Use of accelerator based neutron sources



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FOREWORD

With the objective of discussing new requirements related to the use of accelerator based neutron generators an Advisory Group meeting was held in October 1998 in Vienna. This meeting was devoted to the specific field of the utilization of accelerator based neutron generators.

This TECDOC reports on the technical discussions and presentations that took place at this meeting, and at the same time reflects "present status" of neutron generators. Although the participants represented 14 MeV neutron generator laboratories, the conclusions and recommendations are of general validity.

This TECDOC will be useful to those involved in the use of accelerator based neutron generators in different laboratories with different equipment. It will be of special relevance to persons developing new applications for neutron generators in their laboratories.

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CONTENTS

SUMMARY	1
A review of accelerator based neutron sources and their applications	5
Utilization of a pulsed D-T neutron generator	15
Use of a 14 MeV neutron generator for activation analysis	27
Research and development activities of a neutron generator facility	35
Utilization of a sealed-tube neutron generator for training and research	45
Problems related to the optimal use of accelerator based neutron generators	53
Recent works with sealed tube neutron generators	59
LIST OF CONTRIBUTORS	65



SUMMARY

In the past, the IAEA supported the establishment of some twenty-five 14 MeV neutron generator laboratories worldwide. At least in 15 cases, the generator itself has been supplied through the IAEA. About 10 machines are known to be in operation and about six to eight of these are in active use, mainly for teaching and research. Active laboratories with their own developed neutron generators are, for example, the Oktavian facility at Osaka University and the JAERI in Japan, FEI in Obninsk Russian Federation, TU Dresden in Germany, and the Rotating Target Neutron Generator Facility in the USA. In addition, there are many active laboratories in Germany, United Kingdom, the Slovak Republic, South Africa, etc. These neutron generators are used for extensive programmes covering the acquisition of basic nuclear data as well as a number of technological applications.

In order to promote the optimal use of 14 MeV neutron generators, a meeting was convened in Vienna in November 1996. This resulted in the issue of IAEA-TECDOC-913, "A Manual for Troubleshooting and Upgrading of Neutron Generators". This Advisory Group meeting was dedicated to reviewing the present status and applications of 14 MeV neutron generators and other accelerator based neutron sources. Most of the participants have experience mainly in the field of 14 MeV neutron generators. Papers presented by the participants are reported here in their original form.

The following pages summarize the discussions held at the meeting on the current status of accelerator based neutron generators, their use and application in scientific research and technology.

1. Current status of 14 MeV neutron generators

Many laboratories have active programmes in teaching and research based on the use of 14 MeV neutron generators. The most active laboratories, e.g. Osaka, JAERI or Obninsk, use their 14 MeV neutron generators in continuous and pulsed mode to do activation measurements and double differential cross-section measurements. The IAEA has assisted, in the past, several laboratories throughout the world. Around ten of these generators are known to be in operation and roughly six of them are in active use. They are used mainly for teaching and research.

Many laboratories in developing countries have active programmes in teaching and research. The Chiang Mai University in Thailand has published over twenty papers based on the use of their neutron generator. In Indonesia more than 22 undergraduate and post-graduate theses have been produced. The neutron generator in Morocco is used for research and post-graduate education. The neutron generator in Peru was in use for routine analysis for industry, particularly for the determination of nitrogen and phosphorus in foodstuffs, until a major breakdown in 1991. The Kossuth University in Debrecen, Hungary, has constructed three different neutron generators; in addition, it is also regularly using cyclotron and tandem accelerator based neutron sources with deuterium gas targets. The number of scientific publications and different theses on the work carried out at this laboratory is over two hundred. At the IAEA sponsored Conferences on Nuclear Data for Science and Technology, held every third year, there have been significant contributions from more than 6 neutron generator laboratories in developing countries. 14 MeV neutron generators are used in a number of other countries; to give one example, the Octavian facility at Osaka University in

Japan has a very extensive programme covering the acquisition of nuclear data as well as a number of technological applications.

There are several reasons why some neutron generators are not fully utilized. These include lack of infrastructure, such as appropriate radiation shielding and loss of adequately trained technical and academic personnel. Much of the equipment is relatively old and there is a serious lack of spare parts. In a few cases there is a critical lack of locally available knowledge and experience in the field of accelerator technology. An Internet website, the Neutron Generator Club, seems to be beneficial in establishing links between neutron generator users in order to help in exchanging technical and scientific information.

2. Review of current research activities and applications

The 14 MeV neutron generators manufactured originally for neutron activation analysis (NAA) are utilized for nuclear structure and reaction studies, nuclear data acquisition, radiation effects and damage studies, fusion related studies, neutron radiography, in nuclear safeguards as well as the use of microsecond pulsed techniques for borehole logging. The traditional applications are the NAA of N, O, Mg, Si, Al, P, Fe, Cr, the analysis of light elements by the detection of neutron induced prompt radiation (gamma, inelastically scattered neutrons or charged particles). The in vivo determination of Ca, Na and N in human beings and the nanosecond pulsed prompt gamma analysis are quite new fields of applications. The contraband and explosive materials detection, on-line analysis in the cement and coal industry by the utilization of small size neutron generators are new fields. New results related to the bulk and assay analysis of geological samples and fast neutron backscattering analysis for carbon, nitrogen and oxygen are promising fields of application.

The small accelerator based neutron sources are useful not only for basic nuclear research and chemical analysis but also for contraband detection, resonance neutron radiography and accelerator based boron neutron capture therapy. The pulsed beam prompt gamma ray for contraband detection/identification is an example of the use of a tandem accelerator based neutron source. Accelerator based neutron sources allows the use of isomeric states for on-line analysis, the determination of hydrogen concentration and depth profiles, neutron therapy and dosimetry, and the transmutation of nuclear waste. They are useful tools in the nondestructive and non-intrusive analysis of chemical content inside abandoned ammunition. This may also give the possibility for identification/detection of land mines.

Treatment protocols for neutron therapy are in use at various centers. Spallation neutron sources are used in routine investigations of material structures using inelastic scattering and neutron diffraction. Neutron sources based on RFQ linacs enable high neutron yield through higher energy protons and deuterons at high currents. Electron linacs are utilized for photo-neutron production.

3. Technical advances

Recent technical development related to the 14 MeV neutron generators can be observed in the field of sealed tube neutron generators; long lifetime neutron tubes (thousands of hours), short pulse length and miniature sized neutron tubes.

There are technical advances related to production of monoenergetic neutrons by heavy ions. Continuous work is done on target developments, especially in the field of windowless gas targets. The neutron yield is increased by high current cyclotrons where higher neutron yield comes through higher energy protons and deuterons at high currents. The use of spallation sources shows some benefit compared with other accelerator based neutron sources.

4. Emerging trends

The new measuring methods in the utilization of neutron sources are the depth profiling of hydrogen by backscattering and proton recoil; the imaging using resonance neutron radiography or associated particle method. This requires further development of the associated particle method for D (d,n) neutrons. The pulse-height response spectrometry became standard method for neutron spectrum measurements. A new method is the biological applications of the multiple foil activation technique (in neutron dosimetry) and the directional proton recoil neutron spectrometry. The gem stone enhancement by accelerator based neutron induced radiation damage is an interesting new application of accelerator based neutron sources.



A REVIEW OF ACCELERATOR BASED NEUTRON SOURCES AND THEIR APPLICATIONS

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Abstract

Accelerator based neutron sources or neutron generators have a long history in the application of nuclear techniques. It has been clear from the discovery of the neutron that there are many useful potential applications that could use neutrons. These range from the determination of elemental composition using neutron activation analysis to neutron radiography and neutron scattering in the characterization of materials. In mineral exploration, and more particularly in oil exploration, sealed tube neutron generators have found an important niche. Techniques such as neutron thermalization activation and neutron die-away are all routinely applied in these fields. These generators usually make use of tritium targets and deuteron beams in the reaction $T(d,n)^4He$.

1. INTRODUCTION

Neutron activation has applications in laboratory analysis as well as in on-line determinations with the potential for process control, sorting and the monitoring of grade in mineral processing. Both the conventional and the prompt gamma forms of activation analysis have important roles here. Perhaps the most important factor that has frustrated the more widespread application of neutron based techniques is the limited intensity that is available with commercially available neutron sources. This applies to both isotopic neutron sources and to commercial accelerator based neutron sources or neutron generators.

Isotopic sources using alpha particles emitters and beryllium are usually limited to outputs in the region of a few times 10^6 n s⁻¹ into 4π and even ²⁵²Cf sources that are available commercially typically do not exceed the few mg range, with total outputs of only 10^9 to 10^{10} n s⁻¹ into 4π . These isotopic sources have fundamental limitations in terms of the maximum practical intensity obtainable in terms of factors such as price, the storage of the helium gas emitted (in the case of Am-Be), shielding and transportation. For these reasons such sources will probably always be of limited usefulness and useful applications making use of neutrons in small laboratories will depend on accelerator based sources, i.e. neutron generators.

2. NEUTRON PRODUCING REACTIONS

There are a number of reactions that can be used to produce neutrons with a charged particle beam. These are described in briefly in the following paragraphs.

 $T(d,n)^4H$

This reaction has a large resonance at low deuteron energy. For this reason it was used in the classical version of the "neutron generator", i.e. a low energy (often about 150 kV) positive ion (deuteron) accelerator with a target consisting of tritium occluded in titanium on a

copper backing. The accelerator was usually an electrostatic machine and was capable of accelerating up to a few mA of beam. In practice, it was often not possible to achieve this level of target current because of ion source limitations and, particularly, because the target could not withstand the heating at high beam currents and as a result the lifetime was severely limited.

This reaction has a Q value of 17.6 MeV and this approach produces the 14 MeV neutron generator. The outputs of these machines were usually practically in the region of a few times 10^9 into 4π . Although much higher outputs were often claimed for these machines it was not as a rule possible to achieve them as a matter of routine because of limitations on target lifetime (as was mentioned above). Another problem in the use of this reaction with normal pumped accelerators is the hazard posed by the release of tritium from the target. For this reason sealed tube generators have been developed and have found numerous applications in borehole logging. With these machines outputs of 10^8 n s⁻¹ are routinely achieved but I have the (subjective) impression that there are severe problems in pushing the technology to 10^{10} n s⁻¹ into 4π . Although a number of attempts have been made I am left with the impression that these machines are expensive to re-furbish and maintain and are not reliable.

The high energy of the neutrons means that many reaction channels are open. Although this has proved important in certain cases (e.g. the determination of oxygen) it can also prove to be a grave disadvantage because interference proliferate.

$D(d,n)^3He$

This reaction is not particularly useful at very low deuteron energies (at the energies used in conventional neutron generators) the yield is about two orders of magnitude less than the $T(d,n)^4$ He reaction) but it has great advantages at higher deuteron energies. With a gas target this reaction is capable of reasonably high outputs. For example take a 5 cm thick target at 3 bar, with 5 MeV deuterons incident on it. The thickness of the target is about 900 keV and the neutron output should be about 1.7×10^{10} n s⁻¹ sr⁻¹ in the forward direction. This is at 300 kV with 100 μ A of target current.

The neutrons would have an energy spread of about 900 keV from 7.3 to 8.2 MeV. This output is enhanced in the forward direction (forward output is about 5 times the output in the backward direction) because of kinematical effects (so-called kinematic collimation). It is equivalent to the forward intensity from a conventional (isotropic) output of about 10^{11} n s⁻¹ into 4π . In order to achieve the equivalent of the "magic" figure of 10^{12} n s⁻¹ into 4π we would need a target current of 1 mA under the same conditions.

At an accelerator energy of 2 MeV, the differential laboratory cross-section in the forward direction has dropped from 76 mb to 43 mb so the output would be about half (57%) at the same beam current. In this case the maximum neutron energy would be 5.2 MeV while the energy spread would be greater for the same target thickness.

9
Be(d,n) 10 B

In contrast to lithium, beryllium has characteristics that favour its use as a target. It has one of the highest melting points of the light metals (1280°C) and has excellent thermal conductivity. It is also chemically inert in comparison to lithium. The reaction ⁹Be(d,n)¹⁰B is

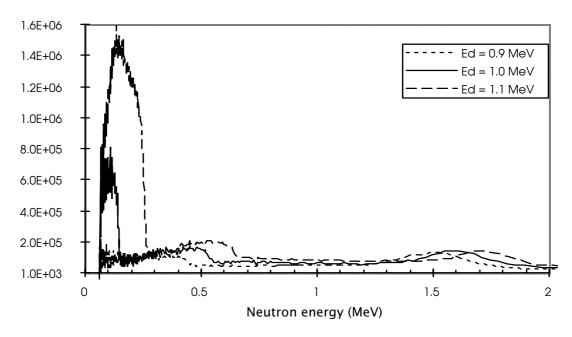


FIG. 1. New data for neutron energy spectra (0.1 to 2 MeV only shown), for deuteron energies close to the threshold for excitation of the 5.11 MeV state and showing the relative intensity and growth of the low energy spectrum

one of the most prolific reactions for low energy accelerators and standard curves [1] show that the neutron output from this reaction is second only to Li(d,n) for incident particle energies between about 1 and 3 MeV. For these reasons this reaction is widely used for the generation of neutrons using small positive ion machines, such as cyclotrons and RFQ accelerators.

Since ${}^9\mathrm{Be}(\mathrm{d,n}){}^{10}\mathrm{B}$ is strongly exoenergic, with a Q value of 4.36 MeV, the deuteron accelerator with a beryllium target is normally considered primarily as a source of fast neutrons. In reality the situation is more complicated and under certain circumstances this reaction may offer itself as a relatively intense source of fairly slow, easily thermalized neutrons. This is because of the intense neutron groups produced when the levels at 5.1 to 5.2 MeV in ${}^{10}\mathrm{B}$ become energetically accessible.

Figure 1, above, shows neutron spectra measured by the time of flight technique for this reaction at three different deuteron energies. The measurements were carried out by J. Guzek and they show clearly the growth of a strong low energy neutron group as the deuteron energy is increased through 1 MeV [2]. This could be of interest in the development of an intense source for boron neutron capture therapy (BNCT)

⁷Li(d,n)⁸Be

This reaction has a very high positive Q value (15.03 MeV), and data obtained by J. Guzek with a thick lithium target show two strong high energy neutron groups, together with a lower energy set of groups that appear when the deuteron energy exceeds about 3 MeV. This is a prolific reaction for the production of neutrons but it has the disadvantage of using a lithium target with the associated problems of oxidation and low melting points.

7 Li(p,n) 7 Be

This reaction is known as a source of monoenergetic neutrons when a thin target is used. It has a threshold of 1.7 MeV and it can be used as a source of slow neutrons. This source has been investigated for the selective activation analysis of isomeric states through neutron inelastic scattering with particular reference to the excitation of the reaction 197 Au(n,n') 197m Au in ores, i.e. in the presence of silicon, aluminum and oxygen [3]. This was carried out using an EN tandem Van de Graaff accelerator over the incident proton energy range from 4 to 10 MeV for two different lithium target thickness. The best limit of detection was found to occur for protons with an energy of about 6.5 MeV with a target thickness of 800 mm. At a current of 1 mA this was about 100 ppm, so with a current of 100 μ A it would be of the order of 1 ppm [4].

As it was mentioned previously lithium has severe disadvantages as a target for large beam currents. It would be necessary to have target current of at least a 100 μA at 6 MeV say to obtain a useful limit of detection. This would need a cyclotron or a linac and would require the solution of the problem of cooling the lithium target to prevent melting by the 600 W beam power.

7 Be(p,n) 7 B

This reaction has also recently been studied in some detail by J. Guzek. It can be used as a quasi mono-energetic fast neutron source in the proton range below 4.5 MeV. In the work by Watterson et. al. this reaction was found to be as useful as ⁷Li(p,n) for the production of nuclear isomers.

3. TYPES OF ACCELERATORS

Sealed tubes

These are usually low energy accelerators using deuteron beams and deuterium or tritium targets. They have been very successful for low outputs but attempts to produce high intensity versions have been relatively unsuccessful. They do not appear to be reliable and are expensive to maintain. For example the tube must be returned to the factory for refurbishment.

Electrostatic accelerators

At low energies and for restricted applications these machines have been fairly successful. Many early neutron generators were of this type using Cockcroft Walton power supplies with energies of say 150 kV. Attempts to produce high current electrostatic accelerators have not met with much success as yet but research and development is still under way.

Cyclotrons

It appears that these machines are capable of accelerating fairly high currents. They are manufactured by firms such as IBA in Belgium, General Electric, CTI and Ebco and are available for a variety of energies from about 3 MeV to about 30 MeV. Of course specially



FIG. 2. An example of a neutron generator based on RFQ technology. The LANSAR accelerator (Courtesy of AccSys Systems).

built cyclotrons operate up to energies of several hundreds of MeV but these are a different kind of accelerator, appropriate only for a large national laboratory.

Linear accelerators

Radiofrequency Quadruple (RFQ) linear accelerators or Drift-tube Linacs or a combination of the two are potentially a very good solution to the delivery of large beam currents at energies of a few MeV. A system based on an RFQ accelerator is shown in Fig. 2.

4. TARGETS

One of the most critical limitations in the development of intense accelerator based neutron sources is the target. The development of appropriate targets for high intensity beams is not trivial. For example a 1 mA beam at an energy of say 2 MeV dissipates some 2 kW of power in the target. This may be dissipated in an area of a cm² or so and it poses a considerable problem for target design, even in the case of beryllium. Originally the targets for 14 MeV neutron generators consisted of tritium (or deuterium) adsorbed in a titanium layer on a copper backing. This type of target posed several problems. For example it could not withstand high beam currents and its lifetime was severely limited even under normal use. An additional problem with drift tube accelerators was that the vacuum pumps became heavily contaminated with tritium. With the exception of the contamination problem the same considerations apply to deuterium targets. In this case however the deuterium may be replaced to some extent by the implanted beam.

Gaseous targets present particular advantages for high neutron outputs because the heat can be dissipated by circulation and cooling of the gas. However where a window is used there is a problem because of the power dissipated in the material of the window — leading to

failure of the window. Research in South Africa has been carried out on cooled windows by C.B. Franklyn. This improves the performance but does not appear to provide a final solution for high beam currents.

In recent years considerable research and development has been carried out on windowless gas targets both at MIT [5] and at De Beers in South Africa. De Beers have developed a windowless system that is capable of sustaining a pressure of 3 bar in the target. This has been used to produce a neutron output of over 1×10^{10} n s⁻¹ sr⁻¹ in the forward direction (equivalent to an isotropic output of over 10^{11} n s⁻¹) using a 5 MeV RFQ linear accelerator of the type shown above.

An even more recent promising development is the plasma window. This was originally developed at Brookhaven National Laboratory by A. Hershcovitch [6] for electron beam welding and other applications. Its application to gaseous targets for accelerators is being investigated in a collaboration between R.C. Lanza of MIT, De Beers and our group at the Schonland Center. These developments may well revolutionize the production of neutrons with intermediate energy accelerators.

5. NEUTRON OUTPUTS

The following Table 1 was compiled by J. Guzek and it shows that high neutron outputs are theoretically possible. It would however be interesting to see how many installations there are that regularly achieve these outputs and apply them routinely.

6. APPLICATIONS

The potential applications of accelerator based neutron sources are numerous. The following shows a partial list of actual and potential applications.

TABLE 1. HIGH OUTPUT NEUTRON GENERATORS

Model	Company	Type of accelerator	Accelerated particle	Beam energy (MeV)	Max. beam current (µA)	Neutron output (n s ⁻¹ into 4 π)
TN46	Sodern, France	D-T Tube	d	0.225	10000	4×10^{11}
DL1.5	AccSys, USA	RFG Linac	d	1.5	325	1×10^{11}
PL4	AccSys, USA	RFG Linac	p	3.9	1000	1.3×10^{12}
Cyclone 3D	IBA, Belgium	Cyclotron	d	3.8	2000	5.7×10^{12}
Cyclone 18+	IBA, Belgium	Cyclotron	p	18	2000	2.1×10^{14}

Neutron activation analysis

The use of 14 MeV neutron generators for the fast neutron determination of oxygen is well established. They can also be used for the determination of nitrogen and some other major elements. There are however major problems because of the many reaction channels that are open at these energies and the resultant interference.

At accelerating energies of a couple of MeV, the reaction $D(d,n)^3He$ produces neutrons that are well suited to the excitation of isomeric states from gold, silver, indium and a number of other elements. These often have short halflives and can be used for rapid (possibly on-line) measurements. Conventional thermal neutron activation analysis is also a possibility with an accelerator based source and it appears to this author that a system with a water moderator surrounding an appropriate accelerator target could be an important general purpose analytical tool.

The useful thermal neutron flux is often almost two orders of magnitude less than the conventional 4π output, so to achieve high levels of sensitivity, fairly large (tens of gram samples) in a Marinelli beaker configuration could be necessary. This could bring the concentration sensitivity up to levels that were traditionally available in small samples with nuclear reactors with thermal neutron fluxes of 10^{12} n cm⁻² s⁻¹, particularly with modern, high efficiency Ge detectors.

Neutron induced prompt gamma ray analysis

Applications of this type could be of importance in certain on-line applications and small sealed tube generators with outputs of 10⁷ to 10⁸ n s⁻¹ could be useful here.

Borehole logging

As was mentioned previously, small, sealed tube neutron generators have found important applications in borehole logging, particularly in oil exploration.

On-line analysis

This is an area where accelerators can play an important role, once they are reliable and easy to install and use. A great deal of work in this area was undertaken at Texas Nuclear many years ago but it did not give rise to any widely accepted on-line solutions, as, for example, X ray fluorescence has. Nevertheless I believe that there are many ways that nuclear technology can contribute to more efficient industrial processes, through measurement and control.

Ore sorting

If neutron generators with sufficient intensity become available they could be used for the separation of ore from gangue, or for upgrading ore grades at coarse particle sizes. Figure 3 shows a schematic of how such a procedure could work.

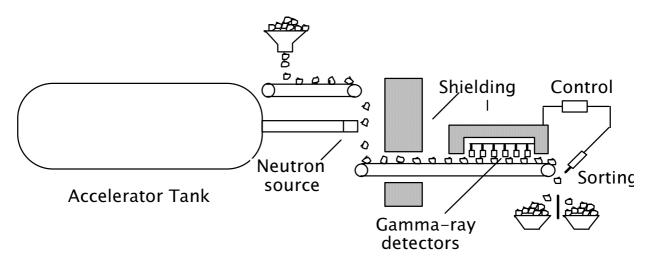


FIG.3. Schematic of a possible implementation of neutron activation ore sorting.

Neutron radiography

One of the problems with neutron radiography in general is the necessity to bring the specimens to a reactor. There has been a great deal of development and research aimed at the production of neutron radiography systems using accelerator based neutron sources. For example there has been the Diane project in Europe. This was based on a sealed tube system.

These could be of great importance in the examination of air frames for corrosion, where the great sensitivity of thermal neutrons for hydrogen is exploited. However the issue of whether the neutron outputs that are available, or that can be developed, are of sufficient intensity, is not yet clear.

Fast neutrons can also be used for the new field or resonance neutron radiography, where resonance can be used in order to produce elemental maps of specimens. In our group a great deal of work has recently been devoted to this technique and its application.

Medical applications

At one time a great deal of work was done on the development of 14 MeV accelerators for neutron therapy. It appears that the energies and intensities available with such machines are inadequate and this development has ceased.

There is however still some interest in the use of accelerator based neutron sources for boron neutron capture therapy (BNCT) [7]. This application is based on the use of drugs containing boron that could be localised in a tumor. Irradiation with neutrons would then deliver a very high LET dose to the tumor through the reaction $^{10}B(n,\alpha)^7Li$. There appear to be two problems in this application. These are the development of drugs that will be localised in the tumor and the development of neutron sources with sufficient intensity and the correct spectral distribution.

Another interesting medical application of neutron generators is the in vivo analysis of for example calcium and nitrogen.

Neutron scattering

Recent work in the USA has indicated that it may prove possible to use small but intense accelerator based neutron sources for materials investigations. This may become possible through developments in modern digital imaging techniques.

7. HISTORICAL REVIEW, NEUTRON GENERATORS IN SOUTH AFRICA

14 MeV neutron generators

Work on the application of neutrons from accelerators started in about 1965 using a 300 kV British accelerator, modeled on a French machine. This had a duoplasmatron ion source. It proved unreliable and its operation was discontinued in 1967.

In 1969 a 150 kV neutron generator was purchased by Mintek (the South African Council for Mineral Technology) from Accelerators Incorporated in the USA. This machine was rated at 5 mA but only once achieved its rated output. It was clear that the technology was adequate for a beam current of 1 mA but not for more. In addition the target lifetime proved to be a problem.

It was first used for the determination of oxygen and a similar machine was purchased by the South African Iron and Steel Corporation, Iscor and used for many years for the determination of oxygen in steel. I understand that this was more recently replaced by a gas analysis system. Nevertheless this remains the most important application of classical neutron generators.

Later it was applied to the determination of silicon, aluminium, magnesium and certain other major elements in rock. However other techniques proved to be superior in this application and interest turned to the use of reactor based neutron activation analysis for the determination of trace elements in rock.

A great deal of work was done using the SAFARI Reactor on the neutron activation of rocks and minerals. This is outside the present area of interest. The fast neutron generator was decommissioned after about five years of operation.

Use of neutron sources

At the University of the Witwatersrand we continued to explore the use of isotopic neutron sources for mineral processing. Research was carried using a 1 mg ²⁵²Cf source as well as smaller Am-Be sources and these definitely have a role to play in on-line analysis in the cement industry and the coal industry. This is however outside the present topic. Work using the tandem accelerator

Work was done on the study of neutron induced reactions particularly for the excitation of isomeric states by inelastic neutron scattering. The tandem accelerator with either a lithium or a beryllium target was used [8] but although some interesting experimental results were obtained, the neutron output is too low to do practical analyses.

Other work carried out indicated the usefulness of neutron activation with an accelerator of this type for the logging of borehole cores [9].

Recent work on fast neutron radiography

In our program investigating the application of nuclear methods in the mining industry we have a collaborative project with De Beers on the use of fast neutron radiography. This project has used the EN tandem accelerator and a 5 MeV single-ended Van de Graaf with a pulsed beam and time-of-flight for the initial investigations. The 5 MeV Van de Graaf is at the South African National Accelerator Centre in Cape Town.

For later more practical work we have used two RFQ linac accelerators. First of all the DL2 with a deuteron beam and a beryllium target and later a 5 MeV DL5 deuteron linac with a windowless gas target. The maximum beam current of this machine is $100 \, \mu A$.

At present the project is concentrated on the development of an RFQ machine with the same energy but capable of delivering several mA of beam current. A great deal of this work has been concerned with examining how rapidly images can be detected, the question of producing an efficient fast neutron imaging detector with a good resolution and the question of the influence of noise, particularly stochastic noise on the detectability of features in neutron radiographs at different signal to noise ratios.

8. CONCLUSIONS

Accelerator based neutron sources will play an important role in the future as analytical tools for radiography applications and possibly in neutron scattering. However the realisation of this potential will depend on the availability of accelerators capable of delivering mA of beam at energies of a couple of MeV easily, reliably and affordably. The achievement of this potential will also depend on the development of target technologies. The most promising of these at present appears to be the use of a windowless target either with a rotating shutter or possibly with the plasma window.

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UTILIZATION OF A PULSED D-T NEUTRON GENERATOR

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Abstract

In the past two decades the IAEA has supported the establishment of neutron laboratories in many developing countries by providing small D-T neutron generators. The neutron generator is basically a low energy (100–400 keV) ion accelerator capable of producing a continuous beam of deuterons with a current in the range between 1-2.5 mA. These neutron generators are primarily intended to be used for fast neutron activation analysis. This paper describes the utilization of a 14 MeV neutron generator in continuous and pulsed beam modes in applied neutron physics program at Chiang Mai University.

1. INTRODUCTION

The Fast Neutron Research Facility at Chiang Mai University is equipped with a nanosecond pulsed 14 MeV neutron generator, a multiparameter data acquisition and analysis system and various radiation detectors. Fast neutrons from the low energy ion accelerator are used in the field of radiation dosimetry, neutron induced nuclear cross sections measurement and elemental analysis. With minor modification, the accelerator can also be used as a gaseous heavy ion implanter.

The neutron generator is not necessarily limited to continuous beam operation. If properly modified, it can also be utilized for pulsed beam operation. The scope of utilizing such a neutron generator in analytical applications as well as in the studies of fast neutron reactions is extended considerably by pulsed beam operation. For example, the measurements of double differential neutron emission spectra at 14 MeV incident neutron energy for several materials related to the fusion reactor development program are best carried out by pulsed neutron time-of-flight (TOF) technique [1]. The feasibility of using a pulsed neutron generator for the measurement of light elements such as carbon, oxygen, and nitrogen has been successfully investigated [2, 3]. The main application field of pulsed neutrons in geology is the well logging analysis where a pulsed accelerator is used to produce 14 MeV neutrons. During the neutron burst, prompt γ rays resulting from neutron inelastic scattering and from reactions induced by high energy neutrons are detected. This method is a well established technique and has been adapted for down hole logging in both the coal and oil industries [4, 5]. Miscellaneous application of pulsed neutron generator has been reviewed by Csikai [5].

2. THE EXPERIMENTAL FACILITY

The Fast Neutron Research Facility (FNRF) operates a 200 kV, 5 mA high stability Cockcroft-Walton type accelerator producing 14 MeV neutrons for applications in fast neutron activation analysis and the study of neutron induced reactions. An associated alpha particle time-of-flight (TOF) spectrometer was established [6] with moderate energy resolution (1.2 MeV at 14 MeV neutron energy) and a well-behaved time independent background. The nanosecond pulsed neutron facility was installed in 1991 by modifying an existing continuous beam accelerator to incorporate beam chopping and bunching devices [7].

Neutron are produced from an AID J25 accelerator by the T(d,n) ⁴He reaction. Continuous deuterium ions (D₁⁺) from a radio frequency plasma ion source are accelerated to 140 kV by a 9 kHz switching frequency Cockcroft-Walton type D.C. high voltage power supply. The deuteron beam that comprises 75% atomic deuterons (D_1^+) and 25% molecular deuterons (D_2^+) is analyzed by an analyzing magnet. The D_1^+ ions are bent 90° to a pulsed beam channel while the D_2^+ ions are excluded by momentum discrimination. Beyond the analyzing magnet, the beam is transported through series of collimating slits and quadruple focusing magnets. The beam is chopped by double plate deflection system and then bunched to pulses with widths of 1.5 to 2.0 nsec at the neutron production target by a double gap klystron buncher. Beam sizes both in horizontal and vertical axes were monitored by means of a crossed wire beam profile monitor. The schematic diagram shown in Fig. 1 is a layout of the beam line components from the ion source to the target. A data acquisition system is controlled by a 16 MB MicroVAX II computer through a multiparameter buffer system (MBS) unit. Each reaction event detected by the main detector is recorded sequentially in list mode on disk. Our off-line analysis software allows dynamic selections for each correlated parameter in contrast to a conventional hardware resolution routine.

3. DOUBLE DIFFERENTIAL CROSS SECTION MEASUREMENTS

Studies of fast neutron induced reactions are of significance for an understanding of nuclear reaction theory as well as for practical applications. For example, the secondary neutron energy and angular distribution from the (n, xn') reactions on certain materials are of importance for the development of fission and fusion reactor systems and other accelerator based applications. The spectra of the emitted neutrons are usually measured using TOF technique.

In Chiang Mai, we have set up a high precision neutron TOF spectrometer system with flight path up to 12 m [8, 9]. Detail on the measurement and data reduction has been described recently [8]. A cylindrical sample about 3 cm in diameter and 5 cm long, was positioned at 90° relative to the incident deuteron beam with its axis along the axis of the beam line as shown in Fig. 2.

The neutrons were detected in a BC-501A liquid scintillating detector of diameter 25 cm and thickness 10 cm. The detector was coupled to a Hamamatsu R1250 photomultiplier tube via a partially coated taper light pipe. It was located at an extended flight path of 12 m inside a well shielded tunnel. Monte Carlo calculation indicates that scattering effect is less than 1% for this collimating system. Time-of-flight measurements were carried out at angles from 20° to 150° in step of 10°. The corresponding energy resolution at 14.1 MeV was about 415 keV FWHM.

The neutron spectrum was determined from the measured TOF spectra in separate energy regions. The γ rejection was observed for each of the TOF regions because the γ rejection technique was pulse height dependent. In consequence of the large size of the neutron detector, neutron and gamma events in the pulse shape spectrum were not completely separated and some neutrons were lost. This loss was estimated to be less than 1%.

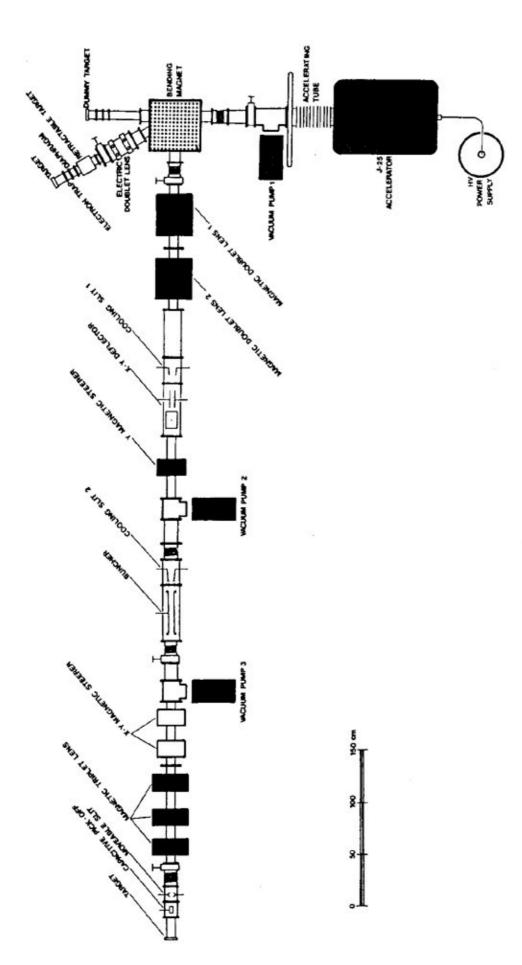


FIG. 1 Schematic diagram of the all pulsed beam line components and their geometrical arrangements.

Several corrections were made to the number of detected neutrons. The most significant one was due to multiple scattering. The overall systematic error for the value of the DDX of ²⁰⁹Bi for example is estimated to be about 11%. The statistical uncertainty varies from less than 1% to about 39% in the discrete region of the spectra of neutrons emitted in backward angles.

In Fig. 3 we show our recent measurement at 20° compared with the result of Takahashi et al. [10]. The two different sets of measurement are in reasonable agreement. Because of better energy resolution, our data reveal more detailed structure in the region between 6 to 12 MeV.

Our angle integrated spectrum is compared in Fig. 4 with the calculated results of Demetriou et al. [11] based on the Feshbach-Kerman-Kooning theory. In their calculation, Demetriou et al. also include contribution from direct reactions that excite low energy vibrations as well as the giant resonance in the continuum, in addition to the incoherent multistep direct reaction, multistep compound and compound nucleus reaction.

The calculated spectrum reproduces the experimental data very well in the regions between 3 to 6 MeV and 9 to 12 MeV. In the region between 6 and 9 MeV where preequilibrium reaction dominates, the two spectra do not agree. The calculated spectrum slightly overestimates the number of emitted neutrons.

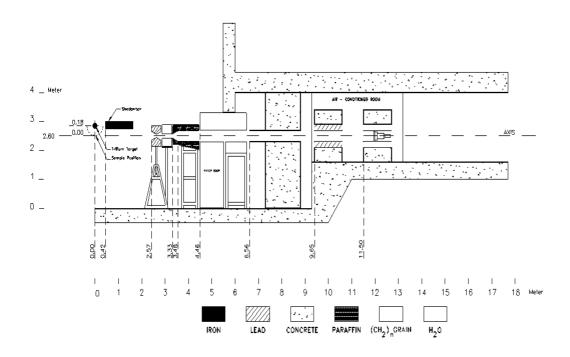


FIG. 2. Experimental arrangement for the DDX measurement [9].

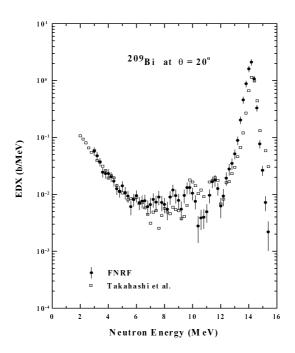


FIG. 3. Differential cross section of ²⁰⁹ Bi at 20° Data of Takahashi et al. [10] are shown for comparison.

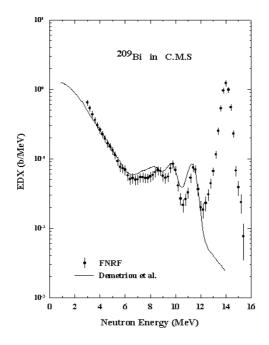


FIG. 4. Comparison of angle integrated spectrum of 209 Bi in the CM system with the calculated spectrum of Demetriou et al. [11].

4. PULSED TIME-OF-FLIGHT PROMPT GAMMA RAY ANALYSIS

A DT neutron generator is now routinely used in a variety of analytical applications, including nonintrusive inspection by means of prompt gamma ray analysis (PGA). Obtaining satisfactory results from this kind of measurement requires an associated trigger signal from either the alpha particle of the DT reaction when utilizing a continuous deuteron beam [12] or the induced signal from a capacitive pick-off when using a pulsed deuteron beam [3]. As is well known, the pulsed neutron time-of-flight (TOF) technique generally provides better signal to noise ratio than the associated alpha particle technique. We thus choose to work with the pulsed beam technique. Also, a pulsed neutron generator is not necessarily a large machine any more [13]. However, the production of wider neutron pulse is normally simpler and cheaper than a narrow one. The aim of this work was to investigate the quality and characteristics of the results obtained using both wide and narrow pulses.

The experimental arrangement is shown in Fig. 5. The gamma ray detector was a 5 inch diameter by 5 inch thick NaI(T1) scintillator. The detector was placed inside a heavy shielding about 2.4 m away from the TiT target and 37 cm from the sample position, as shown in Fig.5. The two parameters (energy-time) data acquisition and analysis system is similar to the one that has been described elsewhere [8]. The threshold detecting was fixed at around 0.5 MeV gamma ray energy.

Our data acquisition system allows energy or pulsed height data of the gamma ray signals that are associated with any interval of the peak of the time spectrum to be selected off-line. Figure 6 shows pulse height spectrum from sample of liquid nitrogen using 2 nsec pulsed neutrons. Each spectrum belongs to different time gating as indicated in the insets. Our experiment indicates that the best signal-to-noise ratio was obtained with narrow neutron pulse (< 5 nsec).

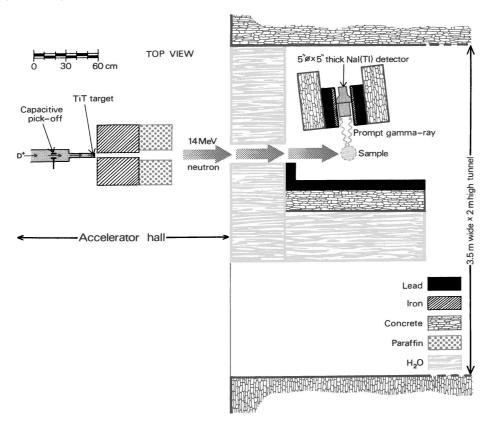


FIG. 5. Experimental arrangements for the pulsed TOF prompt gamma ray analysis [14].

Gamma ray spectroscopy performed using 1.4 kg of C-3 explosive with 2 nsec pulse reveals all expected photo peaks at 1.6, 2.3, 2.8, 3.7, 4.4, 5.1 and 6.1 MeV as shown in Fig. 7.

5. NEUTRON SOURCE AND DOSIMETRY

The neutron generator is normally used to produce monoenergetic 14 MeV neutron from the DT reaction and 3 MeV neutron from the DD reaction. We have investigated the properties of neutrons scattered from a circular surface of rotating paraffin scatterer using the MCNP code and the pulsed neutron TOF measurement [15]. Figure 8. shows the experimental arrangement.

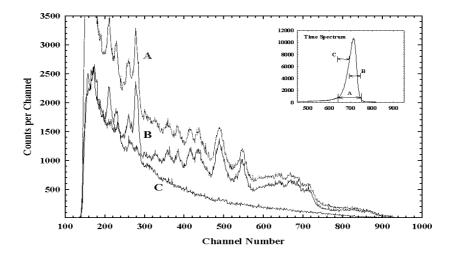


FIG. 6. Gamma ray energy spectrum of 20 kg liquid nitrogen using a 2 nsec width of neutron pulse [14]. Inset: selection of time window on the associated time spectrum (a = 90 nsec, b = 50 nsec, c = 40 nsec).

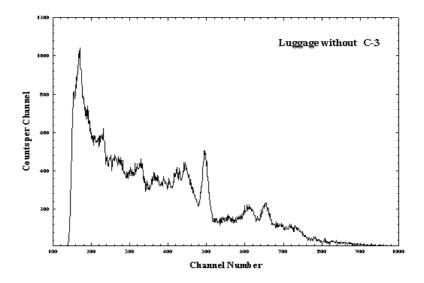


FIG. 7a. Time gated gamma ray spectrum of simulated passenger luggage.

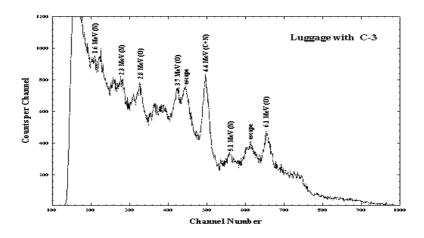


FIG. 7b. Time gated gamma ray spectrum of luggage containing C-3 explosive [14].

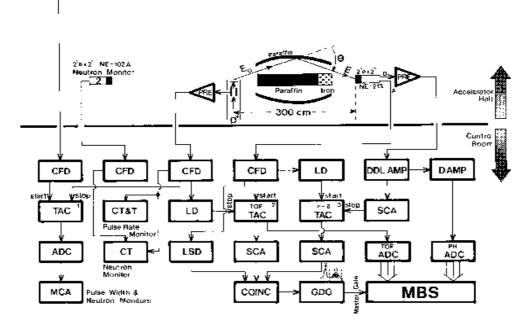


FIG. 8. Experimental set up for elastic scattering of 14 MeV neutron from a circular surface of rotating paraffin scatterer and the two parameter data acquisition system [15].

The neutron pulse height distributions obtained off-line by gating the pulse height events with appropriate TOF events in the specific time window of interest is shown is Fig. 9. It is noted that the yield of $n + {}^{12}C$ elastic scattering is fairly pronounced which can be used for calibration purpose. Elastically scattered neutrons from this type of scatterer has been successfully used for measuring the light output of a small NE-213 detector

Neutrons from the 14 MeV neutron generator can also be used to calibrate a neutron dosimeter. The different LET dependence of the low and high temperature glow peaks of CaF₂. The thermo luminescent material (TLD-300) allows the determination of neutron and gamma dose simultaneously. The method was calibrated for the 14 MeV neutron beams at FNRF [16]. Figure 10 and 11 show glow curves of a TLD-300 dosimeter after ⁶⁰Co and 14 MeV neutron irradiation.

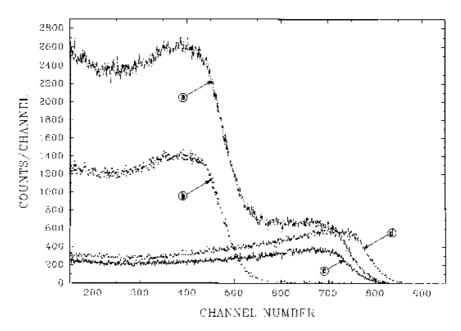


FIG. 9. The pulse height spectrum measured by the 5.08 cm dia x 5.08 cm thick NE-213 liquid scintillation detector. Separate spectra shown are (a) Scattered neutrons from full scatterer, (b) Scattered neutrons from hydrogen elastic scattering in front of the half scatterer. (c) Scattered neutrons from carbon elastic scattering in front of the half scatterer. (d) 14.1 MeV neutrons [15].

The relative neutron responses of both peaks in TLD-300 chips were found to be 0.10 and 0.32. Using this method, various dose distributions of neutron and gamma dose in a water phantom were measured and compared with the results of GM counter measurements and the Monte Carlo calculation as shown in Fig. 12.

6. ION IMPLANTATION

Basically, the 14 MeV neutron generator is a 200 kV ion accelerator using palladium tube to leak deuterium gas into the RF-ion source. At FNRF, we replaced the palladium tube with a thermo mechanical leak valve that enables us to use other gases such as N₂, CO₂, BF₃, etc. Implantation with gaseous ions such as nitrogen, oxygen, boron, argon are possible [17,18]. Surface modification with ion beam based technique has commanded great interest in recent years for improving mechanical, electrical and optical properties of materials [19]. A drift tube neutron generator can be most conveniently modified to be used as an ion implanter concurrently.

7. CONCLUSION

The 14 MeV neutron generator of Chiang Mai University has been utilized in both continuous and pulsed beam modes. Major applications are nuclear data measurement, elemental analysis with prompt gamma ray detection technique, calibrations of neutron detector and TLD-300. With minor modification, the accelerator can be converted into a heavy ion implanter for use to modify the surface properties of materials.

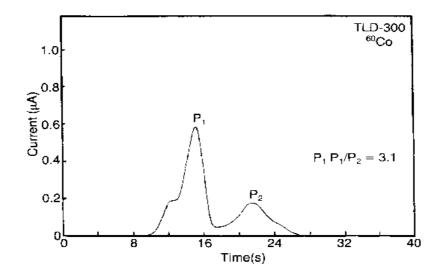


FIG. 10. Glow curve of a TLD-300 dosimeter after ⁶⁰Co irradiation [16].

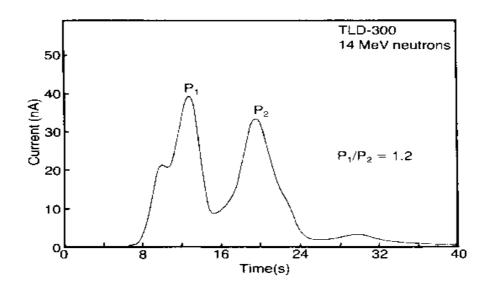


FIG. 11. The glow curve of TLD-300 dosimeter after irradiation by a 14 MeV neutron beam [16].

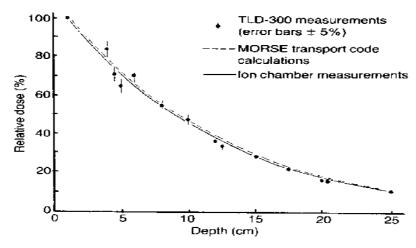


FIG. 12. Comparison of determination of the depth dose $D_{total} = D_n + D_{\gamma}$ of the 14 MeV beam in water by the TLD-300 method, ion chamber measurement, and Monte Carlo transport code calculations [16].

ACKNOWLEDGEMENTS

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USE OF A 14 MeV NEUTRON GENERATOR FOR ACTIVATION ANALYSIS

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Abstract

An overview of applications of the IPEN 14 MeV neutron generator is given. The generator was in active use, for neutron activation analysis, until middle of 1991 when a major breakdown caused serious damage to the vacuum system.

1. INTRODUCTION

Neutron activation analysis in Peru was started in 1978 on a small nuclear reactor of 1W power, provided to IPEN by the Argentine government. Now, the Peruvian Institute of Nuclear Energy has a 10 MW power nuclear research reactor so the neutron activation analysis is carried out mainly with this irradiation facility; in spite of that, elements like N, O, P, Si are better analyzed using high energy neutrons. Our accelerator based neutron generator is a SAMES-AID (Grenoble, France) Model J 25 which was donated to the institute in 1982. The IAEA technical cooperation project IAEA-IPEN PER/1/004 "Neutron Activation Analysis" was formulated for the 1981–1982 period. The machine was working routinely until the middle of 1991, when a major break down occured. Serious damage was caused by target cooling water leaking into the high vacuum part of the system (accelerator tube, ion source, etc.). In October 1996 we invited Dr. Tibor Sztaricskai to revise the general condition of the neutron generator. During the inspection we found that most of the technical problems could be solved by ourselves. At the moment we are looking for the necessary budget to carry out the repair and upgrade of the machine. We would like

- 1. to repair and put our accelerator based neutron generator into operation,
- 2. to collect a good stock of critical materials and spare parts,
- 3. to acquire the necessary know how and tools enabling us to repair the machine in case of troubles or to modify it for new experiments,
- 4. to acquire a high resolution and high efficiency (e.g. with a 160 % relative efficiency HPGe detector) in order to improve sensitivity and save irradiation time,
- 5. to automate the whole system under the control of a personal computer.

Our neutron generator has been used almost exclusively for 14 MeV neutron activation analysis. This report contains information about the most important applications of the technique in our laboratory, extracted from some papers published in Spanish, in Peruvian scientific journals (see references).

2. ANALYSIS OF NITROGEN AND PHOSPHOR IN CEREALS

A method using 14 MeV neutron activation analysis [2,3] was developed for non-destructive simultaneous determination of N and P in cereals. The samples and standards, with mean weights of approximately 2.8 grams, were irradiated in sequence for 5 minutes in a fast neutron flux of approximately 10^8-10^9 n.cm⁻². s⁻¹. After a 30 seconds decay time the samples were measured for 10 minutes. The used reactions are shown in Table 1.

As samples and standards were not irradiated simultaneously, the unavoidable variation of the neutron flux during irradiation had to be corrected for, by calculations using the digitally recorded variation of the neutron flux, monitored by a BF₃ detector. The induced activities were measured using a 3" × 3" NaI(Tl) well type detector. The interference of $^{31}P(n,2n)^{30}P$ and $^{16}O(p,\alpha)^{13}N$ reactions in the determination of Nitrogen was evaluated and corrected for. In the last case it was necessary to analyze the Oxygen in the samples, by the $^{16}O(n,p)^{16}N$ reaction, using 20 seconds irradiation time and 20 minutes counting time, after a 5 seconds decay [1]. The quantity of apparent Nitrogen due the Oxygen of the samples was found to be 0.612% N by gram of O, determined by irradiation of pure glucose standards. The analysis of P is almost free of interference. The accuracy assessment of P analysis was accomplished using the IAEA H5 and H8 reference materials, with a very good agreement with the certified values (see Table 2).

In the case of N analysis, it was necessary to compare the results of the proposed method with the ones obtained by the Kjeldahl's method as can be seen in Table 3, for a typical Andean cereal denominated "Kiwicha".

TABLE 1. THE RELEVANT ANALYTICAL NUCLEAR REACTIONS

ELEMENT	REACTION	% ISOTOPIC	T1/2	σ	E (keV)
		ABUNDANCE	(minutes)	(mb)	
N	$^{14}N(n,2n)^{13}N$	99.64	9.96	7	β^+
P	$^{31}P(n,\alpha)^{28}A1$	100.00	2.25	1.9	1778.8

TABLE 2. P ANALYSIS IN REFERENCE MATERIALS

REFERENCE MATERIAL	FOUND N %	CERTFIED VALUE %
IAEA H5 ANIMAL BONE	10.10 ± 0.30	10.20 ± 0.86
IAEA H8 HORSE KIDNEY	1.10 ± 0.06	1.12 ± 0.06

TABLE 3. ANALYSIS OF N IN "KIWICHA" BY KJELDAHL AND NAA

SAMPLE	N%	N%
#	by NAA	by Kjeldahl's method
1	2.102 ± 0.620	2.080
2	2.000 ± 0.242	1.984
3	2.131 ± 0.476	2.144
4	2.104 ± 0.259	2.128
5	1.973 ± 0.208	2.048
6	2.012 ± 0.408	2.080
7	2.104 ± 0.209	2.096
8	2.051 ± 0.649	2.080

Accuracy was evaluated by repeated analysis of diphenylamine – glucose and $CaHPO_4.2H_2O$ – glucose of known composition (Tables 4 and 5).

The detection limits obtained were 0.275% for N analysis and 0.091% for P analysis. Finally, Tables 6 and 7 show the N and P contents in some other cereals.

3. ANALYSIS OF OXYGEN IN COAL

Oxygen analysis by chemical methods is very difficult, almost impossible. However it can be accomplished in a very convenient way by using Fast Neutron Activation Analysis (FNAA). Oxygen analysis in coal [1] and ash gives a good content estimate of organic Carbon and it is possible to establish a good correlation between Oxygen content and volatile matter. This work describes a non-destructive methodology for quantitative analysis of Oxygen in

TABLE 4. ACCURACY OF P ANALYSIS

SAMPLE #	SPECIFIC COUNT RATE
1	1.260 ± 0.054
2	1.117 ± 0.064
3	1.126 ± 0.066
4	1.41 ± 0.066
5	1.223 ± 0.056
6	1.179 ± 0.061

TABLE 5. ACCURACY OF N ANALYSIS

SAMPLE #	SPECIFIC COUNT RATE
1	3.561 ± 0.393
2	3.350 ± 0.040
3	3.390 ± 0.407
4	3.332 ± 0.412
5	3.441 ± 0.388
6	3.285 ± 0.379

TABLE 6. N CONCENTRATION FOUND IN SOME CEREALS BY KJELDHAL AND NAA

CEREAL	N% by NAA	N % by Kjeldahl's method
Cañihua	1.738 ± 0.126	1.826 ± 0.926
Wheat	1.583 ± 0.286	1.595 ± 0.792
Corn	2.338 ± 0.355	2.326 ± 1.155
Quinua	1.306 ± 0.087	1.393 ± 0.690

TABLE 7. P CONCENTRATION FOUND IN SOME CEREALS BY NAA

CEREAL	P % by NAA
Cañihua	1.386 ± 0.087
Wheat	0.128 ± 0.045
Corn	0.244 ± 0.069
Quinua	0.451 ± 0.038

coals, based on the ¹⁶O(n,p)¹⁶N nuclear reaction using 14 MeV neutrons, produced by the AID J25, accelerator based neutron generator. The proposed method is based on the one developed by MORGAN and EHMAN [5], who registered the irradiation and decay processes in a sequential way. Due to the short lifetime of ¹⁶N, the implementation of this kind of analysis in

laboratories with limited resources is not easy. But, due to our home made interface and software, it was possible to control the whole procedure (irradiation and counting) by a DEC 350 microcomputer. When the sample is in the irradiation position, the computer opens the automatic valve, that intercepts the deuteron beam. When the irradiation is finished the valve is closed and the sample is sent to the measuring position.

Figure 1 shows the block diagram of the counting system, where the neutron yield is monitored by the BF_3 detector. The samples and standards — with weights of approximately 3 grams — are irradiated by 20 seconds. The detection system is initially set for recording the events from the BF_3 detector, giving the starting time a few seconds before the deuteron beam interceptor opened. After the irradiation, the system receives signals only from the gamma detectors. The irradiation and decay processes are set in intervals of 0.5 seconds. Figure 2 shows a typical decay spectrum.

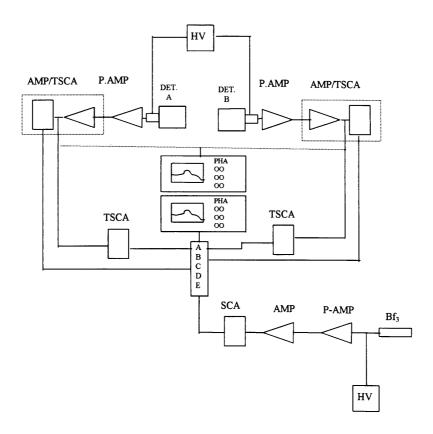


FIG. 1: Block diagram of the integrated measuring system.

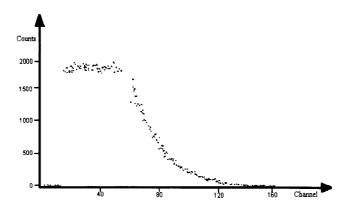


FIG. 2: Irradiation and ¹⁶N decay for a K₂Cr₂O₇ standard.

Due the time dependence of neutron flux is corrected for by utilizing the equation:

$$FK = \lambda \sum_{CI}^{CF} X_i e^{\lambda |T - (CF - CI + 1)| DL}$$

Where:

 λ : = 9.719 x 10⁻² s⁻¹ is the decay constant for 16 N

Xi: Counts in channel i

CI: First channel for neutron counting

CF: Last channel for neutron counting

DL: Time of channel (0.5 seconds)

T: Time at half of channel = (I - CI + 0.5) DL

Table 8 shows the accuracy of this method obtained by repeated irradiation of a $K_2Cr_2O_7$ standard.

Table 9 shows the accuracy of the method, as it was obtained by repeated analysis of several reagents of analytical purity.

TABLE 8. ACCURACY OF O ANALYSIS

SAMPLE	CORRECTED SPECIFIC
#	COUNT RATE
1	1.8500
2	1.8827
3	1.8092
4	1.8835
5	1.8369
6	1.9388
7	1.7588
8	1.8439
9	1.8881
10	1.9854

Table 10 shows the composition of some Peruvian coals studied and Table 11 shows the results of oxygen analysis in coals and in their ashes after heating them to constant weight at $900~\rm C$

TABLE 9. OXYGEN ANALYSIS IN CHEMICAL REAGENTS

REAGENT	OXYGEN	CALCULATED	RELATIVE
	FOUND %	OXYGEN %	ERROR %
UREA	27.25 ± 0.80	26.64	+ 2.29
CALCIUM CARBONATE	47.76 ± 1.40	47.96	- 0.42
TITANIUM DIOXODE	41.19 ± 1.00	40.05	+2.85
SILICIUM DIOXIDE	53.44 ± 0.63	53.26	+0.34
AMONIUM			
MOBLYBDATE	36.58 ± 0.45	36.25	+ 0.91

TABLE 10. COMPOSITION OF SOME PERUVIAN COALS

SAMPLE	H ₂ O	VOLATILE	ASHES	NONVOLA-	SULPHUR %
	%	MATTER %	%	TILE C %	
A-1	5.05	3.92	25.50	65.53	
A-2	5.35	3.17	16.62	75.86	
A-3	4.49	4.61	28.75	62.15	
C-1	5.29	3.57	30.76	60.38	1.58
D-1	2.13	6.42	31.86	59.59	1.30
D-2	6.45	11.38	34.40	47.77	0.94
D-3	3.30	7.92	15.24	73.54	1.06
D-4	4.76	4.30	13.20	77.70	0.80

TABLE 11. OXYGEN ANALYSIS OF SOME PERUVIAN COALS

	O	O		O	O	О
SAMPLE	IN	IN	ASHES	IN NON-	$H_2O +$	ORGANI
	COAL	ASHES		VOLAT. C	ORGANIC	C
	%	%	%	%	%	%
A-1	16.3 ± 0.2	45.9 ± 1.1	25.90	11.9 ± 0.3	4.4 ± 0.3	-0.1 ± 0.3
A-2	12.1 ± 0.2	46.4 ± 1.2	15.19	7.0 ± 0.2	5.1 ± 0.3	0.3 ± 0.3
A-3	18.3 ± 0.3	47.0 ± 1.6	31.64	14.9 ± 0.5	3.4 ± 0.6	0.6 ± 0.6
B-1	47.6 ± 0.6	53.0 ± 1.6	81.68	43.3 ± 1.3	4.3 ± 1.4	
B-2	29.4 ± 0.3	51.6 ± 2.1	47.00	24.3 ± 1.0	5.1 ± 1.0	
C-1	22.4 ± 0.3	55.4 ± 1.0	30.25	16.8 ± 0.3	5.6 ± 0.4	0.9 ± 0.4
D-1	22.2 ± 0.3	53.8 ± 1.2	32.28	17.4 ± 0.4	4.8 ± 0.5	2.9 ± 0.5
D-2	30.4 ± 0.4	53.9 ± 1.7	34.81	18.8 ± 0.6	11.6 ± 0.7	5.9 ± 0.7
D-3	15.1 ± 0.3	53.5 ± 1.2	16.76	9.0 ± 0.2	6.1 ± 0.4	3.2 ± 0.4
D-4	14.4 ± 0.3	51.2 ± 1.6	13.22	6.80 ± 0.2	7.6 ± 0.4	3.4 ± 0.4
·	·			·	·	

There are two possible interference's caused by $^{19}F(n,\alpha)^{16}N$ and $^{11}B(n,p)^{11}Be$, due to the high energy radiation produced by ^{11}Be , but those are rarely found in coals. It was found during the total 45 seconds analysis time, that the accuracy is about 1–3 %

4. ANALYSIS OF AL, SI, FE AND MG IN DIFFERENT POWDERED MATERIALS

A method using 14 MeV neutron activation analysis was developed for non-destructive determination of Si, Al, Fe and Mg in powder materials [4] such as, minerals, coals, catalysts, glass batches, etc.

For the simultaneous analysis of Al and Si samples were irradiated for 5 minutes in a fast neutron flux of approximately 10^9n/cm^2 and measured during 5 minutes after a cooling time of 1.5 minutes. Fe and Mg were simultaneously determined by irradiating 20 to 30 minutes using a home made rotator system for irradiation and measuring during 30 minutes after delay of 27 min. The induced activities were measured by a high resolution intrinsic germanium detector. The accuracy, precision and detection limits obtained are discussed as well as the analytical results for different types of samples.

5. CONCLUSIONS

Accelerator based neutron generators are powerful tools for fast, easy and accurate analysis of important elements and is a very valuable complement to reactor NAA. For an optimal use of this kind of machines it is necessary to get a good stock of critical spare parts and materials. It is also necessary to get the required know how and experience which would allow the experimenter to repair the machine himself and to modify it for the development of new experiments.

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RESEARCH AND DEVELOPMENT ACTIVITIES OF A NEUTRON GENERATOR FACILITY

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Abstract

The neutron generator facility at YNRC is used for elemental analysis, nuclear data measurement and education. In nuclear data measurement the focus is on re-evaluating the existing scattered nuclear activation cross-section to obtain systematic data for nuclear reactions such as (n,p), (n,α) , and (n,2n). In elemental analysis it is used for analyzing the Nitrogen (N), Phosphor (P) and Potassium (K) contents in chemical and natural fertilizers (compost), protein in rice, soybean, and corn and pollution level in rivers. The neutron generator is also used for education and training of BATAN staff and university students. The facility can also produce neutron generator components.

1. INTRODUCTION

Neutron generator facility at Yogyakarta Nuclear Research Center (YNRC) has three main activities. First activity is R & D in the utilization of the neutron generator for nuclear data measurements, and for elemental analysis using a fast neutron activation analysis technique. Second activity is R&D in technological development and fabrication of neutron generator components. Third activity is a training of BATAN staff and education of students from many universities around Java and Sumatra island. The neutron generator facility has been optimally used since 1990. This manuscript will describe detail activities in the neutron generator facility at YNRC.

The utilization of neutron generators is nowadays common not only in nuclear physics but also in other fields like chemistry, biology, metallurgy, geology, radiotherapy, and in the industry [1,2]. During the last decade, nuclear data measurements using a neutron generator has been carried out for fusion, radiation damage, geophysics and dosimetry applications. In chemistry, neutron generators have been used for analysis of chemical products. The fast neutron activation method has also been used to determine protein content, air and water pollution. In metallurgy, neutron generators have been used to determine oxygen and silicon content in stainless steel [1]. In geology neutron generators has been used to determine the content of minerals such as bauxite, manganese, and copper [1].

Based on the BATAN's program, for the peaceful use of nuclear technology for prosperity, one of the duties of Yogyakarta Nuclear Research Center (YNRC) is to get acquainted with accelerator technology and applications. Research and development of an accelerator at YNRC was started in 1983, in the beginning of Pelita IV (the fourth year of the five year development program), 1983–1988. The construction of two accelerators was started during this Pelita; one was an ion implanter and the other one was a neutron generator. These accelerators were basically ready in the middle of Pelita V (1988–1993). However, they are still in need of improvements in order to use them for Research and Development (R&D) and applications.

During Pelita IV, YNRC got a neutron generator from the Isotope and Radiation Application Research Center, one of the BATAN's institutes. In fact, this neutron generator was a grant from IAEA in 1977. The neutron generator is of the type J 25 made by SAMES/France. The SAMES neutron generator was out of order at that time, and it was transported to YNRC at the end of 1987. After about one year of maintenance and repair of the vacuum and high voltage components, this accelerator became a useful tool in the Research and Development (R&D) related to the utilization of neutron generators.

The R&D by the neutron generator have been carried out in Nuclear Physics group of YNRC. The R&D activities can be divided in two subgroups. The first group is doing R & D in nuclear data measurements and a fast neutron activation analysis based on the utilization of neutron generators. This group usually uses the SAMES neutron generator. The second group works on the technological development and manufacture of neutron generator components. These programs are carried out by 1 PhD, 4 MSc, and 6 BSc in physics as well as by 10 electrical and mechanical technicians. A couple of the team has been trained at Jülich Nuclear Research Center, Bhaba Atomic Research Center, Efremov Accelerator Research Center, Institute of Experimental Physics Kossuth University, etc.

The neutron generator facility was also used for the training of BATAN staff and in the education of students from many universities around the Java and Sumatra islands such as: Gadjah Mada University (UGM), Surabaya Technology Institute (ITS), Diponegoro University (UNDIP), Brawijaya University (UNBRAW), Riau University (UNRI), and North Sumatra, University (USU). More than 20 S-1 (bachelor degree) theses have been written using results obtained at the neutron generator facility.

This paper describes some parts of the research and development activities of the neutron generator facility at YNRC. Firstly, it describes the construction of neutron generator components. Secondly it described the utilization of the neutron generator, thirdly the progress and future R&D on neutron generators, and finally different topics related to the neutron generator facility.

2. CONSTRUCTION OF NEUTRON GENERATOR COMPONENTS

This program started in 1983 in the beginning of Pelita IV. The aim of this program was to establish the manufacturing technology of neutron generator components. This group works on the development and construction of neutron generator components. After two Pelita, the construction of a homemade neutron generator was finished. We called it a homemade neutron generator as most of its components are home made except the vacuum pump and the tritium target. This machine consists of a radio frequency deuteron ion source, a high voltage Cockroft-Walton terminal, an accelerator tube, an electrostatic quadrupole lens, a tritium target, and a vacuum system. The scheme of this machine is shown in Fig. 1.

2.1 Ion source

An RF ion source has been developed and constructed. This ion source [3, 4] is basically similar to the Oak Ridge design. This radio frequency ion source consists of a bottle of Pyrex glass 215 mm long and 30 mm in diameter, with a 1.5 mm in diameter and 6 mm long extraction channel.

The ion source can produce a relatively high deuteron ion beam current. At an optimum condition, the radio frequency ion source yields deuteron ion beam current of 2.5 mA. This was achieved at the operating conditions of: 10^{-2} mm of Hg gas working pressure in the Pyrex tube, 50 MHz working frequency, 740 gauss magnetic field, 7 kV extractor voltage and 70 watt total RF energy of oscillator power. The function of deuteron ion beam current vs. the extractor voltage, the input energy of oscillator and the magnetic field are shown in the Figures 2, 3 and 4.

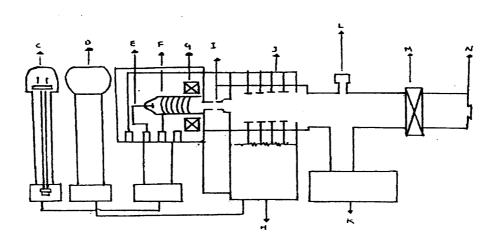


Figure 1. The scheme of the homemade neutron generator Notes: C = insulated power supply, D = HV Cockroft Walton, E = Extraction Voltage, F = RF source, G = Magnet, H = Resistors, I = Beam focusing, J = Accelerator, K = Vacuum pump, L = Vacuum meter, M = Electric Quadrupole Lens, N = TiT target.

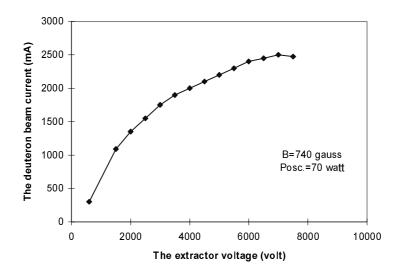


Figure 2. The function of RF ion source output vs. extractor voltage.

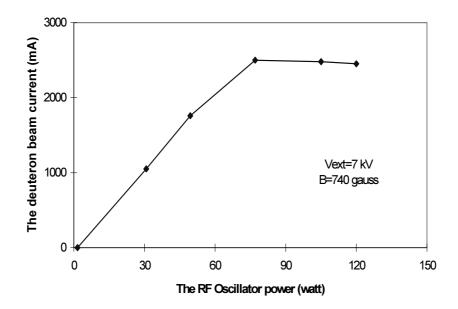


Figure 3. The RF ion source output vs. the RF power.

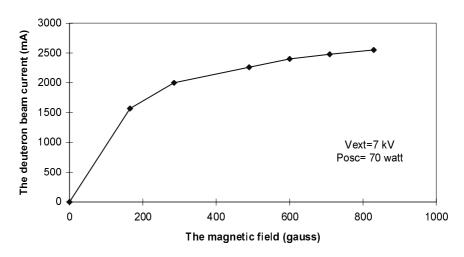


Figure 4. The RF ion source output vs. the magnetic field.

2.2 High voltage Cockroft Walton generator

A 7-stage high voltage Cockroft Walton generator [5, 6, 7] was constructed for the neutron generator using 20 kV/1A high voltage diodes, 0.28 μF/30 kV high voltage capacitors and stainless steel ring electrodes of 82 cm diameter. The dimension of this generator is 70 cm x 80 cm x 190 cm (LWH). The homemade high voltage Cockroft Walton generator supplies the acceleration tube up to 150 kV at a load current of 5 mA. However, the high voltage power supply can not operate at maximum output voltage due to some problems related to the accelerator tube. This generator operates at 125 kV and 3 mA. Recently, the HV power supply has been stabilized by completing the Cockroft Walton generator circuit with a closed loop feedback control system. The main components used for voltage stabilization are a resistor divider as an output sensor; an integrated circuit comparator and a brush motor to control the output of the power supply source. Now, high voltage power supply has a good stability in the load current range of 0 to 5 mA around an output voltage of 130 kV as shown in the Figure 5.

The output high voltage vs. input mains voltage (home electricity) and load currents are shown in Figures 5 and 6.

2.3 Power supplies

To supply the RF oscillator, the beam extracting and focusing systems as well as other equipment located on the high voltage terminal, an insulated motor generator is used [6, 7]. The generator is operating with 1500 rpm, 220 Volt giving 3 kW output power. It is supported by a 30 cm diameter and 1.5 m high PVC tube, and connected to the motor by a PVC rod of 10 cm diameter. The input signal to the Cockroft Walton HV generator is 1 kHz generating a 500 watt output square waves. The power supplies located on the high voltage terminal are controlled by PVC rods rotated by electrical motors on ground potential. The same method is used for controlling the gas inflow to the ion source

2.4 Acceleration tube

The homemade multigap accelerator tube is made of Plexiglas and 20 stainless steel electrodes. These electrodes were glued by using special epoxy. The diameter of the accelerator tube is 100 mm. The diameter of the inner electrode is 60 mm and this electrode

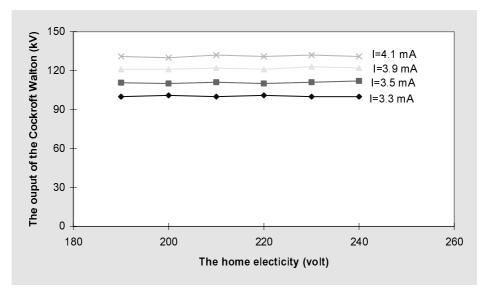


Figure 5. The output of Cockroft Walton HV PS vs. mains voltage.

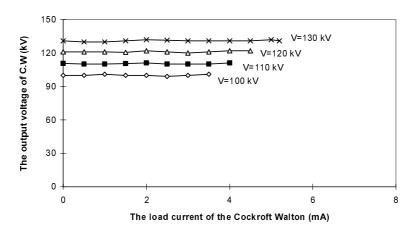


Figure 6. The output voltage of Cockroft Walton HV PS vs. load current.

can yield a high gradient electric field. The gap between the electrodes is 5 cm. The accelerator tube can operate only up to 130 kV and 1.5 mA of deuteron beam current due to the heat produced on the electrodes. This heat can crack the accelerator tube, so during operation, the accelerator tube should be cooled. We have difficulties to get aluminum or ceramic rings to replace the Plexiglas in order to increase the operating voltage of the accelerator tube.

2.5 Electric quadrupole lens

The quadrupole lens [8] consists of four perpendicularly crossed hyperbolical electrodes. Each of the electrode pairs are connected to a positive and a negative voltage regulated between 0 and $\pm 30 \mathrm{kV}$. The size of the quadrupole lens unit is 100 mm in diameter and 170 mm long. The lens electrodes are 45 mm long and 25 mm wide. The lense can focus a deuteron ion beam on a tritium target of 2–3 cm in diameter.

2.6 Ion beam current measurement

A Faraday cup consisting of collectors, feedthrough and current meters measures the ion beam current. The Faraday cup is made of copper with a plate diameter of 4 cm.

2.7 Vacuum system and tritium target

The vacuum system is using a rotary pump and a turbomolecular pump bought from EDWARD VACUUM Company providing vacuum up to 10⁻⁶ mm of Hg. Tritium targets in the form of TiT are procured from USA.

3. THE UTILIZATION OF THE NEUTRON GENERATOR

The YNRC, neutron generator group also carries out research in the field of nuclear data measurements and neutron activation analysis. This group is using the French neutron generator SAMES J 25 granted by the IAEA. This machine produces a fast 14.7 MeV neutron flux in the order of 10^9 n/cm²sec at optimum conditions. At present, the application of this neutron generator is focused on nuclear data measurement related to the re-evaluation of scattered neutrons, nuclear activation cross-section with the aim to get more systematic data for a certain nuclear reactions such as (n,p), (n,α) , and (n,2n). We have also done a lot research related to neutron activation analysis i.e. analyzing Nitrogen (N), Phosphor (P), and Potassium (K) in chemical and natural fertilizers (compost), analyzing protein in rice, soybean and corn as well as analyzing the pollution level in city rivers.

3.1 Analysis of protein content in rice and soybean

The nuclear method [9, 10] was introduced to the users especially to the agriculture researchers as an alternative method. They usually use a chemical procedure, the so-called Keydall method for determination of protein content. The nuclear method has advantages over the chemical method. The nuclear method can be used for destructive as well as nondestructive analysis but the chemical method is always destructive. In this work the ¹⁴N(n,2n)¹³N reaction was used. The activation time was 30 minutes; the cooling time was 3 minutes and the counting time was 30 minutes. Using the gamma peak of 511 keV from

(n, 2n) reaction and the relative method, the protein content in rice was found to be 6.51±0.03% of sample weight. The protein content in white, yellow, black, and green soybeans are respectively 28.9%, 29.5%, 24.12%, and 29.5% of sample weight. They are in agreement with the data obtained by the Directorate of Nutrition of the Department of Health.

3.2 Measurement of phosphorus content in TSP and NPK

In this work [11] the 31 P(n, α) 28 Al reaction was used. By using the gamma peak of 1778.34 keV from the (n, α) reaction and relative method, the phosphorus content was found to be 20±1% of sample weight in TSP and 6.5±0.6% of sample weight in NPK. These data were in agreement with the data given by the manufacturer, i.e. 20.1% for TSP and 6.5 % for NPK.

3.3 Nuclear data measurements

Our neutron generator has been used for reaction cross-section measurements mostly as preliminary research. We have developed a method for cross-section measurements using a two-detector technique [12]. In this technique, the activated sample is simultaneously measured by two gamma detectors, and the reaction cross-section is derived from gamma ray counts obtained by the two detector system. Experimental results show that the two detector technique provides a better accuracy than a one detector system. Some major elements for fusion reactor materials such as Mg, Si, V, Fe, Cu and In have been tested using the two detector technique. The results were compared with other from the literature and found to be in good agreement, as is shown in Table 1. [12,13].

TABLE 1: COMPARISON BETWEEN NEUTRON CROSS SECTIONS MEASURED BY ONE AND TWO DETECTORS (HPGE AND NAI[TL]) METHOD AND THE REFERENCE VALUES.

	Cross Section (millibarn)						
Reaction	Measured values *			Reference values **			
	HPGe	NaI (Tl)	HPGe & NaI(Tl)	1	2	3	
Mg-26(n,α)Ne-23	93.33±3.49	88.78±9.90	91.06±5.25	89.0	84.0±10	77.0±8	
Mg-25(n,p)Na-25	56.60±3.17	51.85±8.67	54.23±4.61	44.9	49.0±20	44.0 ± 5	
Si-29(n,p)Al-29	105.60 ± 4.40	103.74±3.66	104.67±2.86	100.0	120.0 ± 20	120.0±20	
Si-28(n,p)Al-28	240.53 ± 9.76	240.80±6.61	240.66±5.89	235.0	250.0 ± 30	230.0±30	
V-51(n,p)Ti-51	27.71±1.18	27.45±1.12	27.58±0.81	27.0	35.0 ± 5	35.5±5	
$V-51(n,\alpha)$ Sc-48	17.65 ± 0.74	17.65 ± 0.63	17.65 ± 0.48	-	15.0 ± 2	17.0 ± 3	
Fe-54(n,2n)Fe-53g	17.08±0.54	15.18±0.63	16.13±0.42	15	10.5 ± 1	15.5±1	
Fe-56(n,p)Mn-56	109.17±3.47	113.44±4.61	111.31±2.89	103.0	112.0±6	103.0 ± 6	
Cu-65(n,2n)Cu-64	1057.37±29.26	1057.11±24.90	1057.24±19.21	1100.0	913.0±50	956.0±50	
$Zr-90(n,\alpha)Sr-87$	196.22±17.47	193.82±6.93	195.02±9.40	194.0	112 ± 0.3	120±40	
Zr-90(n,2n)Zr-89	778.19±47.15	787.15±22.29	782.67±26.08	770.0	740.0±3	714±50	

- *) Abdurrouf [13]
- **) 1. S.S Nargolwalla, et al [2]
 - 2. M. Bormann, et al, "Table and Cross Section for (n,p), (n,α), (n, 2n) Reaction in the Energy Region of 1 37 MeV", Handbook of Nuclear Cross Section, IAEA Report-156, Vienna, p.87–272, 1974.
 - 3. G. Erdtman, "Neutron Activation Table", Weinheim Verlag Chemie, New York, 1976

4. PROGRESS AND FUTURE R&D ON THE NEUTRON GENERATOR

Recently we finished the set up of the fast neutron spectrometer by using the pulse shape discrimination (PSD) method. In this method the neutron field was measured by using liquid scintillator NE-213, the neutron (corresponding to the proton) and the gamma (corresponding to the electron) pulses were discriminated by using a pulse shape analyzer (PSA). The neutron flux was obtained by unfolding of the proton spectrum.

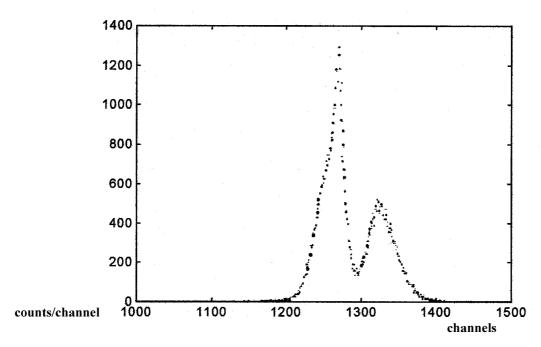


Figure 7 The neutron gamma PSD spectrum from the PHR neutron spectrometer.

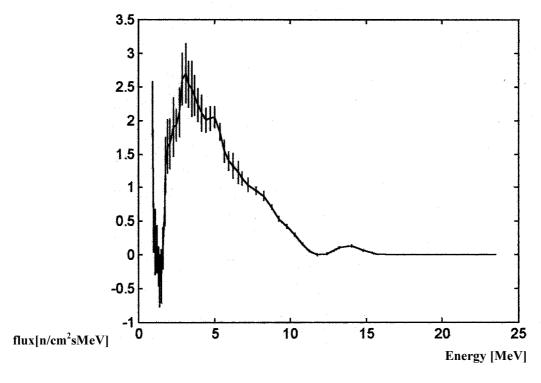


Figure 8. The neutron spectrum of AmBe measured by PSD method.

This setup was tested using an AmBe neutron source with the activity of 10^3 n/s. The output pulse height distribution of the neutron spectrometer is shown in Figures 7 and 8 [14]. This measurement was carried out in normal condition (not in a neutron scattering free) laboratory. The systematic data of the (n,α) reactions were evaluated [15]. The data were determined using the neutron activation method and our results are shown in Figure 9. Combined fast and thermal neutron activation analysis has been applied to identify Fe, Mg, and N elements in downy mildew corn leaves (Sterospora maydis R. Butler) [16]

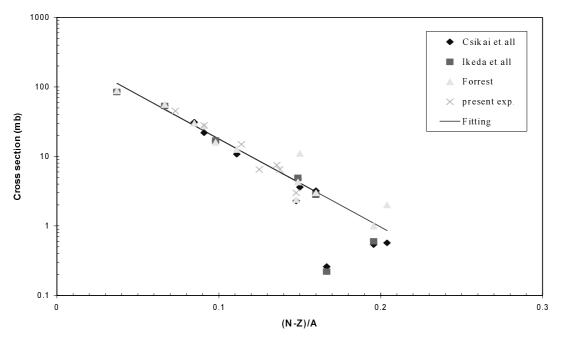


Figure 9: Dependence of (n,a) cross sections on (N-Z)/A asymetry parameter

Fast neutron activation analysis was used to determine the nitrogen content, and thermal neutron activation analysis was used to determine the Fe and Mg contents. Normal corn leaves and downy mildew corn leave were irradiated and analyzed. The experimental results show that the downy mildew corn leaves contain 161.51 to 192.07 ppm of Fe, 170.75 to 272.36 ppm of Mg, and 2541.23 to 2682.61 ppm of N. The normal corn leaves contain 291.48 to 352.66 ppm of Fe, 637.37 to 705.82 ppm of Mg, and 36773.15 to 3745.66 ppm of N. The downy mildew corn leaves has a lower content of Fe, Mg, N than normal corn leaves. Thus the sterospora maydis cause the decreasing of the Fe, Mg, and N content in corn leaves.

In the future, we are going to install the prompt gamma analysis system for bulk assays using the neutron generator as the neutron source. Some instruments for this purpose are ready. However we have not yet decided, whether we are going to use the pulsed neutron source or the continuous neutron source. The pulsed neutron source is better in point of the background, however we do not have a pulsing system for the neutron generator. Another work; we are going to carry out is integral data measurement on some shielding using local components. This measurement will be carried out by using the fast neutron pulse height response spectrometer and the foil activation method. The fast neutron activation analysis will routinely be used by agriculture, biology, and geology researchers.

Besides the above described topics we are also involved in nuclear physics teaching of students from many universities around our institute; supervising them in nuclear physics research related to our neutron generator.

5. MISCELLANEOUS

The neutron generator facility is also used in the training of BATAN staff and in the education of students from many universities around Java and Sumatra island such as: Gadjah Mada University (UGM), Surabaya Technology Institute (ITS), Diponegoro University (UNDIP), Brawijaya University (UNBRAW), Riau University (UNRI), and North Sumatra, University (USU). More than twenty S-1 (bachelor honors) thesis have been written in which the experiments were carried out at this neutron generator facility.

In recent years, we have difficulties in getting tritium targets (TiT) for neutron generators. The SAMES company is not delivering it. We have tried to buy it from a company in the USA but the company wants to sell more than 5 pieces of TiT.

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UTILIZATION OF A SEALED-TUBE NEUTRON GENERATOR FOR TRAINING AND RESEARCH

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Abstract

The development of a program in nuclear science and technology in Nigeria began in 1976 with the establishment of two research centers, namely, the Centre for Energy Research and Training, (CERT), Zaria and the Centre for Energy Research and Development (CERD), Ile-Ife. The choice of Neutron Activation Analysis (NAA) technique as a very effective method of training scientists in basic and applied nuclear research led to the purchase of two KAMAN A-711 Neutron Generators for the two research centers. At CERT, the neutron generator (code named ZARABUNG-1) was successfully installed and the first 14 MeV neutrons were produced through the technical assistance of the International Atomic Energy Agency (IAEA) in 1988 [1]. In 1991, a new tube-head was purchased and installed due to the expiration of the old tube. Following the completion of its permanent site, the neutron generator was re-located from the old site and re-installed at the permanent site of CERT in 1995.

1. INTRODUCTION

Since 1988, the neutron generator has been successfully used for a number of training and technical programs involving participants from Nigeria and other African countries. Similarly, a number of basic and applied research works carried out with the facility have been documented [2–8]. The current area of focus is in the use of facility for short time irradiation especially, the characterization of clays used in the petroleum industry.

2. SYSTEM DESCRIPTION

ZARABUNG-1 is a KAMAN type A-711 neutron generator installed at CERT, ABU, Zaria, Nigeria. It consists of four separate units namely: a sealed-tube portable ion accelerator head, a pressurized tank containing the power supply, a refrigeration-type cooling system and a control console. The units are interconnected by long cables to allow remote, non-hazardous operation.

The advantage of the sealed-tube is that it eliminates the use of components such as pump, power supply and drift tube found in conventional differentially pumped accelerators. It utilizes the ³H (d,n) ⁴He reaction to produce neutrons in a continuos mode with an energy of approximately 14 MeV. Neutrons are produced from the mixed beam of deuterium and tritium gas in the ion-gauge source. From beginning, gas is admitted into the ion source by passing a current of 2–4 A, ionization of the gas takes place in the ion source by the application of a voltage of 5–6 kV. The ions produced are accelerated towards a TiT target by applying a potential of 100–200 kV between the electrodes of the high voltage lens assembly. In order to reduce power dissipation to an efficient level, secondary electrons are suppressed at the target end of the accelerator by placing the lens assembly at a potential of 1kV with respect to the target. When operated as stated above, the beam current is then primarily a function of the pressure within the accelerator column. The beam current must be limited to a value that will not cause the target to over heat. The Penning ion source is cooled by electrically insulating FREON-113 liquid while the target is cooled by circulated water.

3. TECHNICAL AND TRAINING PROGRAMS

After the installation in 1988, the neutron generator has been used for training of undergraduate and postgraduate students at the department of Physics in the field of neutron activation analysis. The following technical and training programs (international and local) involving the use of the neutron generator has been held at our Center. Some of the courses were held under the auspices of the International Atomic Energy Agency (IAEA).

- (1) Africa Regional Training course (RTC) on Application of Nuclear Analytical Techniques in Mineral Exploration, Zaria, Nigeria 28 April to 16 May, 1997.
- (2) National Training Workshop on the Applications of Nuclear Analytical Methods in Crude Oil Characterization 16th 27th June 1997.
- (3) Workshop on the Application of Nuclear Methods in Water Resources Evaluation and Management $27^{th} 29^{th}$ April 1994.
- (4) Utilization of Neutron Generator at CERT Lectures and Practical by G.J. Csikai, IAEA Expert Mission 3rd Oct.–2nd Nov. 1990 [3]
- (5) The Utilization of a Neutron Generator Practical by G.J. Csikai, IAEA Expert Mission 2nd Aug.—12th Sept. 1992. [5]
- (6) Installation of a KAMAN A-711 Neutron Generator by T. Sztaricskai, IAEA Expert Mission (2 weeks) Feb. 1997.
- (7) Installation of a KAMAN A-711 Neutron Generator by T. Sztaricskai, IAEA Expert Mission 23–30 May 1987.
- (8) Installation of a KAMAN A-711 Neutron Generator by T. Sztaricskai, IAEA Expert Mission 22nd April–7th May 1988.
- (9) Relocation and Re-installation of CERT Neutron Generator by T. Sztaricskai, IAEA Expert Mission (1 week) Dec. 1995.
- (10) Repair of CERT Neutron Generator by T. Sztaricskai, IAEA Expert Mission 8–13 Dec. 1996.

4. RESEARCH STUDIES

Most of the operational period of the neutron generator has been spent on trouble shooting than analytical works because of the many breakdowns experienced. Initially, the principal cause of the loss of substantial part of the tube life is due to delay in the installation. The components of the equipment were lying idle for about 7 years before installation in 1988. In 1991, the tube was changed and thereafter it was discovered that the neutron generator could not remain stable for a reasonable length of time at the optimum value of accelerating voltage. This is attributable to the reduction in the tube's voltage hold-off capacity.

The following analytical measurements were carried out to characterize the facility for the analysis of geological and biological samples using fast neutron activation analysis technique.

Characterization of the neutron field produced by the neutron generator was carried out by the activation technique using Al, Nb and Zr foils [4]. In this work an analytical expression was

given for the calculation of the relative flux density. The FWHM of the flux density distributions indicates the use of disk-shaped sample of diameter 2.5-cm. Besides, the average neutron energy was found to be 14.61 MeV using Nb/Zr ratio method.

On the basis of the 27 Al(n, α) 24 Na reaction, the neutron flux density at the sample position is presently 2.8 x 10 6 n cm $^{-2}$ s $^{-1}$. Taking into account a distance of 1.5 cm between the target cooling cap and the sample, the source strength was deduced to be 7.4 x10 7 n.s $^{-1}$ at an accelerating voltage of 130 keV. With the limited life time of the tube and the low neutron flux density, the analytical capabilities of the system via the activation technique was investigated by calculating the detection limits of 20 elements shown in Table 1. Calculation was based on the measured neutron flux at an irradiation time of 10 minutes using a counting time of 5 minutes after a waiting time of 1 minute. The photo peak count of γ line of interest was assumed to be 100 counts for the NaI(Tl) detector while the effect of sample matrix has been neglected. The photo peak efficiency curves of the set-up displayed in Fig. 1 have been determined by standard γ ray sources at different source detector positions.

In Nigeria, the establishment of a Ministry of Solid Mineral Development has led to extensive exploration works. In order to cope with the large number of samples generated, CERT has been commissioned to carry out the analytical works because of the advantages the nuclear analytical techniques have over other techniques. The Country is blessed with large deposits of alumino-silicate ores, which are industrial raw materials. The knowledge of the Al and Si contents of these ores is of immense benefit to mineralogists and industrialists. Consequently, an analytical procedure using the neutron generator has been developed for the simultaneous determination of Al ad Si contents of geological ores. Compared to the procedure already established in our laboratory using a 5 Ci Am-Be neutron source [10], this procedure is rapid and can be used for the determination of Al and Si contents of coals. It is well known that the accurate determination of Al and Si contents of alumino-silicate compounds using neutrons depends on the choice of appropriate experimental conditions because the juxtaposition of the elements on the Periodic Table can cause nuclear interference, see Table 2.

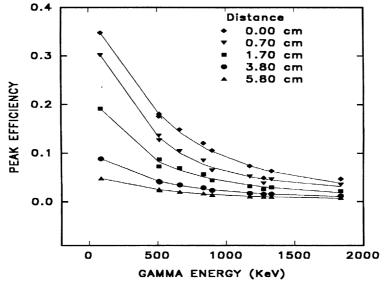


FIG. 1 Photo peak efficiency of the NaI(Tl) detector

TABLE 1. DETECTION LIMITS FOR 20 ELEMENTS FOR THE CERT 14 MEV NEUTRON GENERATOR

Element	Reaction	Half-life	γ-energy (keV)	Det. Lim. (mg)
Na	²³ Na(n,p)	37.24 s	440.0	29.64
Mg	24 Mg(n,p)	15.02 h	1368.5	69.62
Al	27 Al(n,p)	9.46 m	843.7	2.25
			1014.4	7.32
	27 Al(n, α)	15.02 h	1368.5	99.98
Si	28 Si(n,p)	2.30 m	1779.0	1.60
P	$^{31}P(n,p)$	2.30 m	1779.0	3.25
Ar	40 Ar(n,p)	1.35 m	1460.8	11.42
V	$^{51}V(n,p)$	5.75 m	320.8	2.85
Cr	52 Cr(n,p)	3.75 m	1434.1	5.01
Fe	⁵⁶ Fe(n,p)	2.57 h	846.6	25.56
			1810.0	225.10
Co	59 Co(n, α)	2.57 h	846.6	84.47
Zn	64 Zn(n,2n)	38.10 m	511.0	2.89
Cu	63Cu(n,2n)	9.86 m	511.0	0.13
Rb	85 Rb(n,2n)	20.5 m	248.0	0.86
Sr	88 Sr(n.2n)	17.20 s	162.0	48.5
Zr	90 Zr(n.2n)	4.20 m	587.8	6.44
Ag	107 Ag(n,2n)	24.0 m	511.9	6.34
Cd	$^{112}Cd(n,2n)$	49.0 m	245.4	3.01
Sb	112 Sb(n.2n)	15.89 m	511.0	0.76
Ce	¹⁴⁰ Ce(n.2n)	54.0 m	754.2	1.96
Pt	¹⁹⁸ Pt(n,2n)	1.57 h	346.5	0.23

TABLE 2. REACTIONS AND NUCLEAR DATA USED

Nuclear	Half-life	σ	γ Energy	γ-Abund.
reaction		(mb)	(keV)	(%)
27 Al(n,p) 27 Mg	9.46	75	843.7	72.0
	min.		1014.4	28.0
27 Al $(n,\gamma)^{28}$ Al		0.5	1779.0	100.0
28 Si(n,p) 28 Al	2.24	226	1779.0	100.0
56 Fe(n,p) 56 Mn	min.	103	846.8	98.9
· · · · · ·	2.24		1810.7	27.2
	min			
	2.58 hrs.			

Our γ ray measuring system is made up of a 7.6 × 7.6 cm NaI(Tl) detector, which has a resolution of 7 % at 662 keV γ line of ^{137}Cs . It is coupled to computer based multichannel analyzer card system. Because of the poor resolution of the γ ray spectrometer, spectral interference can occur via $^{56}Fe(n.p)^{56}Mn$ especially if the mineral contains iron in large quantities. Consequently, this experimental procedure was developed by taking the advantage of the low neutron flux density value and settling the irradiation, cooling and measuring times to be 5, 2 and 5-minutes respectively. During each irradiation, the primary fast flux of

neutrons was monitored by an Al foil fixed to the sample via 27 Al(n,p) 27 Mg which was then used to normalize the difference between flux reaching samples and standards. Analytical grade Si0₂ and Al₂0₃ powders were used as standards. The reliability of the procedure was checked using Bauxite (BCS-395) and Portland Cement (BCS-372) certified reference materials.

The concentrations of Al and Si in the samples have been deduced using the expression given below.

$$C_{a} = \frac{A_{a} M_{s} A_{fs} M_{fa}}{A_{s} M_{a} A_{fa} M_{fs}}$$
 100 % (1)

Where,

C_a is the concentration in w % of the element in the sample,

 A_a is the γ ray peak count of the element in the sample,

 A_s is the γ ray peak count of the element in the standard,

 A_{fs} is the γ ray peak count of the Al flux monitor of standard,

 A_{fa} is the γ ray peak count of the Al flux monitor of sample,

M_s is the mass of standard,

M_a is the mass of sample,

M_{fs} is the mass of the flux monitor of standard,

 M_{fa} is the mass of the flux monitor of sample.

The results of our measurements are shown in Table 3.

TABLE 3. AI AND SI CONCENTRATIONS IN THE SAMPLES

Sample	Al (D.L. =2.7) w %	Si (D.L. = 0.9) w%
Bauxite std. (BCS-395)	28.2±2.1*	N.D.
Cement std. (BCS-372)	N.D.	9.6±0.6**
Feldspar	12.4±1.8	35.6±1.9
Kaolin	12.2±1.4	34.4±1.8
Bauxite 1	17.8±1.6	19.7±1.1
Bauxite 2	15.4±1.4	19.2±1.1
Bauxite 3	12. 8 ±1.2	20.0±1.1
Clay ball	18.2±1.7	21.7±1.1
Sillimanite 1	20.0±1.3	20.1±1.1
Sillimanite 2	21.7±1.5	19.5±1.1

N.D. Not detectable

The deviations of our measured value from quoted values were found to be 1.6% and 1.3% for Al and Si respectively. This experimental procedure is currently being used for the characterization of bentonitic clay used in the petroleum industry.

^{*} Cited value = 27.7 w %

^{**} Cited value = 9.5 w %

5. THE PROBLEMS AND THE FUTURE

The major problems encountered in the operation of the neutron generator include the lack of spare parts and non-availability of SF_6 gas and FREON-113 in the country. CERT will continue to rely on the goodwill of the Agency with regards to the above. Another problem encountered in the use of the research via the activation technique is the scarcity of certified reference materials. To address this problem, CERT is currently developing in-house standards. The inadequate supply of liquid N_2 for the HPGe detector has been addressed by evolving this procedure involving the use of the neutron generator in combination with the NaI(Tl) detector.

The provisional fast neutron activation analysis experimental arrangement with the 5 Ci Am-Be source at the Center has limited the use of the neutron generator for routine elemental analysis. In this regard, the concentrations of elements like Si and Fe in ores have been determined using not moderated neutrons from Am-Be source.

Presently, a miniature neutron source reactor (MNSR) is being installed at CERT. It is expected that the use of the neutron generator for routine analysis via the fast activation analysis mode will be reduced further following the availability of fast neutron field at the irradiation sites of the reactors. For example, the concentrations of Si, Ti, and Ni in different materials have been determined by placing the samples in movable BN or Cd-shielded vials [11]. It is envisaged that the neutron generator will be used only for those elements that are not suitable with the reactor and the Am–Be neutron source because of the operational cost. Indeed, the Center is also thinking of acquiring a pumped-type neutron generator, which can be used for basic and applied research. The limitation of our present facility in the field cross-section measurements and other fundamental research has hindered our participation in the nuclear data experiments of the Agency despite the availability of expertise at the Center.

6. CONCLUSIONS AND RECOMMENDATION

A 14 MeV neutron generator facility is available at the Centre for Energy Research and Training, Zaria. Despite the operational problems, it is a useful tool for activation analysis and has been used for the training of students, technicians and nuclear scientists from Nigeria and other African countries. It is currently being used for the characterization of geological ores via the determination of their major and minor compositions. We hereby acknowledge the assistance of the IAEA during the installation and subsequently as an analytical tool for training and research.

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PROBLEMS RELATED TO THE OPTIMAL USE OF ACCELERATOR BASED NEUTRON GENERATORS

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Abstract

The status of neutron generator laboratories, in developing Member States, supported by the IAEA is reviewed.

1. INTRODUCTION

During the last couple of decades IAEA has provided under its Technical Co-operation program technical assistance to ca. 20–25 laboratories in developing Member States to help them establish neutron generator laboratories and scientific programs mostly in fast neutron activation analysis and fast neutron physics. The laboratories were provided with funds to purchase neutron generators and additional equipment for scientific programs. Similarly, more laboratories were provided with additional equipment to support neutron generators procured from own or other sources. The efficiency of these investments seems to be very low: most of these neutron generators do not work or are in still-stand due to a lot of problems. The optimal use of the accelerator based neutron generator laboratories depends on several parameters: convenient scientific programs, adequate technical and experimental (accelerator) background as well as on the competency and proficiency of the academic and technical staff. These conditions are mostly not independent, they determine collectively the success of a research laboratory.

2. SCIENTIFIC PROGRAMS IN NG LABORATORIES

The scientific programs of the NG laboratories are the nuclear data study of general or specific interest: increase the nuclear data accuracy of fission, fusion, activation analysis; application of neutron generator irradiation for other scientific purposes (material science, biology, medicine. etc.); nuclear physics and technology teaching [1, 2]. The main tendencies of recent and most important scientific programs of the different neutron generator laboratories discussed during the regular international meetings are as follows:

- 1. Neutron induced nuclear reaction studies,
- 2. Benchmark studies based on pulsed sphere experiments,
- 3. 14 MeV neutron cross section measurements,
- 4. Radiation damage studies of construction materials and components induced by 14 MeV neutrons.
- 5. Biomedical applications,
- 6. Fast neutrons in radiation therapy,
- 7. Charged particle acceleration, PIXE elemental analysis,
- 8. Ion implantation by same accelerator used for neutron production.

The most sophisticated multipurpose neutron generators, like the OKTAVIAN in Osaka demonstrates practically all of the important topics and possibilities related to a neutron generator [3]. Typical large conferences like [4] indicate the main direction in fast neutron related research activities. The improvement of the accuracy of the nuclear data requested by

nuclear fission reactors and the fusion related microscopic and macroscopic data are continuous programs. The study of the neutron physical behavior of bulk samples delivers data for the radiation protection, material analysis, etc..

The scientific programs in less sophisticated neutron generator laboratories are related to the studies of neutron induced nuclear reaction by the activation method; 14 MeV neutron cross section measurements; radiation protection and damage of construction materials and components; fast neutron activation analysis; charged particle acceleration, elemental analysis by PIXE and ion implantation. These programs may utilize successfully the commercial neutron generators with their moderate output parameters. The double differential fast neutron cross section, pulsed sphere benchmark and other time of flight measurements demand much more sophisticated neutron sources and measuring instrumentation than what the originally for activation analysis established laboratories have. The double differential cross sections are measured almost exclusively by laboratories having developed their own neutron generator with nanosecond pulsing.

3. TECHNICAL BACKGROUND: SIMILARITIES AND DIFFERENCES OF THE NEUTRON GENERATOR LABORATORIES

The technical background of NG laboratories can be divided into neutron generators and associated instrumentation. Reviews of neutron generators [5] and related technical problems are summarized in [6–10]. Some neutron generator laboratories have multipurpose, relatively useful neutron generators [11] or more dedicated, DC, microsecond and nanosecond pulsed neutron generators [12,13]. The most sophisticated multipurpose neutron generators like the OKTAVIAN [8] have their own perspective in the form of further development of their performance [14]. The IAEA has provided commercial neutron generators to several laboratories. The most common commercial neutron generators are the SAMES J-25 or J-15. the MULTIVOLT NA-150 and the KFKI NA-4 generators. A couple of KAMAN (TMC), Texas Nuclear, Russian and Toshiba generators [8] are still in use as well. The use of sealed tube neutron generators like the sealed tube KAMAN A-3045-6 has the advantage of its easy to use construction, tritium pollution free operation and the disadvantage of a relatively short lifetime (determined by the decay of the tritium sealed in the tube) and very high operational expenses. The use of a sealed tube neutron generator in "problematic" laboratories has a very low efficiency. The most economical neutron generators are the pumped neutron generators where a fresh tritium target renews the neutron source. In case of the sealed tube type, the neutron yield depends on the age of the tritium contents of the sealed tube. The compactness and the low tritium pollution behavior of the sealed tube enables them to be used in biological and medical laboratories. The use of a sealed tube neutron generator for physical investigation may be limited due to the wide ranging demand of physical laboratories. The most widely used neutron generator for physical investigation is the pumped type. The advantages of the pumped neutron generators are: long life, easy maintenance and repair, ion species selection for monoenergetic neutron production, possibilities for pulsing the neutron source and utilization of them in charged particle reaction and PIXE studies. Disadvantages of these generators are that they require longer time to achieve the operational pressure in the vacuum system, they also require a relatively large room for installation.

The upgrading of present neutron generators in laboratories of more or less developed countries is a function of their scientific programs. The vacuum system, the beam line and the target assembly are usually parts of accelerator based neutron generators that require continuous maintenance. Depending on the requirements and the local circumstances, every alteration of the vacuum system should improve the parameters of the system. An analyzed

beam on the target will result in more monoenergetic neutrons which is essential for fast neutron cross section measurements and for activation analysis. The target assembly is influencing the neutron field around it: the best target assembly for monoenergetic neutron measurements is the wobbling and air cooled type.

The bunching, down to microseconds, of the accelerated beam by ion source pulsing or beam deflection enables activation analysis, nuclear data measurements and studies of neutron diffusion in bulk media. The (fast) neutron activation analysis requires a neutron source (the neutron generator), gamma spectrometers — NaI(Tl) or HPGe detectors — and additional laboratory equipment. For relative measurements, the internal monitorization does not need the knowledge of absolute detector efficiency. However, when using the activation technique for measurements of nuclear data the neutron yield monitoring and the knowledge of the absolute detector efficiency is fundamental.

Nanosecond pulsing of neutron generators is required for double differential cross section measurements and pulsed sphere benchmark experiments. The detectors and their timing requires in this case experienced people and very stable, sophisticated experimental systems. The evaluation of the measured data — especially in the case of proton recoil spectrum unfolding requires a well trained staff with computer knowledge.

In the case of the above mentioned investigation the measuring systems utilize commercial units. The detector arrangements and the interfacing between the experimental set-up and the accelerator are problem specific and are usually developed in the laboratory. This is the point where the knowledge and experience of the staff is fundamental. This background factor determines strongly the total efficiency of the whole laboratory.

The problems related to the repair and availability of spare parts are strongly influenced by the origin of the neutron generator. If the neutron generator is self developed and built, the problems are not so significant: the staff may manufacture spare parts or they know the origin of the different components. In this case there are usually enough spare parts and components around in the laboratory, because during the construction different compatible version of the components were usually manufactured. Usually the constructor of the NG knows exactly the function of different components, making it is easier to locate and replace faulty ones.

The similarities of the accelerator based neutron generator laboratories are:

- Accelerator based neutron production by nuclear reactions,
- similarities in accelerator technology (vacuum and high voltage techniques, etc.),
- possibilities for alteration of accelerator use,
- same or similar commercial equipment.

The differences of the accelerator based neutron generator laboratories are significant depending on if the accelerator is self-constructed or commercial. The construction of accelerators needs a skilled and experienced staff with adequate technological background. The cost of a self constructed accelerator is not in every case less than the cost of a commercial one, but the experience of the staff gained during the design and construction gives a very high benefit for the everyday operation. In case of problems with the neutron generator, the staff may carry out troubleshooting faster when they know the operation and function of each component. The commercial accelerators were delivered ready to use after installation in the laboratory. The operators know the function of the components only

roughly, usually they are not familiar with the operation of the subsystems and they do not have any idea where to find these components on the market. Similarly, the lack of knowledge of the system operation may results in the omission of required maintenance, which may lead to more failures.

4. THE EXPERIMENTAL BACKGROUND

The experimental equipment of neutron generator laboratories are practically everywhere the same: detectors, analogue data processing and test electronics. Differences — depending on the feature of the experiment - are only in detector shields and geometrical layout, experimental halls, etc.. One of the most important parts of an experimental set up is above-mentioned interfacing of the measuring system to the accelerator. This task requires a highly experienced staff.

5. PERFORMANCE OF THE NEUTRON GENERATOR LABORATORIES

The neutron generators with their application capabilities are potentially cost effective equipment in nuclear science programs. This was the reason for the IAEA policy of providing substantial technical assistance to the establishment of NG laboratories and related programs. This program has an amount of IAEA investment (until 1989) in expert services of approximately 110 man-months and US\$ 4,000,000 in equipment [15]. Unfortunately, the efficiency of the investment seems to be very poor.

The survey of the participants in a large conference like [4] may indicate a very small participation from countries where IAEA invested relatively large amounts of money in equipment, fellowships and missions. In the case of the above-mentioned Jülich conference, there were less than 20 participants from IAEA sponsored laboratories albeit the total number of participants from 37 countries was 328, corresponding to 6 % of the participants. Taking into account that most of these 20 participants were co-authors of a common paper or in a work carried out somewhere else, this ratio is even lower. Unfortunately, there exist laboratories without any publications or results related to their IAEA sponsored neutron generator. On the contrary, the OKTAVIAN facility was the basis for 153 papers from 1983 to 1996 [3]. An another figure of merit that may characterize the IAEA investments in these laboratories is the number of operation hours of these neutron generators: this is similarly not high in case of less successful laboratories. In case of sealed tube neutron generator of ca. 200 hours lifetime, the 28 working hours during the last 7-8 years seems not to be a full exploitation.

6. HUMAN FACTORS: EXPERIENCES OF THE STAFF

The staff in the laboratories has different experiences. In the laboratories, where the accelerator and the experimental set-ups are self developed and constructed, the academic and technical staff is well experienced for designing and preparing sophisticated experimental set-ups for carrying out difficult measurements. In laboratories with commercial neutron generators the operators had a relatively short course at the manufacturer on the operation and the actuation of the accelerator. This course was usually during the delivery of the neutron generator. As the building and the radiation shield of the neutron generator sometimes were constructed after the delivery of the generator the operators, mostly technicians, in the meantime left the laboratories. Nowadays, the laboratories have new operators without technical training and experience. This is one of the reasons for their problems and the very low efficiency of these laboratories. Similarly, the operator was sometimes not selected in

accordance with the demands of such a duty: this usually results in a damage to the accelerator. Unfortunately, this is one of the most typical causes of the non-operation of most of the neutron generators.

The fellowships and expert missions from and into the neutron generator laboratories should be analyzed and well evaluated. The technical staff of these laboratories should profit from both. The fellowships in top ranking laboratories do not always help sometimes the fellow, when returning home, may loose his original motivation to do research work in his home laboratory.

7. PROPOSAL FOR THE IMPROVEMENT OF THE EFFICIENCY OF THE AVERAGE NEUTRON GENERATOR LABORATORIES

Depending on the possibilities of different laboratories, each laboratory should determine their own scientific programs on the basis of their intellectual and technical background. As the experimental activity may not neglect their existing technical background, the programs should be adapted to the given technical infrastructure. The technical basis of these laboratories was established during the last decades, so the maintenance, trouble shooting and repair of the experimental basis play nowadays an important role. The training of the present technical staff seems to be actual.

As the present situation of the laboratories are quite different from the condition in the time of the establishment, a new, carefully prepared survey is recommended to be carried out to investigate the present situation related to the equipment and human resources of the of the IAEA established laboratories. This survey should review the present equipment and human situation, discuss the results achieved by the utilization of laboratory investment, analyze the scientific work that has been made and planned to be carried out using the equipment as well as the expected cost of reactivating the laboratories. Experts well skilled in the given field should carry out the characterization of the laboratories. The survey can be accomplished by the study of files of the projects and the publication activity of the institute and it should also involve field studies. This survey may serve to the qualification of the laboratories. The IAEA decision for further investment in these laboratories may be helped by such a study.

The lack of knowledge and experience of neutron generator technology of the present technical staff in the laboratories with commercial neutron generators requires more effective co-operation between these neutron generator laboratories. This is related to the:

- repair, upgrading and operation of existing neutron generators, (Improvement of the vacuum system, ion source, HV terminal, remote control etc.),
- upgrading of the beam line components and related equipment, (Pulsing, use of APM, yield monitoring, beam analyzers, wobbling targets, etc.),
- pool for critical NG components [16],
- Updating the list of suppliers in [4],
- Establishing direct (email) links between the laboratories.

The efficiency of the neutron generator laboratories can be improved by IAEA through common expert missions and regional training courses on NG technology and nuclear electronics. These courses based on the manual [7], and may help to transform the neutron generator laboratories, especially the host into an operating one. In the framework of the course, there is a possibility for common NG and nuclear instrumentation repair that would help the participants to get experienced in these technical fields. As the operators are

electronical engineers or technicians, they may profit a lot from this type of courses. As these courses would be regional courses, they may help to establish the direct links (later the exchange of the equipment and staff) between the institutes in the given region. Location of these training courses should be selected carefully and the host laboratory should organize this course at an acceptable level and the local staff should be a competent for the function. The result expected from such a common expert mission and "instrumentation" training course should be more advantageous than the separate missions and training courses on accelerator techniques and nuclear instrumentation.

8. SUMMARY

The low impact of the neutron generator laboratories supported by the IAEA is motivated by more factors. One of the most important one is a human factor: the skills and the experiences of the technical staff in these laboratories. Some laboratories gave up the experimental works with fast neutron generators, while others try everything without solving the technical problems related to smooth operation of the generators. A survey analyzing the neutron generator laboratory figure of merit may help the decision related to the given laboratory. As the technical staff of the laboratories changed during the last decade, common regional training courses on the maintenance and repair of neutron generators and related electronic instrumentation is warmly recommend.

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RECENT WORKS WITH SEALED TUBE NEUTRON GENERATORS

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Abstract

SODERN is the only European manufacturer of industrial sealed tube neutron generators. In the last decade, we have worked in the field of possible applications for Neutron Generators (NG) which are the detection, analysis and neutron imaging. Our studies and developments are mainly carried out in a co-operation, in which the partner brings its expertise in the application. The responsibility of SODERN is different according to each project: it spreads from the delivery of a neutron generator to that of a whole detection system, including the software transforming roughs measurements into elemental composition.

1. NEUTRON GENERATORS

1.1. The SODITRON

SODERN have some years ago developed a neutron tube, called SODITRON, based on the most recent tube technologies. This tube, sold within an exclusive contract for oil well logging, can be used freely in other fields. It appears to be very reliable and offers a very long lifetime, i.e. thousands of hours, when used with duty cycles greater than about 3 %. These are typically duty cycles used for elemental analysis. These characteristics were the origin for the development of a small NG, the GENIE 16, and a cement analyzer.

1.2. The GENIE 16

The main characteristics are described below:

- Small range neutron output Maximum output: 2×10^8 n/s of 14 MeV neutrons.
- DD tubes can also be used; their neutron yield is approximately 100 times lower.
- The biological shielding is reduced: in case of the maximal output the allowed public dose level is at a distance of about 30 meters from the tube. In many applications, especially when the required output is low, a shielding of less than 1 meter thickness is enough for the above mentioned dose to on the skin from the equipment.
- Unrivaled life time for the tube
 - The lifetime of the tube, due to erosion of the target, has been experienced to be (as expected) inversely proportional to the neutron output.
 - A constant neutron output of 0.5×10^8 n/s can be maintained during 8000 hours before the accelerating voltage is increased up to the values at which the tube operation breaks down.
- Industrial use: The above characteristics, added to an attractive price (between 350 and 510 kFRF according to options) are suited to industrial application. The replacement cost for the tube varies between 120 and 152 kFRF; for the latter cost; the user changes the whole emitting probe in a very simple manner and with absolutely no hazard regarding tritium contamination. The used tube (or emitting probe) is returned to SODERN who takes care of the disposal.

- Portable equipment: Some applications are in-field analysis, and the GENIE 16 meets their requirements. The probe is light (7 kg), with a 10 cm diameter and a length of about 75 cm. All the electronics, together with the cables weight 30 to 45 kg (according to options). The equipment is powered from the mains, and no additional power supply is required.
- Multipurpose equipment: The basic version delivers a continuous emission as chemical sources do, but with the definite advantage of safety, because the output can be stopped when the accelerating voltage is switched off.

For a better analysis, the ion source voltage can be modulated to control the neutron output. A small pulser unit, diameter of 10 cm, can be added to this probe and controlled with an external pulse generator.

An option for the control of the NG through an external computer is available; it is delivered with the necessary software to control all the parameters. This option also delivers the signal for the control of the ion source.

The neutron emitting module has two main versions. In the first one, the tube is insulated by gas (SF6) so it can be changed at a low cost, the spectrum of the emitted neutrons consists of mainly not scattered 14 MeV neutrons. In the second one, the tube is molded into epoxy resin making the replacement of it simpler and the thermalization of emitted neutrons more direct. In this case the heat dissipated by the tube power limits the maximal neutron output.

In conclusion, the proper solution depending on the needs, can be found among the existing options.

1.3. Ng for transuranium detection

For 12 years SODERN have contributed to the detection of fissile material by the Differential Die Away technique. This technique requires short and high intensity neutron pulses: the TN26 neutron tube is well adapted to this specific requirement. Following the GNT 02, the GENIE 26 it is used in active measuring cells, to control the nuclear waste drums or the dissolution of fuel within their hulls in the reprocessing plants.

The SODITRON tube is also used for this application, but its lifetime expectancy is limited to some hundreds hours in this specific mode. As a result of the positive feedback related to the use of the GENIE 26, COGEMA has asked SODERN to study the development of a much more powerful NG which could delive 2×10^9 n/s. The development is now completed and this equipment will be available through COGEMA/SGN for use in the nuclear industry, or directly from SODERN for utilization them in other fields.

2. ELEMENTAL ANALYSIS

For these applications, the contribution of SODERN is not limited to the NG, but also includes the detection and the necessary signal processing software. The interfacing to the above system are the responsibility of the user.

2.1. Bulk analysis

SODERN have worked during some years on an on-line bulk analysis process. The measurement is aimed at quantitative determination of major components, and qualitative detection of some minor ones. Successful studies and laboratory tests were done on aluminum and crude cement.

2.1.1. Cement analyzer

As this market opens in the field of cement industry by analyzers using isotopic sources, SODERN will be able to develop the heart of the cement analyzer. The project is now in its commercial phase: the whole equipment is proposed to the Krupp Polysius engineering company, and it includes SODERN electronics. The POLAB $^{\text{\tiny M}}$ - CNA is a continuous one-line analyzer which characteristics have been demonstrated very recently on a prototype, simulating actual conditions and environment. Table 1. gives the specification on this equipment. All the dynamic tests were performed successfully for different kinds raw material of cement.

TABLE 1. DYNAMIC ACCURACY FOR A MEASURING RANGE

Elements expressed	Test range	(σ) Dynamic
as oxides		accuracy 10 min.
% SiO ₂	0 - 24	± 0.35
$% Al_2O_3$	0 - 6	± 0.30
$\% \text{ Fe}_2\text{O}_3$	0 - 5	± 0.10
% CaO	33 - 56	$\pm \ 0.40$

2.1.2. Basic design of material analyzers

The basic design is the same for different materials to be analyzed, but of course, the definition of each subassembly depends on which elements have to be measured, in order to minimize the background. The physical processes are Fast Neutron Analysis, Thermal Neutron Analysis, and even Neutron Activation Analysis. They are used simultaneously, by means of pulsed neutron output, high efficiency detectors (BGO large size scintillators). Special high counting rate electronics was designed by SODERN with specific software using sophisticated algorithms.

2.1.3. Main characteristics

The cement analyzer and other bulk material analyzers that could be designed have the same basic characteristics:

- fully automatic measurement, without sampling,
- safety related to the radiation hazards,
- relative easy neutron source transport,
- good accuracy,
- low maintenance cost.

Regarding the accuracy, two advantages of the NG compared with the radioactive sources must be emphasized. Firstly, the neutron output can be kept constant during the whole life of the tube; consequently, the need for recalibrating the equipment is much less. Secondly, and because the neutron output is low in order to get a long life-time for the tube, it is always possible to increase for a few minutes and get a better accuracy, typically increased by a factor of 2.

2.2. Detection by using FNA and TNA

Different applications have been studied or are under study. They are based on the intrusive properties of 14 MeV neutrons and on the fact that when the detection time is increased the measurement becomes less quantitative. The measurement of C, H, O, N combining FNA and TNA could have applications for drugs or explosive detection. Due to the short measuring time, the neutron output seems to be necessary for bulk analysis.

An application for the GENIE 16 is the characterization and sorting of chemical weapons: its is possible to sort the ammunition according to the quantity of explosive, but also according to the composition of the contained chemical gases. It should be emphasized that the pulsed neutron emission of the NG allows the detection of very short lived isomers, such as Arsenic. It has been demonstrated that the presence of Arsenic in a shell with 2.5 cm a steel walls, was possible within 5 min.

A promising application is the detection of abandoned land mines. In combination with other techniques which can detect quickly a possible land mine, neutron analysis can confirm in a few minutes, whether the suspected object contains explosives or not. In this application, the main problem is the geometry, because the number of neutrons on the object is low, and the detection efficiency is small, according to the narrow solid angle.

The contribution of SODERN within the different projects, has been mainly directed forwards to a better use of the NG, specially regarding the proper choice of pulse width and pulse repetition rate for optimizing the signal to noise ratio.

2.3. Detection by induced fission

This physical process is used in nuclear industry, as shown in paragraph 1.3. Other applications were presented to IAEA in October 1995, cover:

- determination of natural Uranium in ores, down to 200 ppm, using logging probes equipped with a SODITRON tube,
- inspection of nuclear weapons,
- detection of plutonium pollution, down to 10 nCi / g with a logging probe,
- particularly: different nuclear materials.

In this latter case, it is possible to identify the neutron shielding by neutron capture, and quantities of ²³⁵U as low as 20 grams by induced fission. utilizing portable equipment with a GENIE 16.

2.4. Inspection of fuel storage ponds

A study has been completed with ATEA-Framatome in the frame of a project with Electricité de France. The possibility of detecting defects such as holes, cracks or smaller thickness in the neutron absorbing sheets dividing the storage ponds for used fuel elements in the nuclear power plants, have been demonstrated. Such an inspection asked by the safety authorities now will be possible without the hazards due to the use of radioactive neutron sources.

3. NEUTRON RADIOGRAPHY

A large amount of studies and development has been done in the field of neutron imaging. The process is used for about 25 years, but the objects to be inspected must be brought to existing facilities, that is close to a nuclear reactor. Mobile neutron radiography, using high output NG, has been demonstrated as effective to detect early corrosion, lack of gluing in aeronautics, and to inspect small objects such as turbine blades or pyrotechnic devices.

The market is not big until now, but we have some hopes that a new design, the fan beam geometry allows a much larger inspection zone around the NG target by a slight reduction of the thermal neutron yield.

A powerful NG, the GENIE 46, has been developed as basic equipment for neutron radiography. It delivers a continuous 14 MeV output with a maximum yield of 4×10^{11} n/s, and the life-time of the tube has been experienced to be 500 hours until the neutron output falls down by a factor of two. In industrial applications, this means that, by controlling the accelerating voltage, a constant output of 2×10^{11} n/s can be achieved during about 2000 hours.

In conclusion the GENIE 46 is well suited for Neutron Activation Analysis and can be utilized for many applications in the industry.

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