>).

The stresses induced in the thimbles, combined with those caused by the axial loads, are compared with the design criteria. The beneficial effect of irradiation, which rapidly increases Zircaloy yield strength, is also taken into account. The results of these calculations lead to maximum allowable swelling.







## SENSITIVITY STUDIES ON SWOLLEN RCCA DROP

#### 4.2.3 Swelling kinetics

It is seen that swelling, for the high values obtained when cracks occur, with hard absorber contact against the cladding, depends primarily on RCCA fluence. Neutronic studies are performed and the calculated fluences are correlated with the swelling measured on site.

All these studies are used to define the swelling increase upper bounds per cycle.

#### 4.2.4 Rejection criteria

Taking the maximum permissible diameter increase values and deducing the swelling kinetics times the number of cycles - say one or two - one can infer the swelling limit values. A distinction can be drawn between the RCCA's for immediate rejection, those reloadable for one additional operating cycle and those reloadable for at least two additional cycles.

## 5. DESIGN OF A NEW RCCA

Historically, the first objective of RCCA improvement was the resistance to wear. Then swelling and cracking were found and the remedies now operational to reduce these effects involve:

- reduction in diameter at the bottom of the absorber,
- Helium backfilling to limit absorber temperatures,
- the use of an AISI 316L alloy for the cladding.

#### 5.1 Resistance to wear

As mentioned above, analysis of the wear facies (surface appearance and metallographic examination) leads to the conclusion that the predominant phenomena involve sliding under elevated loads (with some oblique impacts) rather than direct high-energy shocks.

Depending on the wear mechanism, the fact of modifying the surface of one of the two wear partners (surface treatment) reduces the risks of micro-bonding and modification of the superficial elastic limit reduces fatigue wear and flaking. The overall contact energy is unaffected by this treatment and the wear of the untreated partner does not increase; on the contrary, it most often decreases.

#### 5.1.1 Choice of a surface treatment

FRAMATOME, in cooperation with the CEA, undertook a research and development program-aiming to reduce the wearing of stainless steels by replacing them with Inconel 718 or by applying surface treatments. 6 processes were evaluated. After characterization in the as-coated state, the coated samples underwent either 9-month autoclave testing at 330° C or about 3000-hour irradiation in a CEA experimental reactor. At the end of each of these phases, they were subjected to characterization and in-water sliding wear tests at room temperature.

The main results are tabulated below:

Cladding materials (SS 304 tube)	Remarks
Bulk AISI 304	As reference
Bulk Inconel 718	Little improvement of wear resistance
Coating : galvanic chromium plating	Good corrosion resistance Flaking under irradiation
Coating : Electroless Nickel	Tight adherence Good resistance Risk of rise in reactor coolant activity
Coating : chromium carbide by detonation gun (LC1C)	Tight adherence corrosion resistance satisfactory (continuous process tube by tube)
Ionitriding treatment	Good adherence Good autoclave corrosion resistance Treatment by batch (economic saving)
Plasma spraying of chromium carbide	Loose adherence under irradiation Mediocre corrosion performance
Coating : Titanium nitride (TiN) by physical vapor deposition	Poor corrosion performance Loose adherence

At the end of this development phase, FRAMATOME opted for ionitriding, the best tradeoff in terms of efficiency and in-service performance. Furthermore, the principle of charge-based treatment is a major asset. Loop tests and power reactor experiments were undertaken concurrently with additional tests for ensuring the feasibility of the process on industrial scale.

#### 5.1.2 Principle and application of the ionitriding process

Ionitriding is a process for surface nitrogen enrichment, which helps reduce fatigue and fretting wear and is performed by electrical discharge in a rarefied nitrogen-containing atmosphere. In this process, the effect of plasma is to excite the molecular nitrogen into active nitrogen, to depassivate the surface and to heat the workpiece by ion bombardment. For stainless steels, it is possible to carry out surface hardening by simple nitrogen diffusion, without obtaining a combination layer to avoid any excess embrittlement. Industrial expertise in the application of the stainless steel ionitriding process calls for strict control of the temperature and nitriding capacity parameters.

At this time, industrial production is performed in a furnace with a capacity of 200 to 250 tubes per charge. The treatment is applied to welded tubes, on the bottom end plug, whose top section is masked. This leads to enhanced wear resistance throughout the functional part of the rod including the zone close to the bullet head; also, the upper end plug can be welded on a Nitrogen-free area of the cladding. Temperature control is monitored during treatment by twenty thermocouples divided among instrumented tubes.

The control of the Nitrogen gas flowrate, pressure, plasma parameters and thermal cycle leads to tubes nitrided deep enough to be wear resistant but without degradation of the corrosion resistance. All the tubes are examined non-destructively by Eddy Current. On some tubes per charge, metallographic and electrochemical tests are used to ensure compliance with the acceptance criteria of the specifications.

#### 5.1.3 Wear resistance

Accelerated wear tests have been run in pressurized water ( $320^{\circ}$  C - 15 MPa) representing fretting and impact wear. The loop tests are :

FROTTEAU	longitudinal sliding without impact
VIBREAU	predominant unidirectional impacts
AURORE	circular sliding with impacts
A 11 .1 1	1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1. 1

All the loop tests - either sliding or impact - show a drastic improvement in the wear resistance of the cladding (factor >50). When a predominant sliding contact occurs, as is the case with the RCCA's, the improvement in the wear of the partner of the nitrided cladding is also significant (factor 3 to 30).

#### 5.2 Reduction of absorber diameter

The gap at the bottom of the rodlet has been widened by reducing the absorber diameter by a little more than 100  $\mu$ m to significantly delay the time of contact, without noticeably affecting the RCCA worth or the stress applied to the absorber in service. Figure 4 illustrates the effect of the absorber diameter reduction on the swelling kinetics. Three segments represent the absorber diameter variations:

- until contact with the cladding,
- until clad cracking,
- after clad cracking, until violation of the acceptance criteria.



FIGURE 4

#### ABSORBER / CLADDING GAP AND SWELLING EVOLUTION

In reality, the phenomenon is more complex and this variation is not fully linear since it is influenced by:

- the operating mode, which influences RCCA insertion and therefore the heatup and fluence seen by the materials,

- the spatial distribution of the absorber - cladding gap (the off-centering of the absorber in the cladding contributes to a drop in temperature),

- changes in the mechanical properties of the absorber materials arising from those in the chemical composition and its structure.

It can be seen that the reduction in absorber diameter does postpone hard contact with the cladding and provides a lifetime gain.

#### 5.3 Helium backfilling

By increasing the thermal conductivity in the absorber - cladding gap before contact, Helium backfilling reduces the temperature rise due to gamma heating in the absorber. For absorber rods with reduced diameters centered in the cladding in the nominal operating mode, the presence of Helium enables the absorber temperature to be reduced and thus largely offsets the theoretical temperature rise induced by widening the absorber - cladding gap.

The outcome is a decrease in differential thermal expansion and enhanced slumping and creep resistance, phenomena which are both thermally activated.

#### 5.4 AISI 316L steel cladding

Compared with AISI 304L steel used until now, AISI 316L is better suited to ionitriding, but also benefits from slightly enhanced mechanical properties, such as:

- slower decrease in ductility with irradiation down to the same minimum threshold as 304L steel for a minimum fluence on the order of 5 x  $10^{21}$  n/cm<sup>2</sup> (E > 1 MeV) instead of ~  $10^{21}$  n/cm<sup>2</sup>, thus delaying irradiation embrittlement of the cladding,

- smaller variation in mechanical properties with temperature in the 300 - 400 °C range (better stability),

- less irradiation deformation (irradiation creep and intrinsic swelling reduced), which promotes limiting of post-crack swelling.

#### 5.5 FRAMATOME in-reactor experience with new - design RCCA's

These RCCA design changes have been gradually incorporated into reactors on a demonstration, then on an experimental basis, and finally on an industrial scale.

During the demonstration phase (1988), new non-ionitrided RCCA's were placed in parallel core locations which were equivalent from the wear kinetics standpoint. In the 900 MWe reactor, the new RCCA in shutdown position had to be changed twice running due to overstepping of the undercard wear criterion. In cycles 3 and 4, this location was occupied by a nitrided RCCA and later observation revealed no damage. No perceptible wear indication was recorded for the RCCA's treated by ionitriding, either in 900 MWe reactors after 4 annual cycles or in 1300 MWe reactors after 3 extended cycles.

Since the choice was made by FRAMATOME to minimize the effects of wear by applying the ionitriding treatment, 66 nitrided RCCA's have undergone at least 2 cycles in reactor, more than 150 were delivered from 1992 to mid-1993 in France, Belgium and Sweden, and a thousand others are being manufactured.
### APPENDIX

#### **RCCA SURVEILLANCE EQUIPMENT DESCRIPTION**

The equipment consists in a station which is equipped with the necessary accessories for Eddy Current and Ultrasonic inspection. It is composed of :

- an inspection station,
- an Eddy Current equipment,
- a Ultrasonic Testing equipment,
- a visual examination equipment,
- a calibration RCCA,
- computers.

## A.1 Inspection station

The system basically consists of a structure set up in a power plant pool whose mass in air is about 150 kg. Its functions are :

- to guide the RCCA during inspection ; the RCCA itself is moved vertically inside the stand by the plant handling tool,

- to support the equipment used for examinations and measurements.

The station is composed of :

- a frame for installation either on the top of the spent fuel storage racks, or preferably in the fresh fuel elevator,

- a top nozzle designed to fit on to the RCCA handling tool in any of four 90° orientations; it also allows stand handling by the site tool,

- an examination area under the top nozzle into which the EC probe support is inserted,

- a positioning plate for locating UT equipment in any of 6 positions.

### A.2 Eddy Current equipment (figure A1)

Each RCCA rod is moved vertically in a measuring encircling coil connected to a reference coil on the inspection station, near by the measuring coil. The differential voltage obtained at the two coils outputs is acquired at the rate of one measure every 2 mm.

These EC voltages are compared to the calibration EC curves and the curves % Volumetric Cross Section Reduction (% VCSR) = f(elevation) are drawn and printed.

A threshold is set on the EC curves for selection of the zones to be inspected by UT. This EC threshold may vary depending on defect types, their location (elevation, RCCA type) and on RCCA design and history.

## A.3 Ultrasonic Testing equipment (figure A2)

Each RCCA rod zone, selected at the end of EC inspection, is moved vertically in the UT fixture equipped with the UT rotating probe. When the RCCA is at the elevation required for the defect characterization by the UT fixture, the RCCA vertical translation is stopped. A profile measurement is performed with the RCCA motionless. The rotating probe emits Ultrasonic pulses which are reflected by the external cladding surface. The time of flight measurements allow the characterization of the rod deformations (wear or swelling).

Each UT profile is acquired at the rate of one measure every 3°. A UT zone is a collection of about 20 profiles performed with a regular longitudinal pitch along the RCCA elevation (standard pitch is 3 mm). UT data about each profile measurement are processed with the cladding nominal outer diameter and inner diameter.





## EDDY CURRENT EQUIPMENT PRINCIPLE

Profile curves are drawn such as : deformation depth = f (angular position). For each UT inspection zone, the curve % Cross Section Reduction (% CSR) = f (elevation) and the more typical profiles are printed. UT results are not affected by cladding tube coating nor by rod internal design.

The Ultrasonic measuring head is mounted on the positioning plate of the station and, for 17x17 type RCCA's, can reach any of 6 different rod positions (5 for 15x15 type RCCA's).

The 18 other rods of the RCCA (15 for 15x15 type RCCA's) can be reached by rotating the handling tool to any of the 3 other positions. The transducer is rotated by an electric motor and its azimuthal position is determined by an encoder.

The signals from the Ultrasonic channel and encoder are processed by the computer for calibration and readout purposes.

## A.4 Visual examination equipment

A video camera is mounted at the bottom end of a pole placed near by the elevator to verify the RCCA identification before introducing the RCCA in the inspection station. A videoprinter provides a record of the RCCA identification.



## FIGURE A2

## ULTRASONIC TESTING EQUIPMENT PRINCIPLE

## A.5 Calibration RCCA

A short (about 1.5 m long) calibration RCCA is used at the beginning and at the end of each shift, for Eddy Current and Ultrasonic measurement unit calibration.

Each Eddy Current standard allows calibration of the voltages measured on each channel with volumetric cross section reductions (% VCSR).

Among the standard rods, one is equipped with a machined standard which is used for calibration of the Ultrasonic measurement channel.

## A.6 Computers

A first mini computer is used on the floor of the spent fuel pool inside the fuel building, for data acquisition and on line data processing.

A second mini computer is used in an office, outside the fuel building, for extra data processing and data processing final validation. This computer may also be used as a spare unit if the first mini computer breaks down.

A third computer is used for preparing the campaign preliminary report which is supplied to the plant operator before leaving the site.

## A.7 Procedure of inspection

The station is positioned either on the top of a spent fuel storage rack cell, or in the fresh fuel elevator. With the RCCA site handling tool, each RCCA is taken from storage, moved in front of the video camera, then inserted in the station. An up and down motion velocity of 0.5 m/mn on the entire length of the RCCA must be available on the RCCA site handling tool.

All rods of all RCCA's undergoing inspection are first thoroughly examined over their entire length by Eddy Currents. The 24 rods of each RCCA (17x17 type) are scanned 12 by 12 by moving the RCCA up and down twice with the site handling tool (a 90° rotation of this tool is necessary between the two up and down movements).

The 24 EC curves are drawn on line. These curves directly allow the selection of rod zones exhibiting discontinuities by means of thresholds on EC signal amplitudes. At the end of EC scanning, the selected zones are then ultrasonically examined one after the other for exact measurement of wear depth and swelling and for wear shape determination. The number of profiles made by UT depends on the RCCA condition. Computerized evaluation and disposition are performed on line before RCCA removal.

## COUNTERMEASURES FOR PWR CONTROL ROD DEGRADATION IN JAPAN

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#### Abstract

In Japan, the aging degradation of the RCCA (rod cluster control assembly), i.e., fretting wear of the cladding and cracking of the rod tip due to the absorber swelling, was first noticed in 1984. After that, a measuring instrument for the rod diameter have been developed and the onsite inspections have been carried out at almost all the PWR plants in Japan. In addition, a hot cell PIE was performed on selected rodlets of a lead RCCA to confirm the condition and the mechanism of the degradation. Based on these inspection results, replacement criteria and management quidelines for the RCCA have been established, which can be utilized for long-term management in all the PWR plants in Japan. On the other hand, an improved RCCA with hard Cr-plated cladding and thinned absorber material at the rod tip has been developed. On-site wear inspection results for the improved RCCAs show good wear resistance of Cr plating after several cycles of operation. This paper describes the experience and countermeasures for control rod degradation in Japan, divided into the following three parts : (1) Management strategy for the control rod in Japan, (2) Inspection technique for the control rod, and (3) Countermeasures for control rod degradation.

#### 1. INTRODUCTION

In Japan, the aging degradation of the RCCA (rod cluster control assembly) was first noticed in 1984 upon receipt of information on US PWR (pressurized water reactor) plants. The degradation had two modes, i.e., fretting wear of the cladding and cracking at the rod tip, and it seemed to be a potentially serious safety problem inherent in all the Westinghouse type PWRs. To preclude this problem, it was decided to confirm the extent of the degradation of the RCCA and to establish replacement criteria on our own.

A total of 833 RCCAs are in use at 20 PWR plants in Japan. Three types of RCCA, i.e.,  $14 \times 14$ ,  $15 \times 15$ , and  $17 \times 17$ , are used, and each RCCA contains 16, 20, and 24 control rods, respectively. The control rod contains the absorber material (silver-indium-cadmium alloy) sheathed with thin cladding tubes (type 304 stainless steel) as shown in Figure 1.

All the PWR plants in Japan are operated under base load, so that the RCCAs are kept almost fully withdrawn from the core during plant operation.



Figure 1 RCCA specifications.

			Number	Number	Inspection	Measuring	Max. wear	Max. dia.
<u>Plant</u>	Loop	Array	of cycles	of RCCA	<u>cycle</u>	<u>equipment</u>	depth	<u>increase</u>
KMN-1	2	14×14	11	29	7	СР	260µm	30*µm
	(10ft)							
KMN-2	2	$14 \times 14$	13	29	10	СР	420	110
QGN-1	"	4	15	4	10	"	490	110
QGN-2	4	4	10	4	7	"	450	50
SIN-1	4	4	14	"	9	4	460	90
SIN-2	4	"	9	4	5	4	350	60
HTN-1	"	4	4	4	-		-	-
HTN-2	4	4	3	4	-	-	-	-
MANI 2	2	15 - 15	12	6 (520	nala) 8	CP	280	_
KIVIN-5	3	12 X 12	15	10 (San	10 10	CF //	200	-
10781 4				40	10	<i>"</i>	470	50 "
KIN-T	"	"	14		10		480	50
KTN-Z	"	"	14	"	9	"	500	100
KTN-3	3	17 x 17	7	48	4	СР	380	30 *
				4	6	ECT	420	-
KTN-4	4	4	7	4	4	СР	370	20 *
				4	6	ECT	400	-
QSN-1	4	"	8	4	6	CP 460		50 *
QSN-2	4	4	7	4	5	4	410	30
	Λ	17~17	11	53	7	CP	410	40
KON-1	(1141) (1141)		••	55	,	Ç.	410	-0
		"	11	13 (Sam	nle) 6	FCT	220	_
KON-2		·		53	g g		110	- 50 *
GTN 2		1.	c	در <i>بر</i>	2	ECT	240	50
U 118-2	4		U	,	ר ב		200	-
KUNS	1.		7	4		-		-
	., .,	· ·	۲ ۲	., 11		-	-	-
KUN-4	7	~	1	~	-		-	-

Note · Upper head temperature is T-hot in all plants.

· CP means contact type profilometry.

· Mark \* means the maximum value for sampled RCCA rodlets.

#### 2. MANAGEMENT STRATEGY

On-site inspections using the measuring equipment for the outer profile of the control rods, hot cell examinations and out-of-pile tests have been carried out for establishing replacement criteria and management guidelines for the RCCA.

#### 2.1 INSPECTION EXPERIENCE

To confirm the extent of the control rod degradation at Japanese PWR plants, a measuring instrument for the rodlet profile with high accuracy has been developed and on-site inspections of each type of long-operated plants have been performed in sequence, using this instrument. Now RCCAs at almost all the PWR plants in Japan have been inspected at least once. In addition to the measurement, visual inspections using an underwater television camera or a fiber scope have been performed at several plants, and a hot cell PIE (post-irradiation examination) has been performed on segmented rodlet specimens from two lead RCCAs in a specific plant.

#### 2.2 WEAR EXPERIENCE

#### 2.2.1 ON-SITE INSPECTION

Fretting wear was observed at the axial location corresponding to the guide card and continuous guide (sheath and tube) of the RCCA guide tube. Wear was caused by the long-time interaction between the rodlet and the guide tube due to flow-induced vibrations of the control rod. The results of the on-site inspections are summarized in Table 1. The maximum wear depth was 500 µm, but no penetration of the cladding was observed.

#### 2.2.2 HOT CELL EXAMINATION

One specimen was taken from the rodlet with the maximum wear depth in the specific plant. The wear scar of the PIE specimen well corresponded to the shape of the guide card both circumferentially and axially, and had a good agreement with the profile obtained by the on-site inspection (see Figure 2). Its surface was smooth with no fine cracking.

#### 2.2.3 WEAR MECHANISM

Wear is considered to result from flow-induced vibratory contact between the RCCA rodlets and the guide card during plant operation. The envelope of the on-site data shows that the cladding penetration could occur after 7 to 8 years' operation (see Figure 3). The wear rate for  $17 \times 17$  plants except  $17 \times 17$  UHI (upper head injection) was higher than those for other plants because of the difference in the upper core internal design.

The wear rate in the spring plenum region was much lower than that in the absorber region.



Figure 2 Outer profile of wear scar.



Figure 3 Prediction curve for maximum wear depth.





Figure 5 SEM investigation of the fracture surface.

#### 2.3 CRACK EXPERIENCE

#### 2.3.1 ON-SITE INSPECTION

A diameter increase of the rodlet tip was observed, and the results of the on-site inspections are summarized in Table 1. The maximum diameter increase was 110 µm (1 % of the cladding diameter). The outer diameter was larger in the RCCA rodlets with higher neutron fluence (see Fig. 4). Using the fiber scope, an axial crack was observed at the rod tip in some rodlets with the highest neutron fluence and only with an outer diameter increase of more than 0.7 % (see Fig. 4).

#### 2.3.2 HOT CELL EXAMINATION

Three samples were taken from two rodlets, which were RCCA rods with the maximum and the minimum outer diameter increase and with the highest neutron fluence. An axial crack was observed in the rodlet sample with the maximum outer diameter increase. As shown in Figure 5, the fracture surface was fully intergranular and the crack was thought to have initiated at the inner surface of the cladding tube, because the crack was slightly longer at the inner surface of the cladding than at the outer surface. From the result of tensile tests of the cladding tube material, the uniform tensile strain was reduced to about 1.4 % at 320 °C due to neutron irradiation. Moreover, intergranular cracking was observed partially in the specimen used for the slow strain rate tensile test. (1,2)

The increase in outer diameter at the rodlet tip was caused by the swelling of the absorber (Ag-In-Cd alloy)(2) and the diameter of the absorber increased with an increase of the neutron fluence.



Figure 6 Axial variation of diameter change near the rodtip.

Fracture strain of the cylindrical pressure vessel





Proposed critical hoop strain for RCCA cladding



Figure 7 Critical hoop strain for RCCA cladding.

The axial profile of the absorber diameter at the rod tip obtained by the hot cell examination had a good agreement with the outer diameter increase obtained by the on-site inspection (see Figure 6).

#### 2.3.3 CRACK MECHANISM

From the above results, it was considered that the axial crack in the cladding tubes of the RCCA rodlets was caused mainly by an increase in hoop strain due to the absorber swelling and a decrease in elongation due to neutron irradiation.

The failure criterion that cracking caused by the hoop stress of a thin wall cylinder occurs when the hoop strain reaches half of the plastic strain of uniform elongation(3,4,5) is applicable to the RCCA rodlet(6) (see Figure 7). It is consistent with the results of the on-site inspections. The critical neutron fluence for axial crack initiation is estimated to be approximately  $0.8 \times 10^{22}$  n/cm<sup>2</sup> (E>1 MeV) or  $3 \times 10^{22}$  n/cm<sup>2</sup> (E>0.625 eV).

#### 2.4 MECHANICAL INTEGRITY

The performance of the RCCA, i.e., the ability of insertion and withdrawal and support of the absorber, is maintained even if the clad is worn through in the absorber region or an axial crack is caused at the rod tip. Based on out-of-pile tests and analysis, the influence of wear and crack on the integrity of the RCCA was evaluated as follows.

The mechanical integrity of the control rodlet against fretting wear is limited by fatigue due to stepping load. In the case of base load plant operation, the wear limit is evaluated to be 0.9 mm thinning after cladding penetration, i.e., about 44 % loss area of the cladding cross section for the absorber-supported region. On the other hand, 10 % remaining cladding thickness is evaluated to be the wear limit for the spring plenum region.

An axial crack in the rodlet tip does not affect the mechanical integrity of the control rodlet.

#### 2.5 REPLACEMENT CRITERIA

Based on the above investigation, conservative replacement criteria have been determined. The performance of the RCCA is maintained even if cladding is worn through in the absorber region or an axial crack is caused at the rodlet tip mentioned before. In Japan, however, the RCCA is replaced before the cladding is worn through or a crack is initiated from the standpoint of preventive measures.

#### 2.6 MANAGEMENT GUIDELINE

The operating management of the RCCA consists of the repositioning of the fully withdrawn RCCA to move the point of interaction between the control rodlets and the guide tube cards and the replacement of degraded RCCAs. A total of 363 RCCAs have been replaced by enhanced performance type.

#### 2.6.1 WEAR

Based on the wear prediction curve obtained from on-site data, the RCCA is repositioned from 228 steps to 225 steps before the wear reaches 100 % of the clad thickness', then the RCCA is replaced before the same time passes without inspection. The operating time when the wear prediction curve reaches 100 % of the clad thickness is approximately 8 years for  $14 \times 14$ ,  $15 \times 15$ ,  $17 \times 17$  UHI and approximately 7 years for  $17 \times 17$  (see Figure 3).

Wear in the spring plenum region is not critical in spite of the lower replacement criteria because the wear rate is very low.

Actually, more simplified replacement according to the scheme determined in advance (e.g., 10 RCCAs per cycle) is also adopted in some plants.

#### 2.6.2 CRACK

According to the prediction curve for diameter increase obtained from on-site inspection and hot cell PIE (see Figure 4), the RCCA is replaced when its total amount of neutron fluence at the tip reaches the critical neutron fluence for axial crack initiation, i.e., approximately  $0.8 \times 10^{22}$  n/cm<sup>2</sup> (E>1 MeV) or  $3 \times 10^{22}$  n/cm<sup>2</sup> (E>0.625 eV).

#### 3. INSPECTION TECHNIQUE

Two types of measurement equipment have been developed for on-site nondestructive inspection (see Figure 8).

#### 3.1 CONTACT TYPE PROFILOMETRY

Using the contact type profilometry system, the outer profiles of the control rods can be measured by cantilever type springs on which strain gages are attached. In one cycle of measurement this equipment can measure circumferentially 180 points per one cross section, axially four cross sections on each rodlet, and simultaneously all rodlets. Using this equipment, the wear depth and outer diameter of the rodlet can be measured with an accuracy of  $\pm 30 \ \mu m$  (3  $\sigma$ ). The outer profile of the rodlet cross section and axial distribution of the rodlet diameter obtained by this equipment had a good agreement with those obtained by the hot cell examination (see Figures 2 and 6, respectively). A total of approximately 13,000 RCCA rodlets at 15 plants have been measured.

#### 3.2 EDDY CURRENT TESTING

Using the ECT (eddy current testing) system, the wear depth and loss area of the cross section of the cladding tube in contact with the guide card can be measured by pancake type sensors and an encircling type sensor, respectively. To obtain high accuracy, circumferentially 10 pancake type sensors per rodlet should be set and axially 1 mm pitch data can be measured on each rodlet. Besides, the existence of cracking can be confirmed by this equipment.

Using this equipment, wear depth can be measured more economically and quickly with an accuracy of  $\pm 60 \ \mu m$  (3  $\sigma$ ) and loss area of the cross section of the cladding tube with an accuracy of  $\pm 3$  % for the absorber region and  $\pm 4$  % for the plenum region. A total of approximately 5,000 RCCA rodlets at 4 plants have been measured.



## Figure 8 Measuring equipment.

#### 4. COUNTERMEASURES FOR DEGRADATION

To preclude the potential safety problem induced from the cladding wear and cracking, the RCCA is managed according to the guideline mentioned before. In addition, improved RCCAs have already been developed and increasingly used for replacement.

#### 4.1 DESIGN OF IMPROVED RCCA

Hard chrome-plated cladding is applied for improving wear resistance. The long-term wear test shows good wear resistance of Cr plating. For enhancing crack resistance, the outer diameter of the absorber is reduced at the rodlet tip. Enlarging the gap between the cladding tube and the absorber delays the time when the absorber comes into contact with the cladding by the absorber swelling due to neutron irradiation and results in the prevention of the cracking. These improvements reduce the cladding wear and the possibility of cladding cracks, and extend the operational service life.

The improved RCCA rodlet is shown in Figure 9.

			Number	Improv	ed RCCA	Number of	
Plant	Loop	Array	of RCCAs	Number	Max. cycle	Inspected RCCAs	
KMN-1	2 (10ft)	14×14	29	12	3		
KMN-2	2	4	11	5	3		
QGN-1	"	"	11	29	5		
QGN-2	"	4	4	15	4		
SIN-1	4	4	11	26	5		
SIN-2	"	4	4	29	3		
HTN-1	11	4	11	29	4		
HTN-2	4	4	4	29	3		
KMN-3	з	15 x 15	48	34	Δ	з	
KTN-1	"	· · · · · · · · · · · · · · · · · · ·	40	31	5	5	
KTN-2	4	11	"	23	5		
KTN-3	3	17 x 17	48	24	2	12 *	
KTN-4	4	4	4	19	2	7*	
QSN-1	4	4	11	26	2		
QSN-2	4	4	4	20	2		
KON-1	4	17 x 17	53	22	5		
KON-2	11	"	4	30	4	3	
GTN-2	"	11	4	18	3	12 *	
KON-3	4	11	4	53	2		
KON-4	4	4	4	53	1		
Total				527	(Max. 5)	37	

## Table 2 Application of Improved RCCA

\* Measured by ECT





Improved rodlet

## Figure 9 Improved RCCA rodlet.

#### 4.2 PERFORMANCE OF IMPROVED RCCA

More than 500 improved RCCAs are now in use at almost all the PWR plants in Japan with a maximum of 5-cycle operations. In all 37 improved RCCAs at 5 plants have been inspected by the measuring equipment after one- or two-cycle operations and showed good wear resistance. Non-detectable wear was measured in all the measured RCCAs (see Figure 3).

#### 4.3 REMEDIAL ACTIONS

From the viewpoint of preventive measures, a follow-up program for the integrity of the conventional RCCA and the performance of the improved RCCA by periodical on-site inspections using a more economical and quick ECT system is in progress. In addition, a hot cell examination of improved lead RCCA rodlet samples is also planned to confirm the effectiveness of the crack resistance.

On the other hand, another coating material for improving wear resistance is under investigation for further service life extension.

#### 5. SUMMARY

- Operating management guidelines for the RCCA degradation have been established based on the on-site and the hot cell examination. The RCCA is managed very conservatively from the viewpoint of preventive measures.
  - Wear : Management of plant operation time Replacement before cladding penetration
  - Crack : Management of neutron fluence Replacement before crack initiation
- The following two types of measuring equipment have been developed.
  - Contact type profilometry system for measuring the outer profiles with high accuracy
  - ECT system for quick and economic inspection of the cladding wear
- 3) The improved RCCA with hard Cr-plated cladding and reduced absorber diameter at the rod tip has been developed and been monitored. Improved RCCAs have been verified to have good wear resistance from the result of on-site inspection after 1- or 2cycle irradiation.

#### 6. ACKNOWLEDGMENTS

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## A SIPPING TECHNIQUE TO IDENTIFY LEAKING RODS IN RCCAs

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## Abstract

This paper describes an original method developed by CEA and EDF in order to identify leaking rods in RCCAs with silver-based metallic alloy as neutron absorber material. The process consists of an etching of the absorber surface by means of a slightly aggressive solution. This involves the dissolution of a small quantity of silver in the solution. After the test, solution is recovered and sampled, and silver is dosed. A sipping prototype has been assembled, and three on-site demonstration campaigns, with one during the planned shutdown period of the plant, have proved the good reliability of the method and the industrial feasability of this new technique.

## 1. INTRODUCTION

In most PWR reactors, Rod Cluster Control Assemblies (RCCAs) are made up of rods with silver-based metallic alloy as neutron absorber material. After a few years of utilisation, periodic on-site examinations have shown that RCCAs can be affected by damages such as :

- cladding fretting wears, at location of the continuous guideway or guide plates,

- absorber swelling with cladding cracking at the bottom of the rod.

If the fretting wear still goes on, it is possible to obtain a perforation of the cladding and an opened surface of absorber material in contact with primary coolant. Silver alloy itself can be weared out and released into the primary circuit. This release can involve important consequences on dose rates for workers, due essentially to the long half-life of activation isotope Ag 110m (250 days) and its numerous gamma rays at high energy.

In order to prevent such an event, CEA and EDF have developed an original method for detection of leaking control rods in a RCCA, based on a sipping technique, and a sipping prototype has been assembled.



Figure 1 : RCCA's Sipping Test · process



Figure 2 : RCCA's Sipping Prototype : Global design

## 2. DESCRIPTION OF THE PROCESS (See figure 1) :

The RCCA is enclosed in a sipping cell and each control rod is placed in contact with a slightly aggressive chemical liquid solution. Stainless steel constituting the clad is inert towards the product. When a leaking rod is present, an etching of the absorber surface allows a small quantity of alloy components to dissolve in the solution.

After the test, solution is recovered in a storage tank and sampled. Detection of components of the alloy by means of a physical or chemical method allows to detect the existence of a leak on the tested control rods. This result can be completed by a gamma spectrometry on the sample, in order to obtain informations about the location of the defect.

With the aim to have short test times, chemical reaction is enhanced by increasing solution temperature.

This process is patented in several countries.

## 3. DESCRIPTION OF THE RCCA'S SIPPING PROTOTYPE (See figure 2) :

The sipping prototype consists of a mobile device installed in the fuel building of the plant. Three parts can be distinguished :

- \* sipping cells,
- \* connecting mast,
- \* command control cabinet.

## 3.1 Sipping cells :

The sipping cells are located in the spent fuel pool or cask loading pit (see figure 3). Two cells operate simultaneously and allow a test during the handling of an another RCCA.

A cell comprises 24 "guide tubes" (see figure 4), one for each rod, and a central tube for the incoming and outgoing of the solution. The central tube is connected to the guide tubes by means of a flow diffuser, located at cell bottom.

The gap between rods and guide tubes and the volume of the diffuser are minimum in order to minimize quantity of solution used. This allows a high sensitivity and minimize liquid wastes. Each test needs around 4 litres of solution.

At the upper part of the cell, a void volume contains the RCC spider body. It is used during testing as a gaseous plenum to drive off the pool water after cell closing, or to recover the solution in the storage tank after the test.

Heaters are disposed around a metallic body. This body transfers the heat and allows the guide tubes to have an uniform temperature.

Leaktightness towards pool water is ensured with an inflatable seal.



Figure 3 : Cells and connecting mast assembled in cask loading pit

## 3.2 Connecting mast and connecting lines :

Connecting mast consists of two removable sections vertically assembled in the pool, between the surface and the cells *(see figure 3)*. Each section includes inside connecting lines for :

- \* chemical solution,
- \* electrical power,
- \* air control and pressurization, and thermocouples wires.

At each section end, a connecting box is used for the assembly. The connecting mast is slightly pressurized with air to avoid water inlets.

The control command cabinet is connected by flexible lines to the first connecting box.



Figure 4 : RCCA's Sipping prototype : Sipping cell



Figure 5 : Command control cabinet on the operating floor

## 3.3 Command control cabinet (see figure 5) :

A command control cabinet on the operating floor is devoted to

- \* preparation, injection into cell and recovering of the solution,
- \* heating command and control,

\* monitor advancement of each sipping test sequence by means of a programmable controller,

- \* power and fluids alimentation,
- \* liquid and gaseous wastes orientation.

After recovering, the solution in the storage tank is sampled in a glove box.

A mimic panel and a control desk allow to operator to monitor the whole test development.

The prototype has been designed by CEA, Advanced Technologies Directorate, at the Nuclear Research Center of Grenoble.

## 4. DESCRIPTION OF A SIPPING TEST SEQUENCE :

Each elementary sequence of the test carries out itself automatically The passing from one sequence to the next is manual

## 4.1 Test preparation :

RCCA is placed in the sipping cell. After cap closing, the pool water is drived off by pressurization of the cell plenum. During this time, solution preparation is conducted in a special tank. When the cell is empty, solution is transferred into the central tube. The diffuser transmits an equal volume of solution in each guide tube. The quantity of it is adjusted in order to bathe all the rod. Heating operates continuously, and consequently solution reaches rapidly the right temperature in the cell.

## 4.2 Sipping test :

The test itself consists of the etching of the surface absorber. A mixing of solution is carried out during all test, by means of a cyclic pressure change applied to the central tube : this tube is alternately empty or full of solution. This involves a solution level change in the guide tubes, and a displacement of the liquid in front of the control rod. Due to the slight gap between rod and guide tube, this displacement presents a large amplitude.

In case of defective control rod, this mixing avoids redeposition of dissolved metallic components near the leak or on the inner surface of the guide tubes, and allows to homogenize dissolved metallic elements in the guide tube.

## 4.3 Solution recovering and sampling :

After testing, a pressurization of the plenum cell flushes the solution through the central tube to the storage tank, where it is homogenized.

One or several samples are taken at different levels of the storage tank. Results already obtained show a very good homogenization of solution in the storage tank.

The rest of the solution is flushed to waste. There is no problem of compatibility with other plant wastes, due to their slight quantity and chemical activity.

Guide tubes, tested RCCA, lines and storage tank are flushed with demineralized water. The quantity needed is the same than the one of solution.

Before clap opening, the inflatable seal is depressurized, and cell filled with pool water.

## 4.4 Dosing of absorber alloy components :

Dosing of absorber alloy components is carried on Silver, the main element of the alloy. The used technique is Atomic Absorption Spectrophotometry in a separate laboratory.

Other techniques are possible, such as electrochemical dosing with electrodes, on the spot or in a laboratory.



Figure 6 : Results of the on-site demonstration campaign performed during the planned shutdown period

## 5. DEMONSTRATION CAMPAIGNS :

Two on-site demonstration campaigns with selected rejected RCCAs have been performed in June and October 1991 out of critical path. A good agreement has been found between these results and those of UT and EC tests. Several leaking RCCAs have been easily identified, and corresponded with the most weared RCCAs.

A third on-site demonstration campaign, during a planned shutdown period in January 1992, has proved the industrial feasability of this new technique. This campaign was performed in a short time : less than three days to check all RCCAs of a 1000 MWe reactor, without incidence on the outage critical path and with generation of a little quantity of liquid effluents.

Figure 6 shows results obtained during this third campaign (53 different RCCAs have been tested): Silver concentration in the storage tank is plotted versus the number of test. Background level observed on tight RCCAs is due to external deposition (contamination) of absorber alloy during the precedent cycle. The background values are low and constant in time, and no significant contamination of sipping prototype has been observed. Seven RCCAs are easily identified as containing leaking rods, and one classified doubtful.

## 6. CONCLUSION

An original sipping technique has been developped by CEA and EDF in order to detect leaking control rods in RCCAs with silver-based absorber alloy. Based on a chemical surface etching of the alloy, the process is easy to operate and gives unambiguous results. The industrial feasability of a complete control campaign during a short planned shutdown period of a power plant has been proved.

# CONTROL MEMBERS OF WWER-440 AND WWER-1000 POWER REACTORS

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## Abstract

The design and operating experience with WWER-440 and WWER-1000 power reactor control members and neutron absorbing materials is described in this paper. In addition, the radiation stability of the cladding steel of control rods and factors affecting service life and service life extension of control assemblies are discussed. Investigations of hafnium, dysprosium titanate and EP-630U alloy are presented as well as the properties of a number of neutron absorbing materials fabricated in different forms. The results of these investigations support the current utilisation of the materials used in WWER power reactors for both control and shut down.

## 1. The present report outlines:

- the design of WWER-440 power reactor control members;
- radiation stability performance of SBJ-2 boron steel;
- materials and design of absorbing components of WWER-1000 control members;
- substantiation of control members' service life and service life extension.

# 2. WWER-440 automatic control assemblies (ACA) absorbing adaptors and SBJ-2 boron steel radiation stability

2.1. 37 dual-function ACAs are utilized in the power control and scram systems of WWER-440 power reactors at NPPs operated in Russia, Ukraine, Finland, Hungary, Bulgaria, the Czech Republic and the Slovak Republic. Fig. 1 shows the design of such an ACA.

The hexagons are produced by precision casting with consecutive machining of the end faces only. The machined product is polished electrochemically.

2.2. Table 1 shows the results of investigation of the hexagon itself and the specimens (L = 50 mm, cross-section 6x6 mm) cut out from the top and bottom end faces of the hexagon.

Based upon the results of the investigation and some other experiments (after irradiation of the specimens in the SM-2 research reactor) the maximum permissible value of the fluence of thermal neutrons affecting the SBJ-2 steel at the bottom of the hexagon was determined to be  $3.5 \times 10^{21} \text{ n/cm}^2$ .



Figure 1

#### Table 1

The fundamental properties of Boron steel SBJ-2 before and after irradiation (absorbing part of ARK; WWER-2;  $\Phi$  maximum - 3,19x10<sup>21</sup> n/cm<sup>2</sup> (thermal); NPP "Rheinsberg")

2. Decrease in density [%]: +  $\Delta V$  [%] = 1,4 - 1,7% (or swelling)

3. Increase in separation of opposite sides of hexagon  $\Delta S$ : = 0,16 mm

4.	Microhardness of t	the basic metal matrix Hµ:
	initial state:	~ 1560 MPa
	irradiated steel:	~ 2300 - 2520 MPa

- 5. Burnup of B<sup>10</sup>: Near external surface: 26,7 - 33,5% Middle of the hexagonal wall: 5,2 - 6,5% Internal surface of the hexagonal wall: 14,2 - 17,8%
- 6. The structure under irradiation does not change But bubbles of He are seen around Borides of Cr, Ni, Fe (under magnification x 400 and above).
- 7.
   Bend strength (σ<sub>B</sub>):

   Initial metal:
   628 863 MPa

   After irradiation:
   1100 1540 MPa
- Plastic sag during bend test:
   Initial metal: 0,055 0,193 mm
   After irradiation: 0
- 9. Decrease in steel density after irradiation and annealing at 800°C for 3 hours +  $\Delta V_{anneal} = 4,29 4,68\%$

Table 2 Relative physical efficiency (ρ) of different absorbing materials (AM) under the conditions found in WWER-1000 reactors						
Туре	Absorbing Material	ρ <sub>i</sub> <sup>±1</sup> [%]				
n, α	B <sub>4</sub> C (pellets; natural; 1,8 g/cm <sup>3</sup> ) - standard	100				
n, α	B <sub>4</sub> C (extruded rod; natural; 1,93 g/cm <sup>3</sup> )	~ 100				
n, α	B <sub>4</sub> C (pellets; natural; 2,4 g/cm <sup>3</sup> )	104				
n,α	B <sub>4</sub> C (pellets; 40% B <sup>10</sup> ; 2,3 g/cm <sup>3</sup> )	117				
n, α	B <sub>4</sub> C (pellets; 80% B <sup>10</sup> ; 2,3 g/cm <sup>3</sup> )	122,5				
n, α	$TiB_2$ ; $CrB_2$ (pellets; natural; ~ 4,5 g/cm <sup>3</sup> )	96 - 98				
n, α	B-alloy SBJ-2 (rods; $2\%B_{nat}$ ; ~ 8 g/cm <sup>3</sup> )	71,5				
n, γ	$Al_2O_3 + Eu_2O_3(1 \text{ g/cm}^3)$ - extruded rod	72,5				
n, γ	$Al_2O_3 + Eu_2O_3(1,5 \text{ g/cm}^3)$ - extruded rod	78				
n <i>,</i> γ	$AI_2O_3 + Eu_2O_3(2 \text{ g/cm}^3)$ - extruded rod	85				
n, γ	$Eu_2O_3$ (pellets ~ 7,4 g/cm <sup>3</sup> )	111,5				
n, γ	Hf-Zr (5%) - rod	79				
n, γ	Ni-In (10%) - Sm (10%) - Hf (10%) - rod	76				
n, γ	$Sm_2 O_3$ (pellets; ~ 6,8 g/cm <sup>3</sup> )	81				
n, γ	$Dy_2O_3 \cdot 2TiO_2$ ; (pellets; 5,7 - 6,5 g/cm <sup>3</sup> )	76 - 80				
n, γ	Dy-metal (rod)	82				
n, α n, γ	$EuB_6$ (pellets; ~ 4,7 g/cm <sup>3</sup> )	112,5				

Such a value of the fluence makes it possible to operate ACAs in WWER-440 power reactors for 12-15 years (2-3 years in the automatic control regime and the rest of the time in the emergency protection regime).

# 3. WWER-1000 - design and operational experience of absorbing components (ACs) and control members

3.1. During the initial stage of design work, experiments were performed to determine the relative nuclear physics efficiency of different absorbing materials in the WWER-1000 power reactor environment. The measurements were carried out using cladding specimens produced from X18H10T steel, 8 mm OD, 0.3 mm wall thickness, with neutron absorber  $L = 50.0 \pm 0.1$  mm,  $OD = 7.0 \pm 0.01$  mm. Table 1 lists the most important results of this experiment and Table 2 lists the relative physical effiencies of different absorbing materials under the conditions found in WWER-1000 reactors.

Table 3 Basic characteristics of "mock-up" Neutron Absorber (NA) elements irradiated in reactor SM-2 to $\Phi^{max} \sim 1,7x10^{21} \text{ n/cm}^2$ (thermal)						
Neutron Absorber Type	Extruded R	od Type	Pelleted Type			
Absorber Material	B₄C	$AI + Eu_2O_3$	B <sub>4</sub> C	Dy <sub>2</sub> O <sub>3</sub> ·2TiO <sub>2</sub>	Ni-In-Sm-Hf	
OverallI density of AM [g/cm <sup>3</sup> ]	1,93	3,63	1,72	4,85	9,28	
Density of B <sub>nat</sub> , Eu, Dy etc. [g/cm <sup>3</sup> ]	B~1,53	Eu ~1,85	B~1,35	Dy ~2,93	Sm - 0,4 In -0,44 Hf -0,9	
+∆d <sub>Clad</sub> (sealed design) [%]	~0,5	0	~1,5	0	0	
He <sup>free</sup> [cm <sup>3</sup> NTP]	~1,8	-	~ 3	_	-	
+∆d <sub>NA</sub> (sealed design) [%]	~0,5	0	~ 5	0	0	
+∆d <sub>Clad</sub> (unsealed design) [%]	~0,5	0	-	0	0	
<ul> <li>Notes:</li> <li>clad of NA: steel type X20H40, Φ 8,0 x 0,5 mm;</li> <li>water parameters in channel No. 10, where "mock-up" designs of NA were irradiated: pressure = 19MPa; temperature = 350°C</li> </ul>						

**3.2.** The first reactor tests of short AC dummies (L = 100 mm, cladding OD 8.0 mm, wall thickness 0.5 mm, material - X2OH4O alloy) were carried out in the SM-2 reactor (water pressure 190 kg/cm<sup>2</sup>, temperature =  $350^{\circ}$ C). The maximum thermal neutron fluence accumulated by AC dummies was  $1.7 \times 10^{21}$  n/cm<sup>2</sup>).

Table 3 shows the principal characteristics of the AC dummies tested.

**3.3.** Based upon the results of further reactor tests, performed in the WWER-2 reactor (NPP "Rheinsberg", Germany), the following AC types were tested in special clusters (10 ACs per cluster) - [see Table 4].

In addition to the tests run for "normal" sealed ACs, during the last stage of the experiments at NPP "Rheinsberg", the working capacity of dummy unsealed ACs was tested (L = 100 mm, hole dia. in cladding = 1 mm).



Such specimens were tested for 5000h in autoclaves in a by-pass loop and directly in the reactor core.

In all cases water parameters were similar - P=70 kg/cm<sup>2</sup>, temperature = 270°C, pH = 8.0 - 8.5. Irradiation does not increase the corrosion of absorbing materials significantly. The results obtained have demonstrated that the leaking ACs deliberately produced with the above-mentioned absorbing materials (AMs) retain their working capacity in typical WWER operating environments.

Based upon the analysis of the results of investigations of irradiated ACs, two types were chosen for WWER-1000 control members:

1. as-drawn ACs of an Al +  $Eu_2O_2$  composite (2 g/cm<sup>3</sup>) in an X18H1OT steel cladding (initial OD 15 mm x 0.8 mm, as-drawn dimensions OD 8.2 mm x 0.6 mm). For Novo-Voronezh NPP to meet the AM efficiency requirement, the AM must be the equivalent of  $\geq$  80% natural B<sub>4</sub> C.


- 1. End cap
- 2. Upper weld
- 3. Compensation volume
- 4. Plug
- 5. B<sub>4</sub>C absorbing material Vibropacked ≈1.75 g.cm<sup>-3</sup>
- 6. Cladding X18H10T steel  $\phi = 8.2*0.6 \text{ (mm)}$
- 7. Lower weld
- 8. Tip

Figure 2

2. as-drawn ACs based on  $B_4C$  (1.9-2.0 g/cm<sup>3</sup>) in X18H10T steel cladding (initial OD 11.0 mm x 0.7 mm, as-drawn dimensions OD 8.2 mm x 0.6 mm) for the control of commercial WWER-1000s.

To avoid NPP unit contamination by radioactive isotopes  $Eu^{152}$  (half-life 13y) and  $Eu^{154}$  (half-life 16y) as the result of an accidental rupture of at least one AC during repositioning of absorbing rods (ARs) of control and safety systems between refuellings, the decision was taken to replace all Al+EU<sub>2</sub>O<sub>3</sub> ACs of control and safety systems with B<sub>4</sub>C ARs, though they have considerably less radiation stability (B<sub>4</sub>C swelling and free helium formation due to B<sup>10</sup> + n  $\rightarrow$  He, Li).

Based on the generalization of test results in WWER-2 and SM-2 reactors on the modified ACs and further investigation of irradiated specimens, the service life was defined as follows:

- year in the automatic control regime,
  - 5 years in the emergency protection regime.
- **3.4.** Fig. 2 shows the design, dimensions and materials of a vibrocompacted AC, which has a number of advantages over previous designs.

#### Table 5

The specification of control rods and test conditions for long-term tests in Unit 5 of Novo Voronezh, WWER-1000 NPP

Type of NA (NA Material; technology)	The number of clusters(rods)	Time and use in power control (PC) or safety control (SC)	Effective power. days	Max. fluence [n/cm <sup>2</sup> ]		The research: " + " complete. "-" planned
				thermal x10 <sup>21</sup>	E>0.8MeV x10 <sup>21</sup>	
B₄C (vibropack)	6 (72)	2y - PC	491	~ 1.5	~3.3	+
"	5 (60)	Зу - РС	793	~2.4	~5.3	+
B₄C(extruded)	41 (492)	7y - SC	2028	(2-5)*	~4.5*	+ * *
Dy <sub>2</sub> O <sub>3</sub> TiO <sub>2</sub> (vibropack)	6 (72)	Зу - РС	813	~2.5	~5.5	+
"	5 (60)	4y - PC	*	×	*	_***
B₄C (vibropack)	7 (84)	Зу - РС	*	~3.2*	~7.6	_***
11	3 (36)	2y - PC + 3y - SC	*	*	*	_***

\* The exact data will be worked out and presented later in the test reports on these NAs.

\*\* The research has been completed and the report is in preparation.

\*\*\* This research is planned for 1994-95.

# Table 6. The basic results of investigations of NAs in the hot cells of Novo-Voronezh NPP and Research Institute. NAs used in the Automatic power control subsystems of Unit 5 of Novo-Voronezh NPP (NAs - vibropack type, based on $B_4C$ and $Dy_2O_3$ ·TiO<sub>2</sub>).

Characteristics	NA Ba	NA, Based on $Dy_2O_3$ ·Ti $O_2$ $\tau_{PC} = 3$ years	
	$\tau_{PC} = 2 \text{ years}$	$\tau_{PC} = 3$ years	
Common condition of NA	$\Delta d = 0$ ; All rods are intact; ther of corrosion products; thickness		
Max. Burnup, B <sup>10</sup> (%)	- 32,5	~42,6	Isotopic composition of Dy will be determined at 0-750 mm distance from the bottom of the NA
Production of free He within NA (cm <sup>3</sup> NTP)	70-130	202-248	-
Pressure of gas under clad of NA (atm) -20°C -350°C	5,0-9,3 10,6-19,8	14,4-17,7 30,5-37,6	-
Condition of absorbing materialFor mechanical properties of irradiated clad, NAs were cut into circular specimens. The powder of $B_4C$ was cleaned from all specimensAt ~3 Close the str and no ( $\Delta d = 0$ )		At ~300mm from the bottom of NA the B <sub>4</sub> C was sintered; it was in close contact with the clad, but the stress in the metal was low and no creep was observed ( $\Delta d = 0$ ).	see $B_4C$ with $\tau_{PC} = 2$ year
Mechanical props. of clad most irradiated (bottom of NA):- $\sigma_{\rm B}$ (kg/mm <sup>2</sup> ) at 20°C $\sigma_{\rm B}$ (kg/mm <sup>2</sup> ) at 350°C	80,2-84,0 44,6-64,0		~102 ~68
σ <sub>0,2</sub> (kg/mm²) "	47,2-51,9 28,4-29,3		-
δ <sub>ο</sub> (%) "	10,0-17,3 4,9-9,2		1,4-1,8
Characteristics	NA Based on B₄C		NA, Based on $Dy_2O_3$ . Ti $O_2$ $\tau_{PC} = 3$ years
Conclusions about NA condition	The clusters are far from their limits.	The clusters are close to their limiting conditions.	Clusters may be exploited in the PC- regime for an extra 1-2 years.

- **3.5.** Tables 5 and 6 list the basic results of ACs investigated after service life tests in the WWER-1000 reactor, Unit No. 5 of Novo-Voronezh NPP. As a result, it can be stated that in the process of AC modification, the following measures are expedient:
  - a) utilization of a cladding material with less radiation embrittlement under prolonged neutron irradition;
  - b) utilization of an  $n,\gamma$  neutron absorber (e.g.  $Dy_2O_3 \cdot TiO_2$  or Hf) instead of  $B_4C$  in the bottom part of AC due to its considerably higher radiation stability (no out-gassing, high structural and volumetric stability).
- **3.6.** During operational experience of  $B_4C$ -based ACs in commercial WWER-1000 reactors at NPPs in Russia and the Ukraine no failure of any of these components during 95 reactor-years at 17 NPP units has yet been observed.

In 1993, on the basis of the above-mentioned data on  $B_4C$ -vibrocompacted ACs, the absorber assembly service life in the control and safety system (cross-member and 18 ACs) was increased to 2 years in the automatic control regime.

Work is in progress to validate the safe extension of the service life of these items to 6-8 years in the emergency protection regime.

#### 4. Further improvement of WWER-1000 power reactor control members

It is evident that it is absolutely necessary to use an n,  $\gamma$  - absorbing material instead of B<sub>4</sub>C in the bottom part of the AC which bears the highest radiation load. Thus, it becomes necessary to increase the AC diameter, otherwise it is impossible to provide for the required physical efficiency, because only Eu-based absorbing materials can compete with B<sub>4</sub>C in this aspect, but they are highly radioactive. Along with the requirements for high physical efficiency and radiation stability, n,  $\gamma$  - absorbers at the bottom of ACs should possess high corrosion resistance and should not produce  $\gamma$ -active daughter isotopes when interacting with neutrons. For AC cladding a structural material with higher radiation stability able to retain a plasticity ( $\delta_0$ ) of not less than 3 - 4% in the environment of high neutron irradiation doses should be used.

Fig. 3 shows two variants of AC designs with n,  $\gamma$ -absorbers.

Compared to currently produced components, the new AC designs possess the following advantages;

- efficiency is 14-18% higher;
- reliability in operation, because the area under the heaviest irradiation contains structural and absorbing materials with a higher radiation stability.
- service life is equal to reactor service-life (about 30 years), including 2-3 years in automatic regime and the rest of the time in the emergency protection regime.

The basic factors which support the decisions taken are the following:

1) dysprosium titanate  $(Dy_2O_3 \cdot TiO_2)$  compared to the ln-Cd-Ag alloy commonly used in absorber assembly clusters has the following advantages:

- a) "burn-out" of dysprosium isotopes (see Fig.4) is slower, than the "burn-out" of indium, cadmium and silver isotopes:
- b) it has no long-lived  $\gamma$ -active isotopes, such as Ag-110,
- c) with equal physical efficiency, it has a rather high radiation stability and corrosion resistance. In addition, because of its lower density, heat generation is lower.



Figure 3



2) metallic hafnium has the advantage of being both an absorbing and structural material. Consequently, because of a greater effective diameter, one can provide for the efficiency of the bottom part of the AC to be within 90-100% of the efficiency of the top part (boron carbide in cladding), despite the fact that:

$$\frac{\rho_{_{Hf}}}{\rho_{_{B_{4}C}}}=0.8$$

In addition the utilization of hafnium without cladding results in its temprature being practically equal to the temperature of the reactor primary circuit coolant and the strong oxide film formed on AC surface (5-8  $\mu$ m) makes a good barrier against hydrogenation of the metal proper.

3) EP-630U alloy (see section 5.3) has a high mechnical strength, which allows for a smaller AC cladding thickness and makes it possible to increase the absorber diameter, i.e. to increase the efficiency compared to X18H10T or 304 steels.

The high resistivity of this alloy to radiation embrittlement increases the life expectancy of an AC and absorbing assembly to between 15 (minimum) and 30 (maximum) years.

5. Basic results of investigations of the properties of Hf,  $Dy_2O_3$ .TiO<sub>2</sub> and EP-630U alloy

Below, the main results of the work which supports the utilization of these materials in reactor are presented.

#### 5.1. Hafnium

- **5.1.1.** The relative physical efficiency of hafnium is equal to 80-85% of  $B_4C$  (1.8 g/cm<sup>3</sup>) with the natural isotopic content of  $B^{10}$ .
- **5.1.2.** The chemical compositions of two commercially produced grades of hafnium are listed in Table 7.
- **5.1.3.** Radiation stability of hafnium was investigated using specimens irradiated in SM-2 and BOR-60 reactors up to fluences of:
  - thermal neutrons:  $\sim 7.2 \times 10^{22} \text{ n/cm}^2$ .
  - with  $E \ge 0.1$  MeV: ~ 2.4 x 10<sup>22</sup> n/cm<sup>2</sup>.

These investigations of irradiated specimens and AC dummies have demonstrated:

- Hf has high structural and volumetric stabilities (+ $\Delta V \leq 0.6\%$ );
- an oxide film (5-8  $\mu$ m) protects hafnium against hydrogenation by atomic hydrogen formed by the radiolysis of primary circuit water: HfH<sub>2</sub> < 3 x 10<sup>-2</sup>%;
- mechanical characteristics of hafnium are quite good:  $\sigma_B$  and  $\sigma_{0.2}$  increase by factors of 1.5-2, while plasticity retains a reasonably high level ( $\delta_0 = 2-4\%$ ).
- **5.1.4.** The corrosion resistance of pure metallic hafnium in conditions corresponding to WWER-1000 primary circuit water after 15000h testing is found to be very high, approaching the "absolute resistance" class.

Elements	% weight				
	GFI-1 (lodine)	GFE-1 (Electrolytic)			
Hf	Base (≥99,8)	Base			
Zr	≤1,0	≤0,6			
N	≤0,005	≤0,003			
Fe	≤0,04	≤0,02			
Si	≤0,005	≤0,004			
Nt	≤0,05	≤0,002			
Ті	≤0,005	≤0,001			
Al	≤0,005	≤0,005			
Са	≤0,01	≤0,01			
Mg	≤0,004	≤0,004			
Mn	≤0,0005	≤0,0005			
С	≤0,01	≤0,006			
Cr	≤0,003	≤0,001			
Nb	≤0,002	≤0,002			
Мо	≤0,1	≤0,01			
0	≤0,01	≤0,03			
W	≤0,01	≤0,01			
Cu	≤0,005	≤0,005			

 Table 7

 Chemical composition of Hf - grades

Under irradiation hafnium retains its high corrosion resistance: after 26800h a dense oxide film is formed (5-20  $\mu$ m). The corrosion rate after the first 4000h equals zero, and + $\Delta$ m does not exceed 0.8%.

#### 5.2. Dy<sub>2</sub>O<sub>3</sub>.TiO<sub>2</sub> (dysprosium titanate).

-

- **5.2.1.** The relative physical efficiency (compared to  $B_4C_{nat.}$ ; 1.8 g/cm<sup>3</sup>) in a WWER-1000 reactor environment is equal to 76-83% (for Dy within the range 2.5-5 g/cm<sup>3</sup>).
- **5.2.2.** Dysprosium titanate has been commercially produced for several years already (it is used as an absorbing material in the "stripe" ACs in the latest modification of the rods in the automatic control and emergency protection systems of RBMK-1000 and RBMK-1500 reactors).
- 5.2.3. The high radiation stability of dysprosium titanate is supported by:
  - tests and further investigations of spent ACs (after 3 years of operation in the automatic control regime at Unit No. 5 of Novo-Voronezh NPP);

- test and investigations of irradiated pellets in the SM-2 and BOR-60 test reactors ( $\Phi_{E0} \sim 2.4 \times 10^{22} n/cm^2$ ):  $+\Delta d_{AM} \langle 1\%$  (due to thermocyclic cracking);
- -  $\Delta V$  of the grid ~ 2%; structure and phase composition are practically unchanged.
- **5.2.4.** The corrosion resistance of AC dummies in a WWER-1000 primary circuit water environment during 10000h testing is rather high.

$$+\Delta m < 1\%;$$
  
+ $\Delta d_{AC} - 0.$ 

#### 5.3. EP-630U alloy

5.3.1. Chemical composition: Chromium 42% (by weight); Molybdenum 1%; Nickel the remainder.

# 5.3.2. Mechanical characteristics of the material in the pre-irradiated state:

Test temp. (°C)	σ <sub>e2</sub> (MPa)	σ <sub>a</sub> (MPa)	δ, (%)
20	$520\pm23$	876±16	$46,7\pm 3$
350	$388 \pm 42$	$702 \pm 57$	$41,5 \pm 5$

#### 5.3.3. Post-irradiation mechanical characteristics:

 $\Phi_{E>0.1MeV} \sim 3 \times 10^{22} \text{ n/cm}^2$ 

c (MPa)	at 20° C	750
	at 350°C	540
		070
(MPa)		870
	at 350°C	660
······································		
\$ (0()	at 20° C	30%
0 <sub>0</sub> (%)	at 350°C	20%

# **OVERVIEW OF THE EDF APPROACH CONCERNING RCCA PROBLEMS**

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#### Abstract

Since 1988 EdF has decided to inspect the RCCA of all the nuclear plants. This decision has been reinforced by two incidents which occurred in 1988 and 1989 where one rod broke by excessive wear.

The inspection allowed to identify two problems:

- clad wear due to fretting,
- clad swelling which could induce cracks.

To address these problems essentially three actions are initiated:

The first concerns the immediate requirements and consists in defining a RCCA management strategy based on mechanical calculations. This strategy includes periodical controls, axial repositioning and specific rejection criteria.

Second concerns some design improvements of RCCA. Regarding the wear problem, various hardening methods of the clad have been investigated (nitride, chrome carbide, chrome). For the swelling, an increase of the gap between the absorber and the clad should delay their interaction.

Third, is a long term work and more ambitious approach. It consist in a large R&D program concerning the study of the phenomena involved in three wear process. The aim is to built a full set of models and to gather it in a computer code, for predicting RCCA wear.

The state of these actions is the following:

The first action has allowed Utilities to operate their power plants in safety conditions, even with standard RCCA.

In the second, out of pile tests have demonstrated the potential efficiency of various remedies. These remedies are progressively introduced, now about 180 new design RCCA are in reactors. The first controls confirm the good behaviour of these solutions.

The third action is still in progress. EdF has planned to qualify the code in 1995. Then it will be possible to perform analytical studies to investigate modifications on the design of RCCA or internal guide tubes.

# 1. Introduction

The RCCA system ensure important safety functions as reactivity control in normal conditions , quick insertion of anti reactivity in incident and accident conditions and safety state in all the shutdown conditions. These functions has to be preserve in any case. Particularly no important damage could be accepted.

	CP0 - 900MWe	CPY - 900MWe	PQY - 1300MWe
Number of plants	6	28	20
Plant type	17x17 - 3Loops	17x17 - 3Loops	17x17 - 4Loops
Hat temperature	T hot	T cold	T cold since 1992
Guide tube type	Deep beam 125"	Deep beam 125"	Inverted hat 96"
Number of RCCA	48	53	65
Number of rods	24	24	24
Clad thickness	0,47	0,47	0,98
Absorber material	AIC	AIC	AIC and B4C
Tubing material	304L	304L	304L

# TABLE 1. MAIN PLANT & RCCA DESIGN FEATURES

Wear and swelling problems has been encountered in all reactors and appear to be potential limiting phenomena. Industrial controls are now perform regularly and they give a large amount of results which is very useful to know where the damages are located and to assess the kinetics of the phenomena. In addition hot cells examinations give some indications on the mechanisms and on the damages' characteristics.

Based on these findings, EdF starts three actions. The first one is to define a management strategy in order to continue to operate the reactor in safety conditions despite the existing phenomena. The second is to look for new RCCA design among those proposed by the suppliers. The aim of the last is to develop a computer code to predict the wear phenomena in order to improve the design of internal guide tubes, particularly for new power plants. All these actions are presented in the next paragraphs.

The main design features of the EdF plants, internal guide tubes and RCCA's, which have some importance regarding RCCA problems, are gathered in table 1.

# 2. Feed back experience

At the very beginning RCCA clad wear has been considered as a potential problem but essentially because of the sliding against the internal guide tube during the step by step motions. As load follow operating conditions could emphasise the sliding wear (some RCCA could accumulate a total of steps motion exceeding 500 000) EdF decided to examine some RCCA to assess the amount of the wear. The very first RCCA control on EdF reactors had been performed in 1984 on BUGEY4 plant. On this plant, the wear appeared very low like in many others T hot reactors in the world. However others controls done in 1986 on six T cold 900 MWe reactors showed a lot of wear particularly in front of the guide tube cards on low duty RCCA. On 1300 MWe RCCA's the most affected zone regarding the wear is the tip. These observations indicates that the main wear mechanism is the fretting induce by hydraulic forces inside the guide tube of the upper internals, and it has been confirmed by out of pile test (see § 5). As 900 MWe and 1300 MWe guide tube have different design the hydraulics force and so the wear kinetic are not the same.



Figure 1

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In 1988 - 1989 two incidents occurred respectively in DAMPIERRE 1 and GRAVELINES 4 (CPY plants) where one rod failed in front of 7th card (total number of cards is 8 on 900 MWe). Expertises, carried out on the two rods, revealed: the mechanism of the failures which is fatigue damage and the specific action of the AIC rod upper end which increases the bending stresses [1].

Since 1984, more than 100 RCCA control campaigns has been perform on the 900 MWe and 1300 MWe EdF reactor. The results shows a large scattered in wear from one reactor to an other or from one position in the core to an other one. Nevertheless some trends emerge concerning the worst position in the cluster, the evolution of the kinetic and the various shapes of the wear. For example the single rod vanes are the more affected position. More ever the kinetic decreases from one cycle to the next and higher is the kinetic, higher is the decrease of the kinetic.

With the same equipment used for wear control some clad swelling has been identified in 1989 on 1300MWe reactors and in 1990 on 900 MWe reactors. Such swelling are located at the bottom of the rod and is correlated, in part, with the amount of neutron captures [figure 1]. In fact the swelling of the clad is due to the settlement of AIC during the step by step motions and also to the volume increase induce by transmutation of indium into tin. The local increase of the clad diameter has to remain compatible with the assembly guide tube to avoid any burst of the assembly dash-pot or RCCA jam.

In addition the swelling is generally associated with clad cracks. The potential consequences of unwatertight clad are Ag 110 m contamination for all shutdown RCCA filling with a AIC rod. But in 1300 MWe reactors the shutdown RCCA's contain also B4C pellets which is a soluble material in specific conditions [2]. Hot cells examinations, carried out on several swelling rods, demonstrate the absence of contact between water and B4C due to the strong binding of the absorber and the clad in the swelling zone. This establishment is up to now limited to the R group (temperature control) and additional examinations are required to extend the conclusion to the others groups.

#### 3. RCCA management on 900 MWe and on 1300MWe reactors

#### 3.1 Description

The numerous wears find on RCCA leads EdF to define a management strategy in order to avoid unacceptable incidents like those appeared in 1988 but also to take into account the swelling problem. For 900 MWe reactors the management strategy has been completely established in 1990 while for 1300 MWe reactors started in 1992.

In the two cases the management strategy has three aspects:

- a full set of rejection criteria based on mechanical studies
- a frequency control
- a procedure for RCCA positioning

Those three aspects are linked. In any cases shuffling is not permitted, this is due to the fact that wear kinetics used to define the criteria are based on wear measurements obtained cycle after cycle without any modification of the RCCA-guide tube couple. On the contrary axial repositioning is used systematically with three allowed level for 900 MWe and five for 1300 MWe. This procedure spreads on independent zones the wear and increase the life time of RCCA's but it does not improve the situation regarding wear on the tip (worst zone on 1300 MWe) because the assembly guide tube is a continuous element.

Frequency control is based on the wear and swelling kinetics. The control is performed every three cycles on 900MWe reactors and every two cycles on 1300 MWe reactors. The inefficiency of axial repositioning for wear tip and the recent modification from T hot to Tcold upper internal conditions on all 1300 MWe, explain the difference between the two situations.



Figure 2



Figure 3



Figure 4

The rejection criteria are defined taking into account the calculated limiting wear or swelling, the kinetic of the phenomena before the next control and the axial repositioning procedure (just for wear).

On 900 MWe RCCA's only contain AIC absorber material so the criteria has to prevent from RCCA rod failure. For 1300 MWe the potential risk of B4C dissolution requires to avoid any leak. To determine the wear limits it is necessary to assess the actual forces acting on the component and to know the actual wear shapes. The first are measured on a specific mock up and the second come from hot cells examinations. An another important parameter is the maximum number of step by step motions the various RCCA group can done. So, more a RCCA group is concerned by load following, lower is the criteria because of the leading fatigue mechanism.

From mechanical point of view, swelling of the clad in the lower part could increase significantly the over-pressure when the RCCA go into the dash-pot assembly during a shutdown. The RCCA speed at the entrance of the dash-pot and the axial shape as the location of the swelling are the main parameters governing the over-pressure.

# 3.2 Application

The figures 2, 3 and 4 give the number of rejected RCCA by application of these management strategies. In fact as far as swelling is concerned, in 1990 on 900 MWe reactors, the RCCA was rejected when it exceed about half of the criteria limit because of the quite unknown kinetic of the phenomena at that time. For 1300 MWe, as mentioned before the criteria are well established at the beginning of 1992, so previously just empirical criteria are used.

The total amount of rejected RCCA on CP0 reactors (900 MWe T hot) decreases significantly and seems to stabilise to about 30 rejections every year (mean value per reactor: 10 rejected RCCA). No control had been performed in 1991. This decrease could be explain by the fact that swelling was an important cause of rejection in the past and this phenomena present an incubation period (for AIC-clad gap closure) so it take some time before finding swelling on new RCCA.

On CPY reactors (900 MWe T cold) the situation is not so good as in 1993 the total amount of rejections is similar to the 1990's one. Between 70 and 170 RCCA are rejected every year mainly due to wear on the cards (mean value per reactor in 1993: 20 rejected RCCA). The large number of rejected RCCA is mainly due to the conservatism of the criteria but it confirm the high level of wear on this type of reactor.

It is a little more difficult to interpret the evolution of the rejection on 1300 MWe reactors because most of these plants are very recent so the number of control campaign increase continuously. Nevertheless swelling and wear tip appear the main causes of rejection even if some card wear are found on the oldest plants. The mean value of rejection per reactor is about 10 so the situation seems to be similar to CP0 one. But the more important conclusion we can get from the comparison of the results between 1992 and 1993 is that the conversion in T cold of the all the 1300 MWe does not increase the wear as it could be feared, considering the situation on 900 MWe reactors between CP0 and CPY.

# 4. RCCA new design

#### 4.1 Wear remedies

In parallel to the establishment of the RCCA management strategies EdF ask to FRAMATOME to develop new RCCA design to settle or reduce the wear and swelling problem.

In this purpose FRAMATOME set up a R&D program consisting in autoclave wear test in order to select the best wear remedy. Finally FRAMATOME choose the ionic nitrogen treatment for the absence of any adherence problem, the cheap industrial process and its good efficiency regarding the wear even in very conservative fretting conditions.

ENTERING	TYPE	NUMBER	REACTOR	CONTROL	CONTROL	CONTROL	CONTROL
YEAR				89	90	91	92
88	Nitrided	2	CPY	No wear	No wear	No wear	No wear
89	Nitrided	2	PQY	*	No wear	50µm	70µm
89	LC1C	2	PQY	*	360µm*	360µm*	360µm*
91	Nitrided	14	CPY	*	*	*	No wear
92	Chromium	15	CPY	*	*	*	*
93	Nitrided	146	CPY	*	*	*	*
93	Chromium	23	CPY	*	*	*	*

#### TABLE 2. EXPERIENCE ABOUT NEW RCCA DESIGN

\* wear on uncoated area

Others remedies were also proposed by the suppliers as chromium carbide coating and hard chromium coating. Out of pile tests indicate also a good wear resistance of such coating.

EdF decided to test in reactor as soon as possible all these solutions, to assess their actual behaviour. They have been introduced progressively since 1988/1989 for nitrogen treatment and chromium carbide coating and since 1992 for hard chromium [table 2]. All these RCCA are inspected regularly as defined by the general management strategies. And for the very first one systematic ultrasonic measurements have been performed in order to detect the incipient wears

The results show a very good wear resistance behaviour for nitrogen treatment and chromium carbide coating On the sixteen nitrogen RCCA loaded in CRUAS 2 none wear has been detected after one cycle whereas some standard RCCA's have to be rejected for exceeding criteria value after the same time [figure 5]. The very limited wear measured on one rod of a nitrogen RCCA loaded in PALUEL 2 is in the range of the uncertainty and is located on the tip where the treatment is lower due to end effects in the fabrication furnace. Improvement of nitrogen process has been adopted in 1991 and a very good homogeneity of this treatment is now obtained. In the same reactor the wear measured on chromium carbide plated RCCA is also at the lower end in an uncoated zone (few centimetre near the tip are uncoated zone).

No inspection has been performed until now on chromium coating.

In the next years this experience should increased as now the total amount of wear remedies RCCA in reactors exceed 200.



Figure 5

Before concluding on the efficiency of the various wear remedies an other element must be considered: the behaviour of the mating surface represented by the cards in the discontinuous part of the guide tube. These parts are very solid and thick but in any case the guide function has to be ensured. Out of pile test are not easy to carry out because it is difficult to get representative contact forces and displacements. The trend, coming from the comparison of several experimental results obtained in various conditions, shows the more representative are the fretting and the sliding conditions, the less different are the behaviour of the opposite surface (between stainless steel / stainless steel and treated steel / stainless steel).

# 4.2 Swelling remedies

As AIC material is a very soft material at operating temperature (350°C), two modifications could delay the interaction between the absorber and the clad:

- increase of the gap,
- decrease of the spring rod force.

The first one is an easy way to get some delay but it could be useful to combine this modification with a better thermal conductivity gas filling, as helium, to avoid any temperature absorber increase.

The second one is the result of an optimisation study concerning spring force apply all along the irradiation to ensure:

- a good push on the absorber in order to prevent any take off
- no excessive stresses in the absorber which could accelerate its creep.

Since 1992, all the new RCCA put in the EdF's reactors have had such improvements but no feed back experience are available at that moment because of the threshold effect of the clad swelling phenomena.

# 5. A computer code for wear prediction

The two previous actions allow to control the situation after the two incidents occurred in 88 and 89 and improve it by introducing gradually new design RCCA's. But the RCCA monitoring spend money and even more time, during the shutdown period of the reactor. So it could be more profitable to reduce the wear causes itself rather than to take care to the wear. In this way it could be interesting to study a new design for the internal guide tube, for new power plants but also for existing reactors.

This could be done by direct in loop test as FRAMATOME proceeded to define the guide tube of the new french reactors call N4 with his MAGALY loop. On this full scale driving RCCA mock-up various guide tube modifications have been tested and the main contact parameters have been measured in each case. Finally a combination of the more efficiency modifications have been tested in order to ensure an actual benefit.

A complementary approach, which corresponds to an ambitious and long term work, is the assessment of the wear mitigation leaded by any improvement of the guide tube design. The aim of this work is to obtain a comprehensive answer regarding wear problem. To achieve this objective it is necessary to develop a full computer code including models which can simulate all the steps from stimulating sources to the wear.

EdF set up this R&D activity since 1990, partially in collaboration with FRAMATOME and CEA, with a large programme valid for a number of years. The R&D EdF department is in charge of this work.



Figure 6

The programme is divided in three parts:

- study of the hydraulic sources,
- study of the mechanical behaviour of the RCCA rods,
- study of the wear.

All these parts are linked and are based on mock-up test to characterise the phenomena and to validate each modelling steps.

The goal of the first part is to identify the stimulating hydraulic sources and to quantify the hydraulics forces which act on the RCCA rods. The identification has been done on FRAMATOME MAGALY from the 900 MWe guide tube tests (figure 6). Two main hydraulic sources has been found:

- the first in the lower part of the guide tube, at the outlet of its continuous part,
- the second is located at the upper part of the guide tube.

To study these sources, two mock-up are built: GRAPPE 1 for the lower part and GRAPPE 2 for the upper one [figure 6]. Specific analytical methodologies are developed to characterise quantitatively the hydraulic forces in such way that it can use as input in a computer code. In the upper part the source is a combination of fluid elastic forces and turbulent forces while in lower part only turbulent forces are considered. Also some parametric tests are performed to assess the sensitivity of the hydraulic forces to the inlet or outlet flow distribution. This is necessary because of the great uncertainties concerning the flow distribution.

The second part is devoted to the determination of the RCCA behaviour submitted to the previously calculated hydraulic forces. This requires the development of non linear dynamic models with sliding and impact. The MASSIF device allowed to validate the models considering the forces, the displacements and the wear powers in the impact zones. The tests are performed in various conditions (air-water, mono-biaxial stimulation, gap size....).

After completion of the these two parts it is necessary to validate globally the calculations on a new mock-up call MOCHE. This mock-up simulates only one rod and only one support with a gap. On this very simple configuration the comparison between calculated and measured values are very useful.

The third part concerns the wear modelling in order to predict the volume and the depth of the wear starting from the characterised impact conditions obtained in the former part. As the knowledge on wear mechanism is very limited, the approach considered is based on experimental tests.

The simulator VIBRATEAU which is a mono-axial stimulation is used for these tests. It can run in various conditions and even in PWR conditions. For example the big sensitivity of the wear to the temperature has been investigated and confirmed by the results obtained. Considering the simple ARCHARD's wear rule it is possible to determine a wear factor corresponding to the ratio between the power and the wear volume. Bi-axial tests are also foreseen in a near future on a new AECL machine. It should allow to perform more representative tests. So more sophisticated models could be adjusted to get a better prediction.

The connection of all the models inserted in the code ASTER should be completed with the qualification in 1995. This computer code will be used for wear sensitivity studies regarding different parameters as gap size between RCCA clad and card or RCCA axial position or clad material and many others. So it should contribute to define design improvements of the guide tube which whould be easy to adopt on existing reactors and for a more general point of view to optimise the management strategy itself.

# 6. Conclusion

The two problems encountered on RCCA's, the wear and the swelling of the clads, are now well identify and it is now demonstrated that it is possible to manage it. But the rejection rate remains quite high, particularly on CPY. So EdF continue to study to improve the management strategies to reduce the rejection rate but keeping the same safety level.

Remedies regarding these two problems has been introduced in reactors. After four years the ion nitride treatment looks very efficient to reduce drastically the RCCA wear. The exact life time of this remedy has yet to be established and the absence of adverse consequence on the mating surface has to be confirmed. Also the benefit provided by the swelling remedies will be appreciated only in several years.

The best way to solve the wear problem is to eliminate or reduce the causes of the damage. So improvements of upper internal guide tube are necessary for new power plants and also for existing reactors. An approach considered by EdF is to develop a full computer code able to predict the wear. This work is not completed but it already allows a better understanding of the mechanisms involved in the wear phenomena. Studies performed with such code will contribute to improve the design of the existing guide tube.

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# ADVANCES IN CONTROL ASSEMBLY MATERIALS FOR BWRs AND CANDUS

# ADVANCED CONTROL RODS FOR JAPANESE BWR PLANTS

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#### Abstract

Two types of long-life control rod design with pure hafnium absorber were developed in Japan for use at Control Cell locations and were introduced into the commercial BWRs in 1989. One of those is called "the plate type", in which a few pairs of thin hafnium plates are fixed by stainless steel pins in each wing of a cruciform blade. Water between the hafnium plates produces flux trap effect which increases control rod The other design is called "the rod type", in which hafnium rods worth. are hung on the top of each wing of the cruciform blade. The dimensions of the control elements of both designs are chosen considering thermal expansion and irradiation growth of hafnium and the chemical compositions of pure hafnium metal are compliant with the ASTM Code requirement. Thinner hafnium plates or slender hafnium rods in the lower portion of control rod counteract weight problem caused by high specific gravity of hafnium. The important merit of the hafnium control rods is considerably longer nuclear lifetime compared to the boron carbide control rods which were originally used for full core. Hafnium metal, as the absorber, is chemically and physically stable under normal and accident conditions. The corrosion tests showed that hafnium had better corrosion resistance compared with Zircaloy. The irradiation tests showed that irradiation

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growth rate of hafnium was negligibly small. The hafnium control rods in commercial plants have exhibited good performance up to the current maximum 3-cycle irradiation experience.

#### 1. Introduction

Currently, twenty-five commercial BWRs are in operation in Japan.

In the BWRs, cruciform control rods are driven along the gap among four fuel assemblies (Fig.1.1). Boron carbide was originally used as the neutron absorber. In the early 1980's an improved core management was applied to the Japanese BWRs, in which the control rods at specified locations, Control Cell locations, stay in the core during plant operation ; and the control rods at the other locations, non-Control Cell locations, are fully withdrawn (Fig.1.2).

Today, two kinds of materials are used as absorber for control rods in Japanese BWRs. One is boron carbide, and the other is hafnium. The control rods with boron carbide absorber are used for shutdown at non-Control Cell locations, whose design is similar to that in US BWR plants.

Two types of long-life control rod design with pure hafnium absorber, so called "the plate type" and "the rod type", were developed in Japan for use at Control Cell locations and were introduced into the commercial BWRs in 1989.

Longer lifetime of the control rods with hafnium can contribute to reducing the number of control rods to be exchanged and to reducing the amount of radioactive waste accordingly.

#### 2. Advanced control rods

#### 2.1 Plate type hafnium control rod

The plate type control rod is illustrated in Fig.2.1 and its main specification is shown in Table 2.1[1]. A couple of hafnium

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Fig.1.1 Core configuration.

plates are separated from each other with intermediate water gap and fixed by stainless steel pins in each wing of a cruciform blade. To secure reactor shutdown effectiveness, the upper portion of the blade is loaded with pairs of thicker hafnium plates. And the remainder with pairs of thinner plates reduces the weight of the blade. The water gap between hafnium plates is a so-called "flux-trap" region where neutrons transmitted into the blade are moderated in this region and absorbed on the inside surfaces of the hafuium plates (flux-trap effect). The reactivity



(+): Control cell location
+ : Non-control cell location

Fig.1.2 Typical arrangement of control rods in the core.

worth increases with an increase in water gap within the extent of several millimeters. Fig.2.2 shows the flux-trap effect measured by the critical experiments performed at Toshiba Nuclear Critical Assembly (NCA)[3].

Though the reduction in the thickness of hafnium plates results in the decrease in the control worth, the flux-trap effect compensate for it. Consequently, the reduction in the plate thickness was achieved while securing the even control worth.

Hafnium plates with four kind of thinknesses from 1 to 2 mm, are used in a blade. In order to eliminate interference between



Fig.2.1 Plate type hafnium control rod.

		Plate type hafnium control rod	Rod type hafnium control rod	Boron carbide control rod
	Effective length	3.63 m	3.63 m	3.63 m
	Blade thickness	8 mm	7 mm	8 mm
Dimensions	Wing span	250 mm	250 mm	250 mm
	Sheath thickness	1 mm	1 mm	1 mm
Absorber 04 hafnium plates		<ul> <li>3~5 mm<sup>*2</sup> in outer diameter.</li> <li>84 hafnium rods</li> </ul>	<ul> <li>5 mm in outer diameter of stainless cladding tube.</li> <li>84 B<sub>4</sub> C rods.</li> </ul>	
Weight		100 kg	100 kg	100 kg

Table 2.1 Main specification of control rods. \*1

\*1:The above values is for a control rod of BWR 4.

\*2:Dimension for the upper portion is larger and that for the lower portion is smaller.

hafnium plates and stainless steel sheathes due to difference of thermal expansion and irradiation growth, some countermeasures are taken : there are gaps between hafnium plates, and the hole diameter of a hafnium plate is larger than the diameter of a stainless steel pin.



Fig.2.2 Reactivity worth increase with "flux trap"[3].

#### 2.2 Rod type hafnium control rod

The rod type control rod's structure such as handle, tie rod, and sheath, except for the absorber material was designed to be similar to that of existing much experienced boron carbide control rod's as much as possible. The configuration is illustrated in Fig.2.3 and its main specification is shown in Table 2.1[2].



Fig.2.3 Rod type hafnium control rod.

In this type of control rod, the hafnium rods are used in all the absorber material zone, which have much irradiation experience.

The hafnium rod's diameter is 5mm for the upper portion and 3mm for the lower portion. Both portions of hafnium rods are connected with each other by welding in the middle of each blade. Twenty-one hafnium rods are hung on the top of the blade in each wing of the cruciform blade.

As well known, the hafnium density is higher than that of boron corbide, but the maximum weight of the total control rod is limited from the viewpoint of the mechanical integrity. To counteract this problem, slender and hence lighter hafnium rods are used for the lower zone so far as the nuclear worth is secured.

The sheath which is made of thin stainless steel plate envelops the hafnium rods. The centered tie rod connects the upper handle and the velocity limiter which moderates the drop velocity at the time of control rod drop accident.

To control the total weight of the control rod, also the lighter velocity limiter comparing to the boron carbide control rod is used.

#### 3. Properties of hafnium

#### 3.1 Physical properties

Hafnium, which belongs to the 4A group of the periodic table same as zirconium, is obtained as a by-product during the separation of zirconium from raw material. Hafnium has a high melting temperature of 2504  $\pm$ 20K, and undergoes a transformation from a hexagonal close packed (hcp) structure to a body centered cubic (bcc) structure at a temperature above 2000K [4].

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Fig.3.1 Reaction chain of hafnium [5,6].





Natural hafnium is composed of six stable isotopes and its neutron absorption cross section does not decrease markedly after long periods of irradiation because the existence of successive hafnium isotopes with large absorption cross sections permits the neutron absorption by one hafnium atom to result in the formation



Fig.3.3 Effect of iron content on nodular corrosion. (773K, 10.5MPa, 24h, Steam)

of a new hafnium isotope with a significant absorption cross section, as shown in Fig.3.1 [5,6].

From the viewpoint of mechanical design, another advantage of hafnium is gas free characteristic unlike boron carbide which produces helium gas by neutron absorption.

#### 3.2 Corrosion resistance

It is generally known that hafnium has superior corrosion resistance. Fig.3.2 shows the corrosion test results in steam at 773K and 10.5MPa [1]. In this test condition hafnium and zirconium undergo nodular corrosion which is actually suffered in the BWR core. The results shows that hafnium has even better corrosion resistance than Zircaloy-4. And it is also found that addition of small amount of iron improves corrosion resistance of hafnium as shown in Fig.3.3.

Fig.3.4 shows the results of the nodular corrosion test with crevice. It is shown that corrosion resistnce property of hafnium is not sensitive to crevice [1].

Fig.3.5 shows the results of the nodular corrosion test in contact with stainless steel. It is found that contact with stainless steel does not enhance nodular corrosion [1].

Fig.3.6 shows the results of the fretting corrosion test in water at 558K [1]. Plate specimens are contacted with a rotating stainless steel tube by a spring as shown in Fig.3.7. It is shown that hafnium has better fretting corrosion resistance than Zircaloy-4.

#### 3.3 Irradiation characteristics

Dependence on fast neutron fluence of mechanical properties, such as 0.2% yield strength, is shown in Fig.3.8. With increase of exposure, the strength increases and the elongation decreases.

Since hafnium has hexagonal close-packed crystal structure, irradiation growth takes place like zirconium. But the growth rate is negligibly small from the viewpoint of design as shown in Table 3.1.

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Fig.3.4 Effect of crevice on nodular corrosion [1]. (773K, 10.5MPa, Steam)




Fig.3.5 Effect of contact with stainless steel on nodular corrosion [1].

(773K, 10.5MPa, Steam)



Fig.3.6 Fretting corrosion test results [1]. (558K, Dissolved oxygen 8ppm)



Fig.3.7 Fretting corrosion test.





/ Irradiation temperature	:323K	
\ Test temperature	:573K	

Fast neutron fluence (n/m²)	Irradiation growth <sup>*1</sup> (%)	Normalized irradiation growth (%) per 10 <sup>25</sup> n/m <sup>2</sup> *2
1.8 ×10 <sup>25</sup>	0.00	0.00
3.1 ×10 <sup>25</sup>	-0.03	-0.01
1.06×10 <sup>26</sup> [7]	-0.135	-0.01
1.37×10 <sup>26</sup> [7]	-0.27	-0.02

Table 3.1 Irradiation growth of hafnium.

\*1 : Length change of hafnium rods.

\*2 : The value in the left column is devided by fast neutron fluence.

#### 4. Dynamic performance demonstration tests

The dynamic performance was confirmed by the verification tests. The following test data were requested to obtain the license in Japan.

4.1 Control rod drop test

In the event of control rod drop accident, which is deemed to occur very scarecely, the consequence of the accident is affected by the control rod drop velocity.

The purpose of this test is to confirm that the drop velocity of the advanced control rods is within the assumption of safety analysis.



Fig. 4.1 Typical apparatus for control rod drop test [2].

	Drop veloc	Requirement		
	Room temperature	559K <sup>*1</sup>	(m/s)	
Plate type hafnium control rod [1]	0.72	0.85	<u>≤</u> 0.95	
Rod type hafnium control rod	0.73	0.86	<u>≤</u> 0.95	

Table.4.1 Control rod drop test results.

\*1 : The values are derived from the test results at the room temperature in consideration of difference of water density and viscosity.

The test apparatus, illustrated in Fig.4.1, consists of a control rod, four adjacent fuel channel boxes and a control rod guide tube.

The drop test was performed at the room temperature, and the drop velocity at the BWR operating temperature (559K) was evaluated from the test results in consideration of difference of water density and viscosity.

The results in Table 4.1 shows that the drop velocity of the hafnium control rods meets the requirement.

#### 4.2 Scram test

Control rods are required to provide rapid insertion (scram) and safe shutdown so that no fuel damage occurs from any unusual operating transient.

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Fig.4.2 Typical apparatus for scram test [2] .

The purpose of this test is to confirm that the scram insertion time of the advanced control rods meets the requirements.

The test apparatus, illustrated in Fig.4.2, consists of a control rod, four adjacent fuel channel boxes, a control rod guide tube, a control rod drive mechanism and a test vessel.



Fig.4.3 Scram test results of the plate type control rod [1].

(7MPa, 559K)

The scram test was performed at the BWR operating condition, 7MPa and 559K.

The test results in Fig.4.3 and Fig.4.4 shows that the scram insertion time of the hafnium control rods meets the requirement ; within 3.5 seconds at 90 percent of the stroke.



Fig.4.4 Scram test result of the rod type control rod [2]. (7MPa, 559K)

#### 4.3 Scram test in seismic condition

Control rods are required to be inserted under the seismic condition to shutdown the reactor safely.

The purpose of this test is to verify that the advanced control rods can be inserted during an earthquake.

The typical test apparatus, illustrated in Fig.4.5, consists of a control rod, four adjacent fuel boxes, a control rod guide tube, a control rod drive mechanism and a test vessel with a shaking mechanism.



Fig. 4.5 Typical apparatus for scram test in seismic condition [2].



Fig.4.6 Scram test results of the plate type control rod in seismic condition [1]. (Simulated operating condition by adjustment of accumulator pressure)





(Simulated operating condition by adjustment of accumulator pressure)

Though the test was performed at the room temperature, the driving force on the control rod was reduced to simulate scram time at the operating condition.

The test results in Fig.4.6 and Fig.4.7 shows that the advanced control rods can be inserted successfully even under large fuel assembly amplitude.

#### 5. Reactivity control ability

5.1 Shutdown margin

Nuclear reactivity worth of both types of hafnium control rod is designed to be even to that of boron carbide control rod, especially in the top half of the blade. Therefore, the same level of shutdown margin is secured by using hafnium control rods instead of traditional boron carbide ones.

#### 5.2 Scram reactivity

The consequence of operating transient depends on the integrated reactivity from fully withdrawn position to 50% inserted position of the control rod. Since the advanced control rod has the same worth in the upper half as that of a boron carbide one, the consequence of operating transient would not be changed by substituting hafnium control rods for boron carbide ones.

#### 5.3 Influence on the control rod drop accident

The consequence of the control rod drop accident depends largely on the control rod worth, the drop velocity and the scram reactivity.

As for the reactivity insertion by the rod drop accident, the hafnium type one is not greater than boron carbide one. So the

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result of the accident caused by hafnium control rod is not severer.

The rod drop velocity of the hafnium control rod was demonstrated to be well within the supposed value of safety analysis as well as the boron carbide control rod.

The scram reactivity of the hafnium control rod satisfies the safety analysis premise mentioned above.

Therefore, the application of the hafnium cortrol rod does not change the result of control rod drop accident analysis.

#### 6. Experience

Since 1989, two kinds of all hafnium type advanced control rods have been used at the Control Cell locations. Currently in Japan, about one hundred plate type advanced control rods are used in nine BWR plants, and about sixty rod type ones in six BWR plants. Those types of control rods have 3-cycle irradiation experience at maximum.

In addition to the all hafnium type control rod with hafnium and boron carbide as absorber material have been used in Japan. Totally, more than three hundred control rods with hafnium absorber are now used in the Japanese BWR plants, and they have exhibited good performance.

#### 7. Conclusion

Two types of all hafnium control rod were developed in Japan. They are carefully designed taking mechanical, physical, chemical and irradiation properties of hafnium into account, and several demonstration tests showed good dynamic performance.

Irradiation experience of those control rods are now being accumulated in Japanese BWR plants.

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# ABB ATOM CONTROL ROD MATERIALS, EXPERIENCE AND DEVELOPMENT

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Abstract

ABB Atom have since 25 years designed and manufactured control rods for operation in Boiling Water Reactors (BWRs). The basic concept of the control rods comprises boron carbide and hafnium as absorber materials in stainless steel cladding. Mechanical/material limitations have determined the lifetime of the control rods, where Irradiation Assisted Stress Corrosion Cracking (IASCC) is the limiting mechanism.

Development towards a control rod with longer lifetime comprises, among other things, stainless steel cladding less susceptible to irradiation induced sensitization and boron carbide absorber, designed to minimize stresses due to absorber swelling.

Safe and reliable operation of control rods includes, besides high quality control rods, a diligent program of inspections and post irradiation examinations (poolside and hot cell) to determine the control rod operational lifetime and by that an adequate control rod strategy comprising replacement criteria. Modern control rod strategy for safe and reliable operation in Swedish BWRs is described.

## 1 INTRODUCTION

The primary component in the nuclear reactor from the safety point of view is the control rod. To ensure that the reactor can be shut down in a safe way with enough margins, both the quality of the control rods and the operation and replacement strategy is of vital importance.

ABB Atom have since 25 years designed and manufactured control rods for operation in Boiling Water Reactors (BWRs). Most of the rods have been delivered to the 11 BWRs built in Sweden and Finland, from now on referred to as the Nordic BWRs. Between 109 and 169 control rods are contained in the reactor core of these BWRs. In each core about 9 -13 rods are used in so called control cell positions, i.e. they are used to regulate the power during operation. The rest of the rods ( in most cases well above 100) are used in so called shut down positions i.e. they are fully withdrawn from the core during operation. The main task for these control rods are to shut down the reactor at scrams and planned outages. During a period of more than 20 years, control rods in operation have been followed in order to learn the limitations and point out further need of development. These inspection programs, that have been performed in cooperation between the Swedish, the Finnish nuclear power companies and ABB Atom have, besides Post Irradiation Examinations (PIE) of control rod materials in hot cell laboratories, been the basis for inspection-, replacement- and operation criteria. It has also been the basis for work with further development of the control rods towards safer operation and longer life time of control rods.

## 2 EXPERIENCE

# 2.1 All-B<sub>4</sub>C control rods

All the initial cores of the 11 Nordic BWRs had control rods of an all-boron carbide  $(B_4C)$  type called CR70. The design of the control rod is schematically shown in Figure 1. The rod consist of four stainless steel plates mounted to form a cross. In each steel plate about 500 horizontal holes are drilled to contain the absorber material  $(B_4C)$  powder). The cladding material used in these rods was Type 304L stainless steel (SS).

## 2.1.1 Inspection strategy

To study the behaviour and limitations of the control rods, inspection programs were performed. Because of slight differencies in operation, use of control rods, water chemistry etc. it is important that each reactor has its own inspection program to ensure the status in each particular plant.

Earlier in Nordic BWRs very few control rods were used more than one cycle in control cell position. The control rods were moved from these positions during outages and put in shut down position often for the rest of their lifetime. Thus shut down rods were the interesting and important objects to study. The top of shut down control rods, fully withdrawn from the core, is exposed to neutron irradiation during operation especially the handle and the top level of absorber material. The top absorber hole of a shut down rod in a Nordic BWR may be irradiated by upto  $0,6.10^{21}$  thermal neutrons/cm<sup>2</sup> (5-6% <sup>10</sup>B-depletion) each cycle. This means also a fast neutron fluence of about  $0,25 \cdot 10^{21}$  n/cm<sup>2</sup> (>1 MeV) each cycle.

Control rods in shut down position have consequently been found to suffer from cracking in the outer walls of top absorber holes.

## 2.1.2 Post irradiation examination

Two PIEs have been performed in Studsvik hot cell laboratories, where control rods with cracks in the outer walls of top absorber holes have been subjects to examinations. The first rod had been operated most of its life as a shut down rod in Oskarshamn 2 and the calculated <sup>10</sup>B-depletion in the top absorber hole was 53% Ref [1]. Metallographic and fractographic examinations were made in order to determine the cause of the crack failure. These examinations showed that the cracking was intergranular and the conclusion was that irradiation had made the grain boundaries susceptible to corrosion. STEM analysis showed among other things that chromium had segregated from the grain boundaries.



Figure 1 All boron carbide control rod CR70

The second rod had been operated in Forsmark 2 some cycles in control cell position and some cycles as a shut down rod Ref [2]. <sup>10</sup>B-depletion was calculated to 60% in the top absorber hole. Also in this case the cracking was intergranular and the conclusion was that IASCC was the cause of the failure.

# 2.1.3 Consequences of cracking in cladding material

Studies have been performed on ABB Atom rods in order to see if  $B_4C$  is lost due to cracks in the cladding material. Neutron radiography has been used as a technique to determine the status of control rods with cracks Ref [3]. Table I is showing the extent and results of neutron radiographic examinations of CR70 rods. In some cases smaller amounts of  $B_4C$  is lost. The loss was never of any significance regarding shut down margins.

TABLE I	Neutron Radiography of all B <sub>4</sub> C rods	

Reactor		Type Year		No of Blades	Estimated B-10 Tip-depletion %.	*B <sub>4</sub> C Leach-out	
Barsebäck 1		<b>CR70</b>	1985	2	59 - 64	No	
Barsebäck 1		<b>CR70</b>	1986	2	61 - 66	No	
Oskarshamn :	2	<b>CR70</b>	1986	13	61 - 69	2 blades	
Barsebäck 2		<b>CR70</b>	1986	10	68 - 69	4 blades	
Oskarshamn	2	<b>CR70</b>	1987	13	65 - 73	2 blades	

\*Number of blades with minor losses

# 2.1.4 Replacement

Based on inspections, hot cell examination and neutron radiography, operation and replacement strategies are formulated for the BWRs built by ABB Atom. Control rods until today have not reached their nuclear life time and therefore the replacement strategy is focused on the mechanical life time. Thus a typical control rod management in a Nordic BWR is as follows.

- 1 Inspections of lead control rods are continuously performed and inspection limits based on <sup>10</sup>B-depletion values are determined to make sure that rods with cracks will be found as early as necessary.
- 2 If a control rod is found to have a crack/s it is normally replaced. Since examinations have shown that  $B_4C$  is not lost to any significant amounts when a crack occurs, control rods with just one limited crack can be further operated a cycle in shut down position. During this cycle a replacement rod can be provided.
- 3 Rods that have reached the inspection level and still are without defects can be further operated without restrictions, although they shall be inspected on a regular basis.
- 4 Rods that not suffer from cracking have finally to be replaced due to high fast neutron exposure to the handle. This is discussed below in section 2.2.4.

Both control rods in control cell position and shut down rods must be included in this kind of control rod management program although the parameters of interest may vary. For a shut down rod, <sup>10</sup>B-depletion in the tip is the most important value, while it is more adequate to use the <sup>10</sup>B-depletion in the top quarter of a control cell rod as a guideline value.

In the oldest Nordic BWRs which have been in operation for about 20 years, programs that is aimed to in a limited time replace all rods from the initial cores are under progress. The purpose is to replace the rods due to high fast neutron exposures to the handle.

# 2.2 Hafnium tipped control rods

To avoid cracking in the top absorber holes, a hafnium tip design was introduced in the control rods of ABB Atom and this modified rod was called CR82, see Figure 2. This rod is featuring hafnium rodlets in about 20 absorber holes in the tip of the rod. Since hafnium is not swelling due to neutron absorption reactions no stresses are added to the tip of a withdrawn rod during operation. This concept has worked very well and no primary cracks in hafnium tips have occured. Another hafnium tipped rod has 15-20 mm long hafnium plugs at the edges of the control rod wings. This rod is called CR85, see Figure 3. Since the local <sup>10</sup>Bdepletion is higher at the edge of the wing, especially for a control rod in control cell operation, this feature will protect the edge of the wing from cracking. Due to the high density of hafnium it is not possible, because of weight limitations, to have hafnium rodlets throughout the complete rod.

## 2.2.1 Inspection strategy

A similar management program that is used for the CR70 rod is also followed for the hafnium tipped rods. The hafnium tipped rod is not expected to suffer from cracks in the rod tip and they have to a higher degree than CR70 been operated in control cell positions. Accordingly top quarter and average <sup>10</sup>B-depletion values are of most interest when determining inspection criteria.

## 2.2.2 Post irradiation examination

Two PIEs have been performed at Paul Scherrer Institute on hafnium tipped control rods from Oskarshamn 1 Ref [4] and Oskarshamn 2 Ref [5].

The first rod was examined to investigate if hafnium in the top absorber holes had been hydrided by hydrogen diffusing from the reactor water through the stainless steel cladding. The rod had been operated four cycles in the reactor. Examinations were performed by metallography, profilometry and hydrogen gas extraction. It was shown that neither hydriding or swelling had occured. It could be concluded that the amount of hydrogen diffusing through the cladding material during operation is not enough to give significant hydriding of hafnium in the control rod.

The second rod had been operated six cycles in the reactor and in control cell positions for about half of its life time. It had a high top quarter  $^{10}$ B-depletion value. Metallographic and fractographic examinations of four cracks in the rod showed intergranular crack surfaces. It was concluded that the cause of the cracking was IASCC.



Figure 2. Hafnium tipped control rod CR82



Figure 3. Hafnium tipped control rod CR85

Hafnium was studied also during this PIE. This time the hafnium pins had been in contact with reactor water that has intruded through the cracks. Under the crevice conditions inside the control rod cladding, some hafnium pins had been hydrided. Significant swelling of these hafnium pins had occured and caused secondary cracking.

## 2.2.3 Consequences of cracking in cladding material

Neutron radigraphy have been performed on four hafnium tipped control rods with cracks. See Table II. All four of these rods had a high average <sup>10</sup>B-depletion. In no case significant  $B_4C$  losses were found. However it is indicated that the solubility of the  $B_4C$  absorber increases with increasing <sup>10</sup>B-depletion.

# TABLE II Neutron Radiography of hafnium tipped rods

Reactor		Туре	Year	No of Blades	Estimated B-10 depletion %. Top quarter	*B4C Leach-out
Oskarshamn	2	CR85	1991	1	28,8	No
KKL		CR85	1992	2	<b>≈</b> 40	2 blades
TVO 2		CR82	1993	4	37,8	4 blades

\*Number of blades with minor losses

## 2.2.4 Replacement

Control rods with hafnium tips are not expected to have cracks when operated as shut down rods. According to this a question has been brought up to discussion. How long can a withdrawn control rod that will not reach its nuclear life and not reach the mechanical limit, i.e. cracking, be operated before it should be replaced?

A study has been performed where data from operational experiences, hot cell examinations, neutron calculations, material testing of irradiated material etc. have been used as a basis for such an estimation Ref [6]. The study showed that the life limiting part of the control rod would be the handle and limiting operational parameter would be the fast neutron fluence in the handle.

An examination of a control rod handle, of Type 304L SS material, with a fast neutron exposure of about  $9 \cdot 10^{21}$  n/cm<sup>2</sup> (>1MeV) has shown that the fast neutron fluence should not exceed  $10^{22}$  n/cm<sup>2</sup> in the handle, because of safety reasons connected to handling of the rod during outages etc. Experiences have shown that operation and handling of control rods is safe to this level. The time before this level is reached vary depending on where in the core the rod is situated, fuel etc. In the Nordic BWRs shut down rods may reach this level after about 15-25 years, depending on, among other things, the enrichment of  $^{235}$ U in the bottom node.

Thus, it is of vital importance to calculate and follow the fast neutron fluence in the handle of the shut down rod in order to replace the rod at adequate time. Replacement strategy for the hafnium tipped rods, operated considerable time in control cell position, is determined by the levels where cracking is observed to occur.

## 3 MATERIALS

By the inspection programs used it has been discovered that shut down rods fully withdrawn occasionally, during operation have experienced cracking in the walls of the top absorber holes. Cracks in the outer walls of top absorber holes, may appear after about 10-15 cycles in rods operated entirely or most of the time as shut down rods. Post irradiation examinations of such cracks have shown that the crack mechanism is IASCC. In hafnium tipped rods, with high average depletion operated in control cell position, cracks have appeared below the hafnium tip. Also in this case the crack mechanism have been determined to be IASCC.

# 3.1 Irradiation Assisted Stress Corrosion Cracking

IASCC is caused by three factors, which act simultaneously. Fast neutrons (>1 MeV) which sensitize the stainless steel for intergranular cracking, swelling of boron carbide due to the neutron absorption reactions is causing stresses in the cladding material. An oxidizing environment is also necessary for IASCC to occur. The reactor water and especially the core environment is highly oxidizing. Thus, IASCC can be initiated and then propagate in material in contact with reactor water. Later experiences are indicating that an oxidizing environment may not be necessary at higher exposures of fast neutrons

It is today not fully understood what sensitization mechanism/s that is most important. From studies of grain boundaries in irradiated Type 304 SS material we know that fast neutrons (>1MeV) cause segregation phenomenas to occur in the stainless steel such as enrichment of sulphur, phosphorus and silicon in the grain boundaries and simultaneous chromium depletion in the grain boundaries. The latter is known to be detrimental to stainless steel from a related cracking called Intergranular Stress Corrosion Cracking (IGSCC). Depletion of chromium makes the grain boundaries less resistant to corrosion and the crack propagates intergranular. Since anodic dissolution in the crack tip is one part of the mechanism, stress corrosion is supported by oxidizing environment. IGSCC is mitigated or the rate is lowered by shifting to reducing environment. In the same way, IASCC seems to be mitigated in reducing environment, at least upto about  $4 \cdot 10^{21}$  n/cm<sub>2</sub> (>1 MeV). Therefore chromium depletion is strongly believed to play an important role regarding IASCC. When the fast neutron fluence is above about  $4 \cdot 10^{21}$  $n/cm^2$  (>1 MeV), another phenomena besides chromium depletion seems to contribute to the cracking of the material. This latter phenomena is today not identified.

Irradiation tests of stainless steel materials, especially Type 304 SS and Type 316 SS, have been performed in a Swedish BWR Ref [7]. The irradiation have been performed in the core of the plant. After irradiation the specimens have been tested by the so called Constant Elongation Rate Tensile (CERT) technique. The test environment has been water from the recirculation system in the reactor. The results have shown that Type 316 SS is much more resistant to IASCC than Type 304 SS. In figure 4



IGSCC of Type 304 SS in oxidizing environment





Figure 4 IASCC susceptibility Type 304 SS vs Type 316 SS

behaviour of the two stainless steel types are compared. In the diagrams, the y-axis is showing the part of the crack surface where intergranular stress corrosion cracking has occured, in these cases IASCC.

ABB Atom has in the last years replaced Type 304L SS as cladding material in the control rods by Type 316L SS. In the study mentioned in section 2.2.4, it is concluded that fast neutron fluence can be allowed twice as high for Type 316 SS than for Type 304 SS, based on the in-reactor irradiation study mentioned above. This means that a shut down control rod may be used upto 30-40 years. Such rods shall however be followed by inspections regularly. By change of the cladding material, control cell rods, can be operated much closer to nuclear life, typically 40-50 % average <sup>10</sup>B-depletion.

# 3.2 Absorber material

One important factor limiting the life time of a control rod with  $B_4C$  as absorber is swelling of  $B_4C$ , due to neutron absorption reactions. When  $B_4C$  is used as powder in drilled holes or tubes the density of  $B_4C$ , will at the highest be around 70%. Thus 30% of the volume is void. Our understanding is that this void volume is not completely used during swelling of the absorber material. The swelling of  $B_4C$  strains the cladding material and causes stresses. If the void volume could be quantitatively used the stresses should appear later in the control rod life.

# 4 DEVELOPMENT

Reduction of stresses in especially environmental affected material is one remedy against cracking. The hole pattern in the ABB Atom design is now optimized by finite element analysis. By changing hole diameter and distances between holes it has been possible to reduce stresses, due to  $B_4C$ swelling, in the environmental affected surface. See figure 5. Instead, stresses are increased in the wall between the absorber holes, which are not affected by reactor water. Control rods with this feature is now operated in several BWRs. Rods with this new hole pattern will be followed by inspection programs for verification.

A possibility to take advantage of the void volume in order to minimize the negative effects of  $B_4C$  swelling, is to press the  $B_4C$  into pellets of almost theoretical density. This is achieved by the Hot Isostatic Pressing (HIP) technique. Then the void is placed between the absorber material and the cladding.

The use of  $B_4C$  pellets of almost theoretical density is currently under verification in control rods in Oskarshamn 2.



Figure 5 Stress reduction by optimizing hole pattern

# 5 SUMMARY

For safe operation of an nuclear power plant the status of the control rod inventory should be known by using regular inspections. These inspections must be included in a control rod management program that also covers strategies for replacement of control rods according to achieved limits as nuclear life, mechanical life e.g. cracks in the cladding and limits in the structure due to high exposure of fast neutrons (>1MeV).

To completely understand the IASCC phenomena in stainless steels, more efforts are needed. Especially in the field of theoretical studies rather than generating more data. This is not only of importance for the cladding but also for other structural stainless steel parts of control rods and of cause for other internal parts of the core.

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# HIGH TEMPERATURE BORON CARBIDE INTERACTION WITH STAINLESS STEEL IN CONTROL RODS

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#### Abstract

At temperatures > 600°C boron carbide interacts with steel to form low melting ironchromium-borides. In the case of boron carbide filled stainless steel LWR control rods, the reaction is important when looking at severe accidents with core overheating and dry out. The present PSI confirmatory research work was performed for the Swiss Nuclear Inspectorate (HSK). It shows that the B<sub>4</sub>C steel interaction is negligible < 600°C. At temperatures  $\geq$  1000° C on the other hand the reaction proceeds to severe steel degradation and to steel liquefaction even below 1200°C. This process might lead to a recriticality situation in a power plant following a core damage event. The PSI reaction data closely match with B<sub>4</sub>C-steel interaction values from literature.

## 1. INTRODUCTION

Boron carbide is well known for its swelling under high neutron fluence. This swelling can lead to high stress fields within the stainless steel absorber matrix of LWR control rods. Obviously such rods can subsequently be damaged by stress corrosion cracking. [1, 2]. As long as the ambient temperature, however, lies in the range of LWR operation temperature, the interaction of  $B_4C$  in powdered, chipped-, compacted- or pelletized form remains in the mechanical wall interaction, and boron carbide filled absorber rod inner surfaces may indicate some indents. Chemical surface interaction between boron and the steel constituents should not start below 600°C to 700°C [3]. At higher temperature, this reaction becomes progressively important and according to the B, Fe binary alloy system diagram [4] it may lead to the complete steel liquefaction at the eutectic temperature of about 1200°C. Such interaction has to be checked, when looking into severe light water reactor accidents [5] or at lower temperature when dealing with absorber materials for FBR's [6].

In connection with public hearings before issuing an unlimited operating licence for one of the Swiss NPP's, the nuclear inspectorate (HSK) asked PSI for a literature survey and confirmatory research to support literature data about melt down and relocation of control blades during a severe accident with intact core geometry, which could lead to recriticality in a BWR when flooded after overheating and dryout.

To demonstrate the boron steel chemical interaction at high temperature, several experimental concepts, including Na bonding [6], and high applied contact pressures [7], which both seem to enhance the reaction kinetics, as well as compressed  $B_4C$  powder in steel cylinders [5] are described in literature. The experimental conditions selected at PSI were chosen to match as closely as possible the reaction couple geometry in a true ABB-Atom control rod.[8].

B <sub>4</sub>	С	AIS	I 304 SS
Composition:		Composition (wt%)	
$B_4C(wt\%))$	99.1	C	0.002
$B_{2}O_{3}$ (wt%)	0.03	Si	0.08
free boron (wt%)	0.15	Mn	0.75
Fe (wt%)	0.076	P	0.006
Ca (wt%)	0.032	S	0.003
Ti (wt%)	0.016	Cr	18.32
Co and Cu (ppm)	< 5	Ni	10.54
Mg and Na (ppm)	< 20	Co	0.007
powder size (µm)	~ 50 - 100	Ν	0.01
		В	0.0005

# Table I: Chemical Composition of Reaction Couples

# 2. EXPERIMENTAL PROCEDURES

Coarse boron carbide powder and AISI 304 SS in form of an 8 mm steel plate were procured from ABB Atom<sup>1</sup>. The specifications of both products are given in Table I. The 8 mm thick plate was cut into 2 mm thick sheets by electroerosion. Sandwiches with an intermediate boron carbide layer of 2 mm thickness were then fabricated and sealed by electron beam welding. The reaction couples were subsequently isothermally annealed at 1000°C/24h, 1100° C/24h and 1200°C/1.5h respectively using a 25 kW resistance vacuum heating system. At 1200°C the exposure time had to be cut off after 1.5 h due to local melt down of the samples. The furnace temperature control was performed with regulating Pt/PtRh thermocouples localised close to the samples.

After heat treatment the samples were metallographically characterized. For electrolytical (6V) grain boundary etching, the samples were exposed during 30 seconds to oxalic acid (10%). The reaction products were also characterized by x-ray diffraction analysis using  $CuK_{\alpha}$  radiation.

## 3. METALLOGRAPHIC AND X-RAY DIFFRACTION RESULTS

The metallographic analysis clearly demonstrates the chemical interaction of  $B_4C$  with steel. Figure 1 shows the typical texture of the unreacted nondisturbed steel. Figures 2-3 are presenting the reacted specimen texture at the  $B_4C$ /steel interface after heating to 1000°C and 1100°C respectively. Below the reacted surface layer, grain boundary decoration with second phase precipitation in the steel matrix indicates that the reaction is initiated at these locations with highest mobility of the reactants. If the isothermal temperature is increased for 1.5h to 1200°C, nearly the whole steel sample is liquefied and reprecipitates in an eutectic texture with large voids as shown with a bulk phase presentation of such a specimen in Figure 4.

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The reaction products were qualitatively assessed by x-ray diffraction (Figures 5 and 6). The steel/B<sub>4</sub>C interface for this analysis was oriented perpendicular to the incident Cu K<sub> $\alpha$ </sub> beam. The x-ray penetration depth for this experiment is assumed to lie around 0-200  $\mu$ m. At 1000°C and 1100°C respectively the XRD results indicate Chromium-Iron-Borides and remnants of the stainless steel matrix. The relative matrix signal intensity is diminishing when the isothermal annealing temperature is increased from 1000°C to 1100°C. The XRD pattern of the 1200°C reacted sample does not reveal any more metal borides. Obviously the sample has significantly relocated after liquefaction and the metal boride phase is lost within the background of remaining stainless steel reflexes and the newly formed  $\gamma$ (FeNi) phase. Within all samples assessed by XRD no indication of metal carbides was seen.

#### 4. **DISCUSSION**

It has clearly been shown that boron carbide is strongly reacting with stainless steel at temperatures  $\geq 1000^{\circ}$ C. At 1200°C the annealing experiment had to be stopped after 1.5h due to eutectic melting. Hence the melting temperatures of stainless steel (1399°C) and of B<sub>4</sub>C (2350°C) are drastically reduced. The etched metallographic slides clearly indicate that the B<sub>4</sub>C/steel interaction is proceeding along grain boundaries. By variation of the reaction time at constant temperature Hofmann [5] shows that the grain boundary reaction layer thickness growth is a linear function of the square root of time and he is using this indication assuming a grain boundary diffusion process for boron. When measuring the averaged reaction zone



Fig. 1: Undisturbed microstructure of "as received" stainless steel (100:1)



Fig. 2: Microstructure of steel sample at the  $B_4C$ /steel interface after heating for 24h at 1000°C (etched, 100:1)



Fig. 3: Microstructure of steel sample at the  $B_4C$ /steel interface after heating for 24h at 1100°C (etched, 100:1)



b)



Fig. 4: Eutectic Microstructure of reacted steel sample after heating for 1.5h at 1200°C with  $B_4C$  (etched, a) 100:1, b) 200:1)



Peak	2THETA		D	INTEG.I %	Peak	2THETA		D	INTEG.I %
1	32.126	0	2.7837	8.4	10	50.931	+	1.7914	41.5
2	34.961		2.5643	2.7	11	56.736	0	1.6211	16.8
3	37.508		2.3957	11.6	12	63.210	0	1.4698	12.2
4	38.403	0	2.3419	2.6	13	74.966	+	1.2658	37.3
5	41.008		2.1990	2.6	14	76.248	0	1.2476	14.4
6	43.759	+	2.0669	100.0	15	82.152	0	1.1723	12.7
7	44.907	0	2.0167	70.1	16	91.028	+	1.0797	83.6
8	46.307	0	1.9589	7.6	17	96.357	+	1.0336	5.8
9	46.882		1.9362	15.1					

+ (Austenitic) Stainless Steel (JC-PDF 33-397)

o Chromium Boride (JC-PDF 32-277)

Fig. 5: XRD pattern of steel surface layer reacted for 24 h at 1000°C



- Iron Boride (JC-PDF 32463)

Fig. 6: XRD pattern of steel surface layer reacted for 24 h at 1100°C



Fig. 7: Reaction zone growth rates in the B<sub>4</sub>C/stainless steel reaction couple for 800°C to 1200°C [5]



Fig. 8: Boronation reaction depth in stainless steel in function of reaction time and temperature
depth for the grain boundary boration at 1000°C and 1100°C from Figures 2 and 3 and applying Fick's diffusion equation

	with
$D = Do \exp(-Q/RT)$ or	X = grain boundary diffusion depth (cm)
$X^2 = 2 Dt = 2 Do exp (-Q/RT) \cdot t$	D = Diffusion coefficient
	Do = preexponential factor
	Q = apparent activation energy
	T = reaction temperature (K)
	t = reaction time (sec)
	$r = gas constant = 8.314 J/mol \cdot K$

the PSI results lead to a diffusion coefficient of

 $D = 1.2E4 \exp(-3.0E5/RT)$ 

with a preexponential factor Do = 1.2E4 cm<sup>2</sup>/s and an activation energy of 300 kJ/mol.

The PSI values have been added in Hofmann's plot of reaction zone growth vs reciprocal temperature (Figure 7). If these values are used to extrapolate a boronation zone depth after  $B_4C$ /stainless steel interaction for 1 year at 600°C, a negligible reaction depth of 7  $\mu$  is calculated. If on the other hand the diffusion coefficient is used to calculate the reaction zone depth after accidental overheating times in the temperature range of 1000-1200°C (Figure 8) it becomes obvious that serious control blade boronation and an eventual liquefaction and loss of the absorber rod is possible.

In none of the samples annealed at 1000-1200°C and analyzed by XRD any evidence was seen for the presence of carbides in the reaction zone. Although the carbon diffusion coefficient is higher compared to that of boron, carbides are less stable than borides and once formed, borides even act as carbon diffusion barrier since carbon is not soluble within the metal borides [6]. The reason for the occurrence of the  $\gamma$ (Fe,Ni) phase at 1200°C probably lies in the fact that Cr is significantly lost from the austenite for the formation of chronium borides.

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#### ABSORBER MATERIALS IN CANDU PHWRs

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#### Abstract

In a CANDU reactor the fuel channels are arranged on a square lattice in a calandria filled with heavy water moderator. This arrangement allows five types of tubular neutron absorber devices to be located in the relatively benign environment of low pressure, low temperature heavy water between neighbouring rows or columns of fuel channels.

This paper will describe the roles of the devices and outline the design requirements of the absorber component from a reactor physics viewpoint. Nuclear heating and activation problems associated with the different absorbers will be briefly discussed. The design and manufacture of the devices will be also discussed.

The control rod absorbers and shut off materials are cadmium and stainless steel. In the tubular arrangement, the cadmium is sandwiched between stainless steel tubes. This type of device has functioned well, but there is now concern over the availability and expense of cadmium which is used in two types of CANDU control devices. There are also concerns about the toxicity of cadmium during the fabrication of the absorbers. These concerns are prompting AECL to study alternatives. To minimize design changes, pure boron-10 alloyed in stainless steel is a favoured option. Work is underway to confirm the suitability of the boron-loaded steel and identify other encapsulated absorber materials for practical application.

Because the reactivity devices or their guide tubes span the calandria vessel, the long slender components must be sufficiently rigid to resist operational vibration and also be seismically stable. Some of these components are made of Zircaloy to minimize neutron absorption. Slow irradiation growth and creep can reduce the spring tension, and periodic adjustments to the springs are required. Experience with the control absorber devices has generally been good. In one instance liquid zone controllers had a problem of vibration induced fretting but a redesigned back-fit resolved the problem.

#### 1. INTRODUCTION

The CANDU reactor has two major circuits circulating heavy water through the reactor. The primary heat transport water, operating at temperatures between 260°C and 312°C, at pressures up to 10.7 MPa and a pH of 10.0 to 10.5, circulates through the pressure tubes and transports heat from the fuel located inside the pressure tubes, to the steam generators. The other circuit is the moderator heavy water circulating at 65°C to 80°C at a low pressure and neutral pH, into the calandria vessel and through the matrix of calandria tubes which isolate the hot pressure tubes from the moderator, and then into the heat exchangers where the required temperature of the moderator heavy water is maintained. The moderator heavy water contained in the calandria is the environment in which are operated the reactivity mechanisms of the CANDU reactor.

The reactivity control devices in a CANDU reactor consist of reactivity control devices, i.e. adjusters, control absorber, liquid zone control units and poison addition system, and two shutdown units. These are backed by neutron flux measuring devices (flux detectors and ion chamber units). The reactivity control devices span the calandria vessel, vertically and horizontally; those with active mechanical elements operate vertically to take advantage of gravity. The devices operate either from a platform above the reactor or from their support at the shield tank wall (see Figure 1).

The control devices are tubular components, designed to be simple and rugged and highly reliable. All components except the stainless steel thimbles which protect the out of core sections of the devices, can be readily removed and replaced.

Six systems are available for introducing absorber material into the reactor core, and these can be classified as mechanical or non-mechanical:

(i) Mechanical Devices

Shutoff rods (shutdown system #1) consist of thin cadmium elements sandwiched and sealed between stainless steel tubes.

Control absorbers are of similar design to the shutdown rods but with slightly thicker cadmium elements.

The adjusters consist of stainless steel tubes or cobalt pins that are used to axially flatten the flux and are operated inserted into the core.

(ii) Non Mechanical

The liquid zone controllers are the main reactivity control devices which operate by adjusting the neutron absorbing quantities of light water contained in compartments in Zircaloy tubes, to control the flux in different zones of the reactor.

The liquid poison injection system, which injects a solution of gadolinium nitrate through horizontal Zircaloy tubes into the moderator forms the second shutdown system. The gadolinium is extracted by circulating the moderator through ion exchange columns.



Figure 1 An Illustration of a CANDU Reactor Showing the Locations of the Vertical and Horizontal Reactivity Control Devices

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Table	1
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### Reactivity Devices Used in Various CANDU Reactors

	Liquid Zone Controllers	Telescoping Mechanical Zone Controllers	Mechanical Control Absorbers	Mechanical Adjusters	Mechanical Shutoff Rods	Liquid Poison Injection	Boosters	Moderator Dump
Pickering A (1-4)	6	-	0	18	21	0	-	1
Pickering B (5-8)	6	•	4	21	28	б	-	•
Bruce A (1-4)	6	-	4	0	30	7	16	-
Bruce B (5-8)	6	-	4	24	32	8	-	-
CANDU 6's (12)	6		4	21	28	6	-	-
Darlington (1-4)	6	-	4	24	32	8	-	-
CANDU 3	0	4	4	12	24	8	-	-
CANDU 9	6	-	4	24	32	8	-	•

The poison addition system maintains a low boron concentration in the moderator water. The concentration of boron can be reduced when reactivity is low. It is injected into the circulating moderator water, and removed by the purification system. Table 1 details the various reactivity control devices or systems used in the various groups of reactors.

#### 2. PHYSICS OF ABSORBER MATERIALS IN CANDU

From a physics viewpoint there are four properties which have been considered in the selection of the absorber materials used in CANDU reactors. These are as follows: -

#### 2.1 Absorption Cross-Section

The thermal neutron flux in a CANDU reactor is typically  $2.0 \times 10^{18}$  n/m<sup>2</sup>.s at the mid-lattice position in the moderator. At this flux level for an irradiation time of one year, there will be significant burnout of any isotope which has an absorption cross-section greater than 10 barns. There can also be significant transmutation of one element into another over the life of some of the absorber components.

Burnout and transmutation would eliminate the use of strong absorber elements such as boron, cadmium, hafnium or gadolinium for components continuously in-core, such as the adjusters or the mechanical zone control assemblies under development. These devices are relatively weak absorbers and do not provide a large local flux depression. The absorbers can be exposed to high fluxes for relatively long periods of time, and although a weak absorber device with a low concentration of a strong absorber element could be fabricated for this service, the individual atoms would have a high probability of destruction. The destruction of absorber atoms would reduce the macroscopic absorption cross-section of the material with time. There is a 15% drop in the absorption cross-section of type 304L stainless steel over thirty years even though the component element cross-section is less than 10 barns. For hafnium, the drop would be about 70%.

#### 2.2 Activation Products

Cobalt-60 isotope is one example of an activation product which has a long half-life and decays with the release of high gamma energy. The gamma fields can create a radiation hazard which must be considered during flasking operations or during decommissioning activities.

The transmutation process creates new sources of cobalt in stainless steel. Typically the cobalt would be present at low concentrations of about 500 ppm as impurity, but the transmutation of iron-58 forms iron-59, and the subsequent beta decay of iron-59 forms cobalt-59. Although the natural abundance of iron-58 is relatively low the process of transmutation promotes the formation of iron-58 from the lower mass isotopes of iron. After 20 years of exposure to a flux of  $1.0 \times 10^{18} \text{ n/m}^2$ .s the cobalt-60 activity is type 304L stainless steel with no initial cobalt impurity would be indistinguishable from that in a steel with an initial impurity of 200 ppm.

#### 2.3 Nuclear Heating

Energy is released as a result of the absorption process or during the decay of activation products, which then dissipate as heat. This heat may create the potential to melt

the absorber material, e.g., cadmium in the parked shutoff rods which are located in a relatively high neutron flux outside the calandria. The decay energy, in the form of heat, must be allowed for in the flasking of devices for removal.

Nuclear heating has the potential to raise the temperature of shutoff rods to a level at which the auto-ignition of deuterium in the moderator cover gas could occur. The temperature of each component is a function of the nuclear energy deposited in the component and the transfer of heat from the component. The energy released following absorption in boron-10 is carried by two recoil particles namely, a lithium-7 nucleus and an alpha particle. All the energy will be deposited essentially at the point of release. Energy released following absorption in cadmium is by high energy gamma radiation which have a high probability of escaping from a thin walled cylinder. Although the energy released from boron is low the heating problems may be as severe as those with cadmium.

#### 2.4 Nuclear Damage

Some of the nuclear absorption process can form protons as recoil particles. These particles have a short range and they can be stopped by the absorber material. Thus the absorption process can introduce hydrogen and helium into the structure of the absorber. For the fluxes and absorbers used in CANDU reactors this form of nuclear damage is considered unimportant (18 ppm hydrogen and 66 ppm helium after 30 years).

The neutron damage from the fast flux irradiation of the absorbers has also been unimportant because the mechanical duties of the absorber elements are relatively light.

- 3. DESIGN
- 3.1 Description of Reactivity Control Devices
- 3.1.1 Mechanical Control Absorbers (MCA)

The mechanical control absorbers (Figure 2) are under the control of the reactor regulating system (RRS). Should the liquid zone controllers be incapable of responding quickly enough to a demand to reduce power, then the MCAs would be driven in. The MCAs also allow the addition of negative reactivity should high levels of light water develop in the liquid zone controllers.

The absorber elements for the mechanical control absorbers are thin walled cylinders of cadmium sandwiched between type 304L stainless steel tubes (Figure 3). The internal diameter of the absorber element is 114.3 mm. The wall thickness of cadmium is 0.91 mm and each steel tube is 0.76 mm thick. The cadmium thickness was chosen to provide ten mean-free-paths of thermal neutron absorption, so that the absorber material will be black even after intermittent irradiation in the core at high power. The internal diameter was chosen to present the largest surface area to thermal neutrons between the calandria tubes in adjacent columns of fuel channels.

At the top (or trailing end) of each absorber rod there is a support rod to which a cable connecting the absorber to the drive mechanism is attached. The support rod is attached to the absorber rod by means of a stainless steel spider located approximately at the mid-span of the rod. The cadmium sandwiched tubes are welded to the top and bottom surface of the spider rim as shown in Figure 3.



Figure 2 Mechanical Control Absorber Unit



Figure 3 Absorber Elements Formed by Sandwiching Cadmium Between Type 304L Stainless Steel Tubes

The shutdown system 1 shut-off rods use essentially the same design of absorber element as in the MCAs. During normal operation they are parked just outside the calandria. In the event of a trip signal, the shutoff rods are dropped into the reactor core under gravity when the electromagnetic clutches in the shutoff drive mechanisms are de-energized. An accelerator spring, which is compressed by the absorber element when it is withdrawn from the core, imparts an initial acceleration to the elements to reduce insertion time. To withdraw the elements, the drive mechanism clutches are energized and the elements are driven up by motor and parked outside the calandria. Poised in this position, they are instantly available for further use. A magnet in the element support rod, is used to activate the magnetic reed switches in a device called a Rod Ready Indicator. This ready signal, together with the electric output from the drive mechanism potentiometer, indicates the absorber element is availabile for use. A typical shutoff device is shown in Figure 4.

The rods are fully inserted within 2 seconds, and begin to reduce the thermal neutron fluxes in the core several hundred milliseconds after clutch release. Their rapid insertion and consequent reactor shutdown mean the shut-off rods accumulate exposures at the rate of 2 full power seconds per insertion.

The dynamic worth of the shutoff rods, (a measure of the negative reactivity added) is typically -91 mk.

#### 3.1.3 Adjuster Assemblies

The adjuster assemblies are the third absorber system under the control of the RRS. Physically the adjusters are light absorbers and have a large surface area. The mass per unit length was dictated by the need for axial flattening of the neutron flux profile, (see below) and a xenon override capability. The absorber element is either a thin-walled tube and a central shim rod both made of type 304L stainless steel, or a string of bundles of zircaloy-clad cobalt-pencils (Figure 5). The outer diameter of the steel tube is 76.2 mm with a wall thickness of 0.5 to 2.0 mm. In the cobalt adjusters there are 2 to 6 pins of cobalt in a circular arrangement of 51.7 mm pitch; each pencil contains 52.8 grams of cobalt.

The adjuster assemblies are normally fully inserted in the reactor core. They are arranged across three vertical planes along the reactor axis with 7 or 8 adjusters in each plane. In this arrangement the adjuster assemblies provide axial flattening of the fuel bundle powers in a fuel channel. Consequently with the adjusters, the power extracted from each fuel channel can be increased over a channel that has no axial flattening.

The adjusters also provide a xenon override capability. By removal of the adjusters, the operator can correct a fault which prompted the trip and then return the reactor to full power without incurring a poison-out. Typically there is a period of thirty minutes in which the operator can correct a fault and return to power before the growth of xenon-135 adds more negative reactivity than the withdrawal of adjusters can offset.

#### 3.1.4 Liquid Zone Control Units (LZCU)

The liquid zone control system uses light water as absorber in a compartment formed with Zircaloy 2 walls. The rest of the compartment is filled with a helium cover-gas.



Figure 4 Mechanical Shut-Off Rod



Figure 5 Adjuster Unit Assembly



Figure 6 Two Zone Liquid Zone Control Unit

The level of light water in each compartment is altered by changing the water flow to a compartment thereby allowing the compartment to drain or fill (Figure 6).

Typically there are 14 compartments arranged in six LZCU-assemblies. The compartments are distributed throughout the central regions of the reactor core. Each compartment "controls" the flux level in that part of the reactor core. The core is divided into fourteen zones and the power of each zone can, to a degree, be controlled independently of the power in the other thirteen zones.

The liquid zone control system is the primary control system of reactor power. It is under the control of the RRS. The RRS uses the measurements of the local flux from the in-core flux detectors to regulate the flow of light water to each compartment. A demand by the RRS to reduce reactor power globally would prompt an increase in light water levels. A demand to raise power would prompt a drop in light water levels. Ideally the compartments should be 40% to 60% filled to allow addition or removal of reactivity from any part of the core.

For the CANDU 3 reactor under development the liquid zone control units will be replaced by a system of telescoping stainless steel mechanical zone controllers.

#### 3.1.5 Liquid Injection Shutdown System 2 (SDS2)

The second shutdown system is made up of six liquid injection nozzles. These are roughly 50 mm internal diameter Zircaloy 2 tubes and supported horizontally in the calandria. These tubes, which are normally full of moderator heavy water, contain hundreds of small diameter holes which distributes the gadolinium nitrate solution in the calandria.

One end of each tube is connected to a pressurized tank of gadolinium nitrate solution. When a trip signal is received, the gadolinium nitrate is flushed into the calandria through the small holes.

The system is capable of adding -200 mk to -300 mk of negative reactivity. Typically the system adds -55 mk in the first minute and -300 mk over the first hour. The gadolinium is removed from the moderator water by the ion exchange resins of the moderator purification system.

The choice of gadolinium was driven by its large absorption cross section and high solubility in water. A boron salt has been used in some designs. There have been no identified problems with this system. However, one concern is the tendency of the gadolinium to precipitate out of solution when the pD of the moderator heavy water is too high.

#### 3.1.6 Liquid Poison Addition

One of the moderator auxiliary systems is a liquid poison addition system. The system allows the addition of a small amount of negative reactivity to the moderator and consequently globally to the reactor core. The addition of the boron (or gadolinium) is made as a small flow to the suction of the main moderator pumps. A small concentration (0.5 ppm) of boron in the moderator adds -3 to -4 mk of reactivity to the core.

The negative reactivity may be used to control excess reactivity in new fuel (referred to as poison shim). Operating the core with a low concentration of boron in the moderator allows greater flexibility in fuelling operations. (A refuelling operation could be delayed by reducing the boron content in the moderator to zero through a purification system, thus maintaining core reactivity.) The reduction of boron is more attractive than removing a bank of adjusters because there are no changes to the relative flux distribution in the core.

The negative reactivity would also compensate for the loss of reactivity after a poison out or a long shutdown when the xenon-135 decays. Finally the system provides enough poison to the moderator to prevent criticality during a shutdown and guarantee the shutdown.

#### 3.2 Manufacturing

The fabrication process employed in the fabrication of the cadmium sandwich absorber and shutoff rods basically consists of the following steps:

- a. A 99.9 percent pure cadmium sheet is wrapped around the inner cladding tube with the joint between the sheet ends running along the axis of the tube. The cladding tube inside diameter is slightly larger than the required final size.
- b. The inner cladding tube complete with the cadmium sheet is inserted into the outer cladding tube which again is oversized to facilitate assembly.
- c. The cladding tubes are crimped at one end to accommodate the attachment of a pulling mechanism and the cladding tube/cadmium assembly is drawn down to size by pulling it through dies.
- d. The assembly is then trimmed to proper length and the ends sealed by welding and swaging prior to attachment by welding to the support rod.

The cladding tubes are made of type 304L austenitic stainless steel to ASTM A269 or ASTM A270. Initially this material is in a fully annealed condition to facilitate draw down. Fabrication development was required to ensure adequate location of the cadmium with the specified joint gap and internal void.

The finished rods are extensively tested for weld and cladding quality for structural integrity to prevent deterioration of the cadmium which could occur with the ingress of heavy water.

The fabrication of the stainless steel adjuster elements involves a three stage process. First, the type 304L stainless steel is purchased to dimensions calculated by the core physicist based on the nominal chemistry in the material specifications. Second, the actual chemical analysis of the material is obtained and is used to determine the reactivity worth of the element. Third, the centre rod or shim rod (Figure 5) is then sized to fine tune the reactivity worth.

The guide tubes for the shutoff rods, control absorbers and adjusters, which run vertically through the calandria (see Figure 7 and 8) are seam welded tubes made from perforated Zircaloy-2 sheet, (to minimize neutron absorption and preclude voids in the

moderator, while ensuring proper cooling of the in-core devices). These tubes are spring loaded to limit vibration in the circulating moderator water.

The other tubes for the reactivity devices are mostly made from extruded and annealed zurcaloy or stainless steel materials.



Figure 7 Vertical Shutoff Guide Tube Penetrating the Reactor Core Between Rows of Calandria Tubes



Figure 8 View Looking Horizontally Between Rows of Calandria Tubes Showing the Variety of Vertical Penetrations

#### 4. OPERATING EXPERIENCE

The operating experience of the CANDU reactivity devices has been very good. Only a limited number of problems have occurred, predictably related to vibration. In the early prototype reactor Douglas Point, a threaded joint in a flow tube of one booster unit failed permitting the flow tube to vibrate against a calandria tube, eventually perforating the calandria tube. In the same reactor after decommissioning, it was observed that very slight vibration over a 23 year period of operation, caused a booster guide tube to fret a groove and perforate the tube wall at the position where the tube was guided by a support ring.

In one Bruce A unit, the vibration of the liquid zone control unit centring guide (for the internal light water and helium tubes) against the inside wall of the enclosing tube, caused perforation of the tube and necessitated its replacement. This problem is unique to the Bruce A reactors where the moderator heavy water enters the calandria through the booster guide tubes and is discharged into the vessel through nozzles located near the top of the vessel. Vibration of the liquid zone control unit was caused by impingement of the moderator flow from these nozzles onto the wall of the liquid zone control unit.

The Zircaloy-2 guide tubes operate with an axial load applied to them to provide seismic and vibrational stability, by increasing the natural frequency of vibration above that expected in service. The guide tubes are exposed to high neutron fluence during service. We would expect these tubes to elongate slightly from irradiation creep and growth, and the elongation would reduce the spring tension load on the tube. Periodic adjustments of the tension can be done from adjustment nuts at the reactivity mechanism platform. However the elongation rate has been low and few adjustments have been needed.

#### 5. CONCERNS

The most important concern relates to the use of cadmium. The shutoff rods and absorbers have performed well but concerns have arisen from the viewpoint of (a) expense of the cadmium and restrictions on its use and (b) nuclear heating of the bottom of the shutoff rods in the parked position.

Cadmium is receiving attention as a hazardous material and possibly faces restrictions on its usage. A problem of effective disposal or neutralization of the waste from the refining processes used to produce it, adds another concern for the economics of its use in the future. Thus a substitute material is being sought as a back up. The favoured option is type 304L stainless steel alloyed with pure boron- $10^{(1)(2)(3)}$ . A boron-10 concentration of about 2% would be required for control absorbers and shutoff rods. The relatively easy incorporation of boron-10 stainless tubes into the existing design favours the material selection.

Nuclear heating of the cadmium in the parked shutoff rods has previously been a concern but the satisfactory performance has not indicated any need for replacement. The concern relates to the conditions where the moderator level gets too low and the additional neutron flux seen by the bottom of the parked rod produces extra heat. This could lead to the potential melting of the cadmium or the possibility that the rods could reach a temperature which could ignite deuterium gas that could occasionally build up to higher concentrations in the helium atmosphere as a result of radiolysis of the moderator heavy water. While there is no evidence that such conditions have even been approached the use of borated stainless steel may eliminate this concern if the steel is capable of disseminating the heat generated by the boron more effectively.

#### 6. CONCLUSION

The relatively benign environment in which the reactivity devices of the CANDU reactor operate has contributed to a reliable performance record for these devices. The potential future substitution of borated stainless steel for cadmium is under consideration from economic concerns but such substitution will possibly alleviate some potential operating concerns.

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