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Homogeneous Aqueous Solution Nuclear Reactors for the Production of Mo-99 and other Short Lived Radioistotopes

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FOREWORD

Technetium-99m (^{99m}Tc), the daughter of Molybdemum-99 (^{99}Mo), is the most commonly used medical radioisotope in the world. It accounts for over twenty-five million medical procedures each year worldwide, comprising about 80% of all radiopharmaceutical procedures.

^{99}Mo is mostly prepared by the fission of uranium-235 targets in a nuclear reactor with a fission yield of about 6.1%. Currently over 95% of the fission product ^{99}Mo is obtained using highly enriched uranium (HEU) targets. Smaller scale producers use low enriched uranium (LEU) targets. Small quantities of ^{99}Mo are also produced by neutron activation through the use of the (n, γ) reaction.

The concept of a compact homogeneous aqueous reactor fuelled by a uranium salt solution with off-line separation of radioisotopes of interest (^{99}Mo, ^{131}I) from aliquots of irradiated fuel solution has been cited in a few presentations in the series of International Conference on Isotopes (ICI) held in Vancouver (2000), Cape Town (2003) and Brussels (2005) and recently some corporate interest has also been noticeable.

Calculations and some experimental research have shown that the use of aqueous homogeneous reactors (AHRs) could be an efficient technology for fission radioisotope production, having some prospective advantages compared with traditional technology based on the use of solid uranium targets irradiated in research reactors.

This review of AHR status and prospects by a team of experts engaged in the field of homogeneous reactors and radioisotope producers yields an objective evaluation of the technological challenges and other relevant implications. The meeting to develop this report facilitated the exchange of information on the ‘state of the art’ of the technology related to homogeneous aqueous solution nuclear reactors, especially in connection with the production of radioisotopes.

This publication presents a summary of discussions of a consultants meeting which is followed by the technical presentations given by the participants during the meeting. The IAEA thanks all the experts for their contributions to the consultants meeting and for the presentations of their work and detailed discussions based on their experiences which have resulted in the manuscript for this publication.

The IAEA officer responsible for this publication was E. Bradley of the Division of Nuclear Fuel Cycle and Waste Technology. This work also incorporates the collaborative support of P. Adelfang, of the Division of Nuclear Fuel Cycle and Waste Technology and N. Ramamoorthy of the Division of Physical and Chemical Sciences.

EDITORIAL NOTE

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SUMMARY

1. INTRODUCTION

Technetium-99m (^{99m}Tc) is the most commonly used radioisotope in nuclear medicine. It is produced from the decay of its parent ^{99}Mo. Although ^{99}Mo can be produced via different technologies, the primary method for a high specific activity product is irradiation of uranium targets. Uranium-235 in the targets in nuclear reactors undergoes fission reactions yielding ^{99}Mo among other fission products. The use of aqueous homogeneous reactors (AHRs) or solution reactors presents an attractive alternative to the conventional target irradiation method of producing ^{99}Mo in that solutions eliminate the need for targets and can operate at much lower power than required for a reactor irradiating targets to produce the same amount of ^{99}Mo.

As the name implies, solution reactors consist of an enriched uranium salt dissolved in water and acid and contained in shielded tank or vessel. The first solution reactors earned the name 'water boilers' because of the observed bubbling or frothing that results from the radiolytic decomposition of water by fission fragments and subsequent evolution of radiolytic gases (hydrogen and oxygen). Approximately 30 solution reactors have been built world wide and operated over many years since the beginning of modern nuclear programmes in the 1940s and 1950s. Most of these reactors are no longer in service. However, in the 1990s a renewed interest in solution reactors for the production of medical isotopes has prompted several countries, including China, the Russian Federation and the United States of America to initiate programmes to assess the feasibility of utilizing AHR technology for medical isotope production applications on commercial basis.

2. ADVANTAGES OF SOLUTION REACTORS FOR THE PRODUCTION OF MEDICAL ISOTOPES

The use of solution reactors for the production of medical isotopes is potentially advantageous because of their low cost, small critical mass, inherent passive safety, and simplified fuel handling, processing and purification characteristics. These advantages stem partly from the fluid nature of the fuel and partly from the homogeneous mixture of the fuel and moderator in that an aqueous homogeneous reactor combines the attributes of liquid fuel homogeneous reactors with those of water moderated heterogeneous reactors. If practical methods for handling a radioactive aqueous fuel system are implemented, the inherent simplicity of this type of reactor should result in considerable economic gains in the production of medical isotopes. The principal advantages of aqueous fuel systems include:

- Flexibility with respect to reactor parameter variation, fuel selection, and geometry;
- Inherent nuclear safety characteristics;
- Efficient neutron utilization for isotope production;
- Elimination of targets, less uranium waste generated per curie of ^{99}Mo produced, and overall simpler waste management;
- Ability to process other isotopes such as ^{133}Xe, ^{89}Sr, ^{90}Y, ^{131}I more efficiently using off-gas extraction;
- Less capital cost and potential lower operating costs.

2.1. Reactor design flexibility

The flexibility of solution reactor design parameters is an important feature of the AHR concept that allows customized design configurations to meet required isotope production capacity while using proven design and the existing operating practice for the main components and safety systems. These design flexibilities include:

- The thermal power of the rector which can be varied in the range of about 50–300 kW(th).
- Selection of fuel base (UO_2SO_4, $UO_2(NO_3)_2$, etc.), chemical composition (uranium loading, solution pH, etc).
- Thermodynamic operating state (low pressure, temperature) allows flexibility in selection of reactor core geometry to accommodate inherent safety characteristics and to meet specific project safety objectives and requirements.

The greater flexibility afforded by solution reactors with respect to core operating power range is an important advantage with respect to ^{99}Mo production demand. However, power instabilities that result from radiolytic bubble formation and thermal agitation can perturb the surface of the fuel solution as power is raised to the desired levels. This in turn generates reactivity variations that impair automatic reactor control via the measurement channels that enable control rod displacement. As a result the power density becomes an important design parameter which must be controlled to ensure stable operation within the desired or selected power envelope. The use of low enriched uranium (LEU) fuel, which is recommended by the IAEA for the AHR applications, requires a greater volume of fuel and thus results in an increase in core solution height which potentially diminishes the reactivity variations induced by perturbation of the solution surface. Additionally, a non-cylindrical core tank design would probably attenuate the instability phenomena, thus further enhancing safety.

The choice of fuel base and solution composition is contingent on core design, operation and product isotope processing strategy. Traditionally, uranyl sulphate fuel was preferred over uranyl nitrate because of its greater radiation stability. However, irradiated uranyl nitrate solutions have a higher distribution coefficient for ^{99}Mo extraction than irradiated uranyl sulphate solutions; consequently a nitrate fuel base is clearly more advantageous from a processing yield point of view. The fuel concentration is selected to minimize core volume/fissile mass, optimize processing efficiency, or both. The solution pH is controlled within a range that ensures good uranyl salt solubility and at the same time minimizes corrosion effects.

Solution reactors are typically operated at 80°C and slightly below atmospheric pressure. The low operating fuel solution temperature, power density and pressure provides thermodynamic stability and minimizes potential safety risks, yet allow for sufficient flexibility to optimize ^{99}Mo production demands.

2.2. Inherent nuclear safety characteristics

The inherent nuclear safety characteristics of solution reactors are associated with the large negative density coefficient of reactivity in such systems. In essence, during the core operation fission energy is deposited in the fuel solution causing it to expand due to the increase in fuel temperature and the formation of gas bubbles resulting from radiolysis. The formation of radiolytic gas bubbles, along with the increased solution temperature, results in fuel density reduction (volumetric expansion) thereby affecting neutron leakage. Although the negative reactivity addition due to fuel solution temperature increase is relatively uniform, the radiolytic gas generation rate, and therefore the density reduction, is not uniform but is a function of the core power profiles and thus is spatially dependent. During a typical reactivity transient, the fuel solution is displaced from the region of highest power (near the core centre) slightly faster than at the edges thus creating mixing currents. As a result, the reactivity effect resulting from the operation of solution reactors at power may be thought of as the superposition of two effects, namely: (1) an overall uniform volumetric expansion of the fuel solution due to the increase in fuel temperature and the formation of gas bubbles due to radiolysis; and (2) a corresponding density redistribution within the expanding volume in which the introduction of an equivalent void volume displaces fuel from regions of higher reactivity worth to regions of lower reactivity worth. The resulting density reduction is manifested in a large negative coefficient of reactivity which provides a self-limiting mechanism to terminate a reactivity excursion and provides inherent nuclear safety. Relevant experiments in the French CRAC and SILENE facilities have demonstrated these phenomena.

2.3. Efficient neutron utilization for isotope production

In the majority of reactors producing medical isotopes, targets are irradiated in a neutron flux generated primarily by the reactor fuel. Typically, the ratio of reactor power to target power is of the order of 100 to 1, and thus represents a cost in uranium consumption and waste production which must be added to that of the targets. Alternatively, a unique feature of using the solution reactor for medical isotope production is that the reactor fuel and target are the same. Consequently a solution reactor can produce the same amount of ^{99}Mo at 1/100th the power consumption and radioactive waste generation. Thus the potential advantage of utilizing solution reactor technology are lower reactor power, less waste heat, and a reduction by a factor of about 100 in the generation of spent fuel when compared with ^{99}Mo production by target irradiation in heterogeneous reactors.

2.4. Elimination of targets, less uranium generated per curie of ^{99}Mo produced, and overall simpler radioactive waste management

When one considers waste management in terms of both spent reactor fuel and spent target disposition, waste management for the solution reactor is far simpler. A solution reactor has no need for targets and, therefore all processes related to the fabrication, irradiation, disassembly and dissolution of targets are eliminated. Because these target related processes result in the generation of both chemical and radioactive wastes, ^{99}Mo production in solution reactors can significantly reduce waste generation. Since the recovery and purification of ^{99}Mo from conventional targets after dissolution will be quite similar (if not identical) to that of a solution reactor, the solid and liquid wastes produced will be similar, except for the disposition of the resultant uranium. For conventional target-based technologies, uranium from the irradiated targets is stored for disposal or recycling; solution reactor uranium is recycled and only disposed of at the end of the fuel solution's viability (up to twenty years).

2.5. Ability to process other isotopes more efficiently using off-gas extraction

In addition to ^{99}Mo, other radioisotopes used by the medical community can be processed more efficiently from a solution reactor. In particular, the intensive radiolytic boiling which occurs during core operation enhances the off-gassing of volatile fission products from the fuel solution into the upper gas plenum of the reactor. A number of valuable radioisotopes such as ^{89}Sr, ^{90}Y, ^{133}Xe, ^{131}I, ^{132}I, and ^{133}I can be recovered from the off-gas. Higher specific activity of these isotopes in the off-gas makes recovery much more effective compared to traditional uranium target irradiation technology. This is a far more effective way to recover fission ^{89}Sr (about 1000 times) compared to production of this isotope from ^{88}Sr and ^{89}Y target irradiation.

2.6. Less capital cost and potential lower operating costs

Significant isotope production capacity could be developed using one or more small, low power medical isotope production reactors (MIPRs) in the 50–200 kW thermal power range. Each reactor is a simple 100–200 L shielded vessel that operates at near atmospheric pressure and 80°C. The core cooling, gas management and control systems along with their logic control and auxiliary equipment will be relatively small and simple compared to research reactor target systems. Isotope separation, purification and packaging systems should be very similar to target system facilities. The relatively smaller, less complex solution reactor will be less costly to design and construct than much higher power, research reactors used for isotope production.

Potential savings in operating costs compared to current research reactor/target systems can be projected in several areas:

- Since the fuel in a solution reactor is also the target, no separate fabricated target is necessary. While the fuel/target solution may require periodic additions of LEU, it is anticipated that the base load of solution will be useable over a significant period, potentially up to the life of the unit. Enriched uranium targets as fabricated for use in a traditional reactor are expensive consumables.
- Current practice involves transport of the irradiated target from the reactor to a processing facility for dissolution and product recovery. This operation typically involves an intermediate cooling step, transfer equipment and an acid or base process to dissolve the target for product recovery. These steps require personnel and equipment that will not be necessary for a solution reactor system in which fuel solution can be pumped to the initial isotope separation device.
- Reactor control and operation is expected to be simpler than higher powered research reactors which may result in decreased reactor operations staff.
- In a reactor/target facility, only a small percentage of the target is transformed to useful isotopes leaving over 99% of the target material which must be handled as waste or processed to recover uranium. With a solution reactor system, target waste handling, disposition activities and costs are not necessary.

3. SOLUTION REACTOR DESIGN CHALLENGES FOR THE PRODUCTION OF MEDICAL ISOTOPES

Although aqueous homogenous solution reactor technology is well characterized in the research environment, the capability of a solution reactor to perform a medical isotope production mission in a long term continuous steady state mode of operation in the 50–300 kW range is not guaranteed. Specifically, many technical challenges must be addressed in transitioning the technology from research to a commercial industrial environment including:

1. Development of an efficient isotope separation technology that is commensurate with an irradiated fuel solution:
    a. Fuel base selection and optimization of the reactor fuel solution chemical state parameters;
    b. Impact of fission product buildup on product isotope yield.

2. Increased unit power beyond operating experience database:
    a. Potential instabilities may limit power density;
    b. Design may have to compensate for fuel burnup by adding LEU while maintaining pre-established solution uranium concentration and chemical state;
    c. Management of fission and radiolytic gases formed in the reactor core;
    d. Corrosion effects on core material resulting from long term core operation;
    e. Radiological concerns resulting from operation and processing of a fluid (unclad) fuel solution.
3. Licensing of solution reactors for isotope production
    a. First of its kind to be licensed for medical isotope production - No specific regulations exist.

3.1. Isotope separation technology development challenges

One of the key challenges in utilizing solution reactor technology for the production of medical isotopes is the development of an efficient extraction methodology for separating the product isotope from the irradiated fuel solution. Specifically, the effects of radiation and fission product buildup on the separation of ^{99}Mo by an adsorbent agent must be determined. Any new undesired complex chemical species in the product isotope and/or chemical reactions with the solution being processed must be identified. Furthermore, characteristics of the fuel purification systems required to compensate for potential ^{99}Mo processing additives need to be identified and analysed to quantify their effect on reactor operation.

Two fuel solutions are currently being considered for solution reactors dedicated to radioisotope production, namely, uranyl sulphate [UO_2SO_4] and uranyl nitrate [$UO_2(NO_3)_2$]. Both have advantages and disadvantages. Uranyl sulphate's main advantage is that only H_2 and O_2 are formed by radiolytic degradation of the fuel solution. These gases can be recombined to water using a heated catalyst bed, and the condensed water is then returned to the fuel solution. Its disadvantages are related to the chemistry of sulphate and its salts, specifically:

- Recovery of molybdenum from a sulphate solution is not as efficient as from a nitrate solution by anion exchange due to more effective competition to sorption of $(MoO_4)^{2-}$ by the $(SO_4)^{2-}$ and $(HSO_4)^{-}$ than by $(NO_3)^{-}$.
- Solubilities of sulphate salts are generally far less than nitrate salts, and as the fuel ages, the buildup of fission and adsorption products may become high enough to approach solubility limits. Fission products of special concern are Ba, Sr, and rare earth elements.

Uranyl nitrate solutions have superior chemical properties relative to uranyl-sulphate solutions; however radiolytic decomposition of an aqueous uranyl nitrate solution is far more complex than that of the sulphate salt. In addition to production of H_2 and O_2 from water radiolysis, nitrogen and nitrogen oxide (NO_X) gases are also formed from the decomposition of the fuel solution. These additional gases have two detrimental effects to reactor operation, namely:

(1) The off-gas system must be far more complex than that for the sulphate based reactor. These gases must be scrubbed from the off-gas system early. If not, they can foul the catalysis

bed, potentially cause fires in off-gas activated charcoal beds and compromise collection of iodine in the off-gas.

(2) Because the NO_x gases are acidic, there release will raise the pH of the fuel solution. Controlling pH of the solution will require significantly higher acid addition than for the sulphate based reactor and, likely, at far shorter intervals. Fuel solution pH (nominally 1) cannot be allowed to rise above pH 3; if it does, precipitation of uranium and many fission products will begin.

Because both salts have good and bad properties, a decision on which is superior requires additional experimental data and detailed system design. If the NO_x off-gas concerns are easily addressed, nitrate would be the salt of choice. If not, sulphate should be used.

With respect to ^{99}Mo recovery/extraction from the fuel solution, a number of issues must be considered in developing a viable process for use with a solution reactor. Firstly, the selected uranium concentration in the fuel solution is a compromise between reactor design optimization and ^{99}Mo separation efficiency. In essence, a lower uranium salt concentration in the fuel solution results in a larger K_d for Mo(VI) and therefore a more effective and efficient recovery of ^{99}Mo. As a result, the size of the recovery column can be smaller making washing of impurities more effective, obtaining a more concentrated product solution of the raw molybdenum from the column. However, a higher concentration of uranium in the solution will minimize the reactor fuel solution volume leading to a more compact reactor. Optimization of these two opposing factors is a challenging design study.

The pH of the fuel solution must also be considered. Specifically, the lower pH, avoids the hydrolysis of the uranium and fission products metals which could lead to precipitation of hydrous metal oxides; however, if the pH is too low, the ion exchange resin is the less effective. A pH of 1 is a good compromise. The pH must also be optimized to keep corrosion of the reactor vessel and ancillary structural materials to a minimum. The concentration of the uranium salt also effects solution density, and therefore the radiolytic gas bubble formation and dispersion.

As discussed previously, another concern is the chemical stability of the fuel solution induced by a high radiation environment without introducing new undesired complex chemical structures in the product isotope and/or chemical reactions with the solution being processed. Furthermore, the potential problems caused by the buildup of adsorption and fission products, their effect on reactor operation and the subsequent recovery system is another challenge which must be addressed. In addition, the effects of corrosion product buildup, materials leached from the recovery system and chemical additions must also be analysed and optimized. If the fission product buildup and/or corrosion product effects are important, a means to clean up the fuel solution in concert with waste-management and economic considerations must be devised.

3.2. Increased unit power beyond solution reactor operating experience database

Historically, solution reactors have been used either in a research capacity to: (1) study nuclear kinetics phenomena associated with nuclear excursions; (2) as a neutron generator to study the effects of irradiation on materials; or (3) to generate radioisotopes. As a result, most reactor operations were transient in nature, or limited with respect to steady state operation.

When operated at steady state, stable core operation could be sustained as long as the maximum average power density did not exceed approximately 1.8 kW/L.[1] Physically, the radiolysis gas and vapour that form at high power densities create bubbles that migrate to the surface of the solution. The resulting perturbations at the liquid surface may cause reactivity variations, as well as waves and sloshing effects due to reflection off the reactor vessel walls making it difficult for the automatic rod control system to maintain steady state power conditions. These phenomena are closely related to power density and need to be examined carefully to avoid potential power instabilities or uncontrolled power transients due to a supercritical configuration of the solution in the core tank. The design of the core tank may also need to be reconsidered. These instabilities, while detrimental to predictable production operations, pose a relatively small risk provided the reactor vessel design can accommodate potential pressure transients due to liquid perturbations.

At the present time, the only large scale experiment on the use of a solution reactor in steady state operation stems from the Russian ARGUS reactor, which operates at a maximum power density of 1 kW/L of solution. The French SILENE reactor is designed to operate in steady state mode at about 0.3 kW/L, but tests performed in the past at 10 times higher levels were unsuccessful due to power instabilities. Since the objective of future AHRs is to operate at power densities up to 2–2.5 kW/L, tests are being considered to verify that this power density can be achieved.

The overall design of a LEU based solution reactor for the production of medical isotopes must accommodate a methodology to ensure that sufficient LEU fuel, and therefore excess reactivity, can be maintained to compensate for inherent negative reactivity effects, fuel burnup effects (i.e. fuel depletion and fission product buildup) and must ensure reactor criticality over the operational cycle without changing the fuel concentration or chemical state. The ^{235}U content may have to be replenished to maintain production capacity by adding LEU uranyl salt solution without altering optimized fuel solution parameters (concentration, chemical state, etc). Typically, this requires a slight volumetric increase in fuel solution per refuelling cycle. As a result, the control system design must be flexible enough to compensate for the required excess reactivity and volumetric adjustments, and must be properly allocated among several different control mechanisms to ensure flexible operational control over the operating cycle within the constraints imposed by safe reactor operation while at the same time possess sufficient passive and/or active engineering features so that the safety functions can be achieved in all operational states and accident conditions.

Another challenge associated with long-term continuous steady-state mode of a solution reactor operating the range of 50–300 kW is radiolytic gas recombination management and solution pH control. As discussed earlier, uranyl sulphate is stable in an irradiation environment, thus non-volatile; however uranyl nitrate is unstable in an irradiation field and decomposes to N_2, O_2 and NO_x. The hydrogen and oxygen resulting from water radiolysis can be recombined to water and returned to the core to maintain solution balance, such as pH and uranium concentration. Unfortunately, N_2, O_2, and NO_x can not be recombined to nitric acid and returned to the reactor fuel solution, so if not removed from the system, the gas pressure will increase over time and the acidity of the fuel solution will be decreased. If the acidity of

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[1] 1.8 kW/L is based on operating experience of two 50 kW reactor, one at Walter Reed in the early 1960s and one at Armour Research Foundation at the Illinois Institute of Technology over the same time frame. Experiments from SUPO and KEWB indicated that operation at core power densities below 2 kW/L may be required to ensure core thermal stability.

the fuel solution is permitted to decrease below 0.001 mol/L, precipitation of $UO_2(OH)_2$ will occur, potentially resulting in excess corrosion. It is therefore necessary to discharge the additional noble gases from the reactor gas loop and to concurrently makeup the lost nitric acid. If this method is adopted to maintain the reactor chemistry, an online radioactive gaseous waste processing capability will be needed, adding to the cost of the AHR and increasing the potential for a system leak. Consequently, an online method to recombine the N_2, O_2, and NO_x gaseous products to a nitrate and therefore allow retention of the radiolytic gases in a closed system configuration will need to be developed if uranyl nitrate is to be used as the fuel base.

One of the traditional challenges associated with fluid fuel reactors is that the presence of fission radiation increases the rate of corrosion of exposed metal surfaces which in turn affects permissible wall power density, and therefore impacts the allowable average power density within the reactor. Radiation may profoundly alter the chemistry of the fluids through the formation and decomposition of various chemical species, especially when uranyl nitrate is used. Similarly, the corrosion and physical behaviour of materials may be changed by radiation damage and transmutation. Materials of construction must be assessed as to their chemical and nuclear compatibility with long term continuous operation in a corrosive, irradiation environment.

Finally, radiological safety of the AHR reactor and the isotope recovery system is of paramount importance and thus is ensured by the design and layout of equipment, proper organizational measures as well as a provision for multibarrier radioactive product retention systems, including pressure reversal. Safety analysis has shown that for all realistically conceivable initial events and accident sequences, radioactive products are maintained within design limits. As a result no radiological impact to the surrounding population and operations personnel is expected for design basis accidents. Nevertheless postulated beyond design basis accidents with essential loss of fuel solution may pose local radiological concerns for repair and maintenance personnel in the recovery system compartments.

3.3. Licensing of solution reactors for isotope production

Since no operating license applications involving solution reactor facilities for isotope production have been submitted, world wide nuclear regulatory bodies have not developed regulations that address solution reactors for commercial isotope production. Two solution reactors, however, have been licensed in the USA (by the Atomic Energy Commission) but these were not commercial isotope production facilities. Current US Nuclear Regulatory Commission regulations address power reactors, 'commercial' reactors and research reactors. Hazard analyses for solution reactors have indicated significantly lower hazard to workers, surrounding populations and the environment than those reactors currently addressed by regulatory bodies. New regulations appropriately addressing specific hazards associated with solution reactors for commercial isotope production will be necessary. Until these regulations are formulated and issued, it is feasible to address these facilities in a manner similar to current research reactor standards.

4. STATUS OF SOLUTION REACTORS FOR MEDICAL ISOTOPE PRODUCTION

MIPRs are under development in China, the Russian Federation and the USA. Two fundamental technologies have been patented in the Europe, the Russian Federation and the USA. These are solution reactors using LEU solutions of (a) uranyl nitrate salt and (b) uranyl sulphate salt as the fuel. The ARGUS reactor, a 20 kW(th), high enriched uranium (HEU) solution reactor has been operated as an experimental development activity by the Kurchatov Institute in the Russian Federation. Irradiated solution from this unit was processed to separate and purify ^{99}Mo to European and US pharmacopeia standards. It should be noted that minimum pharmacopeia purity requirements may not be sufficient for specific formulations used in the eventual medical imaging procedure.

Fundamental research on hydrated metal oxide sorbents continues both at Argonne National Laboratory in the USA, and at the Kurchatov Institute and Ural Technological University in the Russian Federation. Four sorbents have been considered for molybdenum recovery: alumina (the classic inorganic sorbent for molybdenum recovery from acidic solutions), PZC (polyzirconium compound, Kaken Co., Ltd., Hori, Mito-shi 310-0903 Japan developed for replacing alumina in ^{99m}Tc generators for low specific activity ^{99}Mo), and two sorbents specifically designed by Thermoxid (Thermoxid Scientific and Production Company, Zarechnyi, Russian Federation) for recovering ^{99}Mo from homogeneous reactor fuel solutions.

Alumina would only be usable for very dilute uranyl nitrate fuel solutions and is far less effective than the other sorbents in all situations. The Thermoxid sorbents have been tested extensively by the Kurchatov Institute, Ural Technological University, and Argonne National Laboratory. In this regard, a great deal of data for the Thermoxid sorbents has been collected using tracers in simulated solutions, however limited data has been collected at the Kurchatov Institute using ARGUS reactor fuel. Preliminary results indicate that the Thermoxid would be effective for both nitrate and sulphate based fuel solutions, even at high concentrations of uranyl salts. They are more effective in nitrate media than sulphate. However, to get data required for design of the recovery system and to minimize technical risk, far more data are required, including sorption behaviour of molybdenum and of other species under realistic conditions and how this behaviour changes as the fuel solution ages. PZC is almost as effective in nitrate media as are the Thermoxid sorbents for molybdenum recovery but is not suitable for sulphate media. A significant disadvantage of PZC for use in a solution reactor system is that it contains chloride, which is a known corrosion agent for stainless steel.

Another important effect that has not been fully characterized is the effect on molybdenum redox chemistry by high radiation fields that will accompany fuel cooled for only a few hours. Because recovery is based on maintaining molybdenum in the (VI) oxidation state, its reduction to lower oxidation states would diminish both its sorption in the loading phase and its stripping from the column in alkaline solution, where the lower oxidation state modybdenum species precipitate in the column as hydrous metal oxides. Limited studies have shown that four hours after irradiation, effects are seen by lowering of ^{99}Mo distribution ratios, especially in sulphate media. Much more experimental work is required to understand and design for this effect.

In summary, the current technology level is well established within the performed research tests. However, the next step is to confirm that this new technology can be used in a day to day reliable production environment. Active participation by both pharmaceutical and commercial nuclear reactor industries will be necessary in order to successfully develop viable commercial applications of this technology. While the advantages are numerous, commercial markets must be involved in the establishment of an evolving technology in place of an existing well developed alternative.

5. CONCLUSIONS AND RECOMMENDATIONS

The meeting participants agreed on the following:

5.1. Assistance

(a) Support to share research information on solution reactors and MIPRs could benefit the further development and eventual implementation of this technology. The participants of the June 2007 consultancy meeting held in Vienna could assist by developing a recommended list of participants as well as exploring funding options depending on the final scope of the project (realizing that coordination meetings to share information are, for example, considerably less expensive than funding research activities). Areas of research and collaboration include:

1. Fuel base characteristics and material properties;
2. Thermohydraulics;
3. Separation and purification;
4. Choice of unit size for various production capacities.

(b) Participants noted limited guidance within national nuclear regulatory programmes regarding the licensing of AHR facilities utilized for the production of medical isotopes. Assistance on this topic could help avoid any licensing challenges.

5.2. Recommendations for organizations within interested member states

(a) Conduct research and technology development activities based on documented technical challenges associated with solution reactor technology. Areas of potential research include:

1. Development of computer codes related to reactor physics and thermohydraulics;
2. Thermohydraulic analyses of the fuel solution, such as density, heat conductivity, etc.;
3. Criticality control;
4. Gas circulation;
5. Material corrosion;
6. Radio-chemistry properties of the fuel solution, etc;
7. Design criteria;
8. Conceptual and final AHR designs.

(c) Conduct research and technology development activities on isotope separation technology:

1. Laboratory experiments;
2. Experiment on the full scale test facility;
3. Experiment with ‘real’ fuel solution fission;
4. Research interface technology coupling solution reactor and separation process.

(d) Perform other research activities:

1. Commercial utilization;
2. Economic analysis on the base of power scale;
3. Market estimate.

(e) LEU should be considered for all solution reactors for medical isotope production. The international community through the GTRI (global threat reduction initiative) and the many national RERTR (reduced enrichment for research and test reactor) programmes have come together to minimize, and to the extent possible, eliminate the use of HEU in civil nuclear programs throughout the world. There is considerable evidence that aqueous solution reactors can be effective using LEU fuel (<20% ^{235}U). It is for the benefit of all that when a solution reactor is proposed for radioisotope production, every effort be made to avoid the use of HEU fuel.

(f) Construct, commission and operate a prototypic solution reactor and recovery system for commercial medical isotope production, incorporating all available information including outcomes of research activities listed above.

(g) Develop an approach to ensure positive business case for commercial entities. The supply of ^{99}Mo to the pharmaceutical industry is a mature business with supply channels and product pricing long in place. Payments to the supplier for products needing ^{99}Mo are managed by various reimbursement agencies, such as the US Medicare/Medicaid programme. Cost of conversion to LEU technology will be difficult to recover, and commercial entities must maintain a positive business case for such burdens. Industry conversion will be greatly facilitated by the ability to recover these costs by some means, such as price increases or subsidies from State governments.

(h) Attempt to coordinate regulatory and pharmaceutical agencies early in the design process. In all planned applications, two regulatory bodies will be involved in review and approval for ultimate AHR operation. In the USA these will be the Nuclear Regulatory Commission on the reactor portion and the Food and Drug Administration with respect to isotope separation, purification and packaging portion, with similar division of oversight in other countries. Coordination of communications, application data and timing with the respective agencies should be attempted to minimize the potential for conflicting requirements where regulatory oversight overlaps. Early communications with regulatory bodies to identify potential areas of overlap is encouraged.

(i) Assess domestic and world demand for solution reactor products and services. It is very important to verify the world demand for each radioisotope, dividing it by regions (viz. Europe, Africa, etc.). This would indicate the required amount of the production facilities per region. However, domestic demand and production capability for the final pharmaceutical product (e.g. ^{99m}Tc generators) within these global regions should be mapped in order to locate future isotope production plants.

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PART II — PAPERS BY THE PARTICIPANTS OF THE CONSULTANTS MEETING

OPTIMIZATION OF 200 kW MEDICAL ISOTOPE PRODUCTION REACTOR DESIGN

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Abstract. One of the primary methods of producing medical isotopes such as ^{99}Mo and ^{131}I is by irradiating uranium targets in heterogeneous reactors. Homogeneous aqueous reactors present a potential alternative to medical isotope production. In response to the global demand for medical isotopes, a concept design of the 200 kW medical isotope production reactor (MIPR) was accomplished by Nuclear Power Institute of China in 2000. Further R&D work was completed in subsequent years, including the optimization of design, reactor thermohydraulic experiments, gas circulation system experiments, etc. Compared with the normal isotope production method such as target irradiation, the MIPR can produce more types of isotope at lower cost and with less radioactive waste generation.

1. INTRODUCTION

The medical isotope production reactor (MIPR) is designed to produce medical isotopes such as ^{99}Mo and ^{131}I. Currently, the principal method of producing medical isotopes is through the irradiation of targets electroplated with enriched uranium in heterogeneous reactors. After irradiation, these targets are transferred to hot cell facilities where the uranium coating in the targets is dissolved, primarily with nitric acid. The dissolved uranium is passed through an alumina column, where the molybdenum is absorbed. The alumina is then washed in multiple steps. The ^{99}Mo is eventually isolated and purified. This traditional method of separation has is high cost, delivers a small yield and results in significant radwaste generation. Instead, the MIPR uses a $UO_2(NO_3)_2$ solution as its fuel. Large amounts of isotopes (for example ^{99}Mo) are generated directly from the fission of the fuel solution, avoiding the need to fabricate a target. Because of its inherent safety, simple production techniques, less radwaste, lower investment and operational costs, the MIPR is a competitive method of producing isotopes.

The MIPR developed by National Power Institute of China (NPIC) comprises two parts (Fig. 1). One part is the reactor system which uses $UO_2(NO_3)_2$ as its fuel. The other part is the radioactive isotope production system that uses special technology developed independently by NPIC. The homogeneous aqueous reactor was developed during the 1940s and has proven to be very safe through 60 years of operating experience worldwide [1–5]. The MIPR uses the $UO_2(NO_3)_2$ solution as its fuel which can be recycled after isotope extraction. The reactor system includes the fuel solution transfer, heat exchanger, gas circulation, I&C and radioactive waste treatment systems. The isotope production system is housed within two hot cells and comprises five production lines.

2. RULES AND SCOPE OF THE MIPR DESIGN

2.1. Design rules

The MIPR is a water boiler research reactor, the design and construction of which will follow the nuclear safety laws and guides of the IAEA and the Chinese nuclear safety authority. The special safety provision and the design criteria dedicated for the MIPR also need to be investigated.

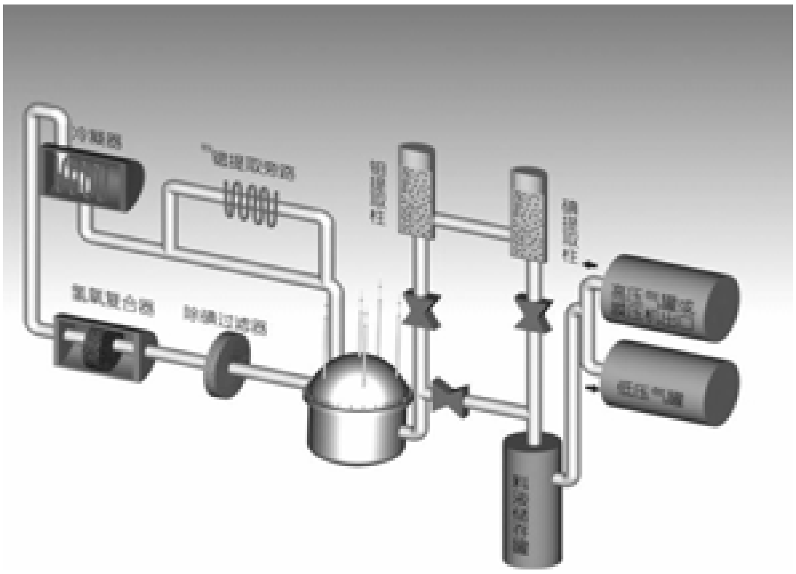

FIG. 1. Sketch of the MIPR.

2.2. Scope of the MIPR

The MIPR is composed of two reactors and their relevant systems, excluding the isotope production system. The reactor power is about 200kW. This project aims mostly at producing the medical isotopes ^{99}Mo and ^{131}I. The main design parameters of the reactor system are given in the Table 1.

Table 1. Main parameters of reactor system.

Item	Parameter/unit
Type of the reactor	Water boiler solution reactor
Fuel	Solution of $UO_2(NO_3)_2$
Enrichment of ^{235}U	90%
Thermal power	200 kW
Primary coolant inlet temperature	~20°C
Primary coolant outlet temperature	~40°C
Operation pressure	0.09 MPa
Average temperature of the fuel solution	<70°C
Power density	2~2.5 kW/L
Original concentration of U	50 gU/L
Concentration of HNO_3	0.1~0.3 mol/L
Inner diameter of the reactor vessel	~700mm
Height of the reactor vessel	~730mm
Absorb material of the control rod	B_4C

2.3. Features of the MIPR

Characteristics of the MIPR

The main characteristics of the MIPR are as follows:

a. It has a large negative temperature coefficient of reactivity which contributes to inherent self-adjustment and reliability;
b. It produces the isotopes while avoiding the need to fabricate fuel, irradiation targets or the subsequent disposal of either;
c. The radioactive waste is contained in the fuel solution, minimizing environmental risk.

Operation and production mode of the MIPR

With the double reactors and their related systems, each of the reactors will run for 100 cycles every year, each cycle includes 24 h power operation and 48 h shutdown. After 1 h shutdown, the fuel solution will be transferred from reactor vessel to the storage pot by the gas pressure difference between the two tanks. During the transfer, the fuel solution will flow through the extraction bed where the isotopes in the fuel solution are adsorbed. After rinsing process, the final product yield is 500Ci ^{99}Mo, and 100Ci ^{131}I. The total annual total products are 100 000Ci ^{99}Mo and 20 000Ci ^{131}I.

3. REACTOR SYSTEM

3.1. General description

The reactor system is mainly composed of a reactor vessel, heat exchanger, gas circulation system, fuel solution transfer system and I&C system. The reactor vessel is a cylinder with a dish bottom. The inner diameter is 700mm and the height is 730mm. The vessel is penetrated by 3 control rod guiding tubes from the upper head through to the bottom head. Some other facilities are also assembled in the vessel, such as the water separator and the heat exchange tube bundle. The general configuration is showed in the Figure 2.

3.2. Reactor vessel

The vessels containing fuel solution are set in the water wells. Four water wells are located in one water pool (two wells for the reactor vessels, the others for the storage pots). Concrete wells of the water are clad with stainless steel. The reactor vessel is a cylinder with a dish bottom and dish top. The $UO_2(NO_3)_2$ fuel solution is located in the lower part and N_2 gas in the upper space. The inlet and outlet of the gas tube are joined to the middle nozzle on the top head of the reactor vessel. The inlet and outlet of the primary cooling water pipes are joined to the heat exchanger tube bundle through the nozzles on the cylinder vessel top. The vessel is penetrated by 3 control rod guiding tubes from the top through to the bottom. In the bottom of the vessel, there is a pipe which is connected to the fuel solution transfer system. The fission heat will be transferred to the primary coolant through the heat exchanger tube bundle which is routed through the fuel solution in vessel.

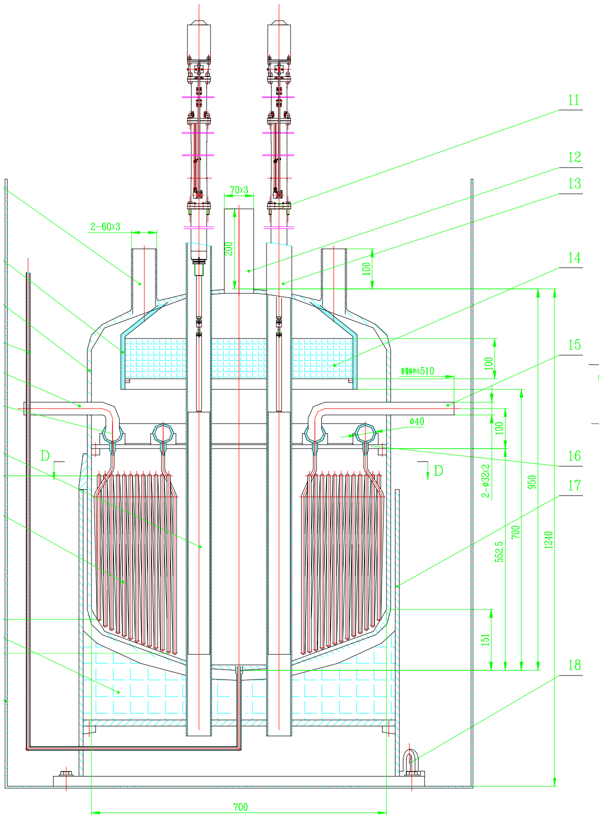

FIG. 2. General reactor configuration.

3.3. Nuclear design

The fuel solution utilizes highly enriched uranium. The volume of fuel solution is about 100 L with an original mass of ^{235}U of 3.8 kg. The concentration of uranium is 50 g/L. The nuclear design was accomplished using MCNP. Assuming the mass of ^{235}U is fixed and the concentration of the solution changes from 40 to 60 g/L, the corresponding K_{eff} calculated as shown in Fig. 3.

From the result, it is clear that with the same mass of ^{235}U, the K_{eff} of the core is changed in the range of 3‰ when the concentration varies within the range of 42~54 g/L. If the error of calculation is counted as 5‰, the reactivity of the core is relatively stable. Therefore, the core will not be affected while it is operated with the concentration of the solution in the range of 42~54 g/L.

3.4. Description of the main systems

Primary coolant system

The function of the primary coolant system is to maintain the temperature of the fuel solution by transferring the heat generated in the core through the heat exchanger to the secondary coolant system.

The primary coolant system is composed of two pumps, one heat exchanger and the related pipes, valves, instruments etc. The flow rate of the primary coolant is 7 t/h.

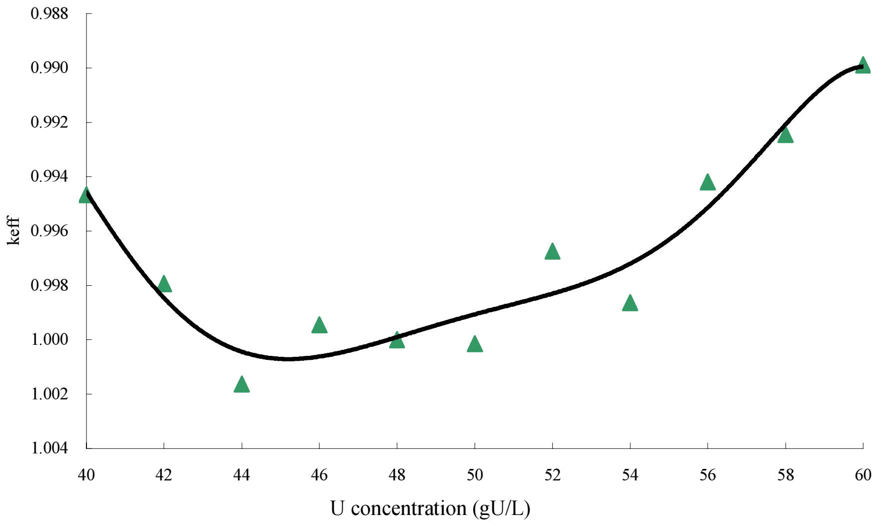

FIG. 3. The curve of K_{eff} changes according to the concentration with the same mass of ^{235}U.

Gas circulation system

The function of the gas circulation system is mainly to recombine the H_2 and O_2 which are decomposed during the fission operation. This ensures the concentration of H_2 is less than the limited concentration, thus preventing an H_2 explosion. The system then returns the water back to the fuel.

The gas circulation system is composed of a water separator, iodine separator, electric heater, H_2 and O_2 synthesizer, spray condenser, coolant tank and stainless steel pipes.

Fuel solution transfer system

The function of the fuel solution transfer system is to transfer the fuel solution between the reactor vessel and the storage pot and to adsorb the ^{99}Mo and ^{131}I isotopes. The fuel solution transfer system is composed of one storage pot, two extraction beds, related pipes, and valves.

N_2 removal and acid makeup system

During the reactor operation, H_2O and NO_3^- will be decomposed, consequently, the chemistry of the fuel solution will be changed and the pH will be increased. It is necessary to control the pH of the solution because higher pH will result in $UO_2(OH)_2$ deposition. Another challenge is how to deal with the N_2 resulting from the decomposition of NO_3^-. From the SUPO reactor, N_2 production is 2.5 mL/(kW·min). In the 200 kW MIPR, the quantity of N_2 is therefore 30 L/h or 720 L/d.

In order to maintain the chemistry of the fuel solution during the reactor power operation, additional nitric acid from the acid makeup tank will be injected into the reactor automatically, the pH and concentration of the fuel solution are then limited in the operating range. The additional N_2 is discharged to the storage tank to stabilize the gas pressure.

Emergency reactor shutdown system

If control rod trip failed to shut down the reactor, H_2 accumulation in the system would pose a considerable risk. In order to manage such risk, a passive method of discharging the fuel solution from the reactor vessel is adopted to shut down the reactor automatically. The emergency reactor shutdown system is the second reactor shutdown system of the MIPR. The main function of the system is to shut down the reactor rapidly by discharging the fuel solution from the reactor vessel if the reactor protection system becomes disabled.

4. ISOTOPES PRODUCTION SYSTEM

The main purpose of the MIPR is to produce ^{99}Mo and ^{131}I which come from the fission products of ^{235}U in the solution. These radionuclides can be adsorbed by acidic granular alumina (Al_2O_3).

The isotope product system includes two large hot cells and five product lines. These product lines are listed as follows:

(1) $Na_2{}^{99}MoO_4$ and ^{99}Tcm production line;
(2) ^{131}I production line;
(3) $^{89}SrCl_2$ production line;
(4) ^{99}Tcm radiolabel production line;
(5) ^{131}I capsule and ^{131}I radiolabel production line.

5. CONCLUSIONS

The medical isotope production reactor is a very promising project. The MIPR can produce many types of isotope used in industry, agriculture and medicine at low cost and on a large scale, and in addition the waste production of the MIPR is much less than that of the conventional isotope production method.

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THE PROGRESS REPORT OF AQUEOUS HOMOGENEOUS REACTOR FOR MEDICAL ISOTOPE PRODUCTION IN CHINA

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Abstract. This paper describes briefly the main progress in development of aqueous homogeneous reactor for medical isotope(^{99}Mo, ^{131}I and ^{89}Sr etc.) production from 1997 to 2007, including the design of a reactor using a $UO_2(NO_3)_2$ solution as the fuel, the purification process of the fuel solution, and the extraction and purification of medical isotopes.

1. INTRODUCTION

In China and other Asian countries such as Japan and South Korea, there is an increasing demand for medical isotopes, especially ^{99}Mo, ^{131}I and ^{89}Sr. Therefore, a project to design an aqueous homogeneous reactor (AHR) using $UO_2(NO_3)_2$ solution as the fuel for medical isotope production started during 1997 in the Nuclear Power Institute of China(NPIC). After about 10 years of effort, the preliminary design of MIPR was completed. The fuel solution purification process, including the extraction and purification process for the principal medical isotopes of interest, were established and demonstrated on a mock-up.

2. MIPR DESIGN AND MAIN SYSTEM VERIFICATION EXPERIMENTS

2.1. Reactor core design

Choice of fuel solution

Either UO_2SO_4 or $UO_2(NO_3)_2$ solution can be used as the fuel solution. The comparison of the main characteristics between two fuel solutions is listed in Table 1. In order to facilitate the purification of the fuel solution and to facilitate the extraction and purification of medical isotopes, the $UO_2(NO_3)_2$ solution was adopted in the design by the NPIC.

Table 1. Comparison of the main characteristics between the two fuel solutions.

Fuel solution	Thermal stability	Radiation stability	Solubility	Neutron absorption cross section	Isotope extraction	Fuel solution purification
UO_2SO_4	<280°C	Good	Pu,Ba,Sr may be deposited	Small	Difficult	difficult
$UO_2(NO_3)_2$	<184°C	2.5mL N_2/kW.min	good	Large	Easy	Easy

Choice of ^{235}U enrichment

Both HEU and LEU can be considered to be the reactor fuel. The comparison of the main characteristics of the fuel solution with HEU and LEU is listed in Table 2. In order to facilitate the design of MIPR and to get high quality ^{99}Mo product, HEU was initially chosen for the MIPR. In the future, after the main problems associated with LEU are resolved, LEU can be used.

Table 2. Comparison of the main characteristics between HEU and LEU as MIPR fuel.

Fuel	^{238}U content	NO_3^- concentration	^{99}Mo extraction	Purification of fuel solution	Nuclear and thermo-hydraulic design
HEU	10%	0.62 mol/L	Easy	Easy	Easy
LEU	82.25%(More ^{239}Pu produced)	1.72 mol/L(More N_2 produced)	Difficult	Difficult	Difficult

Choice of reactor power

In the past, the power of most AHRs was 20kW to 50kW. From an economic perspective, if the design power of MIPR is higher, it is better. In order to meet the needs of the main medical isotopes(^{99}Mo, ^{131}I) in China and other Asian countries, 200kW was chosen as the design thermal power for MIPR. The main constraints relevant to reactor power are listed in Table 3.

Table 3. The main constraints relevant to reactor power.

Main limits to the reactor power	Main measures to increase the power
Power density<2.5kW/L	Increase the fuel solution volume to over 100L, add some metal ions into the fuel solution to avoid UO_2^{2+} precipitation by H_2O_2.
Capacity of recombination of H_2 and O_2 to H_2O	Increase the gas flow in the gas loop and increase the capacity of the recombiner for H_2 and O_2 to maintain the H_2 concentration in gas loop below 2%.
Capacity of the heat produced from reactor operation	Increase heat transfer area to 4m^2, improve the arrangement of heat exchanger coils, increase the flow of cooling water in the exchanger.
Prevent fuel solution from entering the gas loop	Increase the diameter of the reactor core with a large surface area for the release of gas bubbles, increase height of reactor core container and some other means to prevent the fuel solution from entering the gas loop.
Produce a nuclear design to minimize gas bubbles in the fuel solution	Decrease the power coefficient and flatten neutron flux.

The main results of MIPR design and system experiments with mock-up

a. $UO_2(NO_3)_2$ solution: 100L~110L, 3.7Kg~3.8Kg ^{235}U, HNO_3 concentration: 0.2 mol/L;

b. Gas loop: gas flow: >180m^3/h, recombiner capacity: >1L H_2/s.

c. Capacity of the heat exchanger: >200kW.

d. At present, analytical computer programmes can not deal with the content and distribution of gas bubbles produced in the reactor core solution, so the power coefficient can not be calculated. A critical tests will be done in 2007.

3. PURIFICATION OF FUEL SOLUTION

After a period of reactor operation, a lot of fission products (FPs) will be produced. For example, operation at 200kW for one year results in more than 40 g FP. Most of the FP such as Cs, Sr, Ba, Y, Zr, Nb, Ru, La, Ce, Sm etc. can be extracted using a series columns loaded with inorganic ion exchangers. Results of experiments carried out on a mock-up showed that more than 70% of FP could be removed from the fuel solution, while uranium loss is insignificant. After purification, the fuel solution could be reused directly.

4. EXTRACTION OF THE MAIN MEDICAL ISOTOPES FROM THE FUEL SOLUTION

After 24 h operation at 200kW, about 200kCi ^{99}Mo, 400Ci ^{131}I and 100Ci ^{89}Sr can be produced in the reactor through $^{235}U(n,f)$ reaction. ^{99}Mo and ^{131}I in fuel solution can be extracted with one or two inorganic exchange columns with a yield over 80%. After purification, the final yield for ^{99}Mo and ^{131}I can be about 60%. ^{131}I in the gas loop as $^{131}I_2$ can be extracted by activated carbon or some other solid adsorbent. ^{89}Sr produced via the decay of the parent radionuclide ^{89}Kr in the gas loop can be extracted in a bypass gas loop. In NPIC, a project to construct a 200kW MIPR is underway. According to the project, 10kCi of ^{99}Mo, 2kCi of ^{131}I and 400Ci of ^{89}Sr can be produced annually.

PRELIMINARY STUDY OF ^{99}Mo EXTRACTION PROCESS FROM URANLY-NITRATE FUEL SOLUTION OF MEDICAL ISOTOPE PRODUCTION REACTOR

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Abstract. The adsorption and desorption characteristics of Mo on alumina(Al_2O_3) were investigated. The Mo recovery yield decreased with a decrease of the ratio of the height to the inner diameter (ID) of the column, increase of elution rate and increase of the solution temperature, but was not affected by the concentration of HNO_3 between 0.01 mol/L and 0.5 mol/L. An overall recovery yield of Mo of more than 85% was obtained when the ratio of the height to the diameter of the column was greater than 2, at a temperature under 31°C and with an elution rate less than 2 mL/mL/min using two Al_2O_3 columns. On a 1:1 mock-up of the circulation loop for uranyl-nitrate fuel solution transfer and radionuclide extraction in a preliminarily designed medical isotope production reactor(MIPR), using an extraction column loaded with 1.2L Al_2O_3 and a purification column loaded with 64 mL Al_2O_3, Mo was separated from 100L $UO_2(NO_3)_2$ solution containing 5120 g U, 50 mg Mo, 800 mg Sr, 800 mg Cs, 800 mg Ce, 500 mg Zr, 100 mg Ru, 200 mg Te and 200 mg I and in a medium of 0.1 mol/L HNO_3. The overall recovery yield of Mo was 67.7% ± 3.2%(n=3), while the contents of U, Sr, Cs, Ce, Zr, Ru, Te, and I in separated Mo meet the specification of European Pharmacopoeia (EP).

1. INTRODUCTION

Technetium-99m(^{99m}Tc)is the most frequently used radionuclide in diagnostic nuclear medicine. It is used for diagnosis of about 60 000 patients daily in the world [1]. ^{99m}Tc is primarily produced via the decay of its parent nuclide ^{99}Mo, and ^{99}Mo can be produced by ^{235}U fission or neutron irradiation of ^{98}Mo.

Concept designs of solution reactors utilizing uranyl-nitrate or uranyl-sulphate solutions as fuel were proposed in the 1940s, and approximately 30 solution reactors, with a thermal power ranging from 0.05W to 5MW, have been built worldwide [2]. However, these solution reactors were applied not for radionuclide production but for research, and most of them have been shut down. Utilization of solution reactors for radionuclide production was proposed in 1989 [3]. The concept design of a 200kW MIPR was presented at the 1993 American Nuclear Society Winter Meeting [4]. A 20KW solution reactor named ARGUS operated at the Russian Research Center-Kurchatov Institute in 1995 [5]. One of the planned missions of ARGUS is radionuclide production. Compared with conventional (research or productive) reactors, MIPR has the advantages of smaller critical mass, inherent safety characteristics, simplified fuel handling and processing, much lower cost (to produce ^{99}Mo of the same activity, the power, uranium consumption, reactor operation and waste disposal expenses of a MIPR are only 7%, 0.4% and less than 2% of those of a conventional reactor, respectively) [2].

Extraction methods of ^{99}Mo from ^{235}U oxide or alloy targets irradiated by neutrons, including ion exchange, adsorption chromatography, precipitation, solvent extraction and extraction chromatography, have been developed [6–9]. The most widely used material was alumina (Al_2O_3). However, there are a great deal of differences between the production characteristics of MIPR and conventional reactors. The target from a conventional reactor can be dissolved in a solution of small volume (for example, 400 g UO_2 was dissolved in 700 mL HNO_3), while the volume of a MIPR fuel solution for ^{99}Mo extraction is more than 100L. Therefore, it is expected that the chemical behaviours of Mo, U and other fission products such as Sr, Cs, Zr and I etc. in these different solutions will be different from each other.

In this work, the ^{99}Mo extraction process from a uranyl-nitrate fuel solution of MIPR was investigated using stable elements and natural U.

2. EXPERIMENTAL

2.1. Materials and equipment

1) 750GW UV-VIS sepctrophotometer (Shanghai); SHB-III circulating water vaccum pump(Zhengzou); DHL-A flow rate controlling pump(Shanghai); Mettler AE240 balance (Shanghai); DB211XSCB drying oven (Chengdu); Glass columns with I.D. of 8, 10, 25, 30mm.
2) $UO_2(NO_3)_2$ solution: 642 gU/L, PH 1.71, molar ratio of U/NO_3 1.87. Chromatographic alumina: 0.136mm~0.093mm, acidic and neutral (Shanghai).

2.2. Experiment

Activation of Al_2O_3

200 g acidic Al_2O_3 was washed with 300 mL water and decanted 6 times to remove fine particles, and then dried at 110°C and heated up to 500°C for 4 h.

Column separation method

The sample solution was passed through an Al_2O_3 column pre-saturated with HNO_3 solution of the same concentration of the sample solution. The Mo concentration in the effluent was determined by sepectrophotometry to calculate the absorption efficiency. Then the column was successively washed with HNO_3 solution of the same concentration as used for saturation, 5 Al_2O_3 bed volumes of H_2O and 0.01 mol/L $NH_3{\bullet}H_2O$. Finally Mo was eluted with 5 Al_2O_3 bed volumes of 1 mol/L $NH_3{\bullet}H_2O$. The concentration of Mo in the eluate was determined to calculate desorption efficiency and recovery yield of Mo.

Repeat use of Al2O3 column

After the first separation cycle, the column was successively washed with 5 Al_2O_3 bed volumes of 2 mol/L $NH_3{\bullet}H_2O$, 5 Al_2O_3 bed volumes of H_2O, 10 Al_2O_3 bed volumes of 2 mol/L HNO_3 and 5 Al_2O_3 bed volumes of H_2O. Then the column was ready for next separation cycle.

Two-step separation

The eluate of the first separation was adjusted to 0.1 mol/L HNO_3 with HNO_3 concentrate, and then the second separation was carried out. The eluate was collected in fractions and the Mo concentration in each fraction was determined for drawing the elution curves. The experiment conditions are listed in Table 1.

Table 1. Experiment conditions for two-steps' separation of Mo on Al_2O_3.

No	First separation			Second separation		
	Column	Adsorption	Desorption	Column	Adsorption	Desorption
1	54 g Al_2O_3 I.D. 25mm	3500 mL 5.2 μg Mo/mL, 0.1 mol/L HNO_3 0.5 mL/mL/min	300 mL 1 mol/L $NH_3•H_2O$	4.2 g Al_2O_3 I.D. 10mm	250 mL 43.8 μg Mo/mL, 0.1 mol/L HNO_3 1.0 mL/mL/min	24 mL 1 mol/L $NH_3•H_2O$
2	54 g[1)] Al_2O_3 I.D. 25mm	3500 mL 5.5 μg Mo/mL, 0.1 mol/L HNO_3 0.5 mL/mL/min	350 mL 1 mol/L $NH_3•H_2O$	4.0 g Al_2O_3 I.D. 10mm	250 mL 42.1 μg Mo/mL, 0.1 mol/L HNO_3 1.0 mL/mL/min	50 mL 1 mol/L $NH_3•H_2O$
3	35.2 g Al_2O_3 I.D.30mm	300 mL 21.0 μg Mo/mL, 0.1 mol/L HNO_3 0.2 mL/mL/min	200 mL 1 mol/L $NH_3•H_2O$	4.0 g[2)] Al_2O_3 I.D. 10mm	250 mL 10.0 μg Mo/mL, 0.1 mol/L HNO_3 1.0 mL/mL/min	50 mL 1 mol/L $NH_3•H_2O$

Notes: 1) Using the first column of exp. No 1 after washing with 150 mL 2 mol/L NH3•H2O, 300 mL H2O, 100 mL 2 mol/L HNO3, 300 mL H2O and 150 mL 0.1 mol/L HNO_3;

2) Using the second column of exp. No 2 after washing with 10 mL 5 mol/L NH3•H2O, 10 mL H2O, 20 mL 0.1 mol/L HNO3.

Separation of Mo from U, Sr, Cs, I

A solution consisting of 50.0 g/L U and 4.2, 4.0, 4.0, 4.0 mg/L of Mo, Sr, Cs, and I respectively was used and the experiment method is the same as in 2.2.2.

Extraction of Mo from stimulated MIPR fuel solution on a mock-up of circulation loop for fuel solution transfer and radionuclide extraction

On a mock-up of the circulation loop for fuel solution transfer and radionuclide extraction, 100L $UO_2(NO_3)_2$ solution containing 5120 g U, 50 mg Mo, 800 mg Sr, 800 mg Ce, 500 mg Zr, 100 mg Ru, 200 mgTe and 200 mg I and in a medium of 0.1 mol/L HNO_3 was passed through the extraction(1st) column with an ID of 80mm, loaded with 1.2L(about 1Kg)Al_2O_3 and pre-saturated with 10L 0.2 mol/L HNO_3. The column was successively washed with 10L 0.1 mol/L HNO_3, 6L H_2O and 6L 0.01 mol/L $NH_3·H_2O$. Then ^{99}Mo is eluted with 6L 1 mol/L $NH_3·H_2O$. The eluate is adjusted to 0.1 mol/L HNO_3 using concentrated HNO_3, and passed through the purification (2nd) column of 30mm ID, loaded with 64 mL Al_2O_3 and pre-saturated with 500 mL 0.1 mol/L HNO_3. The column was successively washed with 400 mL 0.1 mol/L HNO_3, 400 mL H_2O and 400 mL 0.01 mol/L $NH_3·H_2O$. Finally, ^{99}Mo is eluted with 300 mL 1 mol/L $NH_3·H_2O$.

3. RESULTS AND DISCUSSION

3.1. Adsorption capacity

The total adsorption capacities of activated and untreated Al_2O_3 were 52.9 mg/g and 29.3 mg/g respectively, while the working adsorption capacities were 35.8 mg/g and 14.0 mg/g respectively.

3.2. Effect of HNO_3 concentration, temperature, ratio of height to diameter of the column and flow rate on the separation of Mo

Variation of HNO_3 concentration between 0.01 mol/L and 0.5 mol/L had no observable effect on the separation of Mo (Table 2). Burrill K A and Hurrison R J [10] found that the Mo recovery yield decreased from 85% to 60% with decreasing sample solution pH from 0.9 to 0.1, and they concluded that there was a irreversible adsorption of Mo of about 15%. Different results from various laboratories may be due to the complication of the chemical states of Mo in acidic solution and the difference of the property of Al_2O_3 used. In an acidic solution, particles including $Mo_7O_{24}^{-6}$, $Mo_8O_{26}^{-4}$, $Mo(OH)_6$, MoO_4^-, $Mo_2O_7^{2-}$, and $[MoO_x(OH)_6^{-x}(MoO_3)_x]^{-x}$ (x is variable from 1 to 6) may be present, and $Al\text{-}OMoO_3^-$, $Al_2(MoO_4)_3$, $[AlMo_6O_{24}]^{9-}$ can be formed on Al_2O_3 [11,12].

Table 2. Effect of HNO_3 concentration on the separation of Mo on Al_2O_3..

C_{HNO3}/mol.L^{-1}	0.01	0.05	0.1	0.2	0.5
Adsorption efficiency/%	100.0	100.0	100.0	100.0	98.2
Desorption efficiency/%	96.4	92.3	94.4	90.3	96.1
Recovery yield/%	96.4	92.3	94.4	90.3	94.4

As temperatures increase from 25°C to 90°C, the adsorption of Mo had no considerable change, but the desorption efficiency decreased almost linearly (Table 3). This may be due to a spreading of molybdate as a monolayer on Al_2O_3 with octahedral coordination (i.e. Mo-O-Mo bands) caused by heat, or the occurrence of one-dimensional chains of MoO_2 species, the third O associated with Mo being placed in vacancies of Al_2O_3 [11], which leads to the difficulty of desorption of the absorbed Mo. Regression of the recovery yield vs. temperature shows that a recovery yield of Mo of more than 90% can be obtained when the temperature is lower than 31°C.

Results from different column height to diameter (H/D) ratios and flow rates (Table 4) show that under the conditions of a H/D between 1 and 6, a flow rate between 0.05 mL/mL/min and 2 mL/mL/min, the adsorption efficiency of Mo was not affected. However the desorption efficiency was greatly affected. When the H/D was less than 3 the recovery of Mo decreased with flow rate increase. When the H/D was greater than 3, at a flow rate between 0.5 mL/mL/min and 2 mL/mL/min, the recovery of Mo remained at about 95%. On the other hand, when the H/D was 2 to 3, with a flow rate from 0.2 mL/mL/min to 1 mL/mL/min, more than 90% of Mo can be eluted with 3 Al_2O_3 bed volumes of eluant. When the H/D was 6 and the flow rate between 0.5 mL/mL/min and 1 mL/mL/min, more than 90% of Mo can be eluted with 5 Al_2O_3 bed volumes of eluant.

3.3. Repeat use of Al_2O_3 column

After rounding the third data by a Q test (Q is 0.56, which was larger than $Q_{0.90}$=0.47), the mean recovery yield of seven repeat separations of Mo on an Al_2O_3 column was (96.1±2.6)% (Table 5). Hence an Al_2O_3 column can be used repeatedly, at least 8 times.

Table 3. Effect of temperature on the separation of Mo on Al_2O_3.

T/°C	25	50	75	90
Adsorption efficiency/%	100.0	98.1	98.1	100.0
Desorption efficiency/%	94.4	85.4	78.4	73.1
Recovery yield/%	94.4	83.8	76.9	73.1

Table 4. Effect of H/D ratio and flow rate(V) on the separation of Mo on Al_2O_3.

H/D	V/(mL/mL/min)$^{1)}$	Adsorption efficiency/%	Desorption efficiency/%	Recovery yield/%
1	0.05	99.8	98.3	98.1
	0.5	100	89.4	89.4
	1.0	99.8	85.9	85.7
	2.0	99.8	78.8	78.6
2	0.2	99.8	95.4	95.2
	0.5	100	94.5	94.5
	1.0	100	93.6	93.6
	2.0	100	91.3	91.3
3	0.5	100	95.8	95.8
	1.0	100	94.4	94.4
	2.0	99.8	95.7	95.5
6	0.5	99.6	93.9	93.5
	1.0	99.3	96.0	95.3

Note 1): The flow rate of adsorption, washing and desorption was kept same in a separation run.

Table 5. Mo recovery yield in eight repeat separations.

No	1	2	3	4	5	6	7	8
Recovery/%	97.8	93.0	(86.2)	94.3	97.8	96.3	98.4	95.4

3.4. Two-step separation of Mo and the elution profile

Under the conditions of Mo concentration in a range of 4.2 mg/mL to 43.8 mg/mL, H/D between 3 and 6, ratio of sample solution volume to that of Al_2O_3 column from 6.6 to 50.6, flow rate from 0.2 mL/mL/min to 1.0 mL/mL/min, the overall recovery yield of Mo was more than 85% using two-step separation (Table 6). The first column of experiment No 1 was regenerated and reused in experiment No 2, and the Mo recovery yields of two experiments had no difference. The second column of experiment No2 was regenerated and reused in experiment No 3, and the overall recovery yield in experiment No 3 was more than 95%. These results indicate that the overall recovery yield would be greater than 85% using two-step separation and the Al_2O_3 column could be reused.

Table 6. Two-steps separation of Mo on Al_2O_3.

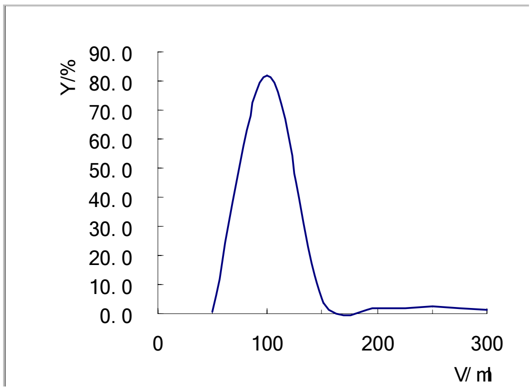
No	First separation			Second separation			Overall recovery/ %
	Adsorption/ %	Desorption/ %	Recovery/ %	Adsorption/ %	Desorption/ %	Recovery/ %	
1	97.3	94.2	91.7	99.3	96.0	95.3	87.4
2	99.6	93.9	93.5	90.5	101.1	90.5	84.6
3	99.8	95.4	95.2	100.0	100.0	100.0	95.2

Figure 2 shows the elution profiles of the two separation steps in the experiment No 1. At the two steps, 87.9% and 91.0% of Mo were eluted respectively.

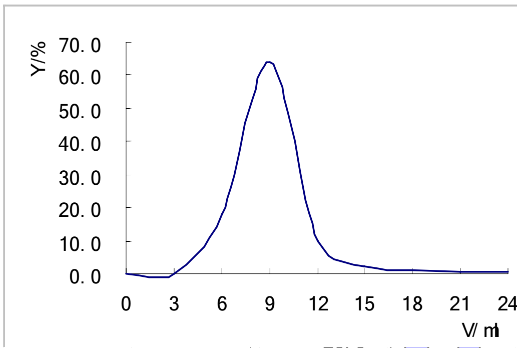
From the above results, it can be concluded that when H/D is 3 to 4, at a flow rate between 0.1 mL/mL/min and 1.0 mL/mL/min, and Mo eluted with 3 Al_2O_3 bed volumes of the eluant, the adsorption efficiency of Mo will be almost 100%, the recovery yield of Mo by one separation run will be greater than 90% and the overall recovery yield using two-step separation will be greater than 80%.

3.5. Separation of Mo from U, Cs, Sr and I

The results of separation of Mo from U, Cs, Sr and I and their comparison with the specifications of European Pharmacopoeia (EP) were listed in Table 7. Although the ratio of Sr^{2+} ion content after separation to that before separation was about 3 times higher than the specification of ^{90}Sr in EP, the radioactivity of ^{90}Sr to that of ^{99}Mo in the final product would be less than 8.7×10^{-9} because the radioactivity of ^{90}Sr in MIPR fuel solution would be less than 1% of that of ^{99}Mo. The radioactivity of α nuclides in the product would be far less than 10^{-9} of that of ^{99}Mo because of that the initial α radioactivity was only about 10^{-6} of that of ^{99}Mo. Therefore, the radioactive purity of ^{99}Mo product can meet the demand of the EP.



a-- Al_2O_3: 54 g; Φ: 25mm ; Mo solution: 3500 mL, 5.2 mg Mo/mL, 0.1 mol/L HNO_3; flow rate: 0.5mL/mL/min.



b-- Al_2O_3: 4.2 g ; Φ: 10mm; Mo solution: 250 mL, 43.8μg Mo/mL, 0.1 mol/L HNO; flow rate: 0.5 mL/mL/min.

FIG. 2. Elution profile of Mo on Al2O3 column.

Table 7. Separation of Mo from U, Cs, Sr and I and their comparison with the specifications of EP.

Element	U	Sr	Cs	I
$F_1/\%^{(1)}$	3.2×10^{-5}	5×10^{-4}	$<8\times10^{-5}$	5×10^{-3}
$F_2/\%^{(2)}$	9.7×10^{-10}	2.5×10^{-7}	$<6.4\times10^{-9}$	2.5×10^{-5}
$S/\%^{(3)}$	$\Sigma\alpha\leq1\times10^{-9}$	$^{90}Sr\leq6\times10^{-8}$	$\Sigma\beta\gamma\leq1\times10^{-4}$	$^{131}I\leq5\times10^{-5}$

Notes:
1) F_1 is the ratio of the content of element in the elate to that added by one-column separation.
2) F_2 is calculated ratio of the content of element in the elate to that added by two-column separation.
3) S is the ratio of radioactivity of the radionuclide to that of ^{99}Mo in ^{99}Mo product in the specification of EP.

3.6. Extraction of Mo from stimulated MIPR fuel solution on a mock-up of circulation loop for fuel solution transfer and radionuclide extraction

The results of Mo extraction from stimulated MIPR fuel solution on the mock-up of the circulation loop for fuel solution transfer and radionuclide extraction showed that the overall recovery yield of Mo was 67.7%±3.2%(n=3) (Table 8), while the contents of U, Sr, Cs, Ce, Zr, Ru, Te, and I in separated Mo meet the EP specification (Table 9).

Table 8. Recovery yields of Mo.

Exp. No.	Recovery yields / %		
	1st column	2nd column	Over all
1	81.0	79.6	64.5
2	77.9	84.4	67.4
3	84.4	86.5	62.3
$\bar{X}$±RSD	81.1±2.7	83.5±2.9	67.7±3.2

Table 9. Ratio of element content in the eluate from extraction column to that added.

Element	Ratio
Sr	$<8\times10^{-6}$
Cs	$<8\times10^{-6}$
Ce	$<8\times10^{-6}$
Zr	$<1\times10^{-5}$
Ru	$<8\times10^{-5}$
Te	$<8\times10^{-6}$
I	5×10^{-3}

4. CONCLUSIONS

Using a two-step separation with Al_2O_3 as the absorbent, ^{99}Mo being adsorbed in 0.1 mol/L HNO_3 solution and desorbed with 1 mol/L $NH_3{\cdot}H_2O$ solution, a overall ^{99}Mo recovery yield of more than 65% can be obtained, while the content of U, ^{89,90}Sr, ^{137}Cs and ^{131}I etc. in ^{99}Mo product can meet the specification of EP.

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FRENCH CEA EXPERIENCE ON HOMOGENOUS AQUEOUS SOLUTION NUCLEAR REACTORS

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Abstract. The CEA experience on Aqueous Solution Reactors results on a large Criticality Accident study programme conducted since 1967 at the Valduc Criticality Laboratory using at first the CRAC facility (1967–1972), then the solution fueled SILENE reactor, still in operation.

1. OBJECTIVES

The objectives of experiments were devoted mainly towards the phenomenology and the radiological consequences of a criticality accident occurring in a fissile solution (physics, detection, dosimetry, airborne releases and emergency preparedness). The fuel used was a highly enriched uranyl nitrate solution because it was available in the laboratory for other criticality experiments.

In the early 1970’s, the idea came in mind to use neutrons generated during the criticality excursions for other applications such as the neutron radiography or the production of short-lived radioisotopes, and to develop the concept of mini-reactors (about 20 litres of solution for the core) operating in a ‘pulse’ mode obtained by small reactivity insertions. In that case the production of neutrons lasts a few minutes and the energy during the pulse is limited (about 5.10^{16} fissions). Several mini-reactors based on this concept were developed in France just for neutron radiography purposes. One of them is still in operation.

2. MAJOR DIFFERENCES

The major differences between all those facilities were as follows:

- The way and the amount of reactivity was inserted
  - Addition of solution for CRAC
  - Rod withdrawal for the SILENE reactor
  - Approach of a reflector for the mini-reactors and constant amount of reactivity.
- The environment
  - Bare core vessel for CRAC
  - Several shields surrounding the SILENE core tank for varying the characteristics of radiation leakage (dosimetry and irradiation purposes)
  - Polyethylene reflectors and water cooling system on mini-reactors.
- The mode of operation
  - Various flow rates for the addition of solution for CRAC

- ‘Pulse’, ‘Free Evolution’ and ‘Steady State’ mode of operation for SILENE
- Always the same pulse for the mini-reactors.

Indeed in CRAC and SILENE most of parameters may vary due to the design and objectives of the programmes conducted. Reactivity for example may rise up to 10 $, $ characterizing the prompt critical state.

As to the mini-reactors designed to always produce the same amount of neutron flux, the characteristics of the pulse are constant.

A last one difference, in the SILENE facility the gases are periodically released through filters and the fuel is also reprocessed by using the PUREX process. For the mini-reactors, the installation was sealed and designed to run several hundred pulses without any fuel reprocessing, just a recombiner to avoid the accumulation of hydrogen.

It must be pointed out that no reactor using a solution as fuel and operating continuously at low power levels was developed at this time because other experimental reactors using more classical fuels were already existing in other CEA nuclear sites (Saclay, Grenoble)

So the CEA competence and knowledge on the behaviour of Uranyl nitrate solution may be worthwhile and useful to contribute to the assessment of the utility of homogeneous Aqueous Solution Nuclear Reactors for radioisotopes production.

The next paper in this report as well as another document in the Bibliography sum up the major results and lessons learned during more than 30 years of criticality excursions involving uranyl nitrate solutions (more than 2000 runs) and the capacities of the solution pulse SILENE reactor still in operation.

CRITICALITY ACCIDENT STUDIES AND RESEARCH PERFORMED IN THE VALDUC CRITICALITY LABORATORY, FRANCE

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Abstract. In 1967, the IPSN (Institut de Protection et de Sûreté Nucléaire – Nuclear Protection and Safety Institute) started studies and research in France on criticality accidents, with the objective of improving knowledge and modeling of accidents in order to limit consequences on the public, the environment and installations. The criticality accident is accompanied by an intense emission of neutronic and gamma radiation and releases of radioactive products in the form of gas and aerosols, generating irradiation and contamination risks. The main objectives of the studies carried out, particularly using the CRAC installation and the SILENE reactor at Valduc (France) were to model the physics of criticality accidents, to estimate the risks of irradiation and radioactive releases, to elaborate an accident detection system and to provide information for intervention plans. This document summarizes the state of knowledge in the various fields mentioned above. The results of experiments carried out in the Valduc criticality laboratory are used internationally as reference data for the qualification of calculation codes and the assessment of the consequences of a criticality accident. The SILENE installation, that reproduces the various conditions encountered during a criticality accident, is also a unique international research tool for studies and training on those matters.

1. PURPOSES OF CRITICALITY ACCIDENT STUDIES

Despite all measures to prevent the risk of criticality, it is impossible to be certain that a criticality accident will not occur when the quantity of fissile material present exceeds the critical mass. What happens then and what are the risks involved? Criticality accident study programmes were started in France in 1967 in order to answer these questions and to improve knowledge about accidents and how to model them, to limit the consequences on public, the environment and installations.

Remember that up to the present day, about sixty accidents have been recorded throughout the world, two thirds of them in research installations and one third in fuel cycle installations, causing the death of about twenty operators [1 and 2].

The purpose of the study on the phenomenology of criticality accidents that can occur in different configurations and how they are modeled in calculation programmes allows to describe the different scenarios that could occur in a given experimental or industrial process with the following objectives : evaluating exposure risks for operators, identifying possible means of detection, studying the long term behavior of the critical configuration, evaluating the consequences of radioactive releases on the public and the environment, if any, and providing information that could help to prepare action plans and crisis management.

In its Valduc criticality laboratory, IPSN has initiated experiments reproducing the criticality accident by divergence of a fissile solution of uranyl nitrate on the CRAC and SILENE test installations.

2. GENERAL PHENOMENOLOGY OF A CRITICALITY ACCIDENT

The criticality accident is the result of an uncontrolled chain fission reaction being started when the quantities of nuclear materials (uranium or plutonium) present accidentally exceed a given limit called the ‘critical mass’.

For neutronics, as soon as the critical state is exceeded, the chain reaction increases exponentially within a time period that depends on the initial reactivity of the system. The result is a fast increase in the number of fissions that occur within the fissile medium, also called the ‘criticality excursion’. This phenomenon results in a release of energy mainly in the form of heat, accompanied by the intense emission of neutronic and gamma radiation and the release of fission gases. The increase in the temperature of the fissile medium usually causes the appearance of neutronic feedback mechanisms that will reduce the reactivity present until the system becomes sub-critical, even if only temporarily. The result is usually the appearance of a power peak.

After the first power peak, radiolysis gas or steam bubbles migrate to the surface such that the resulting negative reactivity effect disappears and the power excursion restarts. This process by which bubbles are formed and then released outside the system causes the oscillating phenomenon usually observed during a criticality accident, see Figure 1.

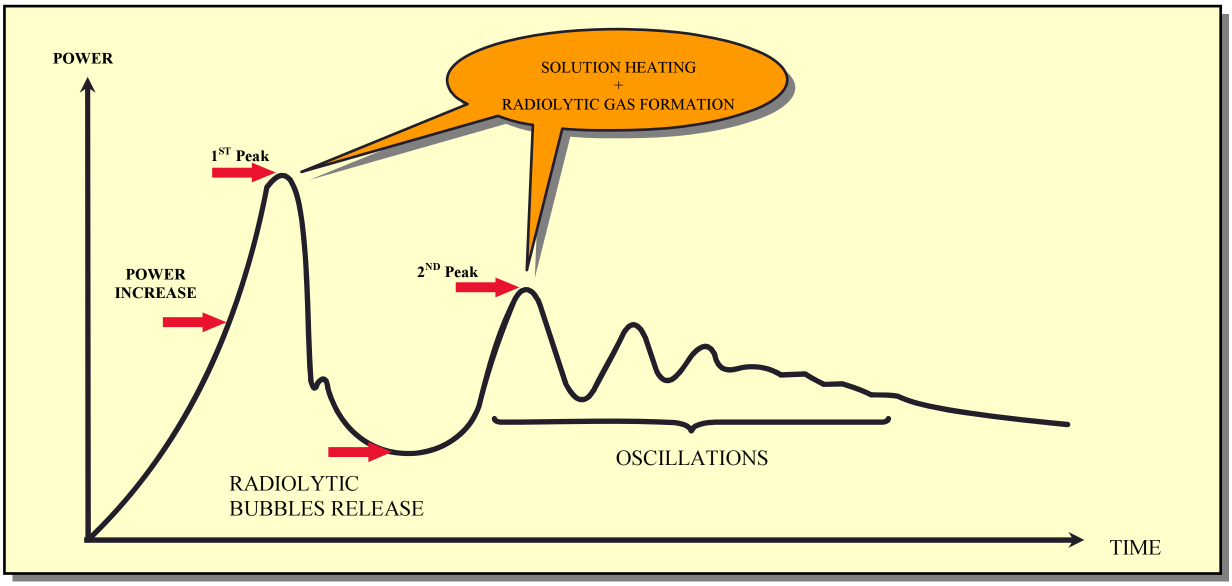

FIG. 1. Typical criticality accident in a fissile solution.

Therefore globally, the behavior of an accidental criticality excursion is defined by the following main parameters:

- The physicochemical nature of the critical fissile medium;
- The overall reactivity of the system;

- The initial intrinsic neutronic source, which is different depending on whether it is unirradiated enriched uranium, or uranium containing plutonium or plutonium;

- Neutronic feedback reactions resulting from effects related to the increase in the temperature of the fissile material, void effects (radiolysis gas bubbles in the case of a solution, steam, etc.) and the installation environment (heat exchanges with outside, confinement of the critical system, etc.).

As confirmed by experiments performed in the SILENE reactor, the combination of the previous phenomena with the initial accident conditions can lead to three types of behaviour (Figure 2):

1. The critical system becomes permanently sub-critical by modifying the configuration (mix, splashing or dispersion of material, modification of the geometry, etc.);

2. The system is made temporarily sub-critical by the increase in the temperature of the fissile material, and in this case the critical reaction will restart after a variable time interval that depends on heat exchanges with the surrounding medium;

Following a large initial reactivity, the system reaches temperatures at which the medium boils and the variation in power then depends on whether the medium is under or over moderated. Thus liquid boiling and the resulting concentration of the fissile solution after boiling can increase or reduce the system reactivity. Therefore the behavior of the critical system during the post-accident phase depends on whether or not the system is confined, in other words whether the steam can recondense and return to the solution, or « open » in which case vaporization or splashing of a solution will enable a return to sub-criticality.

This description applies to typical situations but it is in no way exhaustive since every criticality accident can have unique circumstances, as is clearly demonstrated by looking at accidents that have actually occurred throughout the world and particularly the most recent accident in Tokai-Mura in which a tank cooling device that was becoming ‘critical’ modified the events during the post-accident phase.

3. ACQUIRED KNOWLEDGE ABOUT CRITICALITY ACCIDENTS AND PARTICULARLY ABOUT AQUEOUS FISSILE MEDIA USING THE CRAC AND SILENE FACILITIES

About 70 experiments were performed on the CRAC (Conséquences Radiologiques d'un Accident de Criticité - Radiological Consequences of a Criticality Accident) installation in the Valduc Criticality Laboratory between 1967 and 1972, reproducing criticality accidents in an uranyl nitrate fissile medium [3 and 4]. The studies carried out were continued on the SILENE reactor starting in 1974, and more than 2000 critical exercises have been carried out on this reactor so far [5 and 6].

During the CRAC experiments, reactivity was usually inserted by using a pump to continuously add a solution of uranyl nitrate at a rate of up to 1800 l/h in 30 and 80 cm diameter cylindrical vessels. In the case of the SILENE reactor composed of a 36 cm annular vessel, reactivity is added by extracting a control rod from the core.

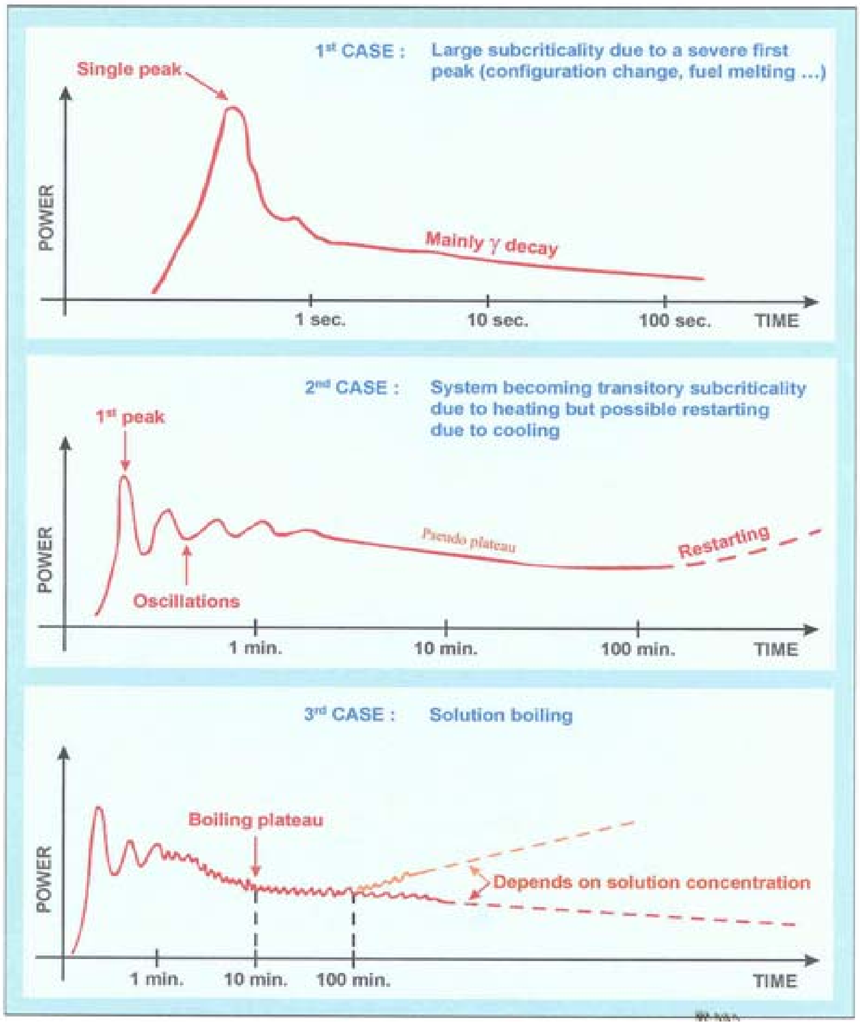

FIG. 2. Post-accident phases of a criticality.

Parameters varied within the following ranges in these experiments representative of accident situations:

- uranium concentration between 20 and 340 g/L;
- potential reactivity ρ less than 10 \$ in a homogeneous system, the dollar \$ being the value of the reactivity corresponding to the ‘prompt’ critical reactivity (also called β);
- reactivity ramps varying up to 2 \$/s;
- variable initial neutronic source.

3.1. Results and practical information about accident physics

The general phenomenology of the sequence of events during a power excursion in an aqueous fissile medium has been described previously, consequently the results and acquired information can be summarized as follows:

The first power peak and associated effects

Tests in a homogeneous medium were sufficient to explore the following domain:

- power period Te varying from 0.9 ms to 4 minutes,
- maximum power ranging from 10^{12} to 3×10^{19} fissions.s^{-1}.

The maximum values of the total energy of the first power peak were observed for the largest volumes (3×10^{17} fissions for a volume of 230 liters). For fast transients ($\rho >> \beta$), the maximum first peak power $\dot{E}$ is varying with the reciprocal period ω as to the following relation: $\dot{E} = C^{te} \times \omega^{1.8}$.

Some of the most important observations were:

- The appearance of a pressure wave for fast kinetics ($T_e < 10$ ms), also causing noise;
- Splashing of the solution under the fast reactions if there is no lid on the vessel used for the experiment;
- A blue light due to the CERENKOV effect concomitant with the occurrence of power peaks, which is the reason for the ‘flash’ concept frequently associated with a criticality accident;
- The strong influence of the intrinsic neutron source on the first power peak, the probability that a power excursion will be initiated being a function of the population of neutrons present in the medium. Therefore, the neutronic kinetics in a medium containing plutonium will not be the same as it would be in a medium containing uranium.

Energy recovered in thermal form

The fraction of released energy that is actually retrieved in thermal form and measured during the CRAC and SILENE tests is of the order of 1.4×10^{11} fissions.cal^{-1} or about 180 MeV retrieved in the form of heat for approximately 200 MeV emitted per fission reaction.

For power excursions subsequent to high reactivity (several $), it was observed during the CRAC and SILENE experiments that the boiling temperature of the solution (about 102°C) was reached for an energy corresponding to about 1.1×10^{16} fissions per liter. These data are valid for a power excursion lasting for a few minutes and for a system without any forced cooling.

Formation of radiolysis gases

Many experiments have shown a rate of formation of radiolysis gases corresponding to 1.1×10^{-13} cm^3/fissions, or about 110 liters of gas for 10^{18} fission reactions. Furthermore, the threshold at which these gases appear has been estimated at 1.5×10^{15} fissions per liter of solution in the studied media [7].

Relation between energy and the solution volume

An empirical relation was determined expressing the variation in the total number of fissions N_f as a function of the volume of the solution (in liters) and the time (in seconds). Based on the most representative CRAC and SILENE experiments in a homogeneous medium [8]:

$$N_f(t) = \frac{t}{3.55\times10^{-15} + 6.38\times10^{-17}\times t} \times V$$

It may be considered as giving an « envelope » value of the number of fissions during a period of a few minutes until the solution boils, the post-boiling phase being dealt with separately depending on the accident scenario considered. In particular, it demonstrates the important influence of the volume of the fissile solution on the energy that could be produced.

3.2. Modelling of accident physics

A fast analysis of criticality accidents that have occurred in the past illustrates the wide variety of situations encountered (different fissile environments, complex configurations, different causes) and the variety of observed effects (power, energy, duration, etc.). This gives an idea of the necessary complexity of calculation models capable of making predictions for all configurations.

Firstly, it should be emphasized that the energy produced during a criticality accident (often expressed in fissions) is very much less than the energy produced following a reactor accident. For example, a criticality accident with 10^{18} fissions releases an energy of the order of about thirty megajoules (1 joule = 3.1×10^{10} fissions).

An examination of accident results shows that the energy can vary from a few 10^{15} fissions to 4×10^{19} fissions for fuel cycle installations, and the power during the first peak can be as high as 10^{20} fissions.s^{-1} for a very short time. The duration can simply be a ‘flash’ of a few milliseconds, or it can continue for tens of hours.

The diversity of these effects is directly related to parameters that affect the accident phenomenology. This is why different accident models were developed, making a distinction between four main environment categories (liquid, powder, metal, fuel rods and water). The following diagram illustrates the common architecture of these calculation programmes (Figure 3).

Accident calculation programmes developed jointly with the UKAEA (CRITEX for aqueous media, POWDER for powders, CHATEAU for immersed fuel rods) can be used to estimate the variation of the power, energy and temperature of the medium as a function of time [9].

3.3. Exposure risks associated with a criticality accident: doses mesaured around the CRAC and SILENE installations

The criticality accident is accompanied by an intense emission of neutronic and gamma radiation with a variable duration varying from a few seconds to several hours, depending on the phenomenology of the power excursion.

The initial radiation field is a complex field of neutrons and gamma radiation with energy of up to about ten MeV. The contribution of neutrons and gamma rays to the total dose is very variable depending on the nature of the fissile material (metal, powder, liquid, etc.), the dimensions and compositions of the system that is going critical, and its environment. Thus, the ratio of the ‘neutrons’ dose to the ‘gamma’ dose expressed in grays can vary from 10 for a ‘metallic’ source to 0.2 for a highly hydrogenated medium, which is a variation by a factor of 50. This neutrons effect is further reinforced if ‘equivalent doses’ expressed in sieverts are compared. As the distance from the source increases, the energy of the radiation field degrades and its intensity drops very quickly and approximately inversely as a function of the square of the distance over the first few meters. For longer distances, radiation propagation laws are more complex due to effects related to the ground and the atmosphere.

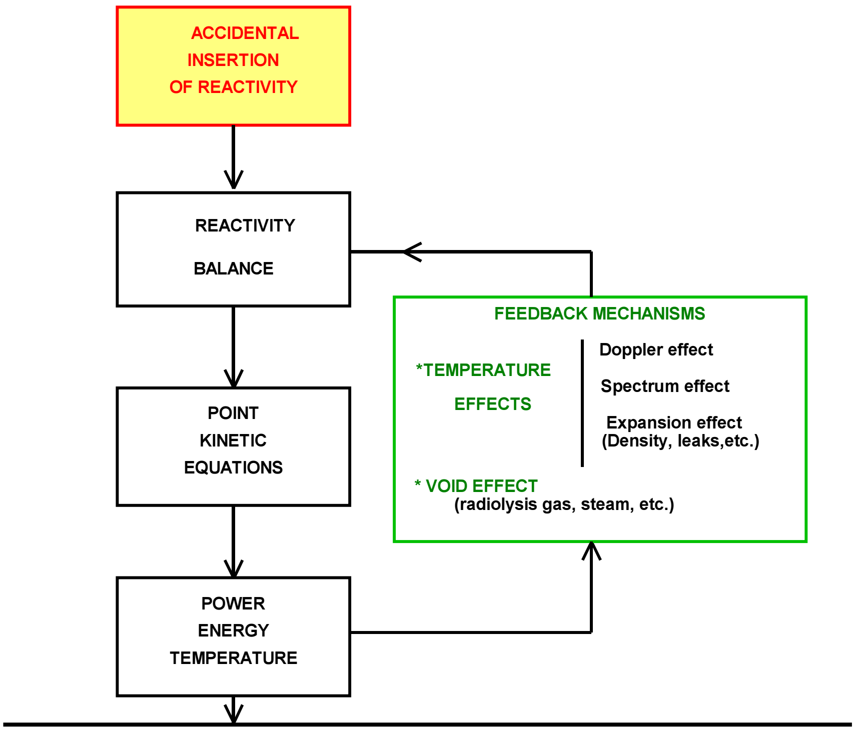

FIG. 3. Common architecture of calculation programmes.

Dosimetry results obtained on the CRAC and SILENE installations [10] must be considered as being representative of the dose to which personnel could be exposed during a criticality accident in a uranyl nitrate solution, the system that becomes critical being located in a concrete room. The number of fissions and the emitted dose are not proportional to each other for sources with very different configurations (variable cylinder diameter and concentration), since leakage radiation depends on the source characteristics. It is found that the dose/fission ratio is maximum for small sources with low concentrations.

The maximum value of the observed total dose during tests on the CRAC and SILENE installations is 5.8×10^{2} Gy at 1m from the centerline of the source, for 10^{18} fissions for a 30 cm diameter cylinder with a concentration of 80 g/L.

For information, doses emitted during the first power peak on the SILENE reactor for 10^{17} fissions at 1 m from the 'core' (40 liters of uranyl nitrate solution) are as follows:

Neutrons:	Dose (KERMA tissus)	≈20 Gy	→	Equivalent dose	≈300 Sv
Gamma:	Dose	≈25 Gy	→	Equivalent dose	≈25 Sv

In summary, it should be emphasized that in terms of criticality accidents, there is no direct proportionality relation between the number of fissions and the dose emitted for different sources. The nature of the fissile medium and its geometric dimensions play an essential role in estimating exposure risks.

Measured doses demonstrate that the risk of exposure is one of the major risks in a criticality accident and the resulting doses can be fatal for personnel working in the immediate vicinity of the equipment concerned.

3.4. Detection of criticality accidents

The purpose of a criticality accident detection system (CADS) is to trigger an alarm as quickly as possible in order to trigger immediate evacuation of personnel at the beginning of a criticality accidental excursion and thus limit exposure risks.

In 1976, the CEA designed the E.D.A.C system (Ensemble de Détection et d'Alarme de Criticité) making use of information derived from the CRAC and SILENE experiments, based on a monitoring unit connected to at least three criticality detectors [11 and 12]. The criticality alarm is only triggered if at least two detectors send an alert signal to the monitoring unit. This alert signal is generated if two conditions are satisfied:

- a predetermined dose limit, usually set to 25 μGy (2.5 mrad), is exceeded;
- a dose rate limit fixed at about 10 $\mathrm{mGy.h^{-1}}$ (1 $\mathrm{rad.h^{-1}}$) is exceeded.

The detection system is based on measuring the total dose due to neutronic and gamma radiation by means of two scintillators, sensitive to these two types of radiation.

Tests carried out in the SILENE reactor demonstrated that the system can be used to detect all types of accidents, in other words power excursions with fast kinetics and with slow kinetics. The EDAC accident detection system can also record and monitor the evolution of the accident by means of criticality detectors, particularly through a remote console placed outside the evacuation area. Its contribution may be essential for management of the post-accident situation and action.

3.5. Estimate of releases of radioactive products during a criticality accident in solution

The SILENE installation was used for an experimental programme to determine the rates of release of fission products (FP) emitted during a criticality accident in an aqueous fissile medium, the experimental conditions varying up to and including boiling of the solution to facilitate the release of fission products [13]. The fission products concerned may be classified into three categories depending on their physicochemical nature, namely rare gases, aerosols, iodine, bromine and ruthenium isotopes.

The main information derived from the SILENE fission products programme is as follows:

Rare gases

The release ratios of rare gases (Xe and Kr) are almost 100% for gases with half-lives of more than one minute. They vary between 10% and 50% for half-lives varying from a few seconds to a minute, and are of the order of 10% for very short half-lives (less than 2 seconds).

Aerosols

The observed size of aerosol particles after boiling is about 0.1 μm, whereas below boiling the size may be about 5×10^{-3} μm. There can be a severe local irradiation risk related to trapping these aerosols on filters.

Iodine, bromine and ruthenium isotopes

In the SILENE experiments, the maximum release ratios observed for iodine for acidity close to 2N, were very much less than 1% for a boiling solution. A release ratio of 10% was measured in the core for a very low solution acidity and a high initial content by load of iodine in the solution.

The maximum emission ratios for other volatile fission products are estimated at 20% for bromine and 1% for ruthenium.

For information, the following table presents quantities of fission products released from the solution, and the consequences of these releases at a distance of 500m along the windward direction under normal diffusion conditions with a wind speed of 5 m/s (DN5), for several « cooling times » of the fission products at the time of the release (Table 1).

Table 1. Risk of exposure to the public

Radionuclides family	Maximum released activity (Bq) and doses (mSv) for 10^{18} fissions at different time		
	10 seconds	1 hour	10 hours
Rare gases, Halogens (apart from Iodine) and Aerosols (Bq)	3.0×10^{14}	2.5×10^{13}	9.0×10^{11}
Iodine (Bq)	1.7×10^{12}	1.8×10^{12}	2.6×10^{11}
Inhalation at 500 m in DN5 (doses to the thyroid and lungs) (mSv)	0.12	0.20	0.12
Direct irradiation by the plume at 500 m and DN5 (mSv)	0.50	0.13	0.004

These values show that the risk of exposure of the public during a criticality accident is low.

3.6. Experiments and exercises carried out on the SILENE reactor for action management

The SILENE reactor is used to provide evaluation data necessary for action management following a criticality accident. The following themes are considered:

- Estimate of the possible dose to a work team during the post-accident phase [9],
- Dosimetry of the criticality accident: SILENE is an international reference source in this subject and has already been used for the purposes of international exercises under the auspices of the AIEA and the CCE [14],
- Radiation instrumentation test during the post-accident phase,
- Fast checking of exposed personnel and dose estimate (sodium activity and dosimeter measurements for example) for an appropriate therapeutic treatment.

4. CONCLUSIONS

The criticality accident studies carried out have improved knowledge in several fields: physics, detection, dosimetry and the release of radionuclides. These results must contribute to a better assessment of the risks of irradiation and contamination associated with a criticality accident and the application of action measures and provisions for crisis management. They emphasize the need to well define intervention plans and to be capable to stop the accident process.

More generally, operating experience with real accidents that have occurred throughout the world confirms that the energy released during a criticality accident is generally limited, but there are severe risks of irradiation for personnel working close to the equipment concerned and lethal doses are possible.

The unfortunate Tokai-Mura accident also demonstrates that the scale of the consequences in terms of the media and acceptability of the nuclear risk may be completely different.

The SILENE reactor is a unique international research installation that can be used for training teams and to maintain the skills necessary for management of action to be taken following a criticality accident.

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MEDICAL COMPLEX FOR RADIOISOTOPE PRODUCTION

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Abstract. The IPPE has been undertaking efforts to achieve practical implementation of the technology applying homogeneous aqueous solution nuclear reactors (HASR) for the recovery of fission radioisotopes directly from the fuel solution for more than a decade. The design of a Medical Complex for radioisotopes production based on HASR of 50 kWth is under development [1, 2]. The reactor capacity ensures the production of at least 500 Ci/d of ^{99}Mo isotope as well as about 100 mCi/d of ^{89}Sr, 130 Ci/week of ^{133}Xe and the mixture of iodine isotopes - ^{131}I, ^{132}I, ^{133}I. The reactor design also includes a neutron beam intended for the treatment of malignancies by neutron therapy methods as well as irradiation rigs for activation analysis located at the central part of the core and at the reflector. A relatively small reactor power - 50kWth – has been selected due to the intention to make the maximum use of the proven design and the operating practice for main components and safety systems of HASRs, as well as to exclude hazardous radiological burden on the personnel and the population under the reactor normal operation and in case of all possible accidents. The Complex designed production capacity is sufficient for meeting the Russian domestic market demands as to the nomenclature of the radioisotopes produced. R&D on the Medical Complex are carried on in cooperation with research institutes and design organizations of the Rosatom and other Russian Agencies: SSC RF-IPPE (Obninsk), 'Red Star' (Moscow), RRC 'Kurchatov Institute' (Moscow), SSDI (Moscow), MRRC RAMS (Obninsk). The planned deployment place of the Complex is one of the existing buildings at the site of the IPPE in Obninsk.

1. TECHNOLOGY OF ^{99}MO PRODUCTION IN HASR

The technology of the production of ^{99}Mo and other reactor isotopes using HASR is as follows [1]:

1) A part of the uranium fuel solution available in the HASR tank is delivered (with a small flow rate) to a special circulation loop and passed through the sorption column so that to recover selectively ^{99}Mo accumulated in the process of ^{235}U fission during the reactor operation at a rated power level. After the ^{99}Mo recovery the fuel solution containing the other fission products is sent back to the reactor tank. Thus, the continuous process of the radionuclide production and recovery is established, and it enhances considerably the efficiency of uranium use in comparison with the traditional uranium target irradiation technology.

2) Intensive water radiolysis taking place in HASR forces the process of volatile fission products release from the fuel solution into the upper gas plenum of the reactor. In the course of the regeneration of the radiolysis products the vapor-gaseous phase is drawn off on a continuous basis, and ^{89}Sr, ^{133}Xe, ^{131}I, ^{132}I, and ^{133}I isotopes are recovered selectively from it followed by the return of the remaining radionuclides to the reactor together with the regenerated water.

3) Then molybdenum and the other isotopes are purified from radioactive and chemical impurities by the standard methods applied in the existing and proven irradiated uranium target processing technology.

2. DESIGN COMPLEX FOR ^{99}MO PRODUCTION BASED ON HOMOGENEOUS AQUEOUS SOLUTION NUCLEAR REACTORS

A design of the Medical Complex for the ^{99}Mo production has been developed at the SSC RF-IPPE for implementing the above described technology. The Complex includes two 50 kWth HASRs and the systems intended for the recovery and purification of ^{99}Mo and other radioisotopes for medical application.

The availability of two reactors makes it possible to ensure required reliability of the production process and to warrant product shipments to the market. The ^{99}Mo purification can be performed using the single common system.

Each reactor production capacity is ~500 Ci/d or 300 kCi/y of ^{99}Mo (as of the moment of production). Each reactor is also equipped with the system for drawing off the vapor-gaseous phase with the view of recovering ^{89}Sr, ^{133}Xe, (^{131}I, ^{132}I, ^{133}I) and some other isotopes.

The design of the reactors also provides for a neutron beam, with parameters required for the treatment of malignant tumors by the neutron therapy method. This design feature makes it possible to perform up to 70 irradiation procedures per day at each reactor. The availability of in-pile irradiation rings makes it possible to perform more than 10,000 activation analyses per year. The main parameters of the complex are given in Table 1.

Table 1. Main technical parameters of the medical complex

Specifications	Units	Value
1. Reactor name		Reactor SR-RN
2. Number of reactors	pieces	2
3. Reactor characteristics		
– power	kWth	50
– fuel		aqueous solution of uranil-sulphate
– core volume	l	20
– uranium enrichment	%	90
– concentration of ^{235}U	g/L	85
– mass of ^{235}U	g	1700
– volume specific power	kW/L	1.5
– average fuel solution temperature	°C	80
– number of control rods	Piece	5
– reactor tank diameter	cm	32
– reactor tank height	cm	72
– reactor tank pressure:	Mpa	
- 'cold' state		0.036
- working state		0.090
– HX coolant temperature, inlet	°C	39
– HX coolant temperature, outlet	°C	74
– Reflector material		graphite
– Reactor life time	yrs	30
4. Production of ^{99}Mo (one reactor)	Ci/d	500
5. Production of ^{133}Xe (one reactor)	Ci/week	130
6. Production of ^{89}Sr (one reactor)	mCi/week	100
7. Electricity consumption	kW hr/yr	790 000
8. Water demand		
– new feed	m^3/d	25
– recirculation	m^3/d	360
9. Reactor operation schedule		
– working days per year	d	330

3. SR-RN REACTOR DESIGN FEATURES

‘SR-RN Reactor’ (solution reactor – radio nuclides, Figure 1) consists of the reactor vessel (tank) filled with the fuel solution, control and protection system (CPS), heat exchanger (HX) for fission and decay heat removal, and system for catalytic regeneration of water radiolysis products. The reactor tank is a welded cylinder with a spherical bottom and a flat cover. The tank is designed to withstand the internal pressure increase. The reactor auxiliary systems include:

- Secondary cooling circuit,
- Spent fuel solution storage system,
- Graphite reflector cooling system, etc.

A helical coil HX and vertical channels, in which regulation and safety control rods move, are installed inside the reactor tank.

The reactor is equipped with two safety control rods (SCR) designed in the form of barrels made of B_4C in steel cladding. Aluminum alloy tube intended for the location and movements of the reactivity compensation rod (CR) made of B_4C is inserted into the central hollow of each SCR. The power regulation rod (RR) made of B_4C, too, is located in the gap between the reactor tank and reflector.

The reactor tank is surrounded with the lateral and bottom graphite reflectors. A guard tank is provided to collect and keep the fuel solution in case of reactor tank break and fuel leakage.

The assembled mock-up of the reactor is shown in Figure 2. Principal flow-sheet diagram of the Complex is presented in Figure 3.

4. SYSTEM FOR CATALYTIC REGENERATION OF WATER RADIOLYSIS PRODUCTS

A specific feature of aqueous solution reactors, which is important from the safety point of view, is the formation of radiolytic hydrogen at the power operation. The problem of elimination of hydrogen is settled by creating a special regeneration system of passive mode of functioning that is continuously under operation. This system design and operation modes have been developed using experience of the research aqueous solution reactors and tested in the process of long-term operation.

The catalytic regeneration system (CRS) is a gravity driven natural circulation circuit. The inlet unit of the CRS is connected to the vapor-air space of the reactor. An electric heater and a special device (‘catalyst of H_2+O_2 recombination’) are installed at the rising section along the flow of the vapor-air medium. The tract turns down after the catalyst. A heat exchanger–condenser is installed in the upper part of the downcomer. The condensate flows down into the reactor tank after this heat exchanger.

5. PHYSICAL FEATURES OF THE REACTOR

The following advantages may be listed as substantial advantages of aqueous solution fuel compared to the traditional fuel pins and fuel assemblies:

- simplicity and low cost of the fuel solution preparation;
- simplified reactor core design;
- favorable operation conditions for the reactor materials at the temperature below 100ºC and at the atmospheric pressure;
- the absence of problems related to peaking effects for power distribution and fuel burn-up;

- elimination of fuel swelling problems;
- small value of Xe-poisoning thanks to free fission gases release from the fuel;
- high efficiency of fuel use;
- high level of the inherent self-protection features.

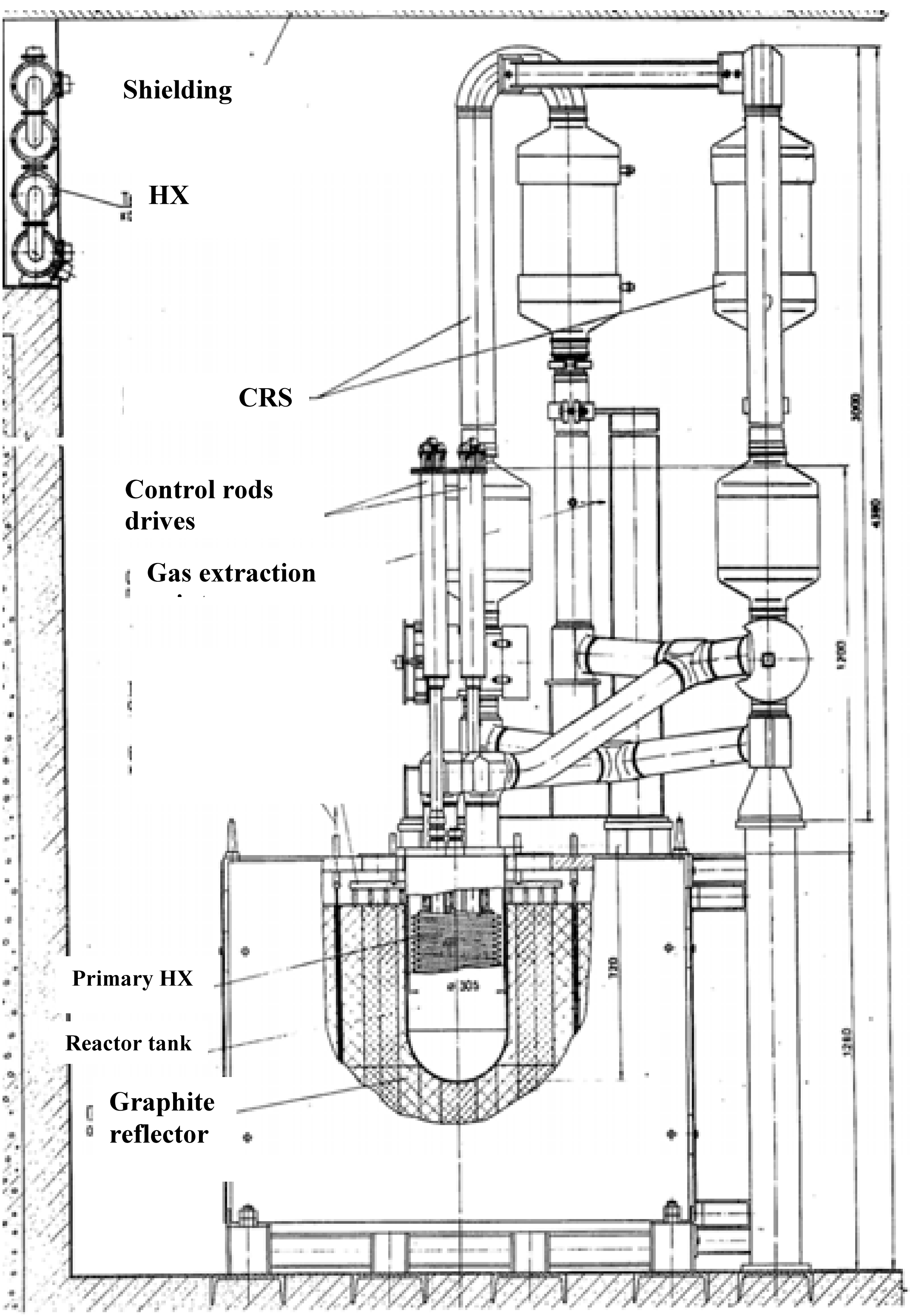


FIG. 1. SR-RN reactor facility.

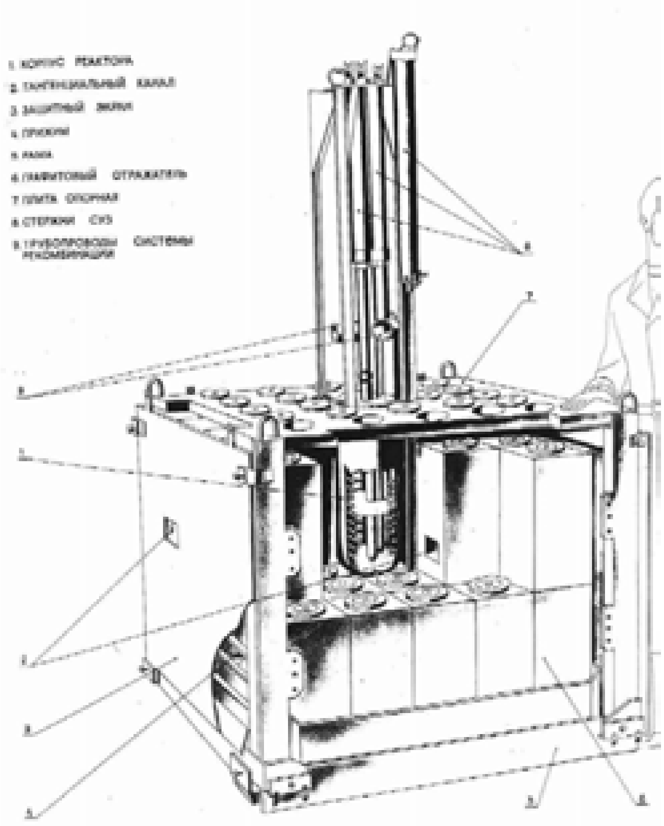


FIG. 2. Assembled mock-up of the SR-RN reactor.

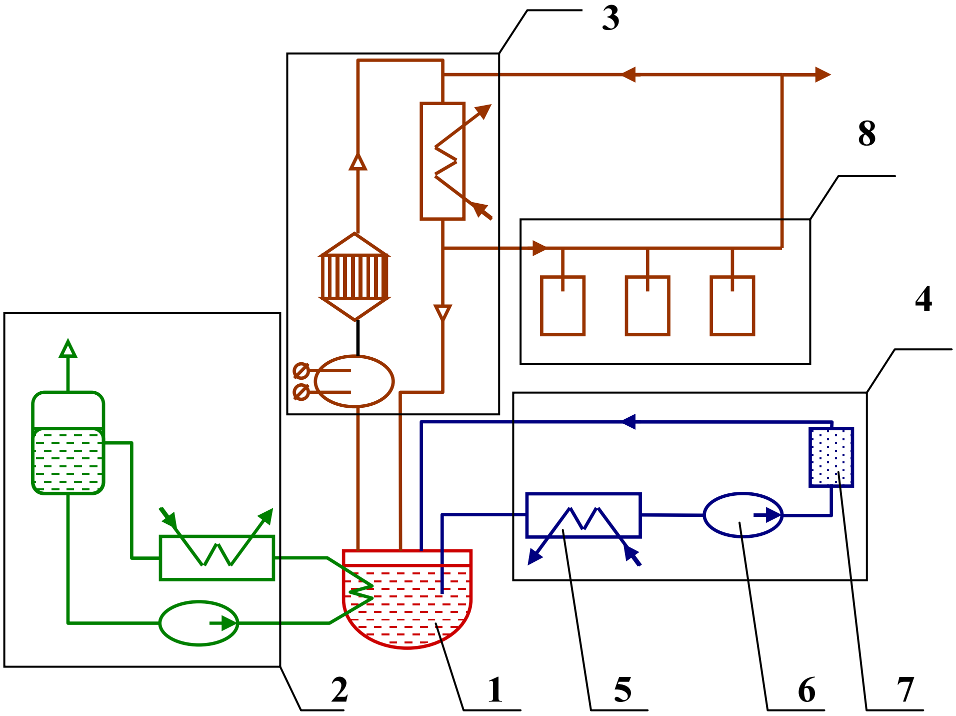


FIG. 3. Principal flow-sheet diagram of the complex.

1) reactor; 2) reactor cooling system; 3) water regeneration system; 4) ^{99}Mo recovery line; 5) cooler; 6) pump; 7) ^{99}Mo extraction column; 8) extraction line for the extraction of ^{89}Sr, ^{133}Xe, (^{131}I, ^{132}I, ^{133}I) isotopes from the fuel vapor-gaseous phase.

Neutron spectrum in the core of the SR-RN is thermal one. Reactivity excess under the cold and operating conditions is compensated by mechanical members of the CPS. The diagram of the HASR core is shown in Figure 4, its main neutronic specifications are given in Table 2.

As the Table 2 proves the reactor has negative temperature and power reactivity coefficients therefore it possesses self-protection features. Besides, the reactor has the following inherent physical safety features. The reactor has the maximum reactivity at the chosen optimum solution volume and uranium concentration. K_{eff} as a factor of ^{235}U concentration in the solution in case of a constant amount of ^{235}U loaded in the reactor is shown in Figure 5. The curve maximum corresponds to the ^{235}U optimum concentrations (g/L).

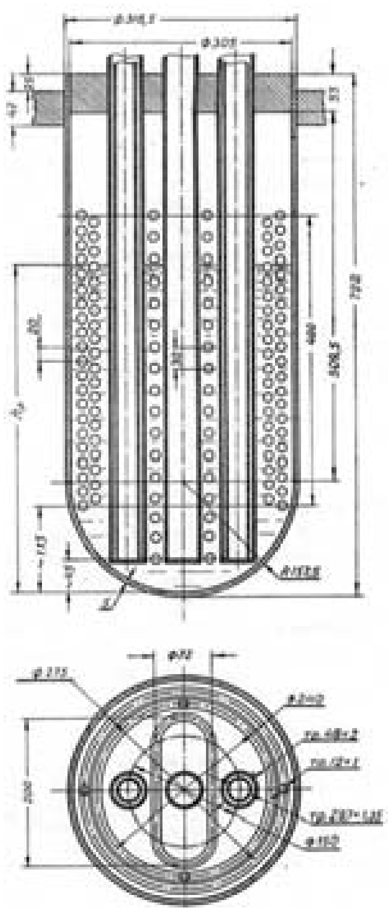


FIG. 4. Schematic diagram of the RR-RN reactor core.

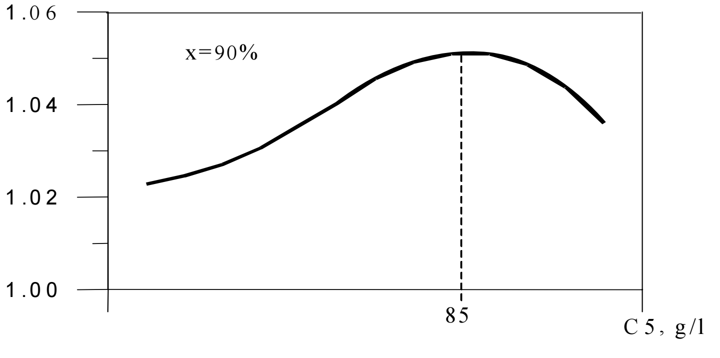


FIG. 5. K_{ef} as a factor of ^{235}U concentration in the solution SR-RN reactor safety.

Table 2. Main results of neutronic calculations

Parameter	Units	Value
Reactivity margin for burnout	β_{eff}	2.9
Reactivity margin for heating from 20°C up to 80°C	β_{eff}	2.4
Reactivity margin for power rise from 0 up to 50 kW	β_{eff}	1.2
Total worth of CPS rods	β_{eff}	7.5
Emergency protection efficiency	β_{eff}	5.0
Control rod efficiency	β_{eff}	0.7
Thermal neutron flux density ($E \leq 1$ eV) in the core center	$n/cm^2 \cdot s$	$0{,}59 \cdot 10^{12}$
Thermal neutron flux density ($E \leq 1$ eV) in the core periphery	$n/cm^2 \cdot s$	$0{,}27 \cdot 10^{12}$
Fast neutron flux density ($E \geq 0{,}1$ MeV) in the core center	$n/cm^2 \cdot s$	$1{,}03 \cdot 10^{12}$
Fast neutron flux density ($E \geq 0{,}1$ MeV) in the core periphery	$n/cm^2 \cdot s$	$0{,}40 \cdot 10^{12}$
Fuel temperature reactivity coefficient	β_{eff}/K	–0.058
Void reactivity coefficient caused by radiolysis	β_{eff}/kW	–0.023
Fuel volume reactivity coefficient	β_{eff}/L	1.0

The dilution of the fuel solution with water, for example, in case of rupture of the HX pipe, decreases reactivity, although it increases its volume. The solution evaporation also decreases reactivity, although it increases uranium concentration in the solution.

The worldwide experience of the operation of the existing aqueous solution reactors (more than 20 unites), the research done at the critical assemblies and reactors developed at the SSC RF-IPPE make it possible to conclude that HASR is highly reliable and safe reactor. Twenty-year operation experience of 'Argus' reactor [3] (SR-RN prototype) at the RRC 'Kurchatov Institute' enabled us to study and learn all dynamic characteristics of the transient processes in the reactor core and the CRS.

Passive methods and systems of protection from the accidents accompanied with heat removal disturbance and the primary circuit depressurization have been used in the SR-RN reactor design along with the inherent safety features:

- in case of failure of the main heat removal system the residual heat is transferred passively to the graphite reflector, and the heat from the reflector is dissipated into the environment;
- the solution working pressure in the reactor tank is lower than the atmospheric one so fuel leakages from the reactor are impossible during the reactor operation.

Absorber rods are used for the reactivity compensation in an aqueous solution reactor as well as in the majority of the other reactors. Reactivity-initiated accidents related to possible withdrawal of the rods resulting in positive reactivity insertion are studied for such reactors.

The analysis of the mode of accidental insertion of positive reactivity of up to 5βeff at the rate of 0.25βeff/s without scram has demonstrated that the power excursion takes place during the first seconds but the pressure inside the tank does not exceed 0.3 MPa. The tank is designed for this pressure and keeps its integrity. If nevertheless the reactor tank is broken the solution is kept inside by a guard tank. Table 3 contains some results of calculations of a number of reactivity initiated (RI) accidents without scram.

Table 3. RI accident calculation results

Value of reactivity insertion, Δρ	Time of reactivity insertion, t, s	Initial power level, % Nnominal	Maximum power excursion, kW	Maximum pressure in the reactor tank, Pmax, MPa
0.7βef	10	0.5%	34.5	0.037
0.7βef	10	100%	105	0.107
5.0βef	20	0.5%	1800	0.210
5.0βef	20	100%	272	0.283

The analysis of an accident during which water supply to the HX cooling the core stops under the reactor operation at the nominal power with failure of the automatic regulator and without scram, has demonstrated that the solution does not boils up during such accident, the reactor power decreases, the pressure in the tank increases slowly and reaches the value of 0.105 MPa during 50 s. Thus, there is a considerable time reserve for restoring the cooling water supply or for the reactor shutdown.

Figures 6 and 7 show the power and pressure changes in the reactor under some accidents without scram. 1) Self-withdrawal of automatic control rod at 100% power; 2) Stop of water flow in the core cooling HX accompanied with failure of the automatic regulator; 3) Self-withdrawal of automatic control rod at 0.5% power.

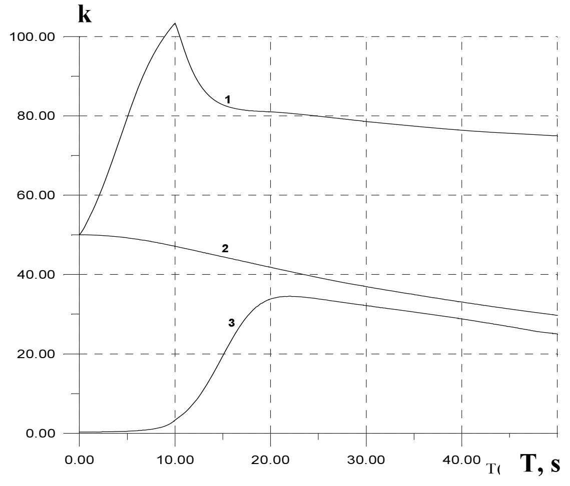


FIG. 6. The reactor power change at accidents without scram.

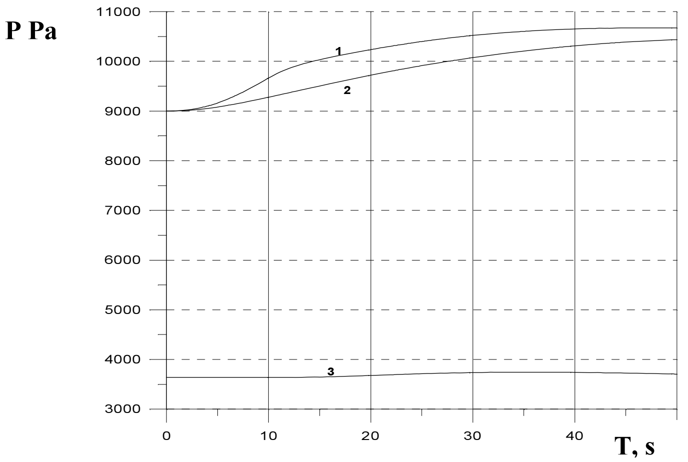


FIG. 7. The pressure change in the space above the core at accidents without scram.

It is evident from the data given in Table 3 that an important feature of the aqueous solution reactor is its ability to self-liquidate power excursions caused by the insertion of a large positive reactivity. This feature has been reliably proved by experiments and used in practice in impulse solution reactors.

The experiments with the positive reactivity insertion were also carried out at the stationary aqueous solution reactor 'ARGUS' [3]. The most dangerous accidents - simultaneous self-withdrawal of the regulation rods without scram - were simulated. The studies demonstrated that even in such cases the self-regulation effects provided the HASR reactor safety. Changes of the 'ARGUS' reactor parameters are given in Figure 8 [1. power, 2. reactivity, 3. solution temperature] in case of the reactivity liner insertion at the rate of $0.09\beta_{ef}/s$. This data was used for verification of the codes used for calculating behaviors of the emergency modes of the SR-RN reactor.

In case of emergency depressurization of the sorption column intended for molybdenum recovery without its disconnection from the reactor the fuel solution is poured from the pipelines into the core. Figure 9 shows changes of the reactor power during the accident under consideration for various designs of the tract.

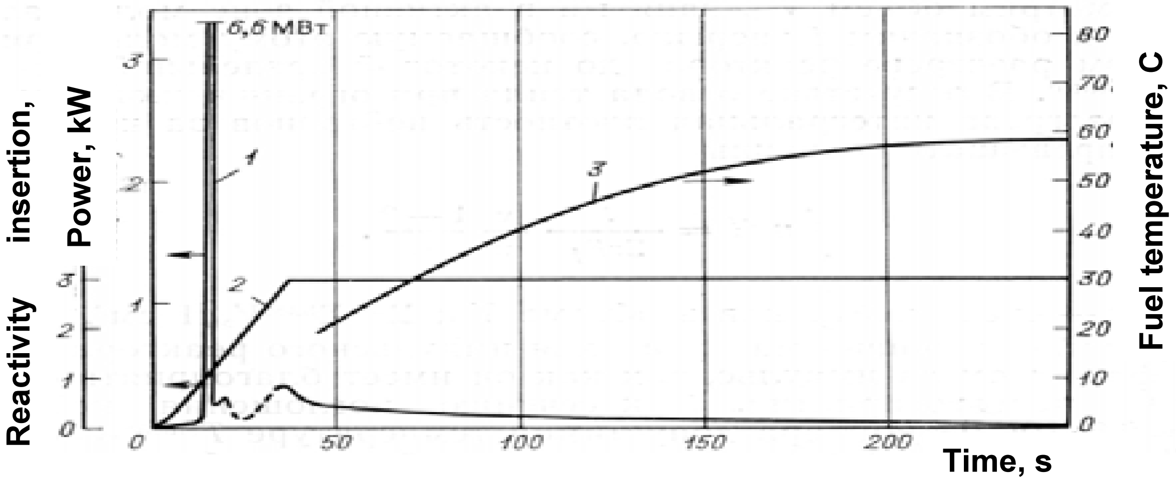


FIG. 8. Changes of the 'ARGUS' reactor parameters in case of the reactivity liner insertion at the rate of 0.09 β_{ef}/s.

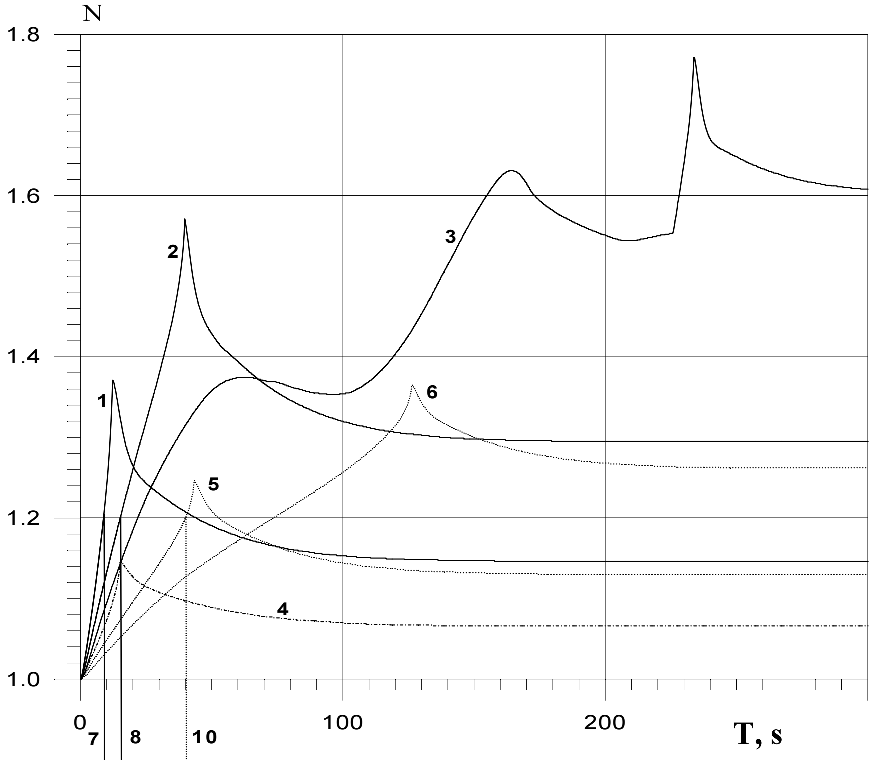


FIG. 9. Relative change of the reactor power during the accident with loss of leak tightness of the sorption columns at 100% power with simultaneous additional failure of the automatic regulator for various designs of the column-reactor tract: 1. pipe of 6mm inner diameter and 10m length; 2. pipe of 6mm inner diameter and 20m length; 3. pipe of 6mm inner diameter and 40m length; 4. pipe of 4mm inner diameter and 10m length; 5. pipe of 4mm inner diameter and 20m length; 6. pipe of 4mm inner diameter and 40m length.

The studies demonstrated that if the length of the column-reactor tract did not exceed 40m (and the pipeline inner diameter was 6mm) the consequences of the column depressurization accident, even without scram, would be limited to the release of gaseous medium into the premise of the column for 20–25 seconds after the termination of pouring of the fuel solution from the tract into the reactor.

The curves 1–6 – without scram. The curves 7–10 correspond to the versions 1, 2, and 5 with scram at the power increase by 20%.

A wide spectrum of other accidents was considered in addition to the above mentioned accidents. No accidents with serious radiation consequences for the personnel and the population have been revealed.

6. ^{99}MO INVENTORY AND RECOVERY

The Complex production capacity greatly depends on its operation mode. Both discrete and continuous operation modes of the ^{99}Mo production are studied. Under the discrete mode the aqueous solution reactor operates at rated power during T_o time period, after that it stops for T_f period needed for the fuel solution 'cooling', filtering into the sorption column, molybdenum washing off and other process operations. Then the cycle is to be repeated.

Under the continuous mode the reactor operates at a rated power, the fuel solution is pumped continuously through the sorption column and after time period T_s molybdenum is washed off the sorption column.

Average weekly ^{99}Mo delivery dependences on the reactor operation time T_o are presented in Figure 10 below with different T_f values under the discrete mode of the Complex operation. The curves 1, 2, 3 correspond to time T_f = 6, 12 and 24 hours. The delivery is proposed on fifth day (in 4 days) after the ^{99}Mo deposition on the sorption column. Data on ^{99}Mo weekly delivery under the continuous reactor operation and at T_s = 24 hours are also presented in the figure for comparison. The reactor facility capacity factor (CF) was taken as 0.9. The data is presented for one reactor facility.

Data on ^{99}Mo production for different versions of the discrete operation mode of the Complex under optimum proportion between T_o and T_f periods is presented in Table 4 below. Data on weekly ^{99}Mo delivery under the Complex continuous operation depending on frequency of molybdenum washing off the sorption column (T_s time period) is presented in Table 5 below.

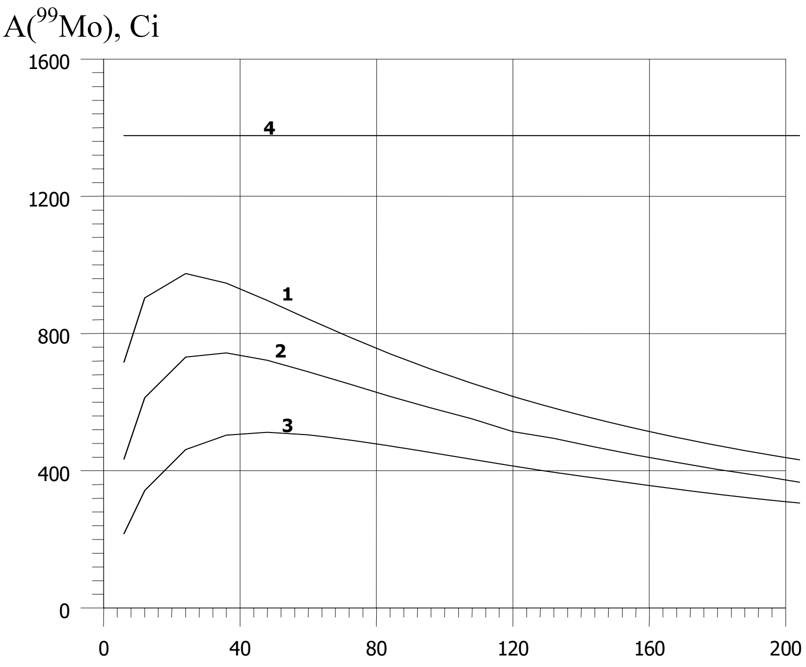


FIG. 10. Average weekly ^{99}Mo production at different operation modes of the Complex (curves 1, 2, 3: discrete mode at Tf = 6, 12, 24 hours; 4: continues mode at Ts = 24 hours).

Table 4. ^{99}Mo production at the discrete operation mode of the engineering Complex (CF = 0.9)

		^{99}Mo activity, Ci		
T_o, hour	T_f, hour	On sorption	In 4 days	
		Column	One shipment	Average per week
24	6	459	167	935
36	12	573	209	732
48	24	593	216	486

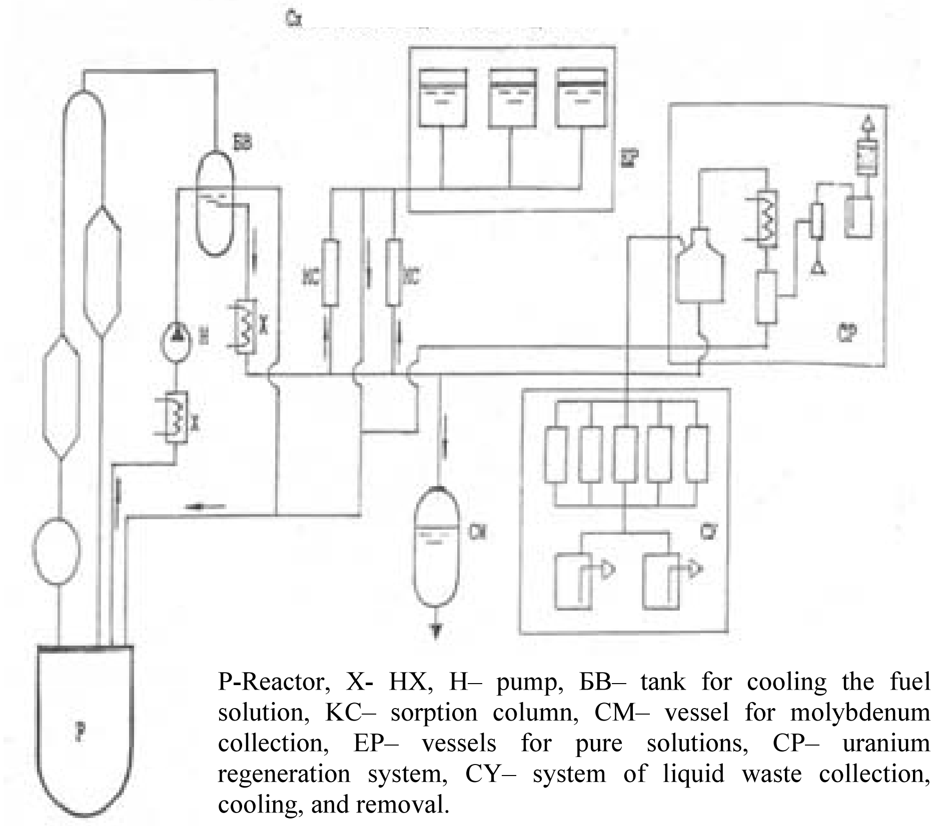
Table 5. ^{99}Mo delivery under the continuous operation of the complex

T_s, days	1	2	3	4	5	6	7
A(^{99}Mo), Ci/week	1350	1205	1076	966	872	791	720

The calculations done demonstrate that the optimum operation mode is continuous operation of the engineering Complex with once per day unloading of ^{99}Mo at each facility.

The ^{99}Mo recovery system provides for two consequential processes: selective adsorption of molybdenum when filtering the fuel solution through the sorbent and selective washing-off of the adsorbed molybdenum after a pre-determined time of the fuel solution filtration. The flow sheet diagram of the ^{99}Mo recovery system is given in Figure 11.

The solution is supplied from the reactor to the column with the sorbent Thermoxide-231 after cooling and without the composition adjustment. Up to 95% of ^{99}Mo and 0.05–0.1% of uranium are sorbed on Thermoxide-231. When the sorption capacity is achieved the column is washed by the distilled water, which is delivered to the liquid waste collector. ^{99}Mo is desorbed by aqueous solution of 0.5M NH_4OH, which volume is ~0.3 liters.



P-Reactor, X- HX, H– pump, БВ– tank for cooling the fuel solution, KC– sorption column, CM– vessel for molybdenum collection, EP– vessels for pure solutions, CP– uranium regeneration system, CY– system of liquid waste collection, cooling, and removal.

FIG. 11. Flow-sheet diagram of ^{99}Mo recovery.

The further affinage of ^{99}Mo is performed by the extraction chromatography method using di-2-ethylhexyl phosphoric acid on a solid carrier (Teflon). It is advisable to perform the operation in a separate box.

The complex of these operations will ensure the production of ^{99}Mo product for medical application, which meets the requirements of a Pharmacopoeia.

After the final analysis the product is dispensed into containers with adequate shield in the unloading box (B(U) type containers, for example, KM-47 manufactured at Czech Republic).

7. RECOVERY OF STRONTIUM-89 AND XENON-133

Strountium-89 and Xenon-133 radioisotopes are recovered from the vapor-gaseous phase of the fuel solution. The isotope recovery systems are connected with the system for catalytic regeneration of water radiolysis products. Strontium-89 as a fission product of uranium-crypton-89 fission fragment is a very valuable product for nuclear medicine. As there is no need to use a 'carrier' in this technology the produced radiopharmaceutical is of very good quality.

The reactor first phase is continuously supplied to the processing line, which includes tanks for cooling for a time required for Kr-89 decay ($T_{½}$=3.15 min.), these tanks are filled up with an aqueous solution for Kr-89 absorption (water, 0.05M HCl). After the processing required the solution is transferred to the box intended for affinage, dispensing and packing of the end products.

The gaseous phase is supplied from the Sr-89 absorption tank to a scrubber filled with NaOH solution for absorbing iodine isotopes. After a certain time period the solution containing the absorbed iodine is transferred to the box intended for iodine affinage, dispensing and packing of the product.

Then the gaseous phase is supplied to a cryogenic trap filled with the activated charcoal for trapping xenon-133. On achieving a required accumulation the trap is heated up to 300ºC and xenon is supplied to a separate box for adjusting its concentration activity and for dispensing.

Therefore, it is desirable to design the arrangement of the whole process in one building with individual work stations for each isotope affinage operations.

8. EXTRACTION OF ISOTOPE MIXTURE: IODINE-131, IODINE-133, IODINE-132 (IODINE-MIX)

The short-lived isotopes – iodine-131, iodine-133, and iodine-132 - are extracted from the vapor-gaseous phase or from the fuel solution and can be used if they are consumed 'rapidly'.

9. RADIOLOGICAL SAFETY

Radiological safety of the SR-RN reactor and the isotope production Complex is ensured by a proven design and layout of equipment, proper organizational measures as well as a provision for multi-barrier radioactive products retention systems:

- Leak-tight design of all equipment excluding radioactivity release to the environment under normal operation.

- Localization of radioactive products inside controller barriers under all realistically conceivable accidents.
- Location of radioactive components and equipment in the hot shielded cells (boxes).
- Special ventilation system with effective clean-up of the air released to high ventilation stack.
- Effective solid, liquid, and gaseous radwaste management system.
- Continuous monitoring of the radiological conditions in the Complex compartments and at site.
- Good established and verified practice is applied to carry out all technological operations.

Safety analysis showed that for all realistic initial events and accident sequences radioactive products are localized under control within design limits. As a result now radiological impact to population and personal is expected. Nevertheless two postulated accidental scenarios (PAS) with essential loss of fuel solution are considered to assess maximal possible radiological consequences of the accidents at the Complex for personal and population:

- Large reactor vessel brake followed by leakage of all fuel solution (20 L) and its evaporation in contact with hot graphite moderator surfaces (PAS_1);
- Pipe brake in sorption column box in the process of fuel solution pumping with failure of the pump to stop which results in 3 l of fuel solution leakage to the box (PAS_2).

Scenario PAS_1 involves the highest radiological impact to population, while PAS_2 scenario gives most serious occupational exposure.

Results of analyses of radiological consequences of the PAS for conditions of deployment of the Complex in the IPPE reactor building 224 are presented in Tables 6 and 7.

Table 6. Population exposure doses

Distance, km	Cloud and Ground Shine (Whole Body), mZv		Inhalation (Child Thyroid), mZv
	10 days	1 year	
0.1	1.0	1.7	--
0.5	0.25	0.34	0.4
1.0	0.20	0.30	1.7
10.0	0.059	0.086	0.61
Regulatory Intervention Level			
– Sheltering			
– Iodine Adm.	5.0		50.0
– Evacuation			100.0
– Relocation	50.0		500.0
		50	

Table 7. Occupational exposure doses

Duration of Inhabitation, min	15
Whole Body (γ-radiation), mZv	120
Skin (β-radiation), mZv	600
Inhalation (Thyroid), mZv	1500

For PAS_1 population exposure in terms of both annual external doses and child thyroid inhalation dose do not exceed regulatory intervention levels for introduction of protective measures.

Results of PAS_2 analysis showed that organizational requirement for exclusion of personal presence in the sorption column box during the process of fuel solution circulation has to be introduced into Complex operating regalement. Access to the box can de allowed after heavy decontamination only.

10. THE DESIGN DEVELOPMENT STATUS

The preliminary design of the Complex consisting of 2 reactors, each of 50 kWth power, includes designs of the reactor and the processing lines: isotope recovery from the fuel solution and isotope recovery from the vapor-gaseous mixture. The development status of these two components of the design differ, and it has been planned to use the existing reactor building and its engineering structures as well as the ready circuit for molybdenum affinage, which is available at the building, the available chemical laboratories and a spectrometric section, and all the necessary engineering networks, which make up 2/3 of the Complex cost by our estimation.

Nowadays a possibility of decreasing the uranium enrichment down to 20% and a possibility of increasing the power up to 80 kW are under consideration.

11. CONCLUSIONS

The design of the Medical Complex on a low power aqueous solution reactor as a basis that is under development and its construction will make it possible to create a modern radioisotope production complex using absolutely new high technologies of recovery of the isotopes that are in the greatest demand directly from the reactor fuel solution.

The distinctive features and the most substantial advantages of the new technology as compared to the traditional target-based technology are as follows:

- use of a low power reactor with a high level of self-protection;
- considerable (by 2–3 orders of magnitude) decrease of the daily volume of radioactive wastes;
- use of almost all ^{235}U in the reactor core for the radioisotope production without target regeneration;
- possibility of production of ^{89}Sr isotope without carrier;

- possibility of application of a low-enriched fuel (<20% in ^{235}U);
- considerable reduction of the construction and operating costs as compared to the traditional target-based technology (the anticipated reduction of the costs of manufacture of the isotopes – by 2÷2.5 times);
- the Complex safety level is considerably higher than that of the existing similar production processes.

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EFFECTIVE METHOD OF ^{99}Mo AND ^{89}Sr PRODUCTION USING LIQUID FUEL REACTOR

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Abstract. Molybdenum-99 is a precursor of technetium-99m applied for diagnostics in the nuclear medicine. About 80% radio-diagnostic procedures in the world are conducted by means of pharmaceutical ^{99}Tc-preparations. ^{89}Sr is used in oncology for anesthesia. As a result we abandon application of anesthetic narcotics. The main method of producing ^{99}Mo and correspondingly ^{99m}Tc is production in the reactor: ^{99}Mo is produced due to fission of uranium-235 nucleus, molybdenum is approximately 6% of fission products. Imperfections of this method are that only target uranium, a small part (~0.4%) of uranium in reactor core, is used for ^{99}Mo production. As a result there are problems: 1) high cost of ^{99}Mo; 2) large quantity of nuclear waste. There are 2 conventional ^{89}Sr production methods: 1) threshold reaction ^{89}Y(n,p) ^{89}Sr needs high-intensive fast neutron reactor and has small cross-section; 2) reaction ^{88}Sr (n,γ) ^{89}Sr for thermal neutrons needs ^{88}Sr enrichment of target and has small cross-section too. Need for high-intensive reactors, fine cleaning of ^{88}Sr target from impurity uranium, ^{88}Sr -enrichment of target cause a low productivity of these methods. Therefore new technologies are necessary for realization such goals:

- to increase uranium economy up to 100%,
- to decrease the necessary reactor power by 100 and more times,
- to use low-enriched uranium,
- to exclude need for target use,
- to decrease quantity of nuclear waste,
- to decrease reactor staff number.

1. NUCLEAR REACTOR ARGUS

The reactor Argus (Figure 1) is unique in the world as a stationary liquid-fuel (solution) reactor. It has operated successfully at RRC ‘Kurchatov Institute’ since 1981 and has great economic and safety indices.

The reactor core consists of aqueous uranyl sulphate solution located in the vessel. Inside the vessel there are mounted vertical ‘dry’ channels: one central channel and 2 symmetric peripheral channels. Inside the vessel there is a coiled-tube heat exchanger too.

The reactor vessel is surrounded with side, top and bottom **graphite reflectors** with the cross section 1300 × 1500 mm and the height 1100 mm.

Gas products of fuel solution radiolysis are regenerated in the catalytic regeneration system. This system consists of a heater, a catalytic H_2-O_2 recombiner and a heat exchanger. This system and the vessel are sealed. Gas mixture flows in the regeneration system under conditions of natural convection.

The system has a condensate accumulator for accumulation of recombined water portion. Later this condensate is used for water flush of the sorption column.

The main parameters of the reactor Argus are shown in Table 1.

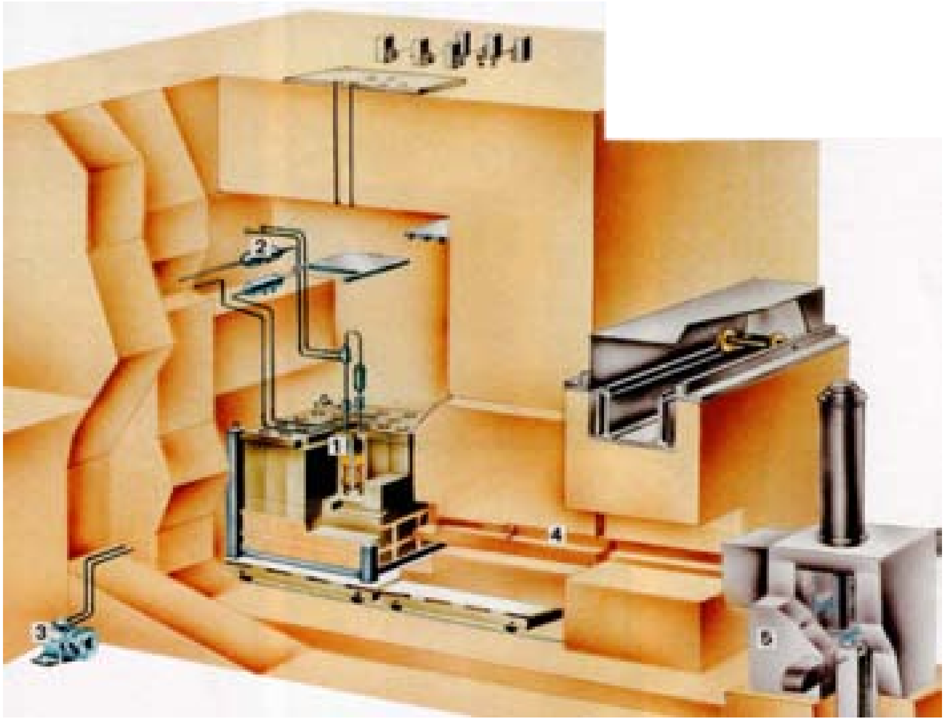


FIG. 1. Reactor Argus.

Table 1. Argus reactor main parameters.

Fuel	Uranil sulphate water solution
Enrichment of uranium	90% U235
U235 concentration	73,2 g/L
Solution volume	22 l
Rated power	20 kW
Thermal neutron flux density	$1/(cm^2 \bullet s)$
– In the central channel	$5 \bullet 10^{11}$
– In the reflector	$(1.0–2.8) \bullet 10^{11}$

2. ^{99}Mo PRODUCTION

We have created a loop to produce molybdenum at the Argus reactor (Figure 2), a process of fuel solution purification, molybdenum separation and molybdenum-99 ready product preparation to produce technetium-99m. The calculated productivity of this system ~80 Ci/week is enough to provide the Moscow region with this isotope.

Demonstration production of ^{99}Mo at the Argus reactor was realized twice. After one operation at 20 kW for five days, there are 708 curies of ^{99}Mo in the 20 liter reactor fuel solution. The growth and decay of this activity as a function of time is shown in Figure 3.

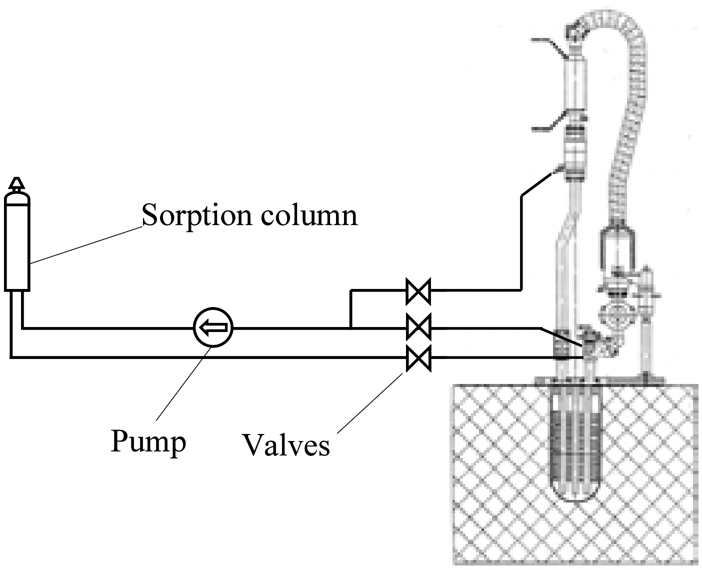


FIG. 2. Reactor Argus with ^{99}Mo loop.

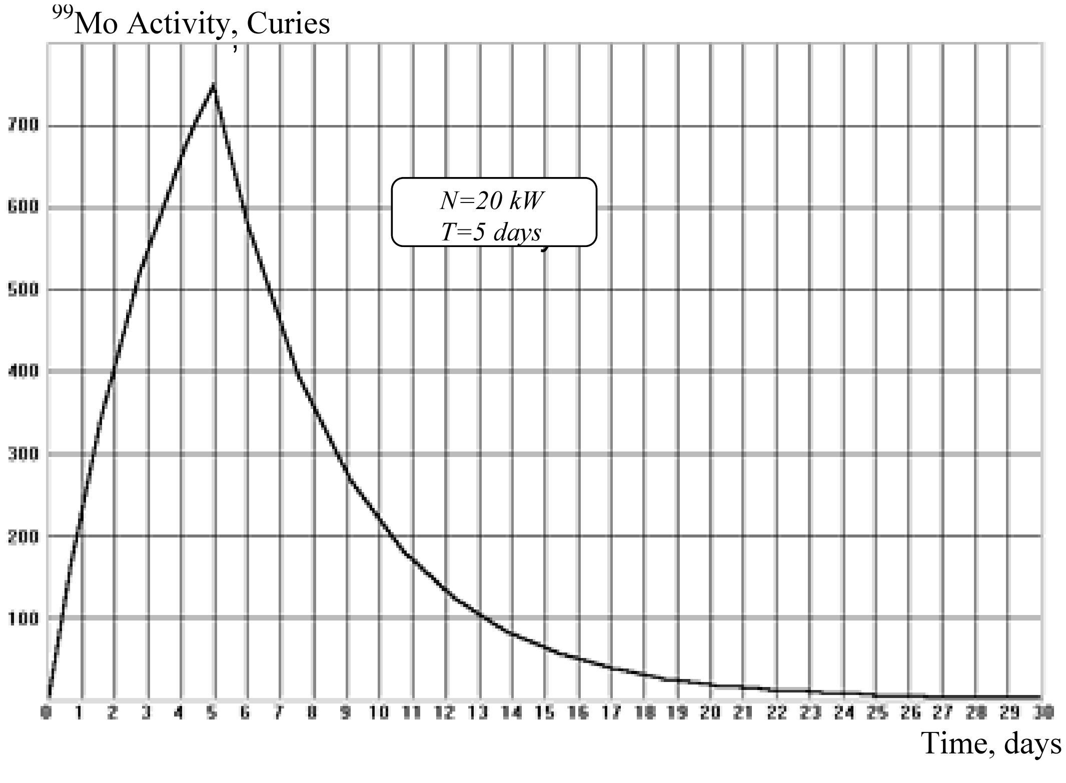


FIG. 3. Relation of produced ^{99}Mo activity and time.

After the reactor has been shutdown and the fission power or radiation has decayed for at least one day, the reactor solution is pumped through a sorption column. The chemicals in the sorption column pass through the uranyl sulphate solution and fission products other than molybdenum. They bind the molybdenum to the sorbent material of the column. This operation takes about 6 hours. As a final step, the column is washed with distilled water. The column containing ^{99}Mo is then disconnected from the extraction loop and placed in a shielded container.

The container with the column is moved to a hot cell. In the hot cell, the ^{99}Mo is recovered from the column with washes and further purified, analyzed and packaged for shipment.

Results of reactor experiments were analyzed by Kurchatov Institute, IPPE, Argonne National Laboratory (USA) and Institute of Radioactive Elements (Belgium). It has been concluded that ^{99}Mo samples from Argus reactor are radiochemically pure, in other words contamination concentrations are lower than safe concentrations.

Results of the ^{99}Mo technology development

- ^{99}Mo extraction and refining process was developed;
- Special equipment for remote operations in hot cells was created;
- Various sorbents were investigated and modified for ^{99}Mo selective extraction from the fuel solution. Inorganic sorbent based on titanium oxide was chosen;
- The demonstration complex for ^{99}Mo production and extraction from the "Argus" reactor fuel solution was created.

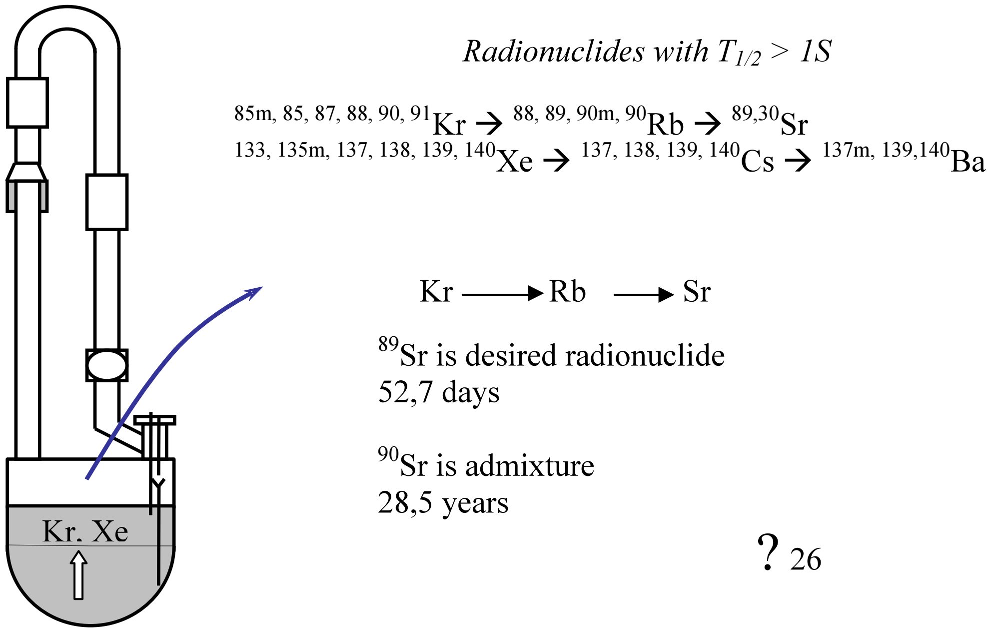
3. ^{99}Sr PRODUCTION

We considered the possibility of ^{89}Sr production by decay of gaseous fission products (^{89}Kr) appearing in the core of the Argus reactor (Figure 4).

The suggested method of the fission radionuclide ^{89}Sr production is based on the effect that its gaseous precursor radionuclide ^{89}Kr goes out of the solution. Experimental studies show that practically all long-lived isotopes of krypton and xenon get to the gas phase of the solution reactors.

As it can be seen in the main decay chains of radioactive strontium isotopes production (Figure 5), the half-life of ^{89}Kr, the gaseous precursor of the end radionuclide ^{89}Sr, is 3.2 minutes and the half-life of ^{90}Sr, the main impurity radionuclide, is much less, it is 33s. Using this circumstance, it is possible to keep and isolate gas volume to achieve the necessary ratio of end and impurity strontium radionuclides gaseous precursors activities and to pump gas mixture through filters to the accumulation reservoir where ^{89}Sr will be accumulated after precursors decay.

Experiments with the following flow diagram were conducted to confirm the possibility of extracting ^{89}Sr from the air mixture of Argus reactor. The reactor operated at power 20 kW for 20 minutes. Then gas mixture was kept for 5–6 minutes and pumped through the column filled with Raschig rings. The ^{99}Mo loop of the reactor was used in the gas-pumping mode for this purpose. Time of gas mixture pumping was 5 minutes. It was enough to fill the column. The column was removed 2–3 days later after gaseous fission products decay. The filling extracted from the column was washed with hydrochloric acid solution to wash out ^{89}Sr and accompanying fission products.



Radionuclides with $T_{1/2}$ > 1S

$^{85m, 85, 87, 88, 90, 91}Kr \rightarrow {}^{88, 89, 90m, 90}Rb \rightarrow {}^{89,30}Sr$

$^{133, 135m, 137, 138, 139, 140}Xe \rightarrow {}^{137, 138, 139, 140}Cs \rightarrow {}^{137m, 139,140}Ba$

Kr → Rb → Sr

^{89}Sr is desired radionuclide
52,7 days

^{90}Sr is admixture
28,5 years

? 26

FIG. 4. ^{89}Sr-production at Argus reactor.

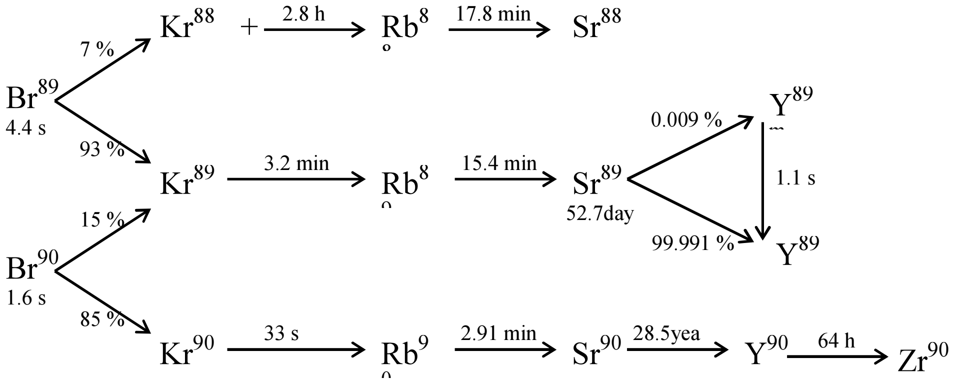


FIG. 5. Decay chains of fission products with atomic mass 89 and 90.

When ^{235}U nucleus decays yield of chain with atomic number 89 is about 5%, so a large quantity of ^{89}Kr and ^{89}Sr is produced. However, necessity to keep air mixture for 5–10 minutes (depending on requirements to purity by ^{90} Sr radionuclide) causes that activity of the end radionuclide ^{89}Sr would reduce too (Figure 6). After 10 minutes ^{90}Kr concentration is much less than ^{89}Kr concentration.

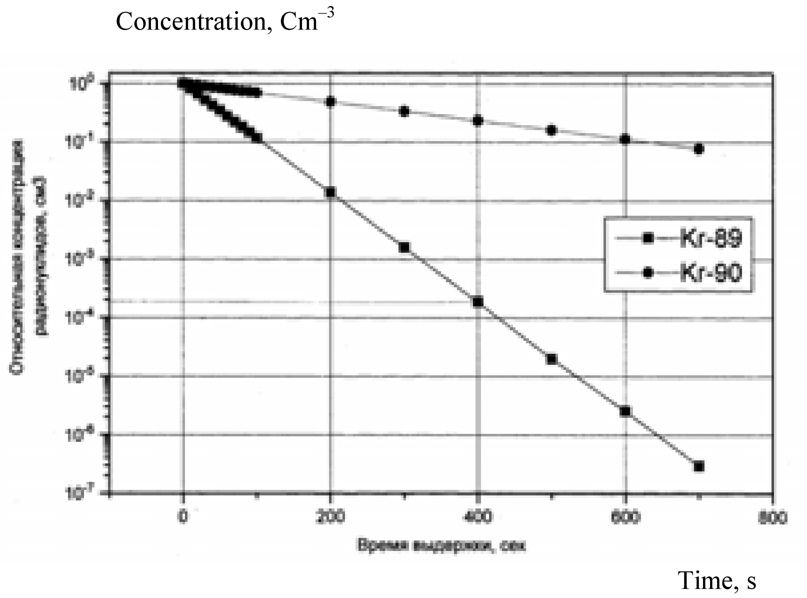


FIG. 6. Decay of ^{89}Kr and ^{90}Kr.

The results of the experiments allowed drawing conclusions as follows:

1. The mechanism of ^{89}Sr delivery to the column is based on its gaseous precursor ^{89}Kr transfer (not its fission product ^{89}Rb or ^{89}Sr);
2. ^{89}Sr activity in the column is 100–1000 times more than activity of contaminating elements with mass 137, 140 etc.;
3. Gas flow filtration very much reduces activity of attendant FP without reducing ^{89}Sr activity;
4. ^{89}Sr purification on chromatographic column with the resin DOWEX 50x8 or the craun-ether Sr-resin very much reduces ^{137}Cs, ^{140}Ba, ^{140}La concentrations in ^{89}Sr chloride solution.
5. Concentration of contamination radionuclide ^{90}Sr in final product ^{89}Sr is less than 5·10–4%.

According to conducted calculations and experiments, the yearly productivity of ^{89}Sr is 3–4 Curies/kW year if air mixture is taken continuously (150 l/h) and if the reactor operates at power 250 days per year.

According to the experiments with single-taken air mixture, the design-experiment value of ^{89}Sr productivity is ~1 Curie/kW year.

We considered a gas loop scheme evacuating the gas-air mixture continuously. It is demonstrated that such a scheme with contnuous evacuation makes it possible to realize the process of ^{89}Sr precipitation.

Productivity of the loop with continuous gas-air mixture evacuation was evaluated. It is shown that the rate of ^{89}Sr production in the loop changes rather significantly (10–30 mCi/kW·day). Meanings of ^{89}Sr production rate reduce as the half-outlet period grows and the flow rate reduces. The necessary delay time for decay of ^{90}Kr (^{90}Sr precursor) is ~12 minutes; then activities relation ^{90}Sr/^{89}Sr 10^{-8} is achieved. If the grade of purification from ^{90}Sr reduces from 10^{-8} down to 10^{-4} the ^{89}Sr production rate will grow about four times and the delay time will reduce approximately twice.

The complex will make it possible:

- To work out the optimum technology, to conduct endurance tests;
- To ensure ^{89}Sr ^{89}Sr production up to 60–80 Ci per a year.

It is expected that the ^{89}Sr production cost by using the Argus reactor will be lower than the traditional methods' production cost.

4. LINES OF FURTHER INVESTIGATIONS

As a result of completed studies we have today the reactor Argus with two experimental loops: first - for ^{99}Mo production from liquid fuel, second – for ^{89}Sr production from gas phase (Figure 7).

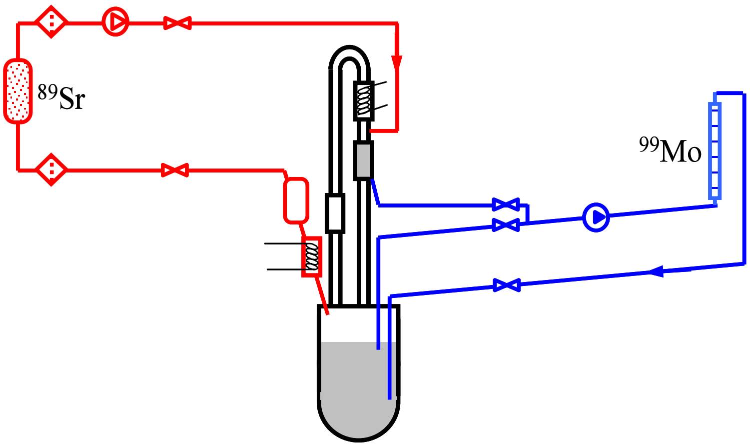


FIG. 7. Reactor Argus with 2 loops.

Lines of further investigations are:

- Optimization of medical isotope production technique;
- Reactor Argus conversion into LEU fuel;
- Creation of nuclear-chemical facility prototype with power about 100 kW

For further design studying we have proposed a concept of nuclear-chemical system of total power 100 kW with two solution Argus type reactors (Figure 8).

Main parameters: Power 2×50 kW = 100 kW Fuel UO2SO4, 20% ^{235}U
Yearly capacity 20 kCi ⇒ ^{99} Mo 250 Ci ⇒ ^{89}Sr

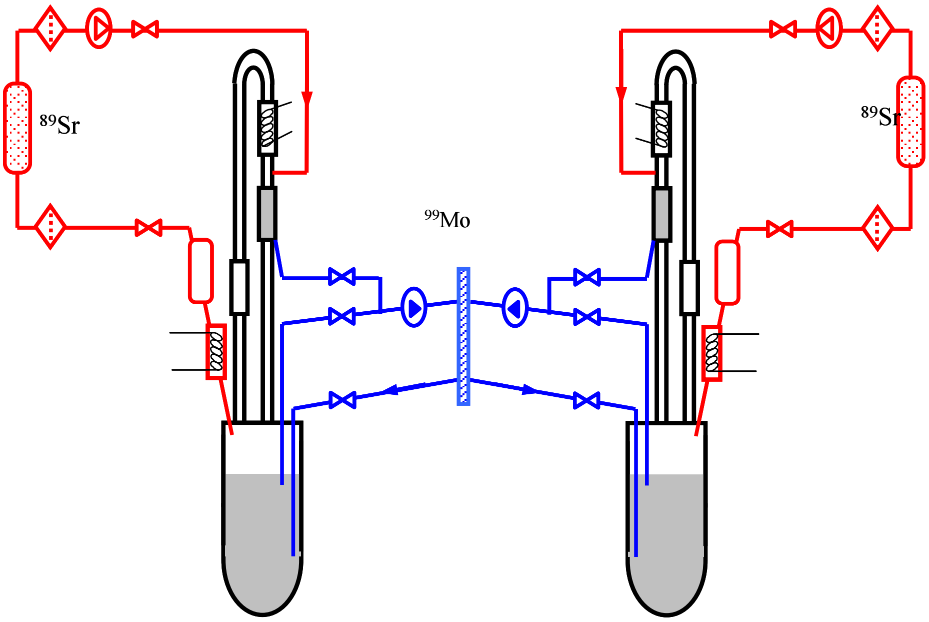


FIG. 8. Nuclear-chemical system of total power 100 kW.

5. CONCLUSIONS

- A liquid fuel reactor enables high-yield extraction of fission radionuclides from reactor fuel solution and gas-air medium.
- An experimental complex was created to develop radionuclide production methods on the basis of Argus reactor.
- The developed technology has advantages over traditional technologies. The main advantages are a gain of uranium economy up to 100% and a considerable reduction of reactor power, radwastes and radiation doses.
- Scientific and technical results make possible starting design of a nuclear-technological complex on the basis of liquid fuel reactor of increased power for radionuclide commercial production.

EVALUATION OF RADSORB, ISOSORB (THERMOXID) AND PZC AS POTENTIAL SORBENTS FOR SEPARATION OF ^{99}Mo FROM A HOMOGENEOUS-REACTOR FUEL SOLUTION

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Abstract. A key to using a homogeneous solution reactor for producing ^{99}Mo is recovering the ^{99}Mo from the fuel solution with high efficiency. Four sorbents were tested for this purpose: alumina (the classic inorganic sorbent for Mo recovery from acidic solutions, PZC (developed by KAKEN for replacing alumina in ^{99m}Tc-generators for low-specific activity ^{99}Mo), and two sorbents specifically designed by Thermoxid for recovering ^{99}Mo from homogeneous reactor fuel solutions. The following results are preliminary, but provide guidance on important parameters for choosing the sorbent, form of the uranium fuel solution (nitrate or sulphate), and the effects of radiation on the stability of the Mo(VI) oxidation state in the solution.

1. EXPERIMENTAL

Materials. ^{99}Mo was obtained by stripping a ^{99m}Tc generator (Bristol-Myers Squibb, North Billerica, MA) with 1.0 M NH_4OH, bringing the eluate to dryness and redissolving in 0.1 M HNO_3. The depleted $UO_2(NO_3)_2 \cdot 6H_2O$ (UNH) was obtained from Argonne's stocks and used without further purification. UO_2SO_4 was prepared by dissolving UNH in sulphuric acid and bringing the solution dryness three times. The Thermoxid sorbents, Radsorb and Isosorb, available as spherical solids of approximately 0.2–0.4 mm in diameter, were used as received from Thermoxid Scientific and Production Company, Zarechnyi, Russian Federation. The polyzirconium compound (PZC) sorbent was used as received from Kaken Co., Ltd., Hori, Mito-shi 310–0903 Japan. The alumina was obtained as Acid Alumina AG4, 100–200 mesh (Bio-Rad Laboratories) and was used as received. All other reagents were analytical grade and were used without further purification.

Batch partioning measurements. The sorption of metal ions by the inorganic ion-exchange sorbents from aqueous solutions was measured by equilibrating a 1 mL volume of a tracer-spiked aqueous solution of appropriate concentration with a known weight of sorbent. A one-hour mixing time was used for the uptake equilibrium measurements. After equilibration, the solution was withdrawn and filtered using a syringe fitted with a 0.2 µm pore size, PVDF Membrane Filter (Fisherbrand). Blank experiments indicate that the filter does not uptake ^{99}Mo. Duplicate experiments indicate that the reproducibility of the measurements was generally within 5%; however, the uncertainty interval may be higher for the highest and lowest K_d values. Good activity balance was observed for all experiments.

Counting of aqueous samples was performed on a Minaxi Autogamma counter (gamma emitters) or a Packard model liquid scintillation counter. ^{99}Mo was quantified by measurement of its 739 KeV and 778 KeV γ-rays. The activity of ^{99}Mo in each sample was corrected for decay.

The extent of radionuclide uptake was expressed in terms of a distribution coefficient, K_d, defined as follows:

$$K_d = \left(\frac{A_o - A_s}{W}\right) \bigg/ \frac{A_s}{V}$$

Here, Ao and As represent the aqueous phase activity (cpm) before and after equilibration, respectively; W is the dry weight of the sorbent (g) and V is the volume of the aqueous phase (mL). The amount of sorbent used was generally kept at 10 (± 1) mg in order to leave a measurable activity in the aqueous phase, this amount of sorbent represents a large excess relative to the amount of radionuclide present.

Uptake kinetics measurements. The rate of radionuclide uptake on the sorbents was measured by equilibrating a series of equal weight (10 ± 1 mg) samples of sorbents with a tracer-spiked aqueous solution. Time zero was taken as the time at which a known volume of the tracer-spiked solution was introduced into the vials containing the sorbent and stirring commenced. At various time intervals, the aqueous phase was withdrawn from a test tube (thus establishing final) and filtered.

2. RESULTS

Kinetics of molybdenum uptake. Figure 1 shows the kinetics of uptake of trace levels of ^{99}Mo by the two Thermoxid sorbents (Radsorb and Isosorb) and PZC sorbents from 0.1 M HNO_3 solution. In all cases, the equilibrium sorption achieved within 60 minutes is far greater than the minimum required for satisfactory performance in a packed column. To yield a suitable retention in a column mode, a Kd of only few hundred is required; therefore, a satisfactory uptake in all cases is achieved in only 15 minutes (Figure 1).

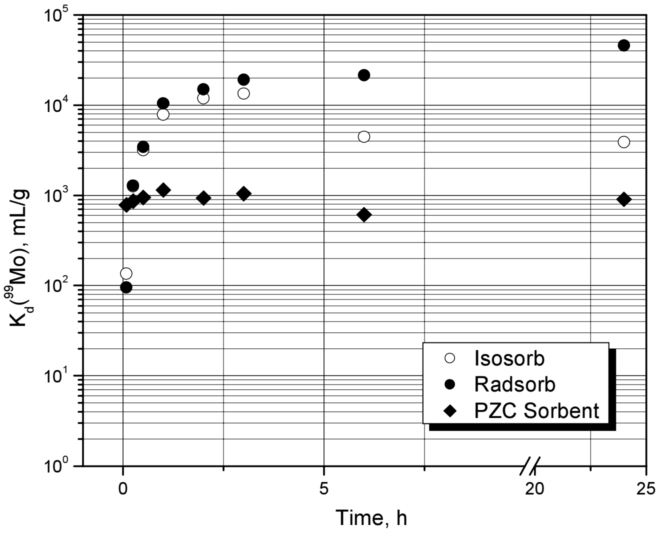

FIG. 1. Partitioning of ^{99}Mo from 0.1 M HNO_3 solutions vs. contact time for the Thermoxid and PZC sorbents.

Evaluation of ^{99}Mo partitioning from HNO_3 solutions. The partitioning (K_d in units of mL/g) of trace levels of ^{99}Mo by the Thermoxid and PZC sorbents was evaluated as a function of nitric-acid concentration (Figure 2). It was found that the uptake of ^{99}Mo at nitric acid concentrations of 0.1 M and lower are practically the same for the Thermoxid sorbents and alumina, whereas uptake of ^{99}Mo by the PZC is significantly lower. At nitric acid concentrations higher than 0.1M, the uptake of ^{99}Mo by the Thermoxid and PZC sorbents is practically the same; whereas, uptake by alumina drops off significantly.

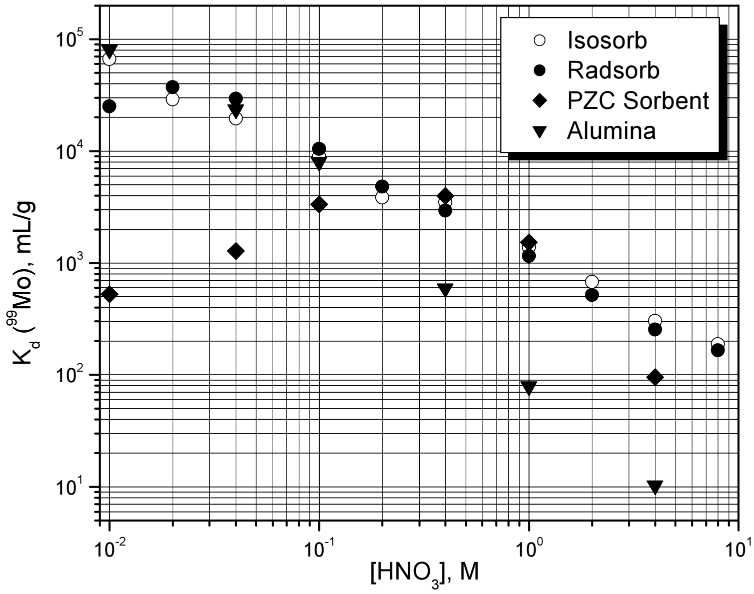

FIG. 2. Effect of increasing nitric-acid concentrations on the uptake of ^{99}Mo by the Thermoxid sorbents, PZC and alumina.

Evaluation of ^{99}Mo separation from uranium nitrate solutions. The efficiency of the sorbents for separation of Mo from solutions containing high concentrations of uranium was evaluated by examining the partitioning of trace levels of ^{99}Mo by Isosorb, Radsorb, PZC and alumina sorbents from 1 M HNO_3 solutions of variable uranium concentrations (Figure 3). It was found that the Thermoxid and PZC sorbents all have a very good retention of ^{99}Mo, up to uranium concentrations of 310 g/L. The K_d values of ^{99}Mo for Thermoxid and PZC sorbents are over an order of magnitude higher than those observed for alumina over the entire range of uranium concentrations investigated. The comparison of the Thermoxid sorbents and PZC reveals the ^{99}Mo is equally well extracted up to uranium concentrations of 150 g/L. At uranium concentrations above 150 g/L, the K_d values for ^{99}Mo by both Thermoxid sorbents are higher than those observed for the PZC sorbent. Retention of ^{99}Mo by alumina from uranium solutions is significantly lower than that observed for Thermoxid and PZC sorbents. The K_d values of <50 over the entire range of uranium concentrations indicate that alumina has a low uptake of ^{99}Mo and is inadequate for separation of molybdenum from a nitrate media based homogeneous reactor containing LEU fuel. Because these data were collected at 1 M HNO_3 rather than at pH 1, take these results with some caution. All K_d values will be significantly higher at pH 1.

Evaluation of partitioning of uranium on Thermoxid sorbents. Figure 4 compares the distribution ratio for ^{233}U to that of ^{99}Mo for the two Thermoxid sorbents vs. the nitric-acid concentration. Uranium has a small but significant K_d value over this range. This distribution and the additional nitrate or sulphate that accompanies the uranyl ion, which also compete with the anionic Mo(VI) species, explain the large suppressive effect that adding uranium salts to the solution has on the K_d for ^{99}Mo.

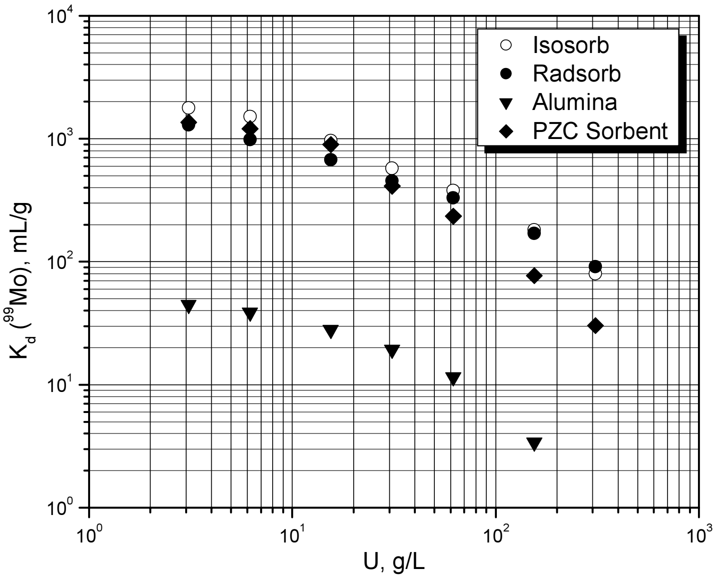

FIG. 3. Effect of varying uranium concentrations on the K_d values of trace levels of ^{99}Mo from 1 M HNO_3.

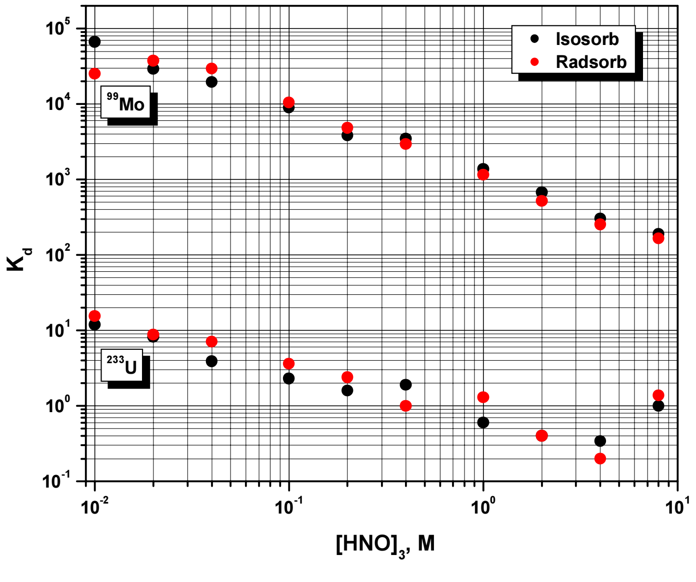

FIG. 4. Effect of increasing nitric acid concentrations on the uptake of tracer-level ^{99}Mo and ^{233}U by the Thermoxid sorbents.

Determination of sorbent capacity. The capacity of a sorbent is an important property because it determines the size of the primary separation/purification column When Kd values are ≥1000. The capacity of the Thermoxid sorbents, Radsorb and Isosorb (0.2–0.4 mm particle size) and PZC (polyzirconium chloride compound) sorbents for Mo from 1 M HNO_3 was tested (Figure 5). The uptake was measured by contacting equal (±10%) amounts of sorbent with nitric acid solutions of increasing molybdenum concentration. Under conditions with no uranyl salts are present, it was found that the PZC sorbent has a capacity of ≥5 meq-Mo/g sorbent. The Thermoxid sorbents, Radsorb and Isosorb have a capacity of ≥2.8 and ≥3 meq Mo/g sorbent, respectively. Sorbent capacity, along with Mo uptake from uranium solutions and kinetic data will be used to determine the size of the primary column needed to separate Mo from a volume of LEU solution.

Evaluation of ^{99}Mo uptake from H_2SO_4 solutions. Figure 6 depicts the uptake of trace levels of ^{99}Mo by the Radsorb, Isosorb, PZC, and alumina sorbents from sulphuric acid solutions. For all sorbents, K_d values of ^{99}Mo from sulphate media are less than from nitrate media; this is explainable by the higher affinity of anionic sorbents for SO_4^{2-} and HSO_4^- anions compared to NO_3^-. Sorption of ^{99}Mo from sulphuric-acid solutions by the Thermoxid sorbents is significantly higher than that by the PZC sorbent. Alumina weakly adsorbs ^{99}Mo from sulphuric acid solutions; making it inappropriate for recovery of ^{99}Mo from a sulphate-based homogeneous reactor fuel.

Evaluation of ^{99}Mo separation from uranium sulphate solutions. To identify the most promising sorbents for the extraction of ^{99}Mo from uranyl sulphate solutions, a candidate media for a aqueous homogeneous reactor, uptake of trace levels of ^{99}Mo from solutions of variable uranium sulphate content at pH 1 by PZC and Thermoxid sorbents was evaluated (Figure 7). It was found that PZC sorbent has low retention of ^{99}Mo when contacted with 10–400 g/L uranium solutions and will not be appropriate for separation of Mo from uranium sulphate solutions. The Thermoxid sorbents have a higher K_d values for Mo (almost 2 orders of magnitude) from uranium-sulphate solutions than does PZC. The molybdenum distribution ratio values for Radsorb and Isosorb from 350 g/L uranium sulphate solutions at pH 1 are 130 and 150 respectively, which suggest that Mo can be separated from uranium sulphate using these sorbents.

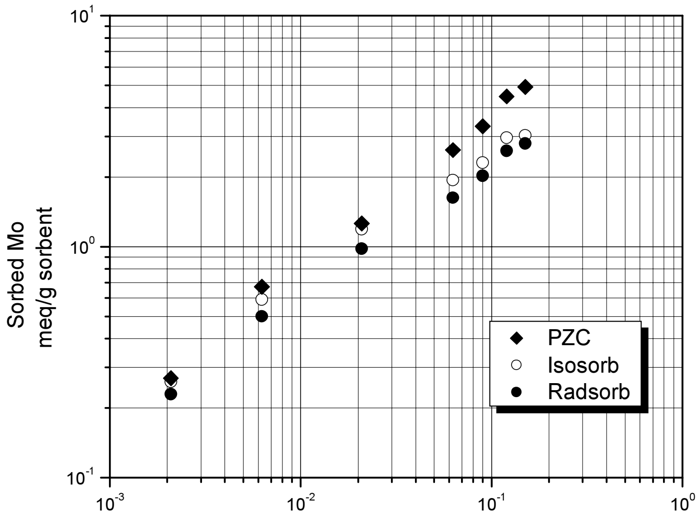

FIG. 5. Capacity of PZC and Thermoxid sorbents for uptake of Mo from 1 M HNO_3 solution.

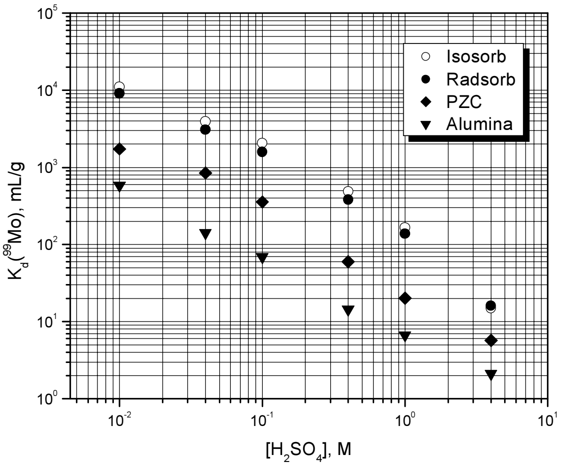

FIG. 6. Effect of increasing sulphuric-acid concentrations on the partitioning of ^{99}Mo by the Thermoxid, PZC, and alumina sorbents.

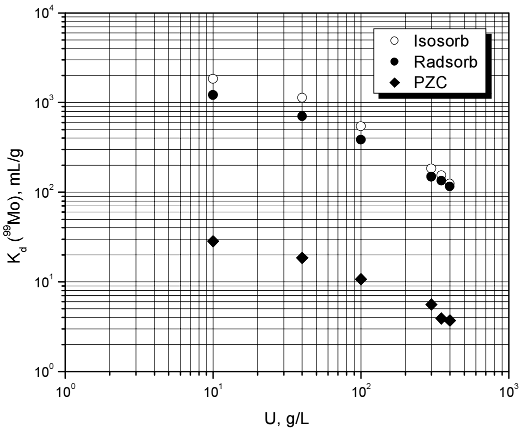

FIG. 7. Effect of varying uranium concentrations on the uptake of trace levels of ^{99}Mo from H_2SO_4 solutions at pH 1 by the Thermoxid and PZC sorbents.

Effect of a radiation field on the sorption behavior of Mo. The effect of fast-electron radiation (3 MeV) on Mo speciation in acidic media and its specific sorption behavior on Thermoxid anion exchange sorbent (Radsorb) has been studied. Solutions of nitric and sulphuric acids with and without corresponding uranyl salts were irradiated in the electron beam at Argonne's Van-der-Graaff generator. The effect of radiation on the oxidation state of Mo is expected to be an important factor for Mo sorption. The solutions irradiated contained 10-mg-Mo(VI)/L, and 300-g-U/L. The initial measured pH was 1.0, and the pH of the solutions did not change during the irradiation. The K_d value for ^{99}Mo was measured for the sorption by Radsorb from irradiated and unirradiated solutions approximately four hours after irradiation (Figure 8). The behaviors of both nitrate and sulphate solutions, with and without uranium were evaluated.

These data show that, after four hours of standing, Mo sorption from nitric acid with no uranium was not affected by irradiation; the K_d from the unirradiated system was 3420, while the K_d from the irradiated system was 3150. As expected (see Figures 2 and 7), the presence of 300-g-U/L decreases the distribution ratios for both irradiated and unirradiated solutions. The K_d is somewhat lower for the irradiated solution (155) than for the unirradiated solution (230).

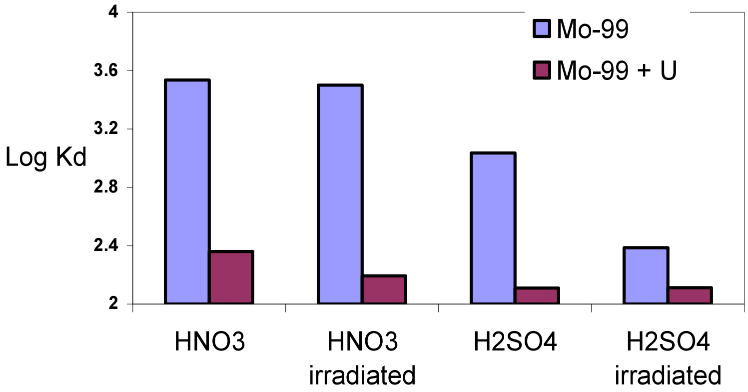

FIG. 8. K_d (Mo) values for ^{99}Mo sorption by the Radsorb in both irradiated and unirradiated nitrate and sulphate solutions at pH 1, with and without 300-g/L U.

The sorption of Mo, in the absence of uranium is less efficient from sulphate solutions than from nitrate solutions. Irradiation caused a decrease of K_d from ~1100 to 240 in the uranium-free sulphate solutions. On the other hand, the K_d values measured for irradiated and unirradiated solutions in the presence of 300 g U/L are identical at about 130.

The decrease of K_d following irradiation in U-free sulphuric acid solutions can be explained by partial reduction of Mo (VI) to Mo (III) or Mo (IV) species, which have low partitioning coefficients on an anion exchanger. In nitric acid, this reduction does not appear to occur under these experimental conditions due to an interaction of a reducing agent with the nitrate or that the Mo (VI) oxidation state was able to re-establish in the time following irradiation, perhaps due to the oxidation of any reduced Mo species by nitrate ion. A similar reaction would not be expected in the presence of sulphate. Further work in this area is required, where partitioning will be measured during irradiation. The four-hour delay between irradiation and measuring partitioning may have given sufficient time for the chemical equilibria to reestablish.

3. CONCLUSIONS

The results of this preliminary study show that use of a sorbent column is feasible to effectively recovery ^{99}Mo from a LEU nitrate- or sulphate-based homogeneous reactor fuel solution. Separation and purification of ^{99}Mo from a nitrate-based fuel solution would be best achieved using either the Thermoxid sorbents (Isosorb and Radsorb) or the PZC sorbent. Separation and purification of ^{99}Mo from a sulphate-based homogeneous reactor solution can only be achieved using Thermoxid sorbents. For all sorbents, nitrate-based fuels will always provide a more effective and efficient recovery of ^{99}Mo from the fuel solution.

ACKNOWLEDGEMENTS

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BWXT SERVICES MEDICAL ISOTOPE PRODUCTION SYSTEM STATUS

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Abstract. BWXT Services, Inc. (BWXS) a subsidiary of BWX Technologies, Inc., one of the Babcock & Wilcox companies, has for some years conducted small scale development of its patented uranyl nitrate Medical Isotope Production System (MIPS). This application of an Aqueous Homogeneous Reactor (AHR) for isotope production was invented by BWXT's Dr. Russell M. Ball in 1997. The MIPS approach targets commercial scale production of medical isotopes, primarily ^{99}Mo, used to supply ^{99m}Tc for diagnostic imaging purposes, using a Low Enriched Uranium (LEU) salt solution as the fuel/target. Recent heightened emphasis on controlling use of Highly Enriched Uranium (HEU) throughout the world in support of nuclear non-proliferation goals has motivated increased commercial interest in this safe and cost effective technology. Conceptual designs for commercial application have been developed and efforts are under way to establish a partnership with an established pharmaceutical firm familiar with current ^{99}Mo production technology and operating practice.

1. FEASIBILITY

AHR's have a long history dating from the beginning of the modern nuclear reactor programme in the 1950's. Over 30 AHRs have been built throughout the world, accumulating over 149 years of combined experience. Two operating licenses have been granted by the US Atomic Energy Commission, and numerous programmes have been established to study criticality in uranium solutions. More recently, the Russian ARGUS reactor programme, in operation since 1981, has demonstrated the successful production of ^{99}Mo and separation to US Food and Drug Administration purity requirements.

Current systems and experience with the conventional ^{99}Mo production methodology, irradiated target-dissolution-separation-purification, are applicable to separation and purification of ^{99}Mo from AHR solution. BWXS's uranyl nitrate solution is very similar to those current systems which dissolve targets in nitric acid. Recent research at Argonne National Laboratory provides insight into performance of new inorganic sorbents.

2. BWXS CONCEPTUAL DESIGN

Current concepts under consideration include a 200 kW reactor, capable of producing approximately 1,100 six day Ci/week of ^{99}Mo and other useful isotopes. An existing containment structure formerly housing a pool type research reactor at the BWXT facility in Lynchburg, VA, is under consideration for an initial commercial facility (Figure 1). The reactor would contain approximately 150 L of LEU solution and would operate at approximately 80°C and atmospheric pressure. A new separation/purification facility is envisioned with hot cell capacity for the several separation/purification/packaging and shipping functions as well as a waste management facility.

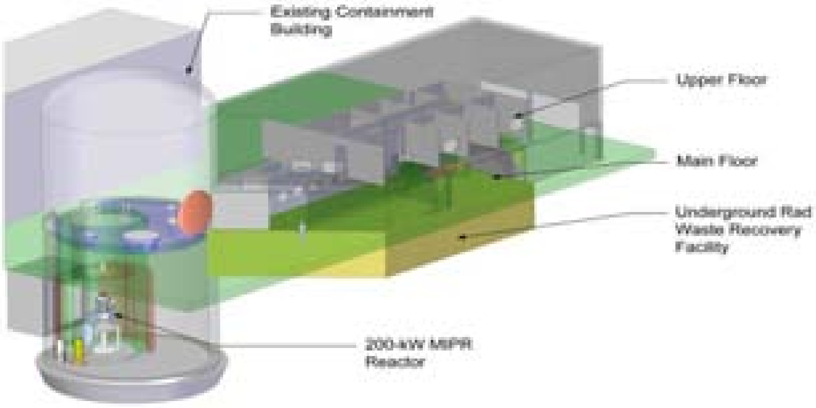

FIG. 1. BWXT commercial facility.

3. PROJECT MANAGEMENT

The project management process established for this programme draws from the US Department of Energy Line Item management approach, incorporating a series of Critical Decision milestones (Figure 2). Each milestone reflects developments to that point which will be reviewed by a select management committee for authorization to proceed to the next phase. The planned development programme anticipates 5 to 6 years duration from full funding to operation. The schedule (Figure 3) indicates. Licensing by the US Nuclear Regulatory Commission and approval to supply radiopharmaceutical products by the US Food and Drug Administration are expected to be major, possibly critical path, activities.

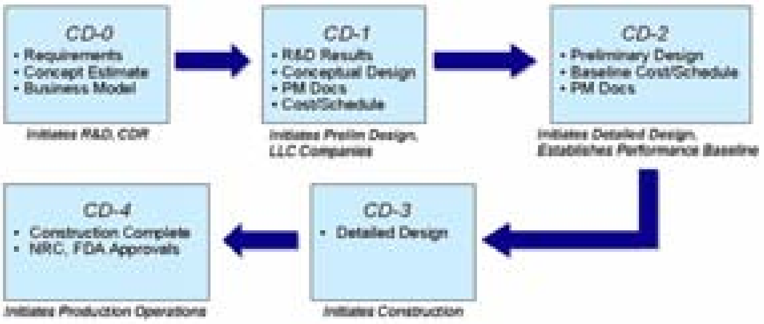

FIG. 2. Series of critical milestones.

FIG. 3. The schedule.

4. LICENSING APPROACH

Solution reactors are not specifically addressed in US regulations, which cover nuclear reactors for power, commercial or research uses. All current non-power reactors in the US are licensed as Class 104 Research facilities. Since a dedicated medical isotope production facility cannot meet the definition of ‘Research’ facility, consideration under Class 103 Commercial facility seems to be the logical alternative. However, specific guidance for such a facility will require further interaction with US NRC. Guidance such as that found in NUREG 1537, Licensing of Research and Test Reactors would seem to be an appropriate methodology.

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