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Editorial

Characterization and examination of radioactive materials is a critical element in a number of priority programmes of the department including waste disposition, national security, nuclear non-proliferation and nuclear facility operations. Quality of a Nuclear Fuel is regarded as 'Conformity to Commitment' at all stages of its conduct initiating from its fabrication, operation in a reactor and reprocessing. Non-destructive techniques are essential for evaluation of the material to meet the desired specifications in finished form as well as while performing in a reactor. Advanced techniques need to be developed for direct measurement of possible defects and to reduce uncertainties in assumptions associated with excessive conservatism. Research focused on the significant long-term effects of aging is needed to engorge advanced techniques, which will have a positive effect on capacity factors. This bulletin is aimed at giving a range of techniques employed in the fuel cycle using nuclear radiation.

IANCAS compliments Dr.G.K.Gubbi, Guest-Editor of this bulletin for having chosen experts in the field from many agencies to author the articles and critically going through the manuscripts. Special thanks are due to the authors for having taken the trouble in submitting the articles in time.

A special feature on 'IANCAS Roundup' is brough out in this bulletin to help the members in providing feed back and offer suggestions on IANCAS activities such as National Workshops, demonstration programme for young students in schools / colleges and thematic bulletin to make them more effective. Readers are requested to contribute News Items of general interest in Nuclear Science and Technology to be published in the bulletin under 'NUCLEUS' for the benefit of the fellow members. We sincerely hope that this column would encourage readers to interact with IANCAS in a better way.

G.A. Rama Rao

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President's Message

Dear fellow members of IANCAS fraternity,

I am thankful to all the members for selecting and electing me as the President of IANCAS for the term 2003-05. I am delighted to have a large number of members in EC who have varied and significant contributions in their chosen field. IANCAS has been doing yeomen service to the academic community and students by organizing the National Workshops on Radiochemistry and Application of Radio-isotopes at several universities and colleges over several years. For this, IANCAS is thankful to Chairman, BRNS, Chairman, Basic Sciences committee, Secretary and Program officer, BRNS for giving us grants and supporting the activities of IANCAS. IANCAS is much beholden to Chairman, AEC and Director , BARC for their unstinted support and praising IANCAS activities in all forums of scientific information dissemination.

We have already completed 50 such ten day National Workshops. As far as school Workshops are concerned, the experiment on Radioactivity has kindled interest in such a large way that invitations for such programs are pouring in. We plan at least ten school programs in a year. In addition, five to seven school programs are conducted wherever National Workshops are organized. Though these two programs are the backbone of our association, we have to think of innovative ideas to attract talent to man-power deployment in the next decade. While appreciating the efforts of IANCAS in organizing the Workshops at different universities, Chairman, AEC emphasized the need to create a pool of talented young scientists for the long term continued sustenance of our nuclear programme. I urge my fellow members and EC members to ponder over new and innovative ideas to further the cause for which IANCAS was created.

Nuclear power and the by-products, radio-isotopes are going to play crucial role in the years to come in our country. At present India is the only country setting up eight reactors simultaneously (TAPS 3,4; RAPS 5,6;Kaiga 3,4 and Kudankulam 1,2). In addition we are embarking on advanced heavy water reactors with all passive fail-safe features for the utilization of our abundant thorium reserves. Today you see a renaissance in the world in nuclear science and technology. The advanced countries, some of them under green government started to think eco-friendly power generating system and slowly leaning towards nuclear energy, as renewable sources of energy may not be at present realistic. Advanced countries and IAEA talk of IV and V generation reactors. India has set a moderate target of 20,000MW(e) by 2020. In order to realize it, a large scientific personnel input will be required. This can come only from Universities. So IANCAS has to put in more efforts to attract talent from Universities to take forward the departments program. I happened to interact with an European scientist recently. He commented about our wisdom in continuing the nuclear program and increasing the man power in this field. He was lamenting that many of the American and European Universities have no sufficient faculty to teach Nuclear Science and engineering. He mentioned that just like IT revolution, the nuclear science revolution is in the offing and at that time other countries have to look at India for their man power requirement.

Well, friends, this is enough for the first issue of the bulletin from the new EC. I request members to send interesting information on science and technology in general and nuclear in particular to bring it to the attention of the fellow members through the bulletins.

V. Venugopal

From the Secretary's Desk

Dear Members,

I take this opportunity to express my gratitude to all the members for having given me the opportunity to serve the Indian Association of Nuclear Chemists and Allied Scientists (IANCAS) as the General Secretary.

As you are aware, IANCAS is just out of its teens. Established formally in the year 1984, it has come a long way. Today it has 1073 life-members, twenty odd corporate members and a southern chapter which was started in 2002. It is noteworthy that a large fraction of the life-members are from the Universities and non-DAE research Institutes.

The primary objective of the Association is to popularise nuclear sciences in general, and, Radiochemistry and applications of radioisotopes in particular. It is being achieved through National workshops, seminars, school/college workshops, awards and through various publications. The National workshops are the most important activities of IANCAS. It has trained more than 2000 researchers in the area of radioactivity handling and research using radiotracers through 50 National workshops, and, about 10000 students have benefited from school/college workshops conducted all over the country. The relevance of these workshops has increased manifold in the light of DAE's plan to set up a large number of power reactors in the coming years. The objective is not limited to human resource development alone. These workshops provide a platform to project the nuclear power program in correct perspective to a group of opinion makers who act as information multipliers. Also, it has helped in sensitising the young minds to the benefits of R&D activities in nuclear sciences to the society in general.

Our thematic quarterly bulletins and books are in great demand and find overwhelming admiration. Some of the books are even translated to Arabic, Hindi and Tamil! As you are aware, IANCAS has instituted awards for excellence in research for young scientists as well as senior scientists. This has been and will continue to be a motivating factor for researchers in this area.

This success story is the outcome of hard work, perseverance and dedication of members and many of the past/ present office bearers. Today, we have to shun complacency and our priorities should be to sustain and expand the current activities. The Advanced Workshop, which is proposed to be organised, is one of the means to consolidate the gains made so far by IANCAS towards popularising nuclear sciences. Sustaining and improving the quality of our quarterly bulletins in terms of contents and other features is another area that requires your active support, suggestions and constructive feed-back.

I take this opportunity to acknowledge the generous financial support from the Department of Atomic Energy(DAE) through the Board of Research in Nuclear Sciences (BRNS) which has enabled us to work towards achieving our objectives..

I thank you again and look forward to your support.

P.K. Pujari

IANCAS Publications - Price List

IANCAS published books that contain wealth of information on nuclear technology, nuclear materials and applications of radioisotopes and are being made available as a special discount prices for its members.

	Life Member	Non-Member
Fundamentals of Radiochemistry *D.D. Sood, A.V.R. Reddy and N. Ramamoorthy*	Rs.200	Rs.250
Nuclear Materials *D.D. Sood*	Rs.100	Rs.150
Experiments in Radiochemistry *Eds. D.D. Sood, S.B. Manohar and A.V.R. Reddy*	Rs.50	Rs.75
Frontiers in Nuclear Chemistry *Eds. D.D. Sood, A.V.R. Reddy and P.K. Pujari*	Rs.100	Rs.150

Persons desirous of pruchasing these books are requested to send the cost of the books with an additional Rs.40 towards postal expenses through a Demand Draft in favour of IANCAS to the following address:

Dr. P.K. Pujari
General Secretary, IANCAS
Radiochemistry Division
Bhabha Atomic Research Centre
Mumbai 400 085

Forthcoming IANCAS Workshops on Radiochemistry and Applications of Radioisotopes

- 51st BRNS-IANCAS National Workshop will be held at Guru Nanak Devji University, Amritsar during November 10-17, 2003. For details contact :

Workshop Director

Dr. R.K. Bedi
Department of Physics
Guru Nanak Devji University
Amritsar
E-mail : rkbedi@rediffmail.com

- 52nd BRNS-IANCAS National Workshop will be held at Nagarjuna University, Guntur during February 2-10, 2004. For details contact :

Workshop Director

Dr. K. Sambasiva Rao
Department of Biotechnology
Nagarjuna University
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Non-Destructive Assay Techniques in Nuclear Fuel Cycle

Guest Editor

Dr. G.K. Gubbi

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FOCUS

Dr. S.B. Manohar

The performance of nuclear fuel is based on its integrity under hostile operating environment in the reactor. This is achievable with a stringent quality control of the nuclear material while it passes through various phases during fabrication. Non-destructive techniques for quality control of nuclear fuels at various stages of processing are very important in the successful implementation of a closed nuclear fuel cycle. The nuclear material follows a cyclic path of processing as purification, fuel fabrication, utilization in the reactor and ultimately the reprocessing. The quality control and material accounting of fissile materials at each stage assume significance considering their chemical and radiation toxicity. A possible pilferage of plutonium is also of concern in view of its strategic merit.

In mid-seventies development of non-destructive techniques for the assay of nuclear materials was initiated in BARC by Late Dr. M.V. Ramaniah. Several other agencies of DAE have contributed immensely in this field.

Having perfected the techniques covering the entire gamut of nuclear fuel cycle, time is opportune to bring out a bulletin on various non-destructive techniques that are in vogue. I was closely associated with the development of nondestructive techniques using neutron and gamma radiations in Radiochemistry Division, BARC. I am happy that IANCAS has chosen to highlight the nondestructive techniques in the form of a bulletin for the benefit of the experts and general awareness to the members. The present bulletin on 'Non-destructive techniques in Nuclear Fuel cycle', with Dr. G.K. Gubbi as the Guest Editor, is an attempt to emphasize prominent techniques based mostly on nuclear radiations. However, some glimpses of other techniques employing non-nuclear radiations are also briefly described in this bulletin.

The articles presented in the bulletin provide sufficient coverage on the basics of the techniques using several nuclear radiations. The applications of these techniques range from fuel fabrication, operating reactors, post irradiation studies, reprocessing and management of nuclear wastes. These techniques are need based and are vital for ensuring the quality control and safety aspects in the Nuclear Fuel Cycle.

Guest Editorial

Dr. G.K. Gubbi

Radiochemistry Division

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I am happy to be associated with the Thematic bulletin of IANCAS covering various aspects of Non destructive assay techniques used at various stages of nuclear fuel cycle. The techniques make use of nuclear radiations, lasers, ultrasonic and eddy currents. The main emphasis was on the development of techniques based on nuclear radiations. The extensive work carried out using ultrasonic waves, eddy current and lasers is mentioned very briefly.

The Non destructive assay techniques have been used in a broad sense covering quantitative assay and quality control of fuel materials, evaluation of the critical reactor, reprocessing plant components and safe operation of nuclear reactors. The techniques for quantitative assay of nuclear materials and quality control of finished fuel products depend mostly on use of inherent or external nuclear radiations.

The use of neutrons and gamma rays in the development of NDA techniques for the quantitative assay of plutonium and uranium based nuclear materials at various stages of nuclear fuel cycle has been well brought out in the articles by Shri S.P.Dange, Dr. G.K.Gubbi, Dr.A, Ramaswami, Dr.Sarbjit Singh, Dr. B.S.Tomar Shri Rahul Trpathi. And Dr. P.R.Vaidya). The techniques used at Nuclear fuel complex which meets the uranium fuel requirement of the power reactors of India have been described by shri B. Laxminarayrayana and Shri R.K. Srivastava. NDA techniques developed and used for the quality control of plutonium based fuel pins are aptly illustrated in the article by Shri Muralidhar, Ms. K.V.Vrindadevi and Shri H.S.Kamat. Radiography is very important for non destructive evaluation technique in fuel fabrication facilities and different innovative techniques developed indigenously are elucidated lucidly by Shri K.N.Chandrasekharan and S.Majumdar. The performance evaluation of the irradiated critical reactor components, burn up and host of valuable information obtained from gamma scanning of reactor components and fuel pins discharged from power reactors has been summarised elegantly by shri Suresh Gangotra, Shri M. Ouseph and shri K.C. Sahoo. Shri M.S.Randhawa illustrated a typical contribution of NDA techniques for the safe operation of nuclear reactors very crisply. Salient features of calorimetry as a tool for assay of nuclear materials are well presented by Dr.V.Venugopal The complexities and the challenges of Fast Breeder Reactor along with associated of Reprocessing of spent fuel and the NDA techniques developed for meeting a very challenging task are precisely covered by Dr. Baldev Raj, Dr. P.Kalyanasundaram and Shri K.V. Kasivishwanathan.

The science and technology of non destructive assay and evaluation techniques is very vast and the scope of the bulletin is not to give an exhaustive coverage, but to introduce prominent need-based techniques developed for enhancing the performance and safety in nuclear power programme.

I thank Dr. G.A.Rama Rao, Editor, IANCAS for inviting me to be the Guest Editor. Thanks are due to my former colleague, Dr.S.B.Manohar for helpful discussions and also for providing a focus to the bulletin. I gratefully acknowledge Dr. V.Venugopal, President, IANCAS and Dr. V.K.Manchanda, Vice-President, IANCAS, for constant encouragement and support throughout the course of this editorial work.

Neutron Counting Methods and Nuclear Materials Assay

Shri S.P. Dange *joined Radiochemistry Division in 1967 after graduating from 10th Batch of BARC Training School. His main fields of interest are nuclear fission and nuclear reactions and the development of nondestructive assay techniques. He has developed different types of neutron and gamma based systems for the nondestructive assay of plutonium at various stages of nuclear fuel cycle. He has published more than 100 papers in international journals and symposia*

Dr. G.K. Gubbi *obtained M.Sc. (Nuclear Physics) from Karnatak University and joined 11th Batch of BARC Training School in 1967. Initially he worked on the experimental determination of reactor physics parameters for heavy water moderated multi-rod UO_2 clusters using ZERLINA reactor. He was deputed to Australian Atomic Energy Commission for a year under Indo Australian Exchange Program. He was actively associated with the development of gamma scanners for the quality control of FBTR as well as plutonium based fuel elements for Tarapur Reactors. His current interests are heavy ion induced nuclear fission and the development of non-destructive assay techniques at various stages of nuclear fuel cycle. He obtained Ph.D. from University of Mumbai in heavy ion induced nuclear reactions. He has more than 60 publications to his credit in international journals, symposia and technical reports.*

Introduction

Non destructive assay techniques (NDA) based on neutron measurements play an important role in the assay of plutonium in various stages of the nuclear fuel cycle. The NDA techniques find wide ranging applications in quality control of finished products, Nuclear Material Accounting, Process Control and Safeguards. The techniques are direct, fast and amenable to automation. They are versatile with respect to chemical composition and physical configurations, unlike the conventional chemical techniques which are more accurate but slow and need more rigorous sampling. The techniques can be classified as passive or active depending on how the response is obtained. In the passive mode, the radiations from the natural radioactive decay of the isotopes of interest are monitored. In the active mode, the delayed neutrons /prompt neutrons or gamma rays from the samples irradiated by the neutrons are monitored. The passive mode based on neutron counting is best suited for heterogeneous samples of various configurations compared to more sensitive passive gamma counting because of self attenuation problems and low abundance of gamma rays.

Origin of Neutrons in Plutonium Samples

The neutrons emitted by a plutonium bearing sample consist of spontaneous fission (SF) neutrons from even-even plutonium isotopes ($^{238,240,242}Pu$) as well as (α,n) neutrons induced by interaction of alpha particles with low Z elements like, O, C, F, Al etc. The thick target neutron yields per 10^6 alpha

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TABLE 1. Thick target yields from (α, n) reactions

Element	Neutron yield per 10^6 Alphas of energy 4.7 MeV (^{234}U)	Neutron yield per 10^6 Alphas of energy 5.2 MeV (Av. Pu)	Av. Neutron energy (MeV) for 5.2 MeV alphas
Li	0.16 ± 0.04	1.13 ± 0.25	0.3
Be	44 ± 4	65 ± 5	4.2
B	12.4 ± 0.6	17.5 ± 0.4	2.9
C (^{13}C)	0.051 ± 0.002	0.078 ± 0.004	4.4
O (^{17,18}O)	0.040 ± 0.001	0.059 ± 0.002	1.9
F	3.1 ± 0.3	5.9 ± 0.6	1.2
Na	0.5 ± 0.5	1.1 ± 0.5	
Mg (^{25,26}Mg)	0.42 ± 0.03	0.89 ± 0.02	2.7
Al	0.13 ± 0.01	0.41 ± 0.01	1.2

TABLE 2. Nuclear data of Plutonium isotopes

Isotope	SF Half-life (y)	ν (SF)	SF n/g/s	Half life (y)	(α,n) /g/s for PuO_2
^{238}Pu	4.9×10^{10}	2.26	2.59×10^{3}	87.74	1.34×10^{4}
^{239}Pu	5.5×10^{15}	2.2	2.18×10^{-2}	2.4×10^{4}	38.1
^{240}Pu	1.17×10^{11}	2.17	1.02×10^{3}	6.6×10^{3}	141
^{241}Pu	5.0×10^{15}	2.2	5.0×10^{-2}	14.4*	1.3
^{242}Pu	6.8×10^{10}	2.16	1.72×10^{3}	3.79×10^{5}	2.0

*Alpha decay branching intensity : 0.00246%

particles for typical elements are shown in Table 1 [1]. The nuclear data of plutonium isotopes, SF neutron emission rates and the (α,n)neutron yields/g/s for oxide matrix are shown in Table 2. In active neutron interrogation, delayed neutrons and prompt neutrons emitted by the neutron irradiated sample are used for the assay fissile materials.

Neutron Detectors

Neutrons are detected through nuclear reactions which may result in energetic charged particles, capture gamma rays, fast recoils, conversion electrons or fission products. The thermal neutron reactions resulting in charged particles for the three types of detector materials are given below.

$^{10}B + n \rightarrow {}^{7}Li + \alpha$

Q-value = 2.792 MeV (ground state) (6%)

${}^{7}Li^{*} + \alpha$

2.310 MeV (excited state) (94%), σ_{th} =3840 b

$^{3}He + n \rightarrow {}^{3}H + p$

Q-value = 0.764 MeV, σ_{th} = 5330 b

$^{6}Li + n \rightarrow {}^{3}H + \alpha$

Q-value = 4.78 MeV, σ_{th} = 940 b

Most of the neutron sources also emit gamma rays and it is required to discriminate against gamma rays in the detection process. The gas based counters using ^{3}He and $^{10}BF_3$ and fissile materials offer more effective gamma discrimination. The Q value of the reaction is also important which determines the energy liberated in the reaction. The higher the Q value, the greater is the energy liberated and it is possible to discriminate against the gamma rays by simple pulse height discrimination. High efficiency ^{3}He (Nat. Ab = 100%) detectors are feasible because

of high neutron absorption cross section for thermal neutrons and higher possible fill pressures up to 4 atm. Comparatively, BF_3 detectors are less efficient because pulse degradation limits fill pressures (0.2-2 atm max.) and lower absorption cross section for thermal neutrons for ^{10}B (nat. ab. 19.8%). However the Q value for neutron reaction with ^3He is lower than ^{10}B. Hence for neutron measurement of highly gamma active samples, BF_3 detectors are preferred as better gamma discrimination is possible. For the passive neutron assay of plutonium, ^3He counters are used because of availability of high efficiency compact detectors. The neutron absorption cross section decreases rapidly as the neutron energy increases and hence moderation of neutrons is necessary to increase the detection efficiency of the counting system. The neutron detectors are required to be housed in a suitable moderator assembly for enhancing the detection efficiency of the counting system.

Moderator Assembly and Counter Configuration

The important characteristics of good moderator are high moderating ratio, ease of fabrication and low cost. In early development, paraffin was used for the fabrication of moderator assemblies for neutron counters. Presently, High Density Polyethylene (HDPE) sheets are available and moderator assembly is fabricated using this material. Thermal neutron absorption cross section of hydrogen is significant (0.33 b) and hence a thickness of HDPE sheet of about 3 cm is optimum for moderation. A thickness of about 6cm can be used as reflector to enhance the detection efficiency. The design of neutron well counter provides the highest possible counting efficiency and the best configuration for biological shielding. The sample to be assayed is placed in the central well of the counter. An important feature of the well counter is also the reasonably good flat response over the volume of the well, which is important for heterogeneous samples. Typically 6 to 24 counters are arranged in a circular array around the central well [2].

A schematic of the associated electronics of the neutron counting setup is given in Fig. 1.

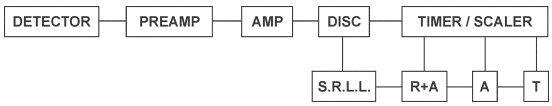

Fig. 1 Fig.1. A schematic of the neutron counting system

Total Neutron Counting

Gross neutron counting is adequate for isotopically well characterised plutonium in homogenous and well defined chemical matrix. For higher amounts of plutonium, multiplication effects come into play because of the fissions induced by source neutrons in fissile plutonium isotopes ($^{239,241}Pu$). The SF neutrons are mainly from $^{238,240,242}Pu$ isotopes and effective $^{240}Pu_{eff}$ mass is defined as

$$^{240}Pu_{eff} = 2.43\ M_8 + M_0 + 1.69\ M_2 \qquad (1)$$

where, M_8 , M_0 , M_2 are the weight percentages of the isotopes, ^{238}Pu , ^{240}Pu and ^{242}Pu respectively. The constants 2.43 and 1.69 account for specific spontaneous fission neutron production in ^{238}Pu and ^{242}Pu relative to ^{240}Pu.The (α,n) yields per g/s are calculated from the knowledge of isotopic and chemical composition of plutonium in the sample. The absolute detection of the efficiency of the counter is determined using the standard ^{252}Cf neutron source. The total neutron counting is carried out routinely for the assay of plutonium in low level nuclear waste packets for taking a decision on disposal or recovery. However in the case of heterogeneous samples, having unknown chemical composition, the (α,n) yields cannot be accurately estimated and coincidence counting of neutrons has to be used for the estimation of plutonium. In uranium compounds, (α,n) reaction due to ^{234}U decay dominates neutron production. Passive neutron total counting is routinely used for rapid verification of low enriched UF_6 cylinders.

Coincidence Counting of Neutrons

Fission events yield multiple neutrons that are correlated in time, whereas (α,n) reactions yield single neutrons that are uncorrelated or random in time. Coincidence counting of time correlated

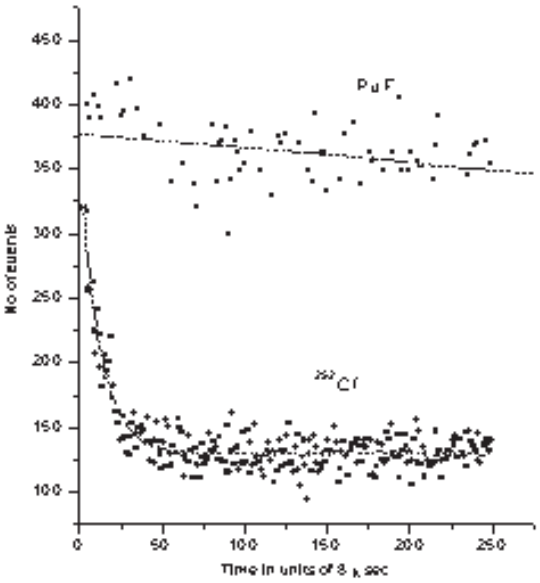

Fig. 2 Rossi alpha curve for PuF_4 and ^{252}Cf neutron sources

fission neutrons is a powerful technique for distinguishing the fission neutrons from (α,n) and background neutrons. The Rossi Alpha distribution [1] obtained from the distribution of arrival times of the neutron pulses from a detector is the basis for obtaining the random (A) and real coincidences (R). This is the time distribution of events that follow after an arbitrarily chosen starting event. If only random events are being detected, the distribution is constant with time. If real coincidences are also present, the distribution is given by

$$S(t) = A + R \exp(-t/\tau) \qquad (2)$$

where τ is the mean life time of the neutron in the counter also called as die away time of the system. A and R are the accidental and the real coincidence counts respectively. S(t) are the number of events at time t. A typical experimental Rossi alpha curve obtained [3] using ^{252} Cf and PuF_4 sources is given in Fig. 2. Die away time depends upon size, shape, composition of the counting system. Typical values are in the range of 30 – 100 μs for neutron Well Coincidence Counters. Therefore the conventional coincidence circuits require large dead time corrections. An alternative approach is the Shift Register Logic [2,4,5] which stores the incoming pulse train for a time G(gate width), so that every event can be compared with every other event for a time G. In effect every pulse generates its own gate; it is not necessary for one gate to finish before the next gate can start. This storage of events eliminates the dead time effect and allows operation at higher count rates.

The Rossi Alpha distribution is perturbed during the dead time of the counter and hence a pre delay of about 8 μs is introduced after the detection of each event. The sum of real and accidental (R+A), only accidental (A) and total (T) are obtained by the shift register based coincidence logic. The difference of (R+A) and (A) gives the real coincidences (R) and is proportional to the effective ^{240}Pu mass and is given by

$$R = m_{240}\, \varepsilon^2\, e^{-D/\tau} (1 - e^{-G/\tau}) \Sigma P(\nu)\nu(\nu-1)/2! \qquad (3)$$

ε = absolute detection efficiency
ν = SF neutron multiplicity
P(ν) = multiplicity distribution
D = Pre delay
G= coincidence gate length
τ = Die away time of NWCC

The statistical uncertainty in the measurement of R is given by

$$\frac{\sigma_R}{R} = \frac{\sqrt{R + 2A}}{R} \qquad (4)$$

Multiplication Correction

For samples containing larger than few tens of grams of plutonium, the SF and (α,n) neutrons may induce fissions in ^{239,241}Pu. The neutron induced fissions may also be caused by slow neutrons reentering the sample placed inside the neutron well counter. These neutrons also have higher neutron multiplicity than the neutrons from SF. Hence they contribute to the enhancement of coincidence response and introduce nonlinearity in the response for larger amounts. A non-multiplying sample is required for determination of the ratio of (α,n) neutrons to SF neutrons which is needed for multiplication correction [6,7,8,9]

TABLE 3. Characteristics of the HLNCC developed at Radiochemistry Division

1.	Well Diameter (ID)	360 mm
2.	Detectors	^{3}He, 25.4 mm dia., 500 mm sensitive length, 4 atm Fill Pressure
3.	No of detectors	24
4.	Absolute Detection Efficiency (T)	16.8 %
5.	Absolute Detection Efficiency (Coincidence) Pu	1.5 %
6.	Die Away Time Dead Time	105 μs 7μs
7.	Gate Width Pre Delay	128 μs / 64 μs 8 μs
8.	Ambient Background	3 cps
9.	[(R+A)-(A)]/A for Random Neutron source (PuF_4)	~1%

Development of Neutron well Coincidence Counters (NWCC)

A nondestructive assay method was developed for rapid characterization of plutonium samples (e.g. oxides, carbides in sealed containers and fuel pins). The method consists of determination of plutonium isotopic composition by gamma spectrometry followed by measurement of spontaneous fission neutrons by High Level Neutron Well Coincidence Counter (HLNCC). The plutonium amounts were determined using the isotopic composition. Plutonium amounts in seven different types of samples and three FBTR fuel pins estimated by this method compared well with the values determined by the chemical analysis. The method can assay samples containing ~2g.of PHWR grade plutonium with an accuracy better than ± 5%, in 3000s of assay time.

The characteristics of the HLNCC are given in Table 3. The linearity of the coincidence response of the HLNCC was established using PuO_2 standards (100-1200g). Assay of ^{233}U in (KAMINI) fuel plates relative to a standard fuel plate was carried out for the quality control. NWCC is being extensively used for the determination of plutonium in MOX blends to ascertain Pu enrichment at the powder mixing stage [10]. It is also proposed by A3F to qualify the sintered pellets by NWCC instead of chemical analysis. The proposal is intended to reduce liquid waste generation, process time as well as man power requirement. The method is also used for rapid identification of Pressurized Heavy Water Reactors (PHWR) and Research Reactor grade plutonium oxide lots [10].

A mobile neutron slab coincidence counter was also developed for in-situ assay of large amounts of plutonium in sealed containers.

Development of a Versatile Data Acquisition System

A PC based data acquisition system is recently developed using Pentium III (700 MHz and 128 MB RAM) and PC compatible PCL 830 ADD ON card [3]. The system is coupled to HLNCC. Neutron pulses from the counter are recorded sequentially along with their arrival time in an array and the data is stored in ASCII format. Software has been developed in visual basic for data acquisition and analysis. The new system is proposed to be a more versatile alternative to the conventional shift register based neutron coincidence counting.

Assay of FBTR Pins and Sub Assembly

A new type of NWCC has been developed in view of large Pu content and geometry for the nondestructive assay of plutonium in FBTR sub assemblies [11]. The reflector geometry is used to reduce the effect of (α,n) neutrons, achieve a flat response over the active region of the fuel sub

assembly. The total and coincidence neutron response as a function of effective ^{240}Pu mass is shown in Fig. 3. The data would be useful for the modification of the system to enhance the performance of the counter for the assay of the fuel sub assembly.

Delayed Neutron Method

The delayed neutron method is a powerful technique for the assay of fissile materials down to nano gram levels. The method is simple, sensitive, matrix independent and amenable to automation. It has wide ranging applications in nuclear fuel cycle and environmental studies. Feasibility experiments for the measurement of ^{235}U and ^{239}Pu in samples containing sub-microgram levels were performed using Pneumatic Carrier Facility (PCF) of CIRUS reactor [12]. A low back ground neutron well counter based on ^{3}He counters coupled to a 4K MCA in Multi Channel Scale (MCS) mode with a suitable dwell time was used to obtain delayed neutron activity as a function of cooling time. (Fig. 4). It was estimated that about 50 ng of ^{235}U and 150 ng of ^{239}Pu could be estimated with an accuracy of ± 10% at 1σ level. The commissioning of PCF at DHRUVA has provided an opportunity to carry out the delayed neutron measurements of fissile materials and the detection limits would be lowered because of higher thermal neutron flux.

Long Counter Development

The plutonium content in 200 L drums needs to be estimated to verify that the drum contains plutonium less than the limits prescribed for safe disposal. The size of the drum is large (900 mm height x 600 mm Dia.). As the distribution of plutonium inside the drum is random, the overall variation in the volume averaged neutron detection efficiency should be better than ± 10%. As the height of the drum is 900 mm, ^{3}He proportional counters of more than 1200mm of sensitive length are required for achieving a flat response of 900 mm. Single long detectors of such sensitive lengths are not readily available. Therefore a novel method of obtaining a counter configuration [13] having the required sensitive length was developed for routine use. The configuration introduces a null sensitivity region (absence of ^{3}He gas) of about 160 mm at the center which helps in flattening the axial response at the

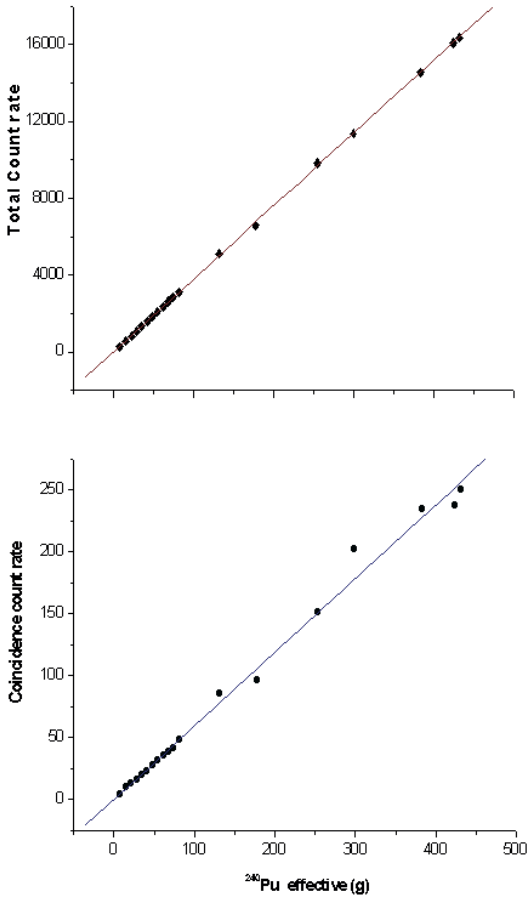

Fig. 3 Variation of coincidence and total count rates as a function of effective 240Pu mass for FBTR fuel pins

moderator assembly. The simulated long counter could be incorporated in the design of neutron well counters for plutonium monitoring of 200 L drums.

Assay Systems Based on Coincidence Counting of Neutrons

Some typical monitoring systems developed else where [1] for non destructive assay and safeguards are described as follows. Inventory Sample Coincidence Counter (ISCC): The portable system is used for the assay of plutonium in solutions, pellets and powders. By suitable moderator design, the system is made relatively insensitive to hydrogen content of the sample.

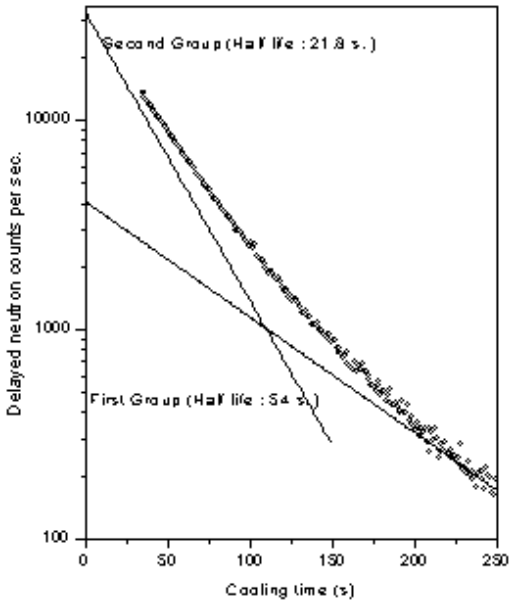

Fig. 4 Non linear fitting of observed delayed neutron activity to sum of two exponentials

Precision is in the range of (4 - 1 %) depending upon the plutonium content (0.5 g – 250 g). Solution Neutron Coincidence Counter (SNCC) developed for online assay of flowing solutions containing high fission product activity. Plutonium nitrate solutions ranging from (0.2-12g/l ^{240}Pu). Multiplication correction is applied for larger samples. The assay values agree within 1.6% relative to chemical methods. Dual Range Coincidence Counter (DRCC)] wide range capability (1-1000 g of Pu) for the assay of plutonium, is achieved by having two removable cadmium sleeves near the ^{3}He detectors. The sleeve is inserted for low efficiency and short die away time and removed for high efficiency with longer die away time.

Active Well Coincidence Counter (AWCC)

Two low intensity AmLi neutron sources of (`10^4 n/s) are placed at top and bottom of sample chamber for sub threshold interrogation of enriched uranium in the sample. Induced fission neutrons in ^{235}U or ^{233}U are monitored for quantitative assay of uranium. AWCC is less sensitive to gamma activity of the sample is applicable to ^{233}U-Th fuel cycle materials. Passive and active Neutron Coincidence Collar is designed for the assay of mixed oxide samples containing large amounts of plutonium. SF neutrons are measured in passive mode. In active mode, passive neutrons are reflected back into the sample to induce fissions. The change in real coincidences (R) normalised to totals (T) is related to fissile content. The method can be used for assay of FBR sub assemblies for total fissile content.

The Neutron Multiplicity Counter

Spontaneous fission rate related to the plutonium amount, Sample Multiplication and the (α,n) reaction rate in the sample are the three unknown parameters to be solved for non destructive assay of plutonium in a given sample without any a priori knowledge about the sample. These three unknowns can be solved exactly by three experimental parameters. The first, second and third moments of the neutron multiplicity distribution are called singles, doubles and triples respectively. Conventional coincidence counting measures singles and doubles. Neutron multiplicity counter [14] gives the third measured parameter "Triples" as well. The neutron detection efficiency for triple coincidences is proportional to cube of gross neutron detection efficiency. Also the detection efficiency should be independent of initial energy of neutrons. Therefore in neutron multiplicity counters, a large number of ^{3}He detectors are arranged in multiple rings to achieve high detection efficiency (50 - 60 %) and flat energy response. Three simultaneous equations setup by three measured parameters and thee unknowns mentioned above can be exactly solved to get the plutonium amount. If multiplication is absent, the gross neutron detection efficiency can be treated as the third unknown parameter. In case of low amounts of plutonium in unknown matrix in large size containers, neutron detection efficiency can be deduced in situ by the multiplicity counter.

Summary

A large number of non destructive assay techniques developed are based on neutron counting methods. The range of performance of neutron based non destructive assay methods is given in Fig.5. The accuracy suffers if there is a large (α,n) back ground and neutron moderating or absorbing materials are present in the sample. New techniques are being developed with more sophistication to reduce the error in measurement for meeting the assay

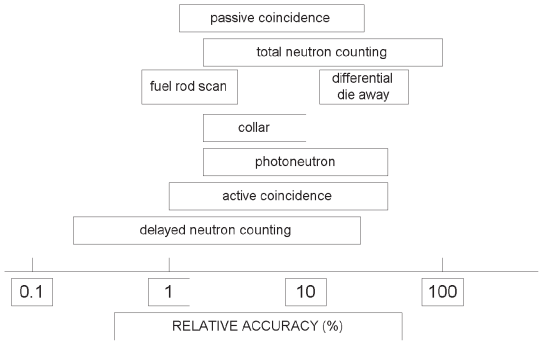

Fig. 5 The range of performance for neutron-based NDA techniques

requirement of special nuclear materials for nuclear safeguards, quality control and nuclear material accounting. The development of the non destructive assay techniques is always need based and expertise has to be developed to meet the specific needs of the users.

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Plutonium Isotopic Composition by Gamma-ray Spectrometry

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One of the most important aspects of the fissile material processing storage is its accounting at every stage. During the preparation of the fissile material in a specified form, e.g fuel for Fast Breeder Reactor containing Uranium and Plutonium as carbide or oxide, Light water reactor fuel containing enriched ^{235}U, mixed oxide (U,Pu) fuel as a replacement for light water reactor, etc, accounting of the fissile material has to be done at every stage. This may require assay of fissile material in various forms, e.g. finished form as fuel rods or pins, waste in the form of solution or solid and any other intermediate form. Nuclear material accounting, therefore, acquires special importance in any laboratory handling fissile materials. The most commonly used method for fissile material accounting is by electrochemical method. However, this method is distructive and time consuming. Often quick results are required on finished products or waste forms. Neutron coincidence counting has proved to be a very useful technique for assay of plutonium bearing samples. The method gives the effective ^{240}Pu mass of the plutonium sample. But determination of the total plutonium amount by this method requires prior knowledge of the isotopic composition of the plutonium sample.

A plutonium sample obtained from irradiated natural uranium as fuel consists of five plutonium isotopes namely $^{238,239,240,241,242}Pu$. Relative amounts of different plutonium isotopes in a plutonium sample depend upon the burn up of the fuel. Plutonium samples are categorised as “low-burn up (research reactor grade)” or “high-burn up (power reactor grade)” sample. High burn-up plutonium

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TABLE 1. Decay characteristics for isotopes useful in measurement of plutonium isotopic composition

Isotope	Half life	Energy (keV)	Intensity gammas/ disintegrations
^{238}Pu	87.74 y	152.8	1.01×10^{-5}
^{239}Pu	24119 y	129.3	6.20×10^{-5}
		161.5	1.30×10^{-6}
		195.7	1.07×10^{-6}
		203.5	5.06×10^{-6}
		345.0	5.61×10^{-6}
		375.01	1.58×10^{-5}
^{240}Pu	6564 y	160.35	4.20×10^{-6}
^{241}Pu	14.348 y	148.6	1.90×10^{-6}
^{241}Am	433.6 y	125.29	3.95×10^{-3}
^{237}U	6.75 days	164.6	4.50×10^{-7}
		208.0	5.12×10^{-6}
		267.5	1.77×10^{-7}
		332.3	2.80×10^{-7}

samples contain 70-80% ^{239}Pu, whereas low burn up plutonium samples contain >90% ^{239}Pu.

At present, the most accurate results on isotopic composition of a plutonium sample are provided by thermal ionisation mass spectrometry (TIMS) in combination with alpha spectrometry. However, this method requires chemical treatment of sample prior to analysis and therefore cannot be used for samples to be assayed non-destructively e.g. fuel pellets or fuel pins. Such analysis is very important for quality control at different stages of fuel cycle. Thus, gamma-ray spectrometry plays a very important role in the area of non-destructive assay [1].

In gamma-ray spectrometric method, plutonium isotopic composition is determined by making use of the characteristic gamma-rays of plutonium isotopes. This method is very fast, reliable and can be carried out on finished forms, irrespective of their physical and chemical form. Plutonium isotopes emit gamma rays having energies from 129 to 670 keV. The useful gamma-ray energies of plutonium isotopes are given in Table 1 [2]. The intensities of these gamma-rays are very low and also there are a number of groups of gamma-rays having near-by energies, which leads to spectral interferences. Presence of ^{241}Am, which is a decay product of ^{241}Pu, makes the situation even worse. Fig. 1 shows the decay scheme of ^{241}Pu, which decays to ^{237}U with a small branching fraction of 2.46×10^{-5}. After approximately 47 days (7 half lives) of the purification of plutonium sample, a secular equilibrium is established between ^{241}Pu and ^{237}U and therefore gamma-rays of ^{237}U can be used for the assay of ^{241}Pu. It is evident from Fig. 1 that both the daughter products of ^{241}Pu, namely, ^{237}U and ^{241}Am decay to ^{237}Np, and they may populate the same excited states of ^{237}Np. Thus, most of the gamma-rays of ^{237}U, which can be used for assay of

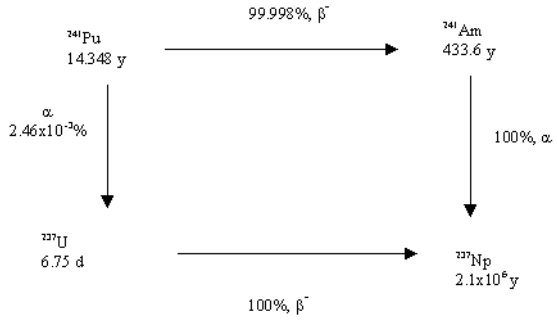

Fig. 1 Decay scheme of ^{241}Pu

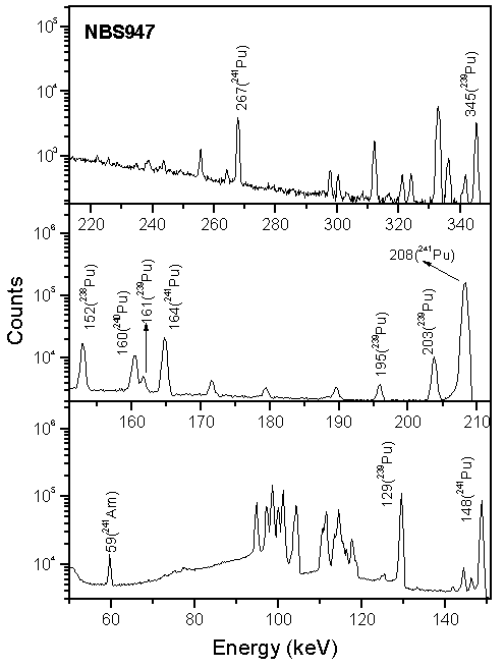

Fig. 2 Typical gamma-ray spectrum of a power reactor grade plutonium sample

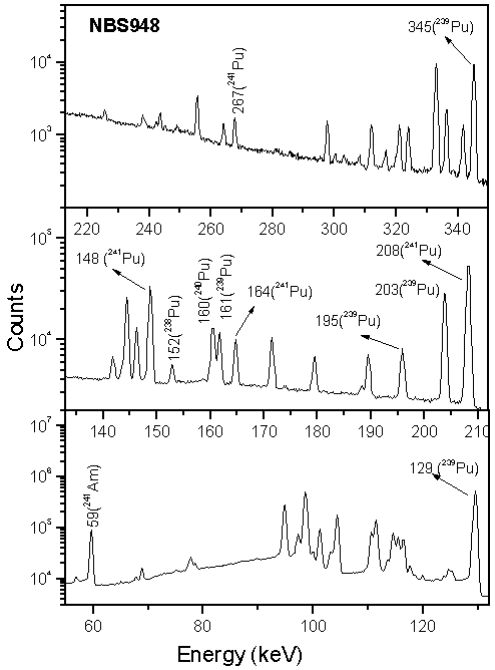

Fig. 3 Typical spectrum of a research reactor grade plutonium sample

^{241}Pu, have interference from ^{241}Am. The gamma-rays of ^{241}Am are also given in Table 1. The problem of spectral interference due to ^{241}Am is more severe in the case of aged high burn-up plutonium samples. Therefore, the measured area under the photo peak should be corrected for spectral interference.

Methodology for Determining Isotopic Composition

The photo peak area of a gamma-ray of isotope 'i' can be written as

$$P_A\left(E_j^i\right) = N^i \lambda^i B_j^i \varepsilon\left(E_j\right) \qquad (1)$$

Where,

$P_A\left(E_j^i\right)$ = photo peak area of gamma-ray of energy E_j emitted from isotope 'i'.

λ^i = decay constant of isotope 'i'.

N^i = number of atoms of isotope 'i'.

B_j^i = branching ratio of gamma ray 'j' from isotope 'i'.

$\varepsilon\left(E_j\right)$ = Efficiency for photo peak detection of gamma ray of energy E_j

Equation (1) can be extended for the atom ratio of the two isotopes 'i' and 'k',

$$\frac{N_i}{N_k} = \left[\frac{P_A\left(E_j^i\right)\varepsilon\left(E_m\right)}{P_A\left(E_m^k\right)\varepsilon\left(E_j\right)}\right] \times \frac{\lambda^k B_m^k}{\lambda^i B_j^i} \qquad (2)$$

In the equation (2), the quantities outside the parentheses are nuclear data, which are given in Table 1. Thus, the determination of atom ratio of two isotopes requires the measurement of two quantities, namely, the photo peak area of the gamma ray and its detection efficiency.

Determination of Photo Peak Area

Typical gamma-ray spectra of high burn up and low burn up plutonium samples are shown in Figs. 2 and 3 respectively. Usually, for determining isotopic composition, the gamma-rays in the region of 120 to 345 keV are used. It is evident from the spectra that, the plutonium isotopes have a large number of closely spaced gamma-lines in this region. Some of these gamma lines are mentioned below.

1. 123.62 keV (^{239}Pu), 124.51keV (^{239}Pu), 125.21keV (^{239}Pu) and 125.29 keV (^{241}Am)
2. 148.57 keV (^{241}Pu), 150.11keV(^{241}Am) and 152.68 keV (^{238}Pu)
3. 159.96 keV (^{241}Pu), 160.19keV (^{239}Pu), 160.28keV (^{240}Pu) and 161.45keV (^{239}Pu)
4. 203.54 keV (^{239}Pu) and 203.87keV (^{241}Am)

Due to low energy gamma ray complexity of plutonium, a HPGe detector (Resolution of 600 eV at 122 keV) with a beryllium window (minimising window attenuation) is an ideal choice for acquisiton of gamma ray spectra of plutonium samples. In order to deconvolute the unresolved multiplets peak fitting softwares are used.

In addition to spectral interferences, there are various summing events, which must be taken care while acquisition of gamma-ray spectrum. There are three major summing events, which are mentioned as under,

1. 59.54 keV (^{241}Am) + 101.1 keV(^{241}Am and ^{237}U) $\rightarrow$ 160.64 keV
2. 59.54 keV (^{241}Am) + 148.57 keV(^{241}Pu) $\rightarrow$ 208.11 keV
3. 129.29 keV (^{239}Pu) + 203.54 keV(^{239}Pu) $\rightarrow$ 332.83 keV

Due to high count rate at 59.54 keV of ^{241}Am in aged samples of plutonium, the first two summing events are very important and can become a major source of error in the analysis. Cadmium foil of thickness in the range of 1-2 mm is placed between sample and the detector to reduce the summing events due to 59.54 keV.

Determination of Photo Peak Detection Efficiency

The detection efficiency of the gamma-ray depends upon several factors such as distance between the sample and detector, sample shape, size, physical form and its composition. Though, the geometrical factors can be ignored in calculating the atom ratios, as they will be independent of gamma ray energy, and therefore will be cancelled in equation (2). However, the effect of physical form and composition of the sample depends upon the gamma ray energy e.g. self attenuation of gamma-rays will be large for lower energy gamma rays compared to higher energy gamma rays. Therefore, the detection efficiency should be corrected for matrix attenuation. This is done by carrying out in-situ efficiency calibration for a particular sample. In this method, the efficiency at a given energy is arbitrarily assumed and efficiencies at other energies are determined relative to the former. The relative efficiency curve takes care of matrix attenuation. Fig. 4 shows relative efficiency curves for a FBTR fuel pin, a FBTR fuel pellet and, a liquid plutonium sample. It is evident from the figure that the correction for matrix attenuation is very important. It should be mentioned here that the gamma ray energies of the two isotopes, which are to be used for determining the isotopic ratio should be close so that the error arising from efficiency fitting will be minimum.

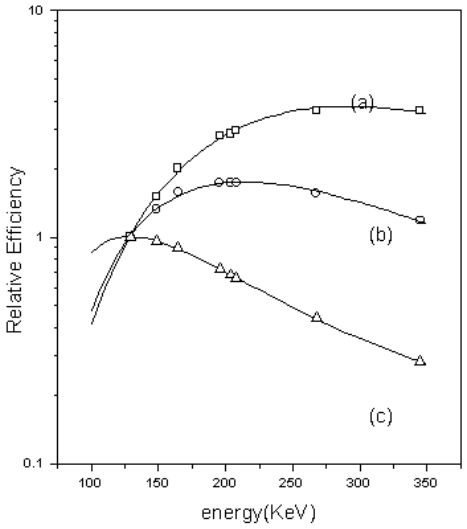

Fig. 4 Relative efficiency curve for plutonium samples in different forms (a) FBTR fuel pin (b) FBTR fuel pellet (c) plutonium in solution

Calculation of Isotopic Ratios

If the isotopic ratios of all the isotopes are calculated with respect to an isotope 'i' then isotopic fraction 'f_i' of isotope 'i' is given by the following relation,

$$f_i = \frac{1}{\sum_{k=1}^{m} \frac{N_k}{N_i}} \quad (3)$$

where, 'N_i / N_k' is the atoms ratio of the isotopes 'i' to isotope 'k' and m is the total number of isotopes. Once the isotopic fraction of isotope 'i' is determined, isotopic fraction of any isotope 'j' can be calculated as,

$$f_j = f_i \left[\frac{N_j}{N_i} \right] \quad (4)$$

The isotopic fractions thus calculated don't include ^{242}Pu. As mentioned earlier isotopic fraction of ^{242}Pu is directly determined using established isotopic correlations. These correlations depend upon the type of the reactor in which plutonium is produced. One such correlation, by Aggarwal et al. [3] is given by,

$$\%f_{242} = M \frac{(\%f_{240})^3}{(\%f_{239})^2} + C \quad (5)$$

This correlation is independent of ^{241}Pu fraction, but the slope M and intercept C depend on the reactor type. Another correlation, as suggested by Gunnink et al. [4] is given below,

$$f_{242} = K \left(\frac{(f_{240})(f_{241})}{(f_{239})^2} \right) \quad (6)$$

This correlation is nearly independent of reactor type. When the isotopic fractions are given in weight percent the constant of proportionality 'K' is 52. One disadvantage of the correlation (6) is that it depends on the isotopic fraction of ^{241}Pu, which decreases by about 5% every year and hence requires the knowledge of the date of fuel discharge. When the discharge time is not known, a partial correction is applied by adding quantities of ^{241}Am to the ^{241}Pu before computing the correlation.

After determining the isotopic fraction of ^{242}Pu, the corrected isotopic fractions are determined using the following equation

$$cf_i = f_i (1-f_{242}) \quad (7)$$

where cf_i is the corrected isotopic fraction of isotope 'i'. The isotopic fraction of ^{241}Am can be calculated using the equation (8), as shown below,

$$f_{Am} = f_i \left[\frac{N_{Am}}{N_i} \right] \quad (8)$$

It should be noted here that this fraction of ^{241}Am is the atom fraction in the sample with respect to the total plutonium and not with respect to the total sample. While calculating the isotopic composition, an assumption is made that the isotopic distribution is constant throughout the volume of the sample.

Measurement Precision

The precision of measurement of isotopic fractions for various isotopes of plutonium by gamma-ray spectrometry depends on the factors such as sample age, amount and type. It also depends on spectral interferences on gamma rays chosen for the determination of isotopic composition. For low burn up plutonium samples, precision of 1.5-10% for ^{238}Pu, 0.2-0.5% for ^{239}Pu, 2-6% for ^{240}Pu and 2-5% for ^{241}Pu is usually achieved. In the case of high burn up plutonium samples, precision of 1-6% for ^{238}Pu, 0.2-1% for ^{239}Pu, 0.5-3% for ^{240}Pu and 1-3% for ^{241}Pu can be achieved. The accuracy is not satisfactory for estimation of plutonium mass in tandem with neutron coincidence counting quality control purpose. However the method is a very useful nondestructive technique for rapid characterisation of plutonium.

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Gamma Scanning Techniques in Plutonium Based Nuclear Fuels Fabrication

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Introduction

The fabrication of plutonium bearing nuclear fuel elements entails nondestructive inspection of the components and intermediate products to ensure their conformance to the stringent specifications required for their optimum performance during operation in Nuclear reactors. One of the important specifications on the fuel elements is the uniform distribution of the fissile material along the length of the active fuel stack, its length and enrichment. The internal arrangement of components in the fuel element also requires to be verified. Gamma scanning techniques are used to ensure that the fuel element meets these specifications.

Plutonium based fuels are being used in our thermal and fast nuclear reactors. Plutonium-Uranium mixed Carbide fuel for the Fast Breeder Test Reactor (FBTR) at Kalpakkam, Uranium-Plutonium (low enriched) Mixed Oxide (MOX) fuel for the Boiling Water Reactors (BWR) at Tarapur and Uranium-Plutonium MOX fuel containing ^{233}U for the experimental Prototype Fast Breeder Reactor (PFBR) sub-assembly are fabricated at BARC.

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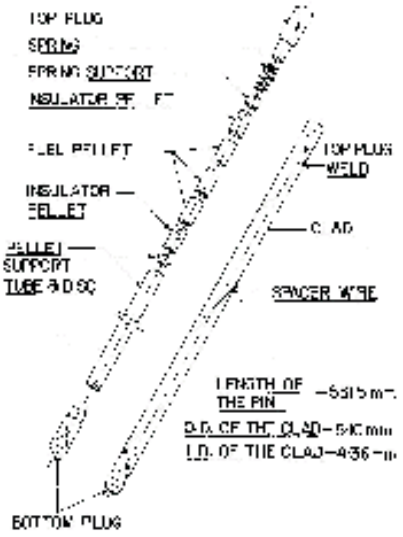

Fig. 1 FBTR fuel pin showing the components

A nuclear fuel element consists of appropriate hardware placed on either side of the active nuclear fuel pellet stack, to hold it in place, encapsulated in a clad–tube by welding end-plugs to both ends of the tube. After encapsulation it is important to confirm the enrichment of the fuel pellets, the uniformity of enrichment along the length of the fuel stack and length of the active fuel stack, nondestructively. The order of all the internal components, from the bottom plug-end to the top-plug end, inside the fuel tube also requires to be confirmed. The nondestructive inspection of these characteristics of the nuclear fuel element is carried out using Gamma Scanning. The term scanning implies the uniform motion of a fuel pin in front of a collimated and shielded detector, so that the spatial distribution of gamma activity along the length of the fuel pin can be studied / recorded.

Gamma scanning is a nondestructive technique in which two types of scanning are possible. One, termed Passive Gamma scanning (PGS), in which one evaluates the signature gamma rays emitted by the fuel and the other, termed Active Gamma scanning (AGS), in which a gamma-emitting isotope is used as a source of gamma radiation and the attenuation of these rays by the fuel is used for evaluation.

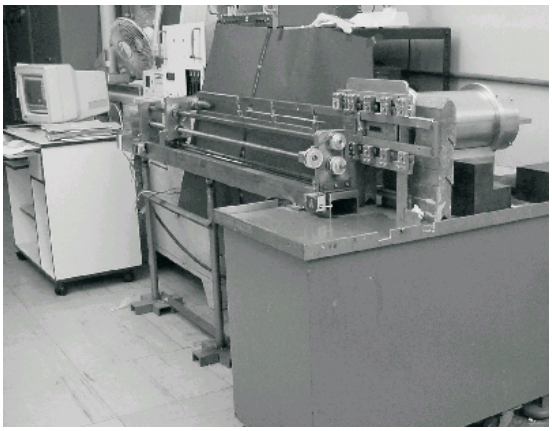

Fig. 2 Passive gamma scanner for active fuel stack

Gamma Scanning of FBTR Fuel Elements

The FBTR fuel pin is a stainless steel tube, sealed at both ends by welded stainless-steel (SS) end-plugs. Inside the tube, from the bottom end-plug, are the SS plenum tube and plenum tube support disc, a uranium carbide insulation pellet, the plutonium-uranium carbide fuel stack, another insulation pellet and finally a SS spring support disc and spring to hold all components in place and the top plug. Figure 1 shows an exploded view of the FBTR fuel pin.

A combination of PGS and AGS is required to monitor the active fuel stack and the internal arrangement of the SS components. The passive gamma scanner developed for the monitoring of the active fuel stack is shown in the Fig. 2 [1]. A differential measurement of plutonium gamma ray activity (80-500 keV) using a 3”x3” NaI(Tl) detector is performed by moving the fuel pin (mounted on the scanner bench) with a uniform speed in front of the detector with a suitable collimator and recording the activity in a multi-channel analyzer in multi-scale mode. A typical activity profile is shown in the Fig. 3. The uniformity of enrichment in the fuel pellets is observed by the plateau of the activity profile while the active stack length (L) is estimated by

L (mm) = FWHM(channels)* dwell-time(s)* speed(mm/sec)

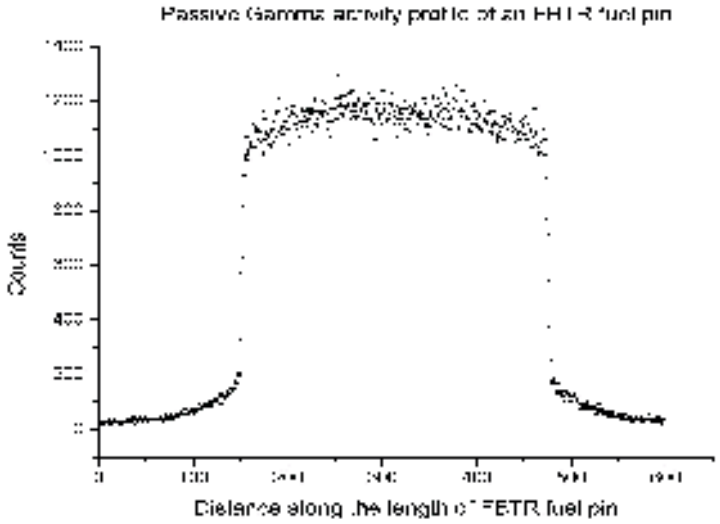

Fig. 3 A typical activity profile

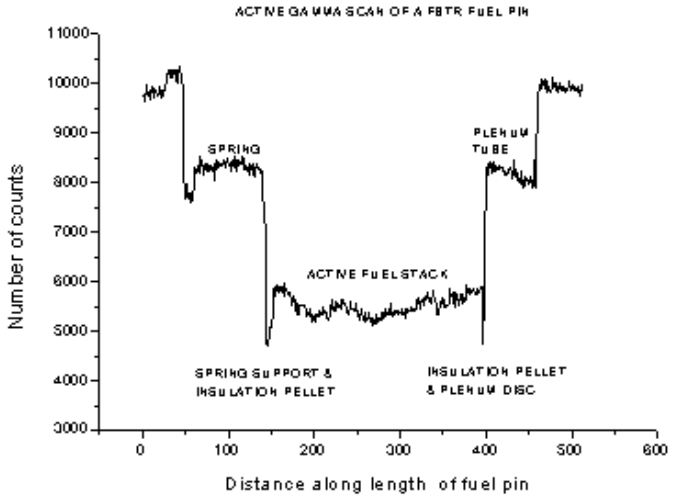

Fig. 4 A typical transmission profile.

The accuracy obtainable in the measurement of the active fuel stack depends upon the precision of speed of movement, width of the collimator and the dwell time. The specified accuracy of estimating the active length to better than 0.5% has been achieved using the gamma scanner.

As only the active fuel stack is examined by PGS, AGS is used to examine the internal arrangement of the hardware components in the FBTR fuel pin [2].

Figure 4 is a typical transmission profile obtained for a fuel pin. The attenuation being distinctively different for the stainless steel hardware, these are clearly distinguished from the fuel pellets. The presence of the insulation pellets is distinguished by the dip in the activity profile.

Gamma Scanning of MOX Fuels

Passive Gamma scanning is carried out on BWR MOX fuel pins and Prototype Fast Breeder Reactor (PFBR) experimental fuel pins fabricated at

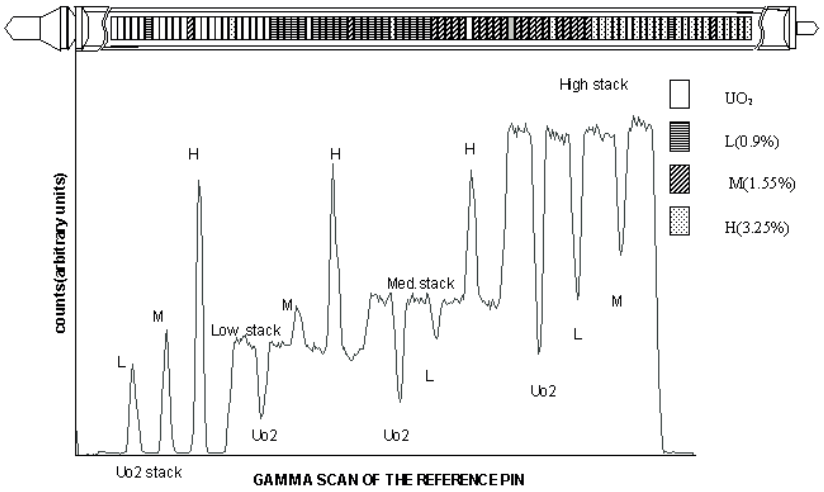

Fig. 5 Gamma scan of experimental pin

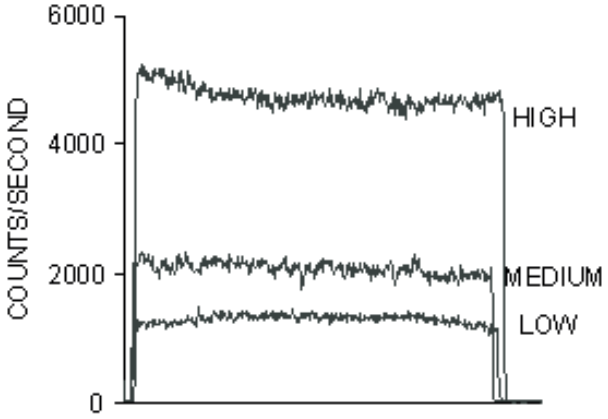

Fig. 6 Gamma scans of MOX fuel pins for BWR

AFFF [3]. The fuel pin scanner (4m long) at AFFF is based on a 3"x3" NaI(Tl) detector and a multi-channel analyzer operated in the multi-channel scaling mode, with an integrated software to control the automatic pin movement and handling of the data acquisition [4,5]. The system has been improvised by introducing an annular NaI(Tl) detector.

BWR MOX Fuel Pins

The configuration of BWR MOX bundle is based on three different enrichments of PuO_2 viz; 0.9%, 1.55% and 3.25%, which are commonly referred as low(L), medium(M) and high(H) enrichments. An experimental pin with pellets of all possible combinations of the three enrichments is used for optimization of the system parameters. The schematic drawing of the experimental pin and its Gamma scan are shown in Fig. 5.

The main objectives are to confirm that a pin has the right enrichment level as indicated by its coded number and that there is no pellet of a different enrichment, also known as a rogue-pellet or anomalous-pellet, within the stack. PGS has been effective in not only monitoring these aspects but also in the study of details like variation of composition, presence of agglomerates etc. Typical scans of BWR MOX pins of low, medium and high enrichments are shown in Fig. 6. Figure 7 shows the

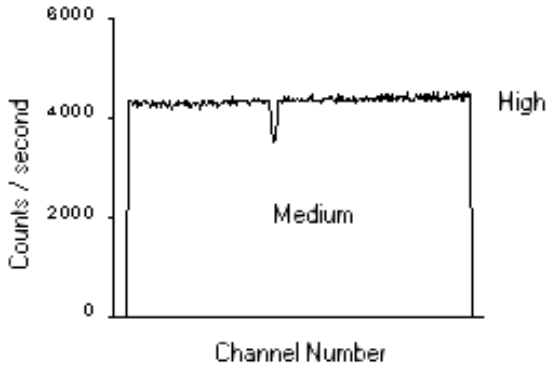

Fig. 7 A medium enrichment pellet in a high enrichment stack

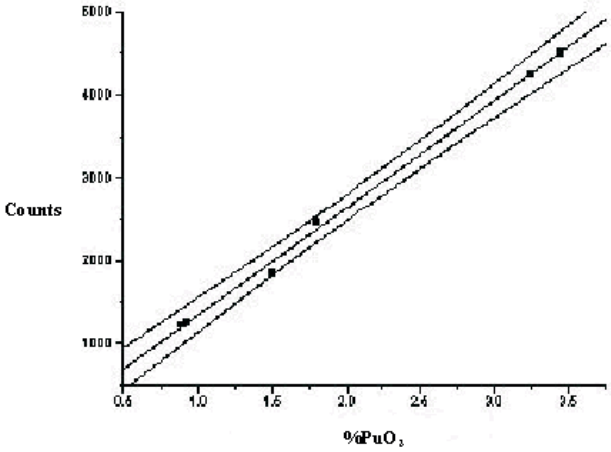

Fig. 8 Calibration graph of PuO_2% enrichment

scan of a pin with high enrichment pellets in which a pellet of medium enrichment (single pellet) had been accidentally placed in the stack during fabrication [6].

Figure 8 shows the calibration graph of PuO_2% enrichment against Gamma counts determined using experimental MOX pellets with marginally varying enrichment of PuO_2 and characterized through chemical analysis. A detection sensitivity of 0.2% variation in enrichment of PuO_2 .in thermal reactor fuels using a conventional detector has been obtained. It is possible to improve the sensitivity of detection of variation in enrichment using a better counting geometry and specifically designed detectors and collimators. The use of an annular NaI(Tl) detector has improved the sensitivity of the system and it has been possible to detect the presence of PuO_2 clusters (about 1mm size) in the fuel pellets [7].

PFBR MOX Experimental Fuel Pins

The PFBR experimental subassembly consists of 37 elements. The stack consists of 30-35 MOX pellets embedded between two natural UO_2 pellets (one each on either end), which serve as insulation pellets. MOX pellets containing 29% PuO_2 and 71% Uranium dioxide, enriched in ^{233}U by 53.5% are encapsulated in a cladding tube made of D9 which are later welded with end plugs.

All the PFBR experimental pins were scanned using 3"x3" NaI(Tl) detector and the pins were found to be free from any anomalies.

Conclusion

Gamma scanning techniques provide a very simple and useful means of inspecting the characteristics of nuclear fuel stack in a fuel element, nondestructively. It has been very effectively used to monitor the plutonium based FBTR fuel elements and plutonium based MOX fuel elements.

Acknowledgements

The authors would like to thank all their colleagues in Radiometallurgy Division and Advanced Fuel Fabrication Facility for their co-operation and help during this work. They would also like to acknowledge and thank Dr. S.B. Manohar and Dr. G.K. Gubbi for their initiative and invaluable contribution in setting up of the scanning systems at both the fabrication facilities. The contribution of Shri A.V. Kathavate and his colleagues from the Reactor Control Division, BARC is also gratefully acknowledged.

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Gamma Based Non-Destructive Assay Techniques

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The emphasis on nuclear power is ever increasing to meet the growing need for energy, leading to rapid growth in handling large quantities of nuclear materials necessitating stringent measures in material management. Non Destructive Assay techniques (NDA) have a very significant role to play in the assay and accounting of the nuclear materials. Excellent review articles are available giving the growth of NDA techniques and their present status [1-3]. Non-Destructive Assay techniques fall in to two major categories. The first, Passive method, involving the detection of spontaneously emitted radiation either neutron or gamma rays by the natural radioactive decay processes. The active method is based on the detection of induced radiation produced by irradiating the sample with an external radiation source. The selection of assay technique for a particular application depends on physical characteristics of the material, chemical composition, isotopic composition and the required accuracy measurement.

Passive Gamma-Ray Measurements

All the isotopes of uranium and plutonium are radioactive and decay by emission of alpha, beta or by spontaneous fission process. The characteristic gamma rays are used for the identification and estimation of the nuclear material. Passive gamma ray techniques can be further subdivided into Low Resolution Gamma Ray spectrometry (LRGS) and High Resolution Gamma Ray Spectrometry (HRGS). The choice of the technique depends on the nature of the application.

NDA Techniques Based on Low Resolution Gamma Ray Spectrometry

Measurement of the absolute intensity of characteristic gamma rays from nuclear material samples can be used to determine the absolute

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amount of isotopes in the sample. The matrix attenuation correction is generally accomplished by measurement of transmission ratios using an external gamma-ray source. A gamma-ray assay of the sample, therefore involves measurement of passive radiation and the estimation of matrix attenuation. The attenuation-corrected gamma-ray assay technique gives accurate assay results on a variety of samples. Now we would see the different NDA techniques developed and in use in BARC, India and other countries.

Plutonium Waste Packet Scanner

A plutonium waste packet scanner system using a 3" x3" NaI(Tl) detector coupled to a single channel analyzer has been developed in Radiochemistry Division for routine use [4]. The 375 keV gamma ray complex of ^{239}Pu was used for its estimation. The rotation collimation technique was used to achieve the flat response with respect to the random distribution of Pu inside the container. The calibration curve was developed for 3 litre container using waste packets containing known amounts of Pu. The attenuation is assumed to be constant for similar packets of soft glove box waste. The detection limit of the scanner was 20 mgs for soft glove box Pu waste in 3 litre container. This scanner is being routinely used for assay Pu in the glove box waste packets. The method is not applicable if the gamma emitters other than plutonium are present in the sample.

NDA Techniques based on High resolution Gamma Ray Spectrometry

The NDA techniques based on high resolution gamma ray spectrometry employ the measurement of the gamma ray using HPGe detector coupled to Multi channel Analyser. The higher energy resolution and better peak to compton ratio enable assay of mixtures of the radio nuclides. Both neutron counting and calorimetric assay techniques need the knowledge of isotopic composition of plutonium in the sample, in order to determine the mass of plutonium. High resolution gamma ray spectrometry is being routinely used for the measurement of isotopic composition of Plutonium in various types of samples. The details are described elsewhere in the bulletin. Some important applications of HRGS based techniques are described briefly.

Hull waste Monitoring System

During the dissolution of spent fuel by chop-leach process in reprocessing plants, small but significant amount of residual fuel remains in the hull because of the incomplete leaching of the crimped hulls. Hence, it is necessary to estimate this material with required accuracy for proper accounting of the nuclear materials. There are various methods to detect and measure the residual fuel in the hull. Active neutron interrogation technique using delayed neutron counting would be the most direct [5]. However, this method would be quite complicated and relatively expensive. Detection of the residual fuel by passive method is based on measurement of spontaneous fission and (α,n) neutrons from transuranic elements or characteristic gamma rays of the fission products in the hull basket. The ideal fission product should have a long half life, high fission yield, convenient high energy gamma ray and should have dissolution properties similar to uranium. The yield of the fission product should be similar for neutron induced fission of ^{235}U and ^{239}Pu. ^{144}Ce, with high cumulative fission yield (~ 6%), has a convenient half life (284 d) and its daughter product ^{144}Pr, which is in secular equilibrium, emits a high energy gamma ray (2.18 MeV) which makes it the most suitable fission product for direct gamma ray measurement. In the case of long cooled fuel, the ^{144}Ce in the hull decays and ^{137}Cs is used as a monitor. The ratio of ^{144}Ce/U or ^{137}Cs/U for the dissolver solution is used to calculate the amount of uranium and plutonium remaining in the leached hull.

A schematic arrangement of the Hull Monitor set up at Kalpakkam Reprocessing Plant [6] is shown in Fig.1. A simulated hull basket was designed and fabricated for determination of absolute detection efficiency for gamma rays of interest using ^{124}Sb as a multi energy gamma source. It was estimated that about 30 Curies of ^{144}Ce-^{144}Pr could be detected in the actual hull basket with an accuracy of ± 10%. As the fuel was long cooled during the dissolution, the ^{144}Ce-^{144}Pr activity could not be detected and only alternative was to use the activity of ^{137}Cs (661.7 keV). A typical gamma ray spectrum of hull basket containing residual fuel is shown in Fig. 2. The axial profile of the activity in the actual hull basket was used to estimate the residual fuel in the hull basket.

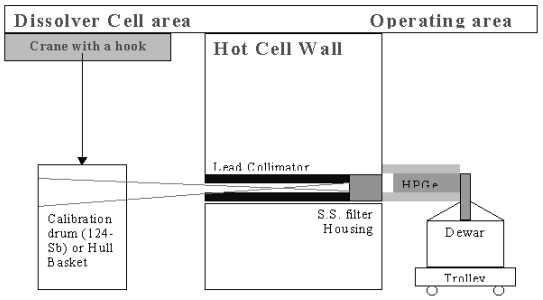

Fig. 1 Schematic diagram of HULL monitoring set up at Reprocessing Plant

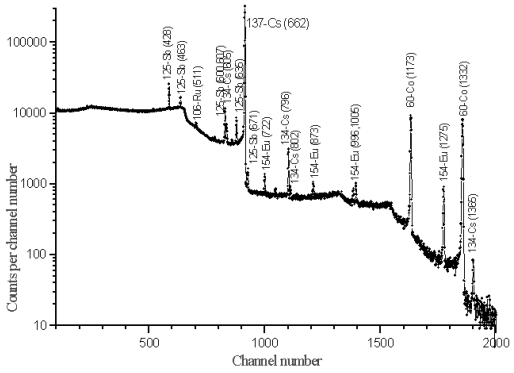

Fig. 2 Plot of gamma ray spectrum of a typical section of the hull basket as seen by the HPGe detector

The performance of the hull monitoring system has been found to be satisfactory.

K-edge Densitometry

Another application of high resolution gamma ray spectrometry is the assay of Plutonium in solution by K-edge densitometry [7]. This technique is used to determine the Pu concentration in solution samples by measuring the transmission at γ ray energies just below and above the K- absorption edge for Pu. Measurements are made with high resolution HPGe detector, external radiation source and a suitable collimator. For K-edge assay of Pu concentration, the sample transmission is measured using radioisotopic sources ^{75}Se (121.1 keV) and ^{57}Co (122.1 keV) which closely bracket the K-absorption edge (121.76 keV) of plutonium. For a sample of path length x, matrix density (ρ_{m}), Pu concentration in the sample (ρ_{Pu}) is given by

$$\rho_{Pu} = \ln (T_L/T_U)/ x \Delta\mu + \rho_m \Delta\mu_m / \Delta\mu \qquad (1)$$

where $\Delta\mu$ and $\Delta\mu_m$ are change in the mass attenuation coefficients for plutonium and matrix respectively at energies just above (E_U) and just below (E_L) the K edge of plutonium. T_L and T_U refer to measured transmission ratios at the corresponding energies. The close proximity of E_U and E_L makes the matrix contribution to the Pu assay negligible. Pu concentration with a precision of 0.5% for 30 min assay time could be obtained in the Pu concentration range of 10 mg/l to 100 mg/l. K-edge densitometers are in routine use for measuring the product solution at reprocessing facilities in Japan (PNC) and USA (Savanna River plant). A prototype K-edge densitometer system was set up at RCD, BARC for the estimation of Pu in solutions [8].

Advanced NDA Techniques

In recent years, more emphasis is being placed on safeguards issues associated with the alpha radioactive waste. The allowed limit for disposal of alpha active waste by International Atomic Energy Agency (IAEA) is 4000 Bq/g. The alpha bearing waste packets are stored in 200 litre drums. Assay of waste can be carried out using neutron counting or passive gamma ray scanning. The detection limits for Plutonium in waste drum are lower for gross neutron counting. Active neutron interrogation technique can be used for low level samples. The segmented gamma ray scanner [9] has been developed for assay of plutonium for specific applications.

Segmented Gamma Ray Scanning System using HPGe Detector

Segmented gamma scanner (SGS) is an axial drum scanner that uses HPGe detectors and appropriate collimators to measure the emission of gamma rays from different horizontal slices (or segments) of the sample within which the activity and the matrix is assumed to be uniform and homogeneous.

The characteristic gamma rays of plutonium are used for the estimation. The attenuation correction is carried out using suitable gamma ray

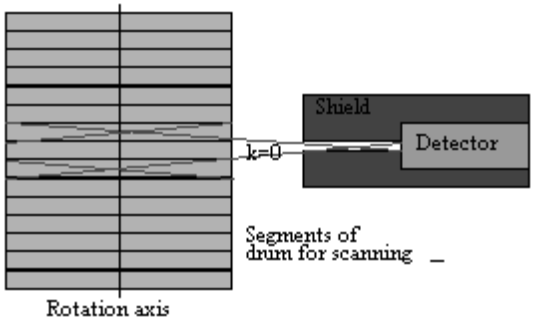

Fig. 3 Sample detector geometry in segmented gamma scanning

sources. A schematic of segmented gamma ray scanning system is given in Fig. 3. Transmission of the gamma rays of ^{169}Yb, ^{75}Se and ^{152}Eu are measured to correct for the self attenuation in the samples.

Tomographic Gamma Scanner

The Tomographic Gamma Scanner (TGS) is a substantial improvement over the SGS. The tomographic gamma scanner corrects for each volume element of the sample. A 3D map of attenuation and source distribution is generated. The award winning Waste Inspection Tomography for

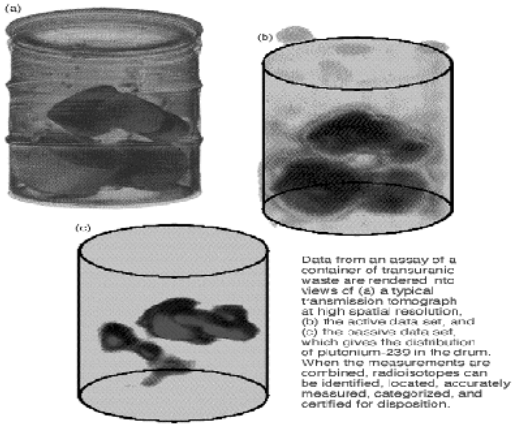

Fig. 4 3D tomographic imaging of the waste drum

Non Destructive Assay (WIT-NDA) was developed at Lawrence Livermore Laboratory [10].

This map can be used for accurate assay of radioisotopes within the drum. Figure 4. shows a typical 3D tomographic imaging of a typical 200 litre waste drum. This system meets the stringent requirements of the waste immobilisation plants in United States [11].

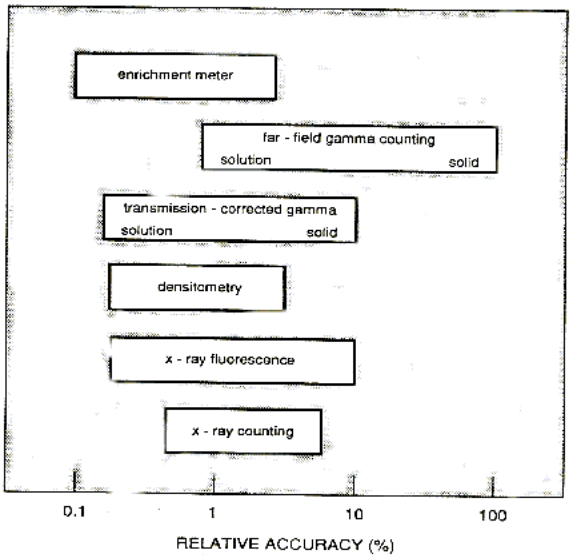

Fig. 5. The range of perfrmance for gamma-ray NDA techiques

Accuracy of Gamma based NDA Techniques

Figure 5 illustrates the range of performance for the most common gamma based NDA techniques [2]. The relative errors range from 1% to 100% depending on the type of sample and assay technique.

Conclusion

Effort has been made to give a glimpse of various gamma measurement based NDA techniques. Gamma Based NDA techniques provide a clear identification of the nuclear species in the sample even in cases where the sample is too dense or too heterogeneous to permit accurate assay of the amount of the nuclear material. For heterogeneous or dense material where gamma ray attenuation is very large to permit accurate attenuation corrections, Neutron based assay techniques may be preferable. Large samples of metals, Oxides and high density scrap and waste require neutron based NDA techniques. In general gamma based NDA techniques, where ever applicable, are more accurate than the neutron based techniques.

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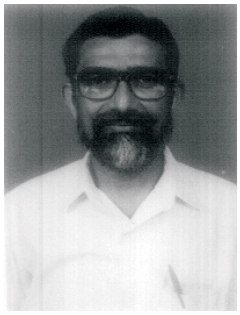
Gamma ray absorptiometry Technique in Nuclear Fuel Fabrication



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Introduction

The process of characterization of nuclear fuels with respect to different properties is called an `assay'. Passive and active assays are both utilized. As described elsewhere in this issue, in passive assay, the radiation emanating from the nuclear material is used. The active assay requires an external radiation source and the transmission properties of the material guide us to the exact nature and/or quantity of the material. Usually uranium based fuels are assayed by active technique and plutonium based systems, by virtue of higher specific activity, can be subjected to passive method.

The term 'active assay' is many times used to mean the technique of interrogation by neutrons where the fission product gamma rays or prompt and delayed neutrons are monitored to measure the quantity of fissile materials. This article only describes the transmission type gamma ray scanning, popularly called radiometry or sometimes 'densitometry'. As the source used emits few well separated gamma rays, scintillation detectors like NaI(Tl) detectors coupled to a single channel analyser can be used. However, the design of equipment lay out is more complex in active assay technique, as may become clear shortly.

Equipment and Instrumentation

The set up for active gamma assay consists of mechanical scanner and computer based gamma spectrometer.

Mechanical Scanner

Mechanical equipment can be of two types. Either the source detector couple or the fuel sample can be moved for the purpose of scanning. The former is required in certain cases of safeguards inspection if the specimen is a vessel or a container, not amenable to movement. In the case of nuclear fuel elements it is convenient to move the element. For cylindrical fuel elements, rotational movement may be necessary in addition to translational (linear) movement. As the collimator aperture is smaller than the diameter of the fuel element, peripheral region of the pin will lie outside the radiation beam. Rotation is necessary to bring this into the field of testing.

The mechanical scanner should satisfy the following criteria for precision in results:

(i) Uniform speed without slippage with minimum vibrations

(ii) Variable rotational and linear speeds.

In view of the above, the modern scanners use stepper motor driven system. They can be directly coupled or through a timer belt. Fig. 1 shows a typical system built on stepper motors and ball screw

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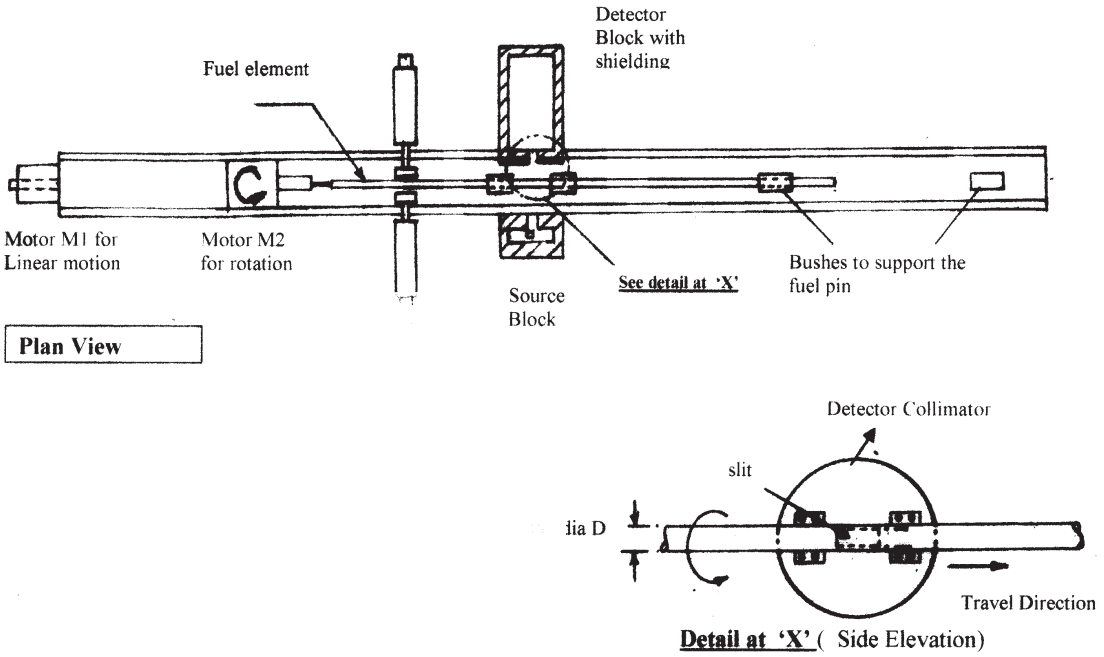

Fig. 1 Schematic of the fuel pin scanner

– LM guide combination. The source and detector are aligned in transmission geometry. The rotary motion is by a separator stepper motor M2, on the shaft of which a holding mechanism is fixed to grip the fuel element. Micro switches are provided at the locations of extremities where the carriage C has to come to rest. Sufficient number of bushes or rollers are provided to contain the vibrations due to motions. But care has to be taken that the bushes or rollers do not damage the surface of the element. Linear speed can be varied from 0.5 mm/sec. to 30 mm/sec. whereas maximum rotary speed can be 10 revolutions per second. The maximum amplitude of vibration was 150 microns as indicated by total indicator reading (TIR).

Interdependence of Operation Parameters

The linear and rotary speeds, collimator dimensions, amplitude of vibration, source strength and gamma ray energy influence the accuracy of results and their interdependence should be understood.

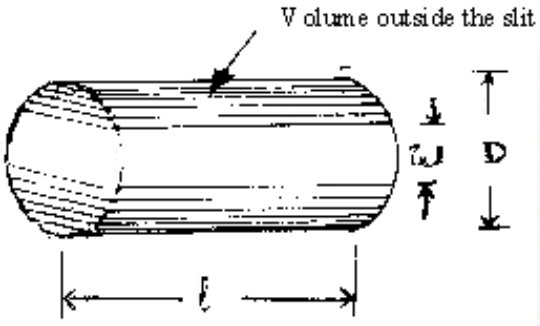

Fig. 2 Segment of fuel pin vis-à-vis collimator slit

Collilmator Dimensions and Vibrations

A typical collimator of length l and width w viewing a segment of a cylindrical fuel pin shown in Fig.2. Longer slit will give higher count rate for better counting statistics, but sensitivity to spatial variation of density is lost if the slit is too long. It should not be too wide to allow external radiation to

enter the detector. This can greatly reduce the accuracy.

A fast rotary movement of the fuel gives rise to the vibrations. Amplitude of vibrations is an important parameter in deciding collimator slit width. Volume of the shaded region will keep changing with the vibration and induce fluctuations in the count rate. This variation depends upon the slit size, fuel pin diameter and the vibration amplitude. For wider slit the count rate variation is larger. For a given fuel diameter D_1, slit width of 0.7 D_1 and vibration amplitude of 500 microns, the volume change is calculated to be about 1.8%. Using appropriate linear attenuation coefficient for the gamma ray (μ), change in count rate due to vibration can be calculated to arrive at the tolerable amplitude of vibration.

Linear and Rotational Speeds

Linear and rotational speeds should be independently changeable. Pitch (p) can be defined as the axial length moved per rotation. If S mm/sec is linear speed and R revolution/sec is rotational speed, the pitch is defined as

$$p = S / R \quad (1)$$

Dwell time (t) in MCS mode also plays an important part in gamma scanning. The dwell time, and pitch are to be optimised so that no volume element of the pin escapes scanning.

Radiation Source

It is well known that the counting statistics improves with the accumulated counts (N). The relative standard deviation is given by

$$RSD = \sigma = \frac{1}{\sqrt{N}} \quad (2)$$

The linear speed S and dwell time t and source strength are to be selected to satisfy the requirement of precision laid down in the specifications.

For uranium and plutonium bearing fuel pin, ^{57}Co is a popular source because of its 122 keV gamma energy which is closer to K-edge of uranium (115.6 keV) and plutonium (121.8 keV). This ensures that the attenuation is high in the fuel and sensitivity to fuel density increases. The gamma ray energy of the source should be such that the transmission is in the optimal band [(1)] of 10 to 37%. The choice of the gamma ray energy depends upon fuel pin dimensions, composition and the desired sensitivity. Active assay has been done using ^{241}Am and ^{169}Yb sources also.

Instrumentation

The data acquisition system is based on the detector being coupled to Multichannel Analyser (MCA) in Multi Scale mode or a single channel analyser and a hard wired MCS unit. Usually active, transmission type systems scan at speeds faster than that employed in passive counting. For keeping the provision of fast counting we have installed a timer counter specially built with dwell time of 20 m sec. The computer based data acquisition system is synchronised with START and STOP modes of scanner movement. The data is transferred to PC and analyzed by a specially written FORTRAN program.

Precision and Accuracy

Every segment of the instrument contributes to the overall performance. The precision results from adequate count rate and design and settings of the mechanical scanner. The accuracy or the closeness to true value is dependant upon the quality of calibration standards, suitability of source energy, resolution of the radiation detector etc. Accuracy can be degraded by systematic errors, sources of which can lie at many places including in the scanner. Precision for the set up at AFD/BARC was determined by scanning the same fuel element a number of times. Two parameters viz., the average linear density (g/mm) and total weight of heavy metal were assessed. The relative standard deviation on these quantities was found to be 0.4 % max for both the parameters. The accuracy is estimated by scanning of well characterised standard fuel pins.

Summary

There are certain application areas where active gamma non-destructive assay is a better option than passive assay, like inventory check or densitometry. Specific advantages of the technique are highlighted. It is shown that the specifications of the mechanical scanning system are quite stringent in active scanning. Inter-relationship of various

design and operational features like collimator dimensions, vibrational stability, linear and rotational speeds, dwell time etc and their influence on the results obtained are also discussed.

Acknowledgement

The author is grateful to the editor and the guest editor of IANCAS Bulletin for the opportunity to share the ideas. Thanks are due to Dr SB Manohar, formerly Head, Radiochemistry Division, Shri SN Betkerur, Shri BK Shah, Shri Sanjoy Das of AFD, Shri KN Vyas, LWRD and Shri S Muralidhar, RMD with whom discussions were held at different stages. The author is thankful to Shri RP Singh, Head, AFD for his for his keen interest in this work.

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Gamma Scanning and Spectrometric studies of Irradiated Nuclear Fuels and Reactor Components

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Introduction

Gamma scanning provides a non-destructive technique of determining burn-up distribution in irradiated fuel pins and cumulative fluence experienced by power reactor components. The axial gamma ray activity distribution of the fuel pin is a monitor of burn-up variation in the fuel stack. Apart from the burn-up profile, the gamma scan also gives a wealth of other information on the fuel stack. The axial burn-up and power distribution, pellet enrichment mix-up, inter-pellet gaps, pellet dimensions, fuel stack length, loss of fissile material, fission product redistribution and fuel temperature profile and pellet cracking pattern are revealed by gamma scanning.

Indian Pressurised Heavy Water Reactors (PHWRs) have pressure tubes made of zircaloy-2 / Zr-2.5%Nb alloys. During irradiation in the reactor, the neutron flux affects pressure tube creep and corrosion properties. Fracture toughness of the pressure tube is affected by the combined effect of hydrogen pick up by the tube and the irradiation damage caused by the neutron irradiation. Hence, to make a comprehensive study of the properties on the pressure tube, a detailed knowledge of the axial variation of the neutron flux is needed. Gamma scanning is carried out on full length irradiated pressure tubes removed from the PHWRs. The gamma scan is a signature of the neutron fluence seen by the pressure tube during its residence in the reactor. This provides vital information required for planning sampling of specimens for subsequent

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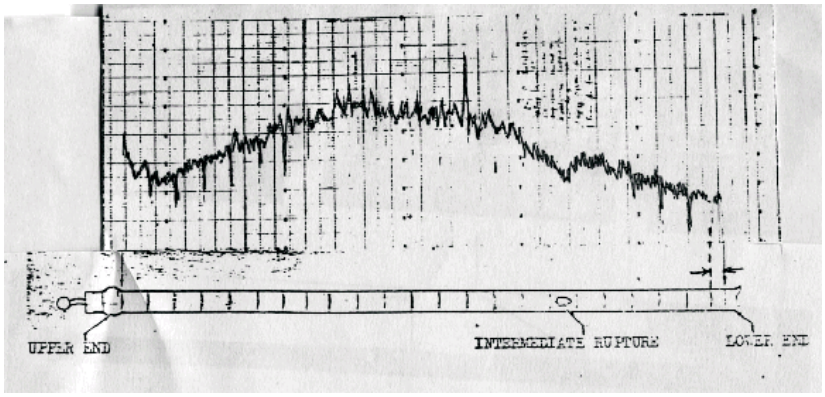

Fig. 1 Axial burn-up profile

investigations and modeling. In addition, the knowledge of activities and decay pattern of various activation products is needed for irradiated pressure tube material handling, storage and disposal. For this purpose gamma spectrometry is carried out on the samples cut from the pressure tubes from operating reactors with varying irradiation and cooling times to generate data on their residual activity.

Gamma Scanning of Irradiated Fuels

Gamma Scanner Set Up

The basic set up for gamma scanning of irradiated fuel consists of a collimator, a scanning stage and a detector assembly. The collimator assembly mounted on 1.2 m thick concrete wall of the hot cells consists of three parts. The defining lead collimator made with a rectangular slit opening of 1 mm x 20 mm size, the back up collimator made of steel with a 20 mm dia. circular hole and a detector housing made up of steel. The scanning stage consists of a fixed guided bed with a long lead screw driven by a reversible motor. The fuel element is mounted on the scanning stage for axial movement across the collimator opening. A second motor fitted on the fuel pin to facilitate rotary motion of the fuel pin. Gross gamma scanning is carried out using a NaI (Tl) detector and a single channel analyzer. Gamma spectrometry is carried out using a HPGe detector coupled to a 8K multichannel analyzer.

Gamma Scanning on Experimental MOX Fuel

The gamma scanning was carried out in Hot Cells Facility on a mixed oxide fuel element (PWL-P1) test irradiated in CIRUS reactor. The fuel element failed during irradiation testing in the loop. The axial burn up profile and the fuel pellet interfaces revealed by the gamma scan of the test pin are shown in Fig. 1. The scan also indicated the region where the fuel had leached out at the clad failed locations. Gamma densitometry was carried out on the same fuel element using ^{60}Co thrust source. Gamma densitometry scan also showed displacement of fuel stack due to swelling of top insulating MgO pellet as well as loss of fuel from the bottom of the fuel pin.

Gamma Scanning on BWR Fuels

Gamma scanning was carried on 18 BWR fuel elements with a burn up range of 5,000 to 29,000 MWD/Te, discharged from Tarapur Atomic Power Station. Gamma scanning of these fuel elements revealed various interesting features (Fig. 2). Fuel pellet enrichment mix up was observed in one of the fuel element. The observed fission product depletion from the defective fuel elements helped in identifying the location of clad defect.

Inter pellet gaps were clearly identified in all the fuel elements. Shortening of fuel column and widening of the inter pellet gaps seen on several fuel pins indicated fuel densification effect.

Significant loss of fuel material over nearly 200 mm of stack length was observed in one of the fuel elements which had developed an axial clad split due to Pellet clad interaction leading to Stress corrosion cracking. Gross gamma activity was used as a measure of total power generated by the fuel

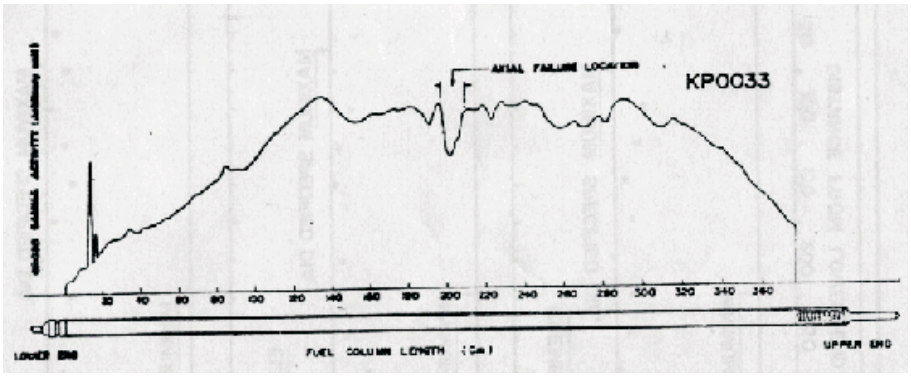

Fig. 2

element and such measurement was used in interpretation of variation of fission gas release in fuel elements.

Gamma spectrometric measurements were carried out at selected locations of the fuel element. Typical gamma ray spectrum showing various fission products like ^{134}Cs, ^{95}Zr, ^{137}Cs and ^{154}Eu is shown in Fig. 3. Activity ratio of $^{134}Cs/^{137}Cs$ was used to generate the axial burn up profile. One such axial burn up profile is shown in Fig. 4.

Redistribution of volatile fission product like ^{137}Cs was observed in the cooler bottom segment of the fuel element. The axial burn up profile and flux profile measurements generated by gamma scanning was correlated with nodular corrosion pattern on the fuel elements.

Gamma Scanning of PHWR Fuel Pins

During the course of Post Irradiation Examination (PIE) of Madras Atomic Power Station (MAPS) fuel bundles gamma scanning of individual fuel pins was carried out. A typical gamma scan of one of the fuel pins from the bundle GX-247 is shown in Fig. 5. The neutron flux peaking at the ends of the fuel column is expected in PHWR fuels. The dips in the profile correspond to the inter pellet gaps of the dished pellets.

Gamma Scanning in the Pool of BWR

An inspection facility for irradiated fuel pins is being set up at the spent fuel pool side of Tarapur Atomic Power Station. The gamma scanning well has provisions for collimating, shielding and

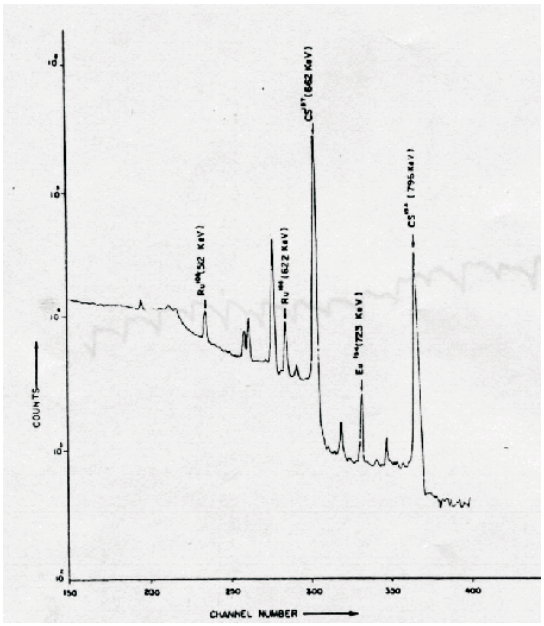

Fig. 3 A typical gamma ray spectrum of a discharged fuel element

adjustments for Cd-Zn-Te detector based probe in the pool. The gamma ray spectra will be acquired using a portable Palm Top based MCA system. Gamma scanning system will be integrated with the master data acquisition system. ^{60}Co and ^{137}Cs sources enclosed inside a dummy pin will be used for energy calibration. The detector system (resolution better than 18 keV at 662 keV) is capable of resolving ^{137}Cs and ^{134}Cs gamma rays. The activity ratio of $^{134}Cs/^{137}Cs$ will be used for the estimation of burn up.

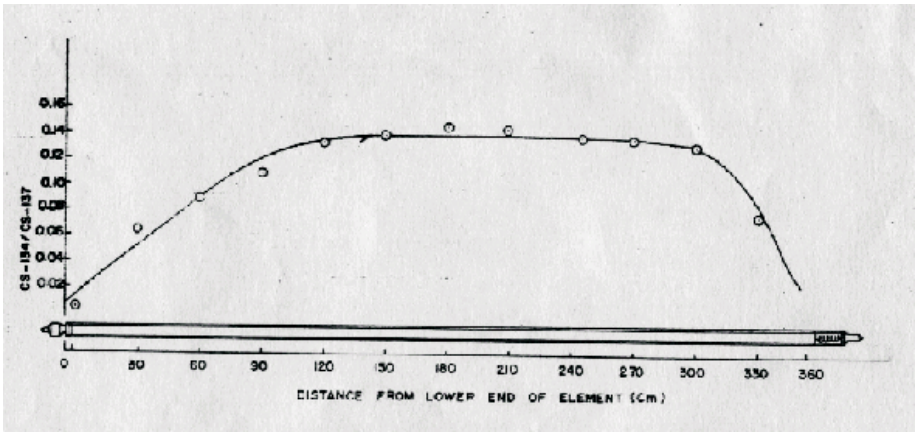

Fig. 4 Axial burn-up profile of a BWR fuel element

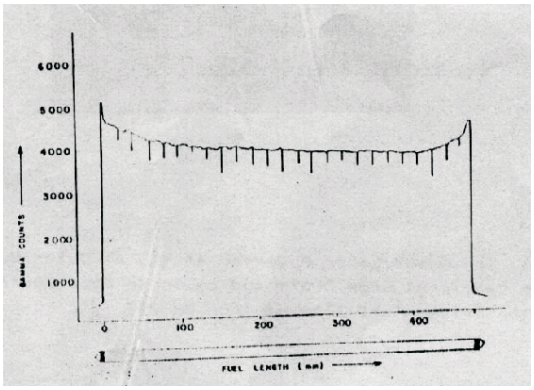

Fig. 5 Gamma scanning profile of a discharged PHWR fuel pin

Gamma Ray Study of Irradiated Pressure Tubes

Zircaloy-2 pressure tubes were used in earlier version of Indian PHWRs. Zircaloy-2 is a zirconium based alloy having 1.5% tin, 0.15% iron, 0.04% chromium and 0.02% nickel as the major alloying elements. During irradiation in the reactor, activation products are formed due to alloying and impurity elements present in the pressure tube. Zirconium has a thermal neutron absorption cross section of 0.18 barns and upon neutron irradiation forms activation product $^{95}Zr/^{95}Nb$. The radioisotope ^{125}Sn formed due to alloying element tin, decays to ^{125}Sb (E_γ = 428 keV, $T_{1/2}$.= 2.77 y)

The knowledge of activity and decay pattern of activation products is needed for planning post irradiation examination on irradiated pressure tube material, handling, storage and waste disposal. Gamma scanning on full length irradiated pressure tubes and gamma spectrometry of various samples from the pressure tubes removed from the reactor as well as scrape samples from pressure tubes from operating reactors has been carried out. These samples correspond to different residence and cooling times and locations in the reactor. A pre calibrated High Purity Germanium (HPGe) detector with IBM PC based 4K Channel MCA system was used for this study

Axial Gamma Scanning of Irradiated Pressure Tubes

Gamma scanning is carried out at discrete locations of the discharged pressure tube in the Hot Cell Facility. A special lead shielded ring having a 5 mm diameter opening is used as collimator. A NaI (Tl) scintillation detector with single channel analyser is used for axial gamma scanning of ^{125}Sb activity. The scans for ^{95}Zr and ^{60}Co activities are also carried out to ascertain that the activation is a consequence of the flux variation and not due to compositional variation along the length of the pressure tube. Fig. 6 shows three typical gamma scans obtained for pressure tubes from different locations in the reactor core and at different irradiation and cooling times.

Gamma Spectrometry of Irradiated Zircaloy Samples

High-resolution gamma spectrometry has been carried out on the samples cut from the pressure

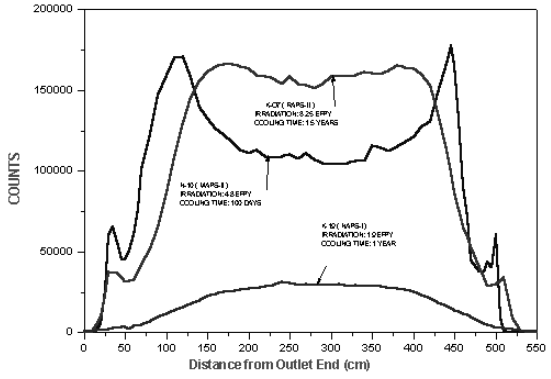

Fig. 6 Axial Gamma scan of pressure tubes

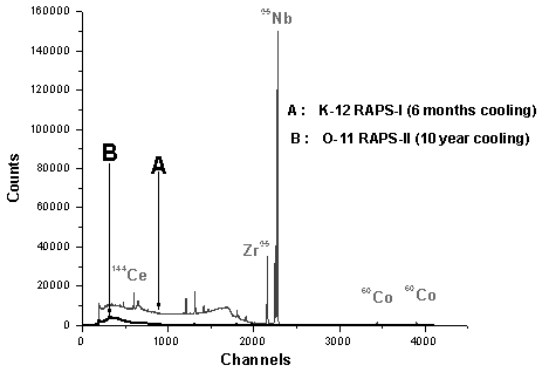

Fig. 7 Gamma spectra of a samples from PHWR pressure tubes

tubes from PHWRs with varying irradiation and cooling times to generate data on their residual activity. The pressure tubes samples corresponded to irradiation times of 7.0 and 6.5 Effective Full Power Year (EFPY) respectively. However the respective cooling times were 6 months and 10 years.

Fig.7 shows the gamma spectra for both of the samples. For coolant channels cooled for six months, the major nuclides present were ^{125}Sb, ^{95}Zr/^{95}Nb, ^{60}Co, ^{124}Sb, ^{181}Hf, and ^{113}Sn. However the residual activity is mainly due to ^{125}Sb and ^{60}Co for the long cooled sample. As seen from the Fig.6, the shape of the axial flux profile along the pressure tube changes with the position of the pressure tube in the reactor. K-19 pressure tube (3EFPY) of Narora Atomic Power Station (NAPS) which occupies a peripheral location in the reactor, shows a typical cosine flux distribution profile whereas N-10(6.5 EFPY), which occupies position near the control rod, shows double hump with a large flux depression at the centre. K-07 is another pressure tube (8.5 EFPY), also from the central location of the reactor Axial gamma scan shows the time integrated flux shape of a typical truncated cosine curve with a small depression in the centre.

Conclusion

Gamma scanning of irradiated experimental MOX fuel pins and fuel pins from power reactors (PHWR and BWR) and zircaloy pressure tubes are carried out at PIED hot cells facility at BARC. Gamma scanning on irradiated fuel pins have generated data on axial burn up distribution, pellet enrichment mix-up, inter-pellet gaps, loss of fissile material and fission product redistribution. Variation in the axial flux profile is observed in the pressure tubes operating at different locations of the reactor core. Measurement of axial gamma profile irradiated pressure tube is used for identifying regions of interest for selection of samples for property evaluation.

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Radiography Techniques for the Non-Destructive Evaluation of Nuclear Fuels

Shri K.N. Chandrasekharan *joined BARC after graduation from Kerala University in the year 1972. He is specialized in Non-destructive Testing and Evaluation of Plutonium based nuclear fuels. He was responsible for the NDE of FBTR, KAMINI and MOX experimental fuels. His current interest is in the neutron radiography and real-time imaging of nuclear fuels. He has more than twenty five publications to his credit in international journals and national and international conferences including technical reports. He is a member of the Institution of Engineers, Calcutta and Indian Society for Nondestructive Testing. Presently he is leading the NDE and Metrology group in the Inspection and Quality Control section of Radiometallurgy Division.*

Shri S. Majumdar *obtained his B.E. (Met) degree from the Bengal Engineering College, Calcutta University and joined Bhabha Atomic Research Centre through its 11th batch of Training School. Since 1968, he has been working in the Radiometallurgy Division and, at present, he is Head of the Radiometallurgy Division. He has devoted his entire working career in research, development, fabrication, charactewrization and out of pile property evaluation of Pu, Th and U based fuels for both thermal and fast reactors.*

Introduction

Radiographic testing involves passing a narrow beam of X-rays or γ rays through the object under inspection and recording the transmitted radiation through the object on a photographic film (Fig. 1). X-rays are differentially absorbed in the bulk of the material due to density and/or thickness variations. The transmitted beam carries information of the volume of the object under test like porosity, inclusion, shrinkage of cavity, piping etc. The exposed photographic film upon development provides a permanent record of the internal details of the object. A porosity being lack of material, absorb less radiation and appear as dark rounded indication where as an inclusion (e.g. tungsten in stainless steel weld) having higher absorption appear as white indication in the radiograph.

Penetrating radiations like X-rays, γ rays and neutrons are employed for the non-destructive evaluation (NDE) of various types of nuclear fuels.

When neutrons are used for radiography, the neutron beam undergoes intensity modulations due to composition and/or thickness changes in the object under test. Since neutrons do not affect photographic film emulsion, the transmitted neutron beam is imaged using a suitable converter screen (having a high neutron absorption cross section) which, on neutron absorption emits photographically detectable radiation such as alpha, beta, gamma or light. This radiation in turn is allowed to fall on a conventional X-ray film or track-etch detector to produce a radiograph.

The mode of attenuation of neutrons by material is totally different from that of X-rays. The latter interacts with the orbital electrons and therefore the attenuation coefficient increases with increasing atomic number. The neutrons on the other hand, interact directly with the nucleus. For neighboring elements and isotopes, the neutron attenuation coefficients are widely different. These

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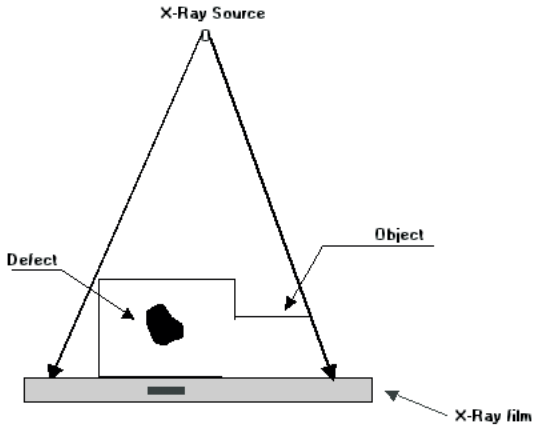

Fig. 1 Radiography Technique

properties make neutron radiography advantageous in certain cases where conventional X-ray or γ ray radiography techniques have limitations.

This article presents the various radiography techniques using X-rays, γ rays and neutrons employed during the inspection and quality assurance of nuclear fuel pins and fuel plates in BARC and highlights the modifications carried out on the conventional radiography technique to meet special requirements of the stringent specifications. Typical results obtained by these techniques during the NDE of nuclear fuels are also presented.

X-ray Radiography

X-ray radiography technique is extensively used for the Non-Destructive Evaluation (NDE) of nuclear fuels. This is a volumetric nondestructive technique and provides a permanent record of the fabricated fuel pin or fuel plate. The main advantage is that this record could be consulted during the post-irradiation examination of these fuels for performance evaluation. Fuel pin end plug welding is one of the most important stages of the FBTR fuel pin fabrication and its quality evaluation is very critical considering the fuel containment throughout the life of the fuel pin in the reactor. Therefore, stringent specifications are laid out for the acceptance of the end plug weld. X-ray radiography technique has been used for the evaluation of the end plug welds [1]. Quantitative estimation of different

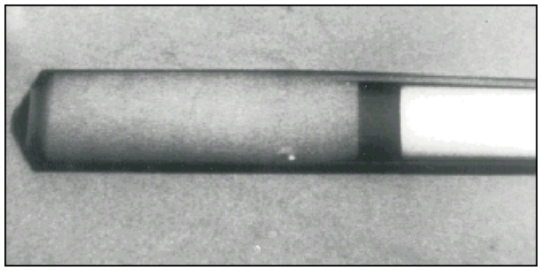

Fig. 2 Tungsten inclusion in FBTR fuel pin end plug weld (SS - TIG weld)

types of defects has been accomplished by employing a suitably designed defect standard and microdensitometric scanning of the radiograph [2]. Fig. 2 presents a typical Tungsten inclusion intercepted in an FBTR fuel pin end plug weld (SS) as seen in the radiograph. Zircaloy-2 end plug welding of MOX experimental fuel pins were also evaluated by this technique.

Gamma Radiography

X ray energy of conventional X-ray machines (~ 420 KV X ray tube) is not sufficient to penetrate high density and high Z fuel materials like UO_2 and $(U\text{-}Pu)O_2$. Because of higher attenuation, the transmitted X-ray intensity will not be sufficient enough to produce a good radiograph. Gamma radiography using ^{60}Co radioisotope source is employed to inspect such materials. Unlike the broad spectrum of X-rays produced by an X-ray tube, γ ray sources emit one or more gamma rays of discrete energies (e.g. ^{60}Co emits 1.17 and 1.33 MeV gamma rays). Because of the high penetrating power, gamma radiography is suitable for the inspection of fuel materials. Identification of annular and solid fuel pellets present in a fuel pin is possible by this technique. Fig. 3 presents a gamma radiograph showing annular MOX fuel pellets inside an experimental fuel capsule fabricated for fuel-coolant-clad compatibility test.

Gamma Autoradiography

Gamma radiation emitted by plutonium in the fuel is utilized for checking the homogeneity of PuO_2 in UO_2-PuO_2 and ThO_2-PuO_2 MOX fuel pellets. In this technique metallographically polished pellet is kept in close contact with a medium speed X-ray film

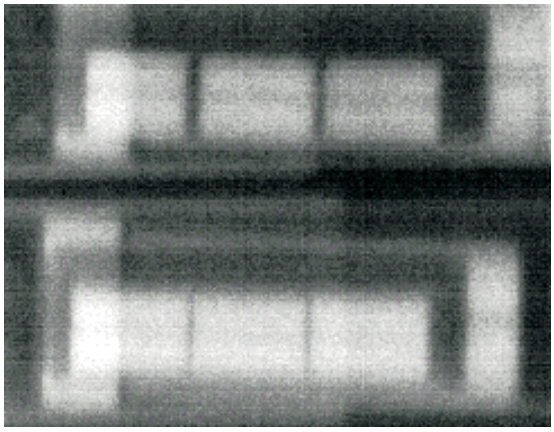

Fig. 3 Gamma Radiograph showing annular MOX fuel pellets

under a constant load for a long duration (~ 30 minutes). PuO_2 agglomerates lying at the pellet surface (or slightly sub-surface) emit more intense gamma rays compared to the matrix providing a dark spot in the processed film. Gamma autoradiograph of a MOX fuel pellet showing PuO_2 agglomerates present in the polished surface of the pellet is presented in Fig. 4. In the as-fabricated fuel pins, PuO_2 agglomerates lying in the periphery of the fuel pellets also could be detected by γ auto radiography. In this technique the fuel pin is wrapped with X-ray film and exposed for a sufficient duration. The developed film revealed dark spots corresponding to PuO_2 agglomerates lying on the cylindrical surface of the pellets.

X-ray Gamma Autoradiography (XGAR)

X-ray gamma autoradiography technique was developed to check the internal components of FBTR fuel pins [3]. This method makes use of a combination of X-rays and γ rays. A sequential X-radiography followed by γ autoradiography without disturbing the pins give a single image in the film. Low energy X-rays cast the outline of the fuel pin and the γ radiation emanating from the plutonium rich fuel column creates fogging in the respective region of the film. As a result uranium carbide insulation pellets at the extremities of the fuel column could be identified and a possible mix-up of non-standard pellets detected. Fig. 5 presents a typical XGAR of a set of FBTR fuel pins showing

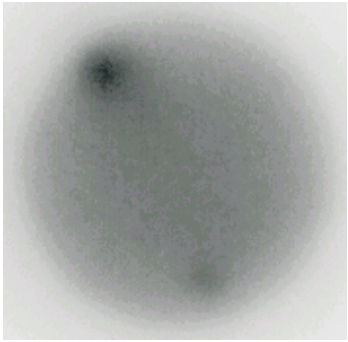

Fig. 4 Gamma Autoradiogrpah of ThO2-4% PuO_2 MOX fuel pellet showing PuO_2 agglomerates (10X)

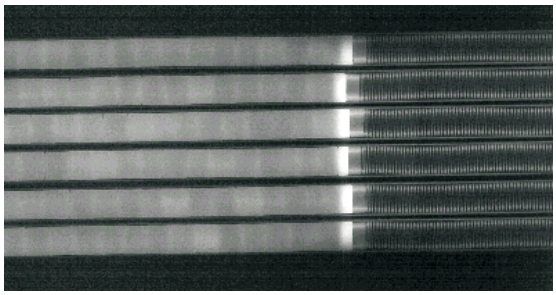

Fig. 5 X-ray Gamma Autoradiograph (XGAR) of a set of FBTR fuel pins showing Mixed Carbide fuel column, Uranium Carbide insulation pellets (white) and SS components

active mixed carbide fuel column and natural uranium carbide insulation pellets alongwith the SS components.

Neutron Radiography

Neutron radiography is a comparatively new nondestructive technique, which provides full penetration radiographs of dense fuel pellets inside the fuel pins and discriminates between neighboring elements of similar atomic number [4]. This unique feature is taken advantage for the evaluation and characterization of MOX fuel pins fabricated for irradiation testing. Neutron radiography was carried out at APSARA, a 400 KW swimming pool type research reactor at BARC. A collimated beam of thermal neutrons from the reactor is used for radiography. The fuel pins were mounted at 60 cm from the collimator followed by a light tight cassette

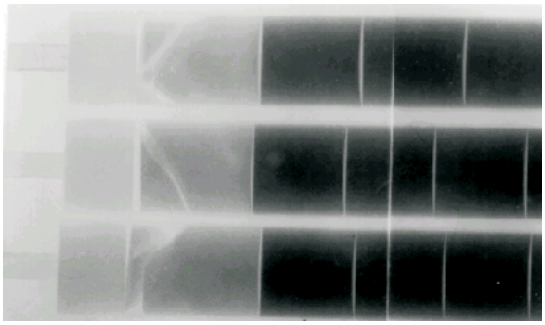

Fig. 6 Neutron Radiograph of a set of PHWR type MOX experimental fuel pins showing cracked UO_2 insulation pellets at the end of the active fuel column

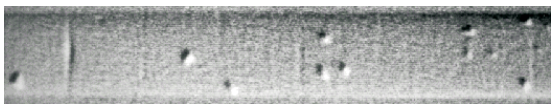

Fig. 7 Neutron Radiograph of an experimental MOX fuel pin showing PuO_2 agglomerates (Digital Image Enhanced Radiograph)

containing X-ray film and/or the converter screen. In direct technique, X-ray film & Gadolinium screen in close contact is exposed to neutrons and in the transfer technique, only Dysprosium or Indium Screen is used. After the exposure, in the former technique the X-ray film is processed to get the radiograph where as in the latter technique the active screen is taken to a dark room and exposed to an X-ray film to get the radiograph.

Neutron radiography was employed for the Quality Assurance (QA) of advanced PHWR type experimental fuel pins containing UO_2-PuO_2 and ThO_2-PuO_2 MOX fuel pellets [5]. These fuel pins contained MOX fuel column with natural UO_2 or ThO_2 as thermal insulation pellets at both the ends of the fuel column. As part of QA of fuel pins, it was essential to check the physical integrity of the fuel pellets, to assure the presence of the thermal insulation pellets at the extremities of the fissile fuel column and to ensure no mix-up of fuel pellets of different species. Neutron radiography revealed all these features present in the fuel pins. Additionally macro-inhomogeneity of PuO_2 in MOX pellets was also detected [6]. Fig. 6 presents damaged UO_2 thermal insulation pellets inside three MOX fuel pins intercepted by neutron radiography. Neutron radiography of PuO_2 agglomerates detected in MOX fuel pellets is presented in Fig. 7.

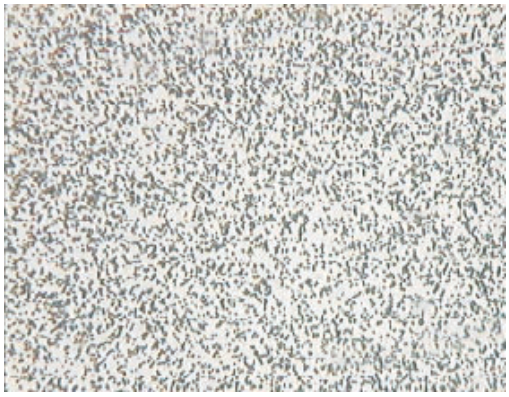

Fig. 8 Alpha Autoradiograph of a MOX fuel pellet showing uniform distribution of Plutonium in the matrix

Alpha Autoradiography

Alpha autoradiography technique makes use of the alpha radiation emanating from Plutonium in the fuel pellet. Metallographically polished fuel pellet is kept in close contact with a solid state nuclear track detector film (Cellulose Nitrate). The alpha radiation falling on the detector generates tracks, which are chemically developed in hot NaOH solution. The radiograph is viewed under a transmission microscope. The etch developed alpha tracks scatter light to reveal the image of the pellet. This technique is used as a process control step during the MOX fuel fabrication [7]. Samples from each sintered lot it evaluated by alpha autoradiography for checking the homogeneity of PuO_2 in UO_2-PuO_2 and ThO_2-PuO_2 MOX fuel pellets. PuO_2 agglomerates present in the polished surface of the pellet also can be detected by this technique. Fig. 8 presents an alpha autoradiograph of a MOX fuel pellet showing uniform distribution of Plutonium in the MOX matrix. Although essentially this technique provides a qualitative data, with proper calibration standards it can be extended to give quantitative information also.

Microradiography

In Microradiography technique the high resolution X-ray radiograph is viewed under a

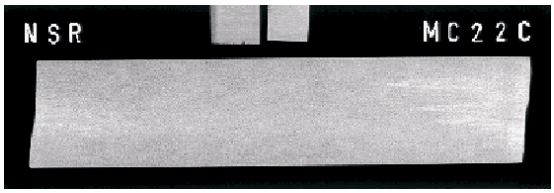

Fig. 9 Continuous Motion Radiograph of KAMINI fuel plate showing Macro-inhomogeneity at the extremities of the plate

transmission microscope at a magnification of 50 X. Small defects, which are obscure to naked eye viewing, will be clearly observed in the radiograph due to the magnification. A mini focus X-ray source with 400 microns focal spot size is used to image the object on high contrast films giving good spatial resolution. Micro cracks and density variations in fuel microspheres and micro inhomogeneity in Al-^{233}U alloy KAMINI fuel were observed by this technique.

Continuous Motion Radiography

In conventional radiography technique, the three components of the system namely X-ray source, object and the film are stationary while taking the exposure. In continuous motion radiography, only the source remains stationary where as both the object and film are moving together across a narrow beam of X-rays. This technique was developed to eliminate X-ray "Beam effect" encountered during the homogeneity evaluation of KAMINI fuel plates [8]. The spatial intensity variations across the incident X-ray beam – the beam effect – during homogeneity evaluation of alloy fuel plate by radiography followed by microdensitometric scanning resulted in an incorrect estimation of fuel homogeneity. The divergent X-ray beam is collimated to a narrow beam using a variable slit made of thick lead sheet placed above the moving stage. The fuel plate and the film are tightly secured on the platform to avoid any differential movement. A smooth continuous movement of the platform is provided by an endless timing belt driven by a small servomotor with reduction gears. The whole set up is shielded with lead sheet to protect the film from scattered radiation. The resultant radiograph is devoid of beam

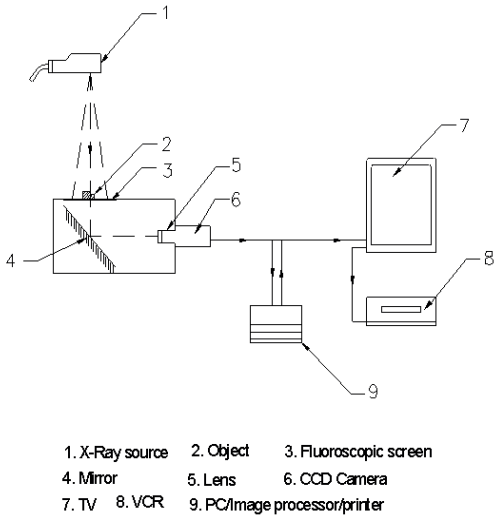

Fig. 10 Filmless Radiography setup (Schematic)

effect and the optical density changes could be directly related to the alloy distribution by comparing with standards. A typical radiograph of KAMINI fuel plate obtained by this technique is presented in Fig. 9.

Film Less Radiography

An X-ray Real-Time Radiography facility based on fluoroscopic screen was developed for the inspection of nuclear fuel plates, fuel pins and light alloy castings. The system consists of a mini focus X-ray source, fluoroscopic screen, a CCD camera with a suitable lens assembly and a TV monitor for the imaging (Fig.10). The transmitted beam through the object is allowed to fall on a fluoroscopic screen and a CCD camera picks up the image and displayed on a TV screen. A videocassette recorder has been interfaced for archiving and for easy retrieval of the X-ray images. The output from the CCD camera can be directly fed to a PC with an image grabber card to freeze any frame of the incoming live X-ray image for digital image processing and hard copy printout making the system a film less radiography facility. Besides being a film less radiography facility, one of its most attractive features is the capability of the system for use in the continuous motion mode radiography. In this mode of radiography, fuel pins or fuel plates of more than half a meter length can be radiographed in a single shot and the moving image can be seen on the monitor or recorded in the VCR.

This is a novel concept where the moving radiographic image can be intercepted at any given location to grab a particular frame of interest for further evaluation by digital image processing.

Conclusion

The various radiography techniques using penetrating radiations like X-rays, γ-rays and neutrons employed for the NDE of various types of nuclear fuels are explained. Typical radiographs obtained during the QA of these fuels are illustrated. Certain modifications incorporated in the conventional radiography techniques to meet special requirements in the fuel specifications are also explained.

Acknowledgement

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Calorimetry as a Tool for Assay of Nuclear Materials

Dr.V.Venugopal *joined the Department of Atomic Energy in the year 1971 after completing the 14th Batch of BARC Training School orientation course in Nuclear Science and Technology. He is currently the Head of Fuel chemistry Division of BARC with 115 scientists and guiding research and development in fuel Chemistry. He is a specialist in the field of high temperature thermodynamics and chemical quality control of Pu based fuels. He has been to Nuclear Research Centre, Julich for 1½ years and worked on the studies of development of nickel based binary and ternary super alloys required for Rocket Jet Nozzle and turbine blades and also on the development of high intensity metal halide vapour lamps at high temperature. He has more than 150 publications in international journals of repute. He has been honoured with NETZSCH-ITAS (Indian Thermal Analysis Society) award for 2000 and a silver medal by the Indian Society for Association of Solid State Chemistry and Allied Scientists (ISCAS) for 2002 for his outstanding contributions in the field of thermal science and solid state chemistry, respectively. He is the regional editor for the Journal of Thermal Analysis and Calorimetry. He is a life member of several professional bodies. He is presently the President of ITAS and IANCAS and Editor of INS News.*

Introduction

The ability to identify and accurately quantify uranium and plutonium is crucial to our national security. The protection, control and accounting of these materials play a central role during preparation, storage, waste assays and processing after dismantlement. A wide range of scientists and technologists are required to accomplish these tasks. The DAE units in India have been using a variety of techniques to quantify Special Nuclear Materials (SNM). The present bulletin deals with different nuclear techniques using active and passive interrogation methods based on nuclear radiations. Nuclear Material Accounting and Control Cell (NUMAC), DAE and Fuel Chemistry Division, BARC have been involved in the measurements and control of SNM. In this article, a brief description of an isoperibol twin calorimeter which has been used for assay of SNM is given. It has a sensitivity of 0.1 K detection. The design of the calorimeter is similar to the ones developed in Mound Laboratories several years before. During the last two decades, several types of calorimeters have been developed by Los Alamos Laboratory for Safeguards and security of SNM. The sensitivities of these calorimeters have been increased by many orders of magnitude by the use of thermistors and thermopiles. Plenty of information on the use of calorimeter for SNM assay is available in the literature [1-5]. Generally no single technique can perform the quantification of all the SNM. The quantification of plutonium based systems are easier than Highly Enriched Uranium (HEU) systems.

Heat production is characteristic of all radioactive decays. Especially Plutonium produces a significant amount of heat from this effect (~2.5 watts/kg) for weapons grade (WG) material and containers of plutonium are typically warm to the touch. The heat output cannot be masked by radiation shielding, not affected by the physical or chemical composition of the sample or other materials (matrix), and it is therefore used as one of the most accurate non-destructive assay methods for plutonium. Some typical thermal out put from different radiation sources is given in Table 1 and shown in Fig. 1 for comparison.

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TABLE 1. Relative thermal power output from different sources

Source	Heat output in W/g
^{238}Pu	0.568
Tritium	0.324
Plutonium (WG and Reactor grade)	0.002 – 0.014
^{239}Pu	0.0019
^{240}Pu	0.0068
^{241}Pu	0.0042
^{242}Pu	0.01
^{241}Am	0.114
Human	0.001-0.002
^{233}U	0.00028
^{235}U(93% ^{235}U, 1% ^{234}U)	2 x 10^{-6}
TRU (100 nCi/g)	3 x 10^{-9}

TRU means Trans-Uranic elements

Traditional calorimeters used for the assay of plutonium involve a heat bath kept at a constant temperature, and two insulated cylinders, immersed in the bath. The principle of the calorimeter is based on heat flow between two identical cavities. The sample is introduced into one of them, and the other remains vacant and serves as a reference arm. Both cylinders are wrapped with resistive wire, and each forms two sides of a Wheatstone bridge. The cylinder with the sample heats up and the resistivity increases, debalancing the bridge. The equilibrium deviation is an accurate measure of the temperature, and hence the sample heat output (power). A typical measurement takes 4 to 8 hours, in order to obtain an accuracy of the order of 0.1%. Such calorimeters have been used extensively to assay numerous plutonium samples. ^{235}U has a much lower heat output compared to ^{239}Pu, making it much more difficult to measure. Work is underway to increase the sensitivity to very small (less than 0.001 degrees Kelvin) temperature changes, which would enable use of calorimeters for assay of ^{235}U in bulk samples. Advantages of calorimetric technique are given below:

(i) The assay technique is non-destructive.

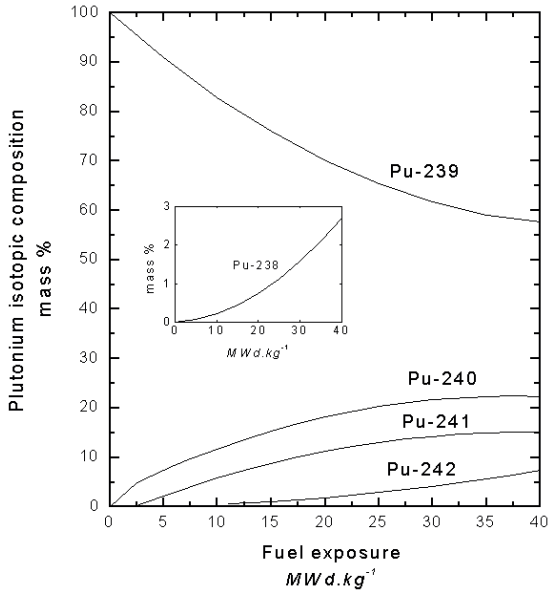

Fig. 1 Plutonium isotopic composition as a function of burnup

(ii) Calorimetric technique is highly precise and, where the isotopic composition of sample is known, highly accurate.

(iii) It can be easily adapted to an automatic data processing system.

(iv) The total power can be determined without sampling and aliquotting.

(v) Calorimetry can distinguish between fissile isotopes ^{235}U and ^{239}Pu because of the difference in half-lives.

(vi) The measurements are based on primary electrical standards which in general are more reliable than counting standards.

Principles of Calorimetric Measurements

If a sample of radioactive material is known to contain only one radio-isotope, the following equation applies:

$$W = (2.11933.10^3 \cdot Q \cdot m)/(T_{1/2} \cdot A) \qquad (1)$$

This above equation is easily derived from $-dN/dT = N\lambda$

Where,
W= power in watts measured by calorimeter
Q = disintegration energy of isotope in MeV
m = mass of the isotope in grams
$T_{1/2}$ = half life of the isotope in years
A = Atomic mass of the isotope in grams

If the disintegration energy and half life of the isotope are known, the mass of the isotope can be determined. The calorimeter thus serves as an assay instrument. If the mass and disintegration energy are known, the half life may be determined by measuring out put power of the calorimeter. If the half life is short enough, the change with time of the power of the sample may be measured calorimetrically and is sufficient to determine half life of the nuclide. The most precisely known half-lives have been determined by this method.

Equation (1) can be expressed in the form:

$$W = 1.60210 \cdot 10^{-13} \cdot Q\, N\, \lambda \qquad (2)$$

Or

$$W = 1.60210 \cdot 10^{-13} \cdot Q\lambda\, N_0 e^{-\lambda t} \qquad (3)$$

Where,
N = number of atoms of isotope at time 't'
N_0 = number of atoms of isotope at time 't = 0'
λ = decay constant of the isotope in sec^{-1}

In actual practice, equations (1) and (2) can be simplified to:

$$W = mF \qquad (4)$$

Where
m = mass of isotope in grams
F = specific power of the isotope in watts/g

The specific power of an isotope can be calculated from equation (1), or determined experimentally utilising equation (4).

The calorimetrically determined wattage (W) in equation (1) through equation (4) assumes that no significant amount of energy of radioactive decay escapes undetected from the calorimeter and that the daughter isotope is stable.

In the case of parent - daughter decay

$$N_1 = N_0\, e^{-\lambda_1 t} \qquad (5)$$

$$N_2 = \frac{(\lambda_1 N_0)}{(\lambda_2 - \lambda_1)}\left[e^{-\lambda_1 t} - e^{-\lambda_2 t}\right] \qquad (6)$$

Where subscripts 1 and 2 represent parent and daughter respectively, and N_0 = number of atoms of parent at t = 0.

The wattage of the sample can then be calculated by adding the wattages of the parent and daughter obtained and by combining equations (2) with equations (5) and (6) as follows:

$$W_s = W_1 + W_2$$

$$= 1.60210 \cdot 10^{-3}\, \lambda_1 N_0 \left[Q_1 e^{-\lambda_1 t} + \left(\frac{Q_2 \lambda_2}{\lambda_1 - \lambda_2} \right) \left(-e^{-\lambda_2 t} - e^{-\lambda_1 t} \right) \right] \qquad (7)$$

Alpha and Beta radiations are completely absorbed by a calorimeter. Gamma radiation is much more penetrating, and the calorimeter must be constructed of sufficiently thick walls so that only a small fraction of the energy may escape. The escaped energy can be calculated or measured with gamma detectors. From these results, corrections may be applied to the observed powers. However, for materials involved in reactor fuel cycle, no such corrections are necessary.

Reactor grade plutonium consists of different compositions of ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{241}Am depending on neutron spectrum, burn-up and the duration of time from americium separation.

When a sample contains a mixture of isotopes, such as reactor fuels, the heat is merely the total of the individual constituents, which may be expressed as follows:

$$W_s = \sum_{i=1}^{n} W_i = \sum_{i=1}^{n} m_i f_i \qquad (8)$$

where n = no. of isotopes of type i.

If the plutonium concentration and isotopic ratios of the material in a sample are known, the quantity of each isotope can be determined. Substitution of $R_i\, m_{total}$ for m_i (mass of a particular isotope) in equation (8) yields:

$$W_s = m_i \sum_{i=1}^{n} R_i\, F_i \qquad (9)$$

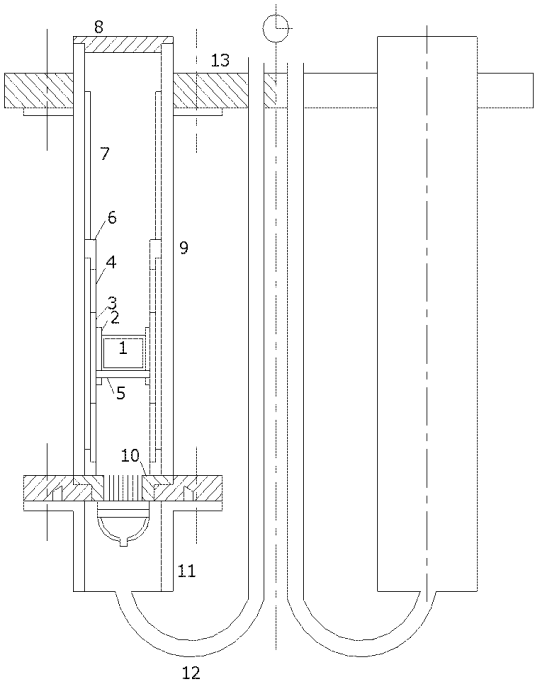

Fig. 2 Mechanical design of the twin calorimeter

Where,
m_i = the total weight of all the isotopes (grams), and
R_i = the isotopic ratio of the i^{th} isotope

The mass of a particular isotope can obtained by rearranging equation (9) as follows:

$$m_i = (R_i W_s)/(R_1F_1 + R_2F_2 + \ldots\ldots R_nF_n) \qquad (10)$$

If the isotopic analysis and calorimetry experiment are carried out in an interval of more than a month, it is necessary to correct for the change of the isotopic ratios. Normally, the uncertainties in the isotopic ratios will be larger than the amount of decay. However, when greater lengths of time are required to obtain better accuracy when ^{241}Pu is present, it is necessary that the isotopic ratios be corrected.

Generally the precision of the calorimeter measurement is greater than the uncertainty associated with the mass spectrometric measurement. A precision of ±0.3% for isotope with 10% concentration and around 1% for isotopes in the concentration range 0.1 to 1% may easily be obtained.

There are two components of heat generation in which the specific power is greater than that of ^{239}Pu. These are ^{238}Pu and ^{241}Am. ^{238}Pu is more serious for two reasons. ^{238}Pu has one mass unit lower than ^{239}Pu and hence will have tailing effect and also has isobaric interference in Mass spectrometry. This error is compounded by the shorter half life of ^{238}Pu whose specific power is 250 times higher than ^{239}Pu. Although this is partially offset by the difference in isotopic abundance ratios, the component of heat generated by the decay of ^{238}Pu present in the sample may be still significant. This effect is less pronounced in Fast Breeder Reactor (FBR) plutonium than in LWR (Light Water Reactor) plutonium, due to the lower fraction of ^{238}Pu present in the sample. ^{241}Am grows from decay of ^{241}Pu and hence a determination of ^{241}Am content is required, preferably at the time of mass spectrometric analysis. Because of the in growth of ^{241}Am, the thermal out put of reactor fuel samples also grows. The rate of this growth offers a method of verifying the ^{241}Pu content.

As an aid in the determination of the quantity of ^{238}Pu and ^{241}Am in the sample, it is suggested that the neutrons both from spontaneous fission and (α, n) reaction, be measured simultaneously along with calorimetric determination.

Design of Assay Calorimeters

The assay calorimeter designed in FCD/BARC is a differential micro-calorimeter. The mechanical design is shown in Fig. 2. The system that absorbs heat consists of an aluminum sample can (1) closely fitting into an aluminum heater form (2). The latter has a small heater (enameled constantan wire of ~250 ohms) wound over it for calibration purposes. The heater form is in turn surrounded by a close-fitting aluminum thermal form (3) which has acrylic tube extensions (4) on either side. The thermal and heater forms are held together by a stainless steel pan (5) which also act as a stop for the sample can. Two resistive coils (double nylon insulated nickel wire, around 600 turns and 1400 ohms each) are wound on 15 cm of thermal form and acrylic extensions. These coils act as temperature sensors of the calorimeter. The complete assembly sliding fit into a brass jacket (9). In the fully assembled condition, the air gap between the jacket

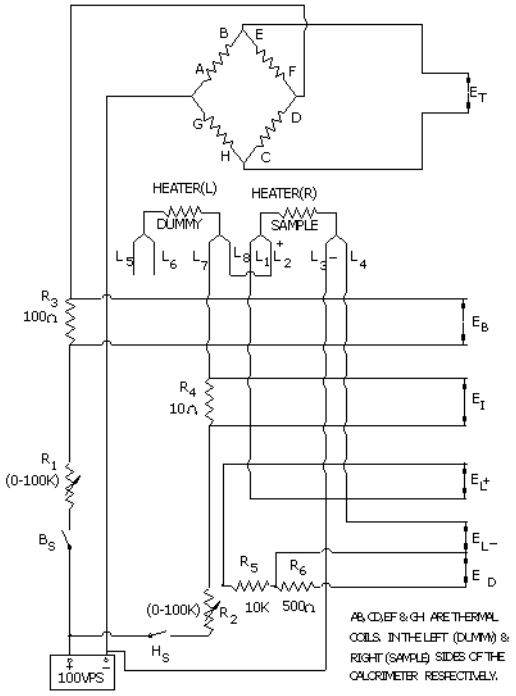

Fig. 3 Electrical circuit diagram of the twin calorimeter

inner wall and the sensor coils is 1.5 mm. Heat is conducted across this barrier gap to the jacket which is maintained at a constant temperature by submerging the calorimeter in a suitable oil bath. Two identical systems are used side by side. One is designated as SAMPLE side and the other as DUMMY side. Four no. of sensor coils are so arranged that the two coils on the same side form opposite arms of a Wheatstone Bridge. The electrical circuit is shown in Fig. 3.

Power to the bridge and the heater circuit is provided by a common 100V, 500 mA D.C. regulated power supply. Initially, all four sensor coils have same resistance and hence the bridge is balanced. If one side is heated either by a source or a heater, the bridge registers an imbalance and the steady state value of out put voltage (ΔE_T) is proportional to the heat input. Calibration is done by measuring ΔE_T as a function of heater power in the SAMPLE side. Bridge current is regulated by R_1 (0-1000 kΩ) and is monitored by measuring the potential E_D across a 100 Ω resistor R_3. The heater current is controlled by R_2 (0-100 kΩ) and is monitored by measuring E_I across R_4 (10Ω). To eliminate small amount of heat generated at the heater leads below the points where potential leads are attached, the current is passed through one pair of leads on the dummy side. The four pairs of leads are exactly identical in length and position; hence the net power delivered to the heater is that measured across its potential terminals. Measurement of E_D, E_{L+} and E_{L-} then permit calculation of heater power (P_H) by the following equation.

$$P_H = [11xE_D - E_{L-} - E_{L+}] \times (E_I/10) \qquad (11)$$

Calibration plot is constructed by taking the values of P_H and ΔE_T. After calibration is done, the same procedure is repeated to measure ΔE_T when the actual assay sample is put into the calorimeter. By using the calibration plot, power out put from the assay sample is calculated and hence the mass of the radioisotope in the assay sample can be determined. At present the calorimeter can assay not less than 3 g of reactor grade PuO_2 with an accuracy of 0.5 per cent. However, work is being conducted to increase the sensitivity of the calorimeter by modifying the mechanical as well as the electrical design of the existing calorimeter for the assay of plutonium in bulk and in waste packets.

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Non-Destructive Techniques in Uranium Fuel Fabrication

Shri B.Laxminarayna *passed B.E (Mech) from Osmania University in 1973 with distinction. After graduating from 17th batch of BARC training school, he joined Power Projects and played an important role in commissioning of RAPS & MAPS. Subsequently, he has joined Nuclear fuel complex in 1981 as production in charge for hot extrusion operations for production of seamless tubes of various materials. At present he is Deputy General Manager for Quality Assurance of Fuels. He has over 15 years of experience in the field of Quality management and played an important role in establishing Quality Assurance systems at NFC. He was deputed to undergo extensive training in Quality control and Inspection of Nuclear Fuels by IAEA. He has to his credit large number national & international papers in field of quality management and Non destructive testing. He has been recipient of Best officer Award for outstanding performance.*

Introduction

Nuclear Fuel has to go through very harsh and severe environment involving high temperature, radiation and corrosive media. Thus it becomes mandatory for the fuel manufacturers to maintain highest quality levels during fuel fabrication. Nuclear Fuel Complex (NFC), Hyderabad produces nuclear fuel and structural components for all Pressurised Heavy Water Reactors (PHWRs) and Boiling Water Reactor (BWR's) in the country. It is a unique organization of its kind in the world, where both zirconium alloys products and finished fuels are produced under one roof. During the financial year 2002-2003, 30,000 nos of PHWR fuel bundles and 110 nos of BWR fuel assemblies were produced; leading to turnover of about 8000 million Rupees. NFC has adopted ISO-9002 as its quality system. Under its aegis a wide array of quality control procedures and inspection tests are carried out so as to ensure that the desired levels of quality are met. Uranium oxide pellets are produced by powder metallurgy route. UO_2 powder is subjected to pre compaction and granulation to produce free flowing granules followed by final compaction to produce green pellets. Green pellets are sintered at temperature of 1700^{o}C in reducing atmosphere to produce high density UO_2 pellets with 95-98% theoretical density. The sintered UO_2 pellets are loaded into thin walled Zirconium alloy tubes and the ends are sealed either by resistance welding in case of PHWR fuel or TIG welding in case of BWR fuel after pressurizing with Helium. The sealed fuel pins are clustered together to form fuel bundles.

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Among the various inspection tests that are carried out, non- destructive tests occupy a special place as they facilitate 100% online and offline assessment of the products. A plethora of Non-Destructive Evaluation (NDE) methods are used at various stages of fuel fabrication. These include Ultrasonic testing, Radiography, Gamma scanning, Beta Backscatter test, Helium leak testing, vision systems both machine and manual systems.

Non Destructive Assay carried on Zirconium Alloy Thin Walled Fuel Tubes

Zirconium alloy tubes are used for the cladding purpose in nuclear fuel. Various NDE tests carried out for the tube evaluation include visual examination, Ultrasonic testing and eddy current testing apart from dimensional measurements.

An indigenously developed automatic and computerized ultrasonic testing machine is used for fuel tube inspection. The system works on tube rotation technique using pulse echo immersion technique. It inspects tubes with speeds up to 2.5 m/min with high sensitivity and reliability. Eddy current testing forms a complementary test for detection of surface and sub surface defects.

Non Destructive Assay carried on BWR Fuel

The BWR fuel is a mechanical assembly of 36 fuel pins each of approximately 4m length in a 6 x 6 matrix. The tests carried out involve Visual inspection and density measurement of UO_2 pellets, Radiography of the welds, Gamma Scanning and Helium leak testing of fuel pins visual and dimensional inspection of fuel pins and fuel assembly.

Density Measurement of UO_2 Pellets

Geometric method is used for the density measurement of UO_2 pellets. It involves obtaining density by taking ratio between mass and volume. An indigenous laser based pellet density measurement station has been developed for measuring the pellet densities. The system utilizes solid- state diode lasers for carrying out dimension measurements. Average diameter is computed by taking three scans at three different locations and a single scan is used for length determination. The weight measurements are carried out simultaneously along with length and diameter. The data is transmitted via RS-232 port to a computer which computes the density. Oracle based software has been developed for maintaining the data and for report generation. The system measures density for about 250-pellets/ hour with accuracies of 0.01 g/cc.

Visual Inspection of UO_2 Pellets

Visual inspection is the most widely used NDE technique during fuel fabrication. Even though visual inspection appears to be very simple but provides unrivalled information regarding the product. It gives unmatched three dimensional resolution, field of view, colour discrimination and provides real time processing.

UO_2 pellets are subjected for 100 % visual inspection for various surface defects. On an average 20 million pellets are subjected to visual inspection annually. Operators are provided with frequent training classes to maintain high standards during inspection and to minimise operator to operator variations. Periodically operators are evaluated by using Juran Meishalmer Index test, where their performance is compared with a standard.In order to minimise the operator strain, automated machine vision systems for inspection of pellets is being developed. In a structured environment, where repetitive operations have to be carried out, such a system is advantageous. The present system is a PC based system and can evaluate one pellet at a time and can compute dimensions, area of cross-section for all the surface defects irrespective of their geometry.

Radiographic Evaluation of Endcap Welds and SS Tie Plate Castings

Tig welded end closure welds of BWR fuel pins and SS tie plate castings are subjected to radiography to ensure their soundness. The radiography of these welds is quite complex, as the ratio of the wall thickness to the diameter of the tube including the endplug is very less, thereby restricting sensitivity.

Special double wall, double image technique with zirconium alloy correction block is used to get a radiographic sensitivity of 1.4%. Welds from 9 fuel pins are radiographed simultaneously. In order to

cover total weld area, radiographic exposure is carried out at three different positions, each 120 degrees apart.

As a supplement to the above test an ultrasonic test method of evaluation is being developed. This method will facilitate detection of even finer defects.

SS tie plates are intricate castings and each BWR fuel assembly has two such tie- plate; one at top and other at bottom; these are used for holding the fuel pins together. These components are radiographed by using single wall single image, double wall double image technique and double wall single image techniques.

Gamma Scanning of BWR Fuel Pins

There are pellets of three different ^{235}U enrichments in BWR fuel assemblies' viz. 1.6 %, 2.1 % and 2.66 %. The fuel design requires that all the fuel pellets in a fuel pin must be of same enrichment. The amount of Gamma radiation emitted by the fuel pellet depends on its enrichment level and this forms the principle for operation of Gamma Scanner. The computerised gamma scanner consists of a NaI(Tl) detector coupled to a multi channel analyser to obtain a digital display of the enrichment levels of the pellets.

Leak Testing of Fuel Pins

In order to ensure leak tightness of the fuel pins, these are tested for leaks by using mass spectrometer based Helium leak detectors operating on contra flow principle. In contra flow technique the Helium atoms enter mass spectrometer against the pumping direction due to its small atomic radius and low mass. Fuel pins are tested for leaks by pressure vacuum testing. The fuel pins pressurised with helium are placed in a evacuated (10^{-5} torr) test chamber. The Helium leak detection system is fully computerised and all operations are performed automatically once the component is placed in test chamber. It also maintains database and gives report in desired format.

Non Destructive Assay of PHWR Fuel

PHWR fuel is based on classical CANDU fuel design. It consists of 19 fuel pins that are clustered to form a fuel bundle. Like in BWR fuel, pellet fabrication forms the first step in fuel fabrication. The NDE tests carried out during PHWR fuel fabrication include visual inspection, Ultrasonic Testing of end-closure welds and graphite coating measurement by Beta ray back scattering technique.

Ultrasonic Testing of PHWR Endcap Welds

The process of sealing the ends of the fuel pin is the most critical job during fuel fabrication as the pin has to contain highly radioactive fission products and is carried out by resistance welding. Due to its complex shape, evaluation of PHWR welds is difficult. Unlike BWR welds, Radiography cannot be used for these welds evaluation, as it does not provide desired sensitivity.

NFC has developed pulse - echo immersion ultrasonic testing method for evaluation of these welds. The system uses 0.2 mm, spot focussed probe of 30 MHz. A motorised system provides both rotary and linear movement to the fuel pin thereby subjecting entire weld area to the UT beam. In order to calibrate the system a fuel pin with naturally occuring leak of 1×10^{-8} std cc/sec and a fuel pin with 0.1 mm diameter laser drilled hole at 45^{o} to the tube axis are used.

So far over half million welds have been evaluated by this method and very high degree of co-relation has been found between the defect signals and defects. Defects are characterised based on the signal shape. Efforts are being made to carry out the signal analysis by using Fast Fourier transforms. The method not only identifies the defective weld but also locates the defect.

Graphite Coating Thickness Measurement by Beta Back Scatter Technique

In order to reduce pellet-clad interaction (PCI) in the fuel, the inner surface of the fuel tubes are given a thin coating of graphite (5 to 9 microns). Coating thickness is very important because a thinner coating will enhance friction between clad and pellet. A thicker coating would lead to poor adhesion. In order to measure the coating thickness; a NDE technique using back scattering of Beta rays has been developed. It works on the principle that the intensity of beta rays back scattered is dependent on the atomic number. However, the method is suitable only when there is at least 20% difference between atomic number of coating material and substrate. When graphite is coated on Zirconium alloy tube, the extent of backscatter reduces as the graphite coating thickness increases. ^{147}Pm is used as a source of Beta rays. GM (Geiger Muller) Counter coupled to a microprocessor is used for thickness measurement.

Vision systems for Tube Inspection and Fuel Bundles

A PHWR fuel consists of 167 Zircaloy components held together with 328 welds. Production of 450 tones of PHWR fuel every year calls for visual inspection of around 5 million Zircaloy components every year. The human based visual inspection poses certain limitations when inspection has to be carried out on such mammoth scale. These deficiencies arise from limitations of resolution, accessibility, fatigue and absence of quantification ability. These problems get further aggravated in inspection of fuel components because of their relatively small sizes, reflective surfaces and the close tolerances required.

In order to overcome this problem, a computer - based vision system has developed indigenously. These systems consist of video camera coupled to PC having video frame grabber card. The lens and illumination is altered depending on the component. For viewing tubes, zoom lens along with fiber optic illuminator is used for capturing the image of the component. In case of PHWR fuel bundles monochromatic halogen illuminator is used along with Telecentric lens. Image analysis software has been developed for making measurements of distances, angles, areas etc in the region of interest.

The system is being used for both process control and inspection. Tube chamfer, bearing pads, spacer pads and end caps are being inspected by using this system. The system has also been coupled to endoscopes for viewing ID defects, uniformity of graphite coating and appendage weld nuggets impressions.

Conclusions

NDE plays an important role both as a tool for process control and inspection. At present NFC is concentrating on automation of most of the techniques so as to reduce manual dependency. The test methods have helped in maintaining fuel quality levels which compares with the best in the world.

Acknowledgments

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Fuel Failure Monitoring System in PHWRs

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Introduction

The fuel used in nuclear power reactors is generally in the form of long cylinders (solid rods or collection of pellets) enclosed within metal cladding. The cladding serves to contain the fuel and the radioactive fission products and to protect the fuel from possible chemical attack by the coolant.

In Indian 220 MWe Pressurised Heavy Water Reactors (PHWR), the fuel consists of UO_2 pellets sealed in Zirconium alloy tubes of 15.2 mm OD, 0.4 mm wall thickness. Nineteen of such 49.5 cms. long rods make up a fuel bundle. Each of the 306 pressure tubes in the reactor holds twelve fuel bundles in heavy water coolant at about 293^0C. Thus at a time 69,768 nos. of fuel rods are loaded in a 220 MWe reactor. In 540 MWe reactors being set up at Tarapore, 37-elements fuel bundles will have overall 1,88,552 fuel rods loaded in 392 channels of each reactor.

Despite stringent quality control requirements imposed at every stage of manufacture and extensive programme of testing being carried out, defects in the cladding can still develop during irradiation of fuel elements over a period of time and also due to thermal stresses caused by unavoidable power cycling. Wear and tear due to fuel movements during loading and shuffling can also result in cladding ruptures. It is, therefore, impossible to guarantee that no penetration of the cladding will occur during reactor operation. The effects of such failures could vary from a minor release of fission products into the coolant to a major incident involving gross release of fission products to the environment.

Consequences of Fuel Failure

A cladding rupture results in the escape of fission products from the fuel to the primary coolant. Continued operation of the reactor with the presence of ruptured fuel elements would cause excessive radioactive contamination of the primary coolant system and make maintenance work around the equipment, piping and tubing difficult. Besides, the increased release of fission products into the coolant, in turn, increases the activity levels in various areas of the reactor building and also the activity discharged to the environment through stack by means of heavy water leaking out of the primary system. Continued irradiation of a defective fuel element may also result in its distortion, thus making its removal from the reactor channel more difficult. Therefore, in order to limit the consequences of fuel failures to an acceptable level, it is desirable, for the safe and healthy operation of a reactor, to detect the presence of a failed fuel, locate its position and remove it from the core as soon after failure as possible.

On-power re-fuelling is a feature of all PHWRs type of reactors which have very low excess reactivity. In this type of reactors, re-fuelling to

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compensate for fuel depletion and for overall neutron flux shaping to give optimum power distribution is carried out with the help of two fuelling machines, which work in unison at the opposite ends of a reactor channel. Because of this feature of PHWRs, on-power removal of failed fuel bundles is possible with the help of fuelling machines. It is therefore desirable to keep a continuous watch on fuel behavior during reactor operation, identify and locate the channel containing failed fuel with the help of a reliable Fuel Failure Monitoring System and remove the failed bundles from the reactor as early as possible to limit the consequences to an acceptable level.

Basic Principle of Fuel Failure Monitoring

All methods employed for fuel failure monitoring depend upon monitoring the primary coolant samples for the presence of fission products. The coolant is always having some fission products inventory from uranium surface contamination of fuel cladding or from previous failures getting re-circulated. At steady operation this inventory achieves an isotopic balance determined by the production rate and the processes operating to remove the fission products from the coolant. When a fuel failure occurs, fresh fission products are released into the coolant, resulting in increase in total inventory and in some cases alter isotopic balance.

Fuel Failure Monitoring in PHWRs is basically accomplished in two phases, viz. (i) detection of failed fuel by globally monitoring the bulk coolant and (ii) its location by individually monitoring reactor channels and identifying channel containing failed fuel.

For detection of any fuel failure, it is essential to monitor increase in inventory in the bulk coolant or a shift in isotopic balance. For location of failure in the reactor channels, it is necessary to measure locally high concentration of particular fission products against an average concentration distributed throughout the coolant.

Detection of Fuel Failure

Indication about any failed fuel in the reactor core is obtained from either or both of the following methods. The reactor building exhaust air is monitored for its activity level. Any increase in the activity release rate is mainly contributed by gaseous fission products and Iodine which have leaked to the primary coolant from the fuel through its ruptured cladding. From the primary coolant, these activities are carried to the air by the leaking heavy water. The monitoring of reactor building effluent is carried out to control the release of radioactive iodine isotopes to the atmosphere and to generate information regarding possible fuel failure.

^{131}I (half life 8.05 days) is the most important nuclide for regulatory requirements and its measurement in the bulk primary coolant gives a reliable indication of presence of failed fuel in the core. Apart from ^{131}I, other isotopes of Iodine and various isotopes of Xenon and Krypton are studied to determine the type of defects in the fuel cladding and to obtain a preliminary estimate of their number.

Location of Failed Fuel

For locating the failed fuel, delayed neutron (DN) activity due to precursors ^{87}Br and ^{137}I (fission products) is monitored in coolant samples drawn from each of 306 reactor outlet feeders. Increase in DN activity above background in a coolant channel indicates presence of failed fuel in that channel.

Principle of Operation

The delayed neutron monitoring system employs detection of delayed neutron emitting fission products in the samples of primary coolant from 306 fuel channels in the reactor. Six groups of delayed neutrons are emitted from certain fission products. The fission products of importance are ^{87}Br and ^{137}I, having half lives of 56 seconds and 22 seconds respectively. The isotopes emitting other 4 groups of delayed neutrons are of little use because of their small half lives, 6.2 seconds and less. The delayed neutron (DN) signal for monitoring the presence of defective fuel is, however, degraded by the background activity generated in the coolant by fissioning of tramp uranium and other non-fission products activities mainly due to neutrons emitted by 4.2 seconds ^{17}N produced in the coolant and photoneutrons produced in D_2O by high energy gamma rays from 7.1 seconds ^{16}N. The effects of ^{16}N and ^{17}N are reduced by selecting suitable sample tube size and delay time from reactor to neutron detector which requires a compromise between

reduced background contribution and the reduced signal.

The neutron signal from a reactor channel, as measured by delayed neutron monitor, varies from channel to channel due to the following reasons:

(i) Variations in detectors sensitivity.

(ii) Variations in the characteristics of electronics system associated with the detectors.

(iii) Uneven reactor flux distribution.

(iv) Variations in sample transport time, which is a function of the sample line length,

(v) Variations in uranium surface contamination.

Variations due to (i) & (ii) above can be eliminated by normalising the DN signal to the average DN signal of the basic channel group to which the relevant signal belongs. Variations due to (iii) are mitigated by grouping the channels in such a way that the channels in each particular group fall in similar flux region, and variations due to (iv) are eliminated by providing pressure compensation tube coils so as to equalise the transport time. With all the assignable causes for variations thus minimised, the only cause for variation may be due to Uranium surface contamination, which in view of the large number of channels, could be assumed to follow a Gaussian statistical distribution pattern. This is established during commissioning with no failed fuel present in the core and a value for the standard deviation σ (SIGMA) is obtained. Then the probability of deviation of the normalised reading from unity by more then 3σ is just 0.0027. This means that any observed variation of the normalised reading by more than a factor of 3σ could not be explained by statistical behavior alone and could, to a high probability, be indicative of cladding failure.

General Description

The delayed neutron monitoring system is provided to give indication of DN activity in the outlet of 306 fuel channels of the reactor. Bi-directional flow of primary coolant in the reactor makes it advantageous to divide monitoring of 306 fuel channels in two locations, one at each end of the reactor, known as North and South DN monitoring rooms. Refer Figure 1. It is not viable, both physically and economically, to provide individual dedicated monitoring electronics for each of the 306 reactor channels. 306 reactor channels are thus covered by grouping them into 34 groups of 9 channels each. 9 channels of each group are connected to one header, which is called sample tube header. Outlet of sample tube header is further connected to one sample monitor tube for monitoring the delayed neutron activity. There are thus 34 nos. of sample tube headers and sample monitor tubes, half of them located in each DN monitoring room. Each sample monitor tube is associated with one neutron detector. By this arrangement, each reactor channel is provided with a sampling tube leading to one of the 34 sample monitor tubes.

Thus, each neutron detector through its corresponding sample monitor tube, is permanently associated with 9 fuel channels so chosen as to be in areas of similar flux. 9 channels in each group are monitored one by one with the help of solenoid valves connected in all the lines. The DN activity in an individual sample line can be measured by valving off 8 of the 9 sample lines connected to each sample tube header.

D_2O coolant samples at temperature and pressure of about 293°C and 87 kg/cm^2(g) respectively are taken from individual outlet feeders before they join the reactor outlet headers and led to the DN monitoring rooms through 10 mm O.D. tubings. The tubing carry the samples from the feeders to the DN monitoring rooms. Each sample passes through a filter and a solenoid valve and connected to a sample tube header where 8 other samples join with it. Outlet sample from the sample tube header is led to the sample monitor tube. 17 sample monitor tubes are housed in a moderator tank in each DN monitoring room. The filters are provided to filter out particulate matter of the corrosion products and crud so that the operation of the solenoid valves is not impaired.

Each sample monitor tube is monitored by a BF_3 counter, mounted directly above it in the moderator tank. Detectors (and also sample monitor tubes) are arranged in a square lattice of 254 mm pitch inside the moderator tanks. Moderator tanks are filled with process water, which serves the purpose of thermalising the fast neutrons for detection by BF_3 counters. Further, the process water

is circulated continuously to remove the heat transferred into the tanks by the hot D_2O samples so as to avoid damage to BF_3 counters. After monitoring, all the 17 samples from sample monitor tubes in each moderator tank are connected to return header. Two return lines carry the combined samples back to the primary heat transport system in downstream lines of north and south steam generators.

The sample monitor tubes are of re-entrant type with small annular space for the sample flow to minimize the contribution of photoneutrons from γ-n reactions of 7.1 sec. ^{16}N on D_2O.

Hydraulic Considerations

The delay time of DN monitoring samples from the reactor must be so chosen as to satisfy two main requirements. Firstly, the delay time must not be so long as to reduce the DN count rate to unacceptable levels. Secondly, the delay time must not be so short as to give a poor discrimination ratio. For good resolution of delayed neutron activity due to the failed fuel, background activity due to ^{17}N and ^{16}N is reduced by providing a delay of about 40 to 50 seconds for the coolant sample before it reaches BF_3 counter.

Coolant sample flow rate should be sufficient to flush out the previous sample from sample tubes and associated equipment as quickly as possible. Other than this the condition that dictates the choice of a particular flow is the size of sample tubing required to give certain pressure drop. The driving head available is about 2.65 kg/cm^2 which exists between channel outlet feeders tap points and the boiler outlet sample return points. The delay time is controlled by selecting 10 mm OD tubing for the main part of the tubing run of about 20 metres from feeder tap points to inlet of filters. Dissipation of the driving head so as to yield nearly identical flow rates and sample delay time in each sample line is achieved by the introduction of additional lengths of 6 mm OD tubings. Individual lengths of 6 mm OD tubing are calculated for each reactor channel in order to get identical flow rate and delay time and, wherever required, additional lengths are provided in the form of coils. With this arrangement, a flow of about 25 cc/sec is obtained with a delay of about 50 seconds when a single sample line is valved-in.

System Electronics

BF_3 neutron detectors and associated electronics for monitoring half of the reactor channels, with primary coolant flow from south to north, are located in north DN Monitoring room. The output signals from the neutron detectors are fed to the pre-amplifiers, outputs of which are further connected to the linear count rate meter units. The analogue signals from the count rate meters are connected to manual data collection unit located in control equipment room. The circuitry associated with the south DN monitors is identical to that of the north DN monitors.

34 analogue signals are also connected to Computerised Operator Information System (COIS) for processing. DN activity data is collected manually with the help of manual data collection unit when the computer is not in operation.

BF_3 counters are selected as neutron detectors in view of their high sensitivity and better gamma discrimination. The sensitivity of the detector for thermal neutrons is about 45 counts per second per unit flux and operates at about 2700 volts. Signal outputs from BF_3 counters are fed to pre-amplifiers, 34 in nos., through 15 meters long super screened co-axial cables. Pre-amplifiers are of charge sensitive type with a charge gain of 0.2×10^{12} volts/coulomb. EHT supply for the BF_3 counters is provided through the pre-amplifiers. One test pulse generator is provided in each pre-amplifier unit for checking the performance of pre-amplifiers and count-rate meters. Count rate meters (CRMs) convert the input pulses from pre-amplifier into 0 to +10 volts analogue signal, proportional to the rate of input pulses. Each CRM board has a discriminator with adjustable threshold level in the range of 0 to -100 mV, which are individually set to respond only to neutron pulses from the detectors. The normal background count rate with no failed fuel is about 5 to 15 counts per second.

High voltage power supply is variable from 2000 V to 3000 V DC, and is normally set at 2700 volts. The supply is provided with over voltage & short circuit protection. Provision is made to switch off the high voltage power supply in 'TEST' mode (i.e. when test pulse generators outputs are fed to

pre-amplifiers). High voltage power supply is rated for a minimum load current of 1 mA at 3 kV.

Low voltage power supply units provide ±15 V DC to pre-amplifiers and ±12 V DC to CRMs.

Collection of DN Activity Data

System operation can be carried out in two modes viz. (a) 'Manual' mode, and (b) 'Computer' mode. Under normal conditions, scanning of DN activity data, its analysis and printing of the results is carried out by computer. When the computer is not working, data is collected manually with the help of Manual Data Collection unit. 'Manual' or 'Computer' mode of operation is selected by the control room operator. DN activity data is normally collected once in a day; and more often when there is some indication of fuel failure as given by high ^{131}I level in the primary coolant and/or high activity (due to gaseous fission products and iodine) in the reactor building exhaust air.

Provision also exists to monitor DN activity of any selected channel continuously with the help of COIS. Once the reactor channel containing failed fuel is confirmed, the failed bundle is identified by continuously monitoring that particular channel during re-fuelling with D.N. Monitoring System.

Conclusion

Fuel failure monitoring system has been playing very significant role in safe operation of all the PHWR type of Indian nuclear power stations. During operation a close watch is kept on ^{131}I level in the primary coolant by monitoring coolant samples once a shift. If it exceeds single digit level (in micro-curie per litre of the primary coolant), action is taken to locate the failed fuel with the help of D.N. monitoring system and remove it from the reactor core as early as possible. From time to time the system has given very reliable indications about fuel failure in all the operating power stations, even at low reactor power levels. The system is also capable of identifying failed $^{232}Th/^{233}U$ fuel bundles, which are initially loaded in the fresh core for flux flattening. Only limitation of the system is that it can not operate when the reactor is shut down.

Nondestructive Science and Technology for Fast Breeder Reactors and Associated Reprocessing Plants

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Introduction

Fast Breeder Reactors (FBR) and associated fuel cycle technologies, constitute important stage in the Indian Nuclear Energy Programme. R&D in the area of Non-destructive evaluation (NDE) science and technology forms an important part of the developmental efforts to ensure the integrity of engineering structures of the liquid metal cooled FBR during its service. Presence of undetected

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defects/flaws in the materials/structures could lead to failures due to their growth and unstable propagation in service, once these defects reach a critical size. In spite of the utmost care taken during the pre-service inspection (PSI) stage, defects/material degradation could occur due to the synergistic effects of stress, temperature, neutron irradiation, environmental conditions, vibration or fretting, which would have detrimental effects, in the overall plant safety and availability.

NDT techniques employed on the pre-service front detect harmful defects and also monitor their growth during the service life so as to prevent leakage of radioactive fluids and to avert catastrophic failure of the components. A variety of NDT techniques are available for detection, location and sizing of defects. In many instances, the well-established NDT techniques for detection and evaluation of defects cannot be directly employed for the inspection of reprocessing plant components, because of limited access and radioactivity considerations demanding innovation in remote application of techniques. Often, through-insulation NDT techniques, non-intrusive immersion methods, high performance sensors and special gadgetry are developed towards meeting the penultimate goal of safe and reliable operation of reprocessing plant components.

This paper focuses on various NDE techniques developed/being developed for the inspection of FBRs and reprocessing plant components. Indigenous developments of techniques suitable for specific components have been discussed. Implementation of NDE techniques for ISI with suitable remote handling mechanisms is also dealt with. Advanced NDT&E techniques and technologies of relevance are briefly described.

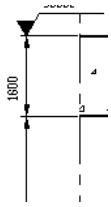
In-Service Inspection (ISI) of main and safety vessels of PFBR

The primary containment structures of PFBR comprise main vessel (MV), safety vessel (SV), and reactor roof structure. The MV and reactor covers, confine the primary coolant and cover gas with any associated radioactivity. The MV supports the primary circuit equipment and core supporting structures. The MV is guarded against any sodium leakage by the outer SV, which is placed concentric to the MV. The interspace between MV and SV is relatively small, typically about 300mm. It is required to carry out periodic ISI of MV and SV welds and visual inspection of outer surface and inner surface of the MV and SV respectively.

Fig. 1 shows schematically a section of the interspace between MV and SV of PFBR with the robotic vehicle and associated inspection modules. The vehicle being developed is a four-wheel drive, remote controlled robotic device to carry non-destructive examination equipment into the interspace between the MV and SV for enabling ultrasonic testing of welds of both vessels and visual examination of external surface and internal surface of the MV and SV respectively. The vehicle has a foldable design to facilitate insertion through the openings provided from the operating floor, into the vessel interspace. Once inserted into the interspace, the vehicle can be spread out by separating one pair of arms using pneumatically actuated cylinder and link mechanism so that its wheels grip the two vessel walls to provide reaction needed to maintain the device in position against gravity and the frictional force required for traction. The vehicle is capable of adapting to the variations in the interspace. This vehicle is designed to move around the cylindrical and curved surfaces in the interspace to facilitate inspection even in the bottom curved regions. The vehicle carries two CCD cameras, one for visual inspection of vessel surface and the other for navigation of the ISI vehicle in the interspace and general viewing of the environment.

NDE Techniques for Steam Generator (SG)

One of the important and critical components in the secondary circuit of PFBR is the SG. The SG is a shell and tube type; counter current flow heat exchanger with sodium on shell side and water on the tube side. Since sodium and water are chemically reactive, any leakage in the system will result in catastrophic failures. Hence stringent NDT of tubes and tube-to-tube Sheet (TTS) welds during manufacture and subsequent ISI of SG tubes was conducted with modified 9Cr 1Mo ferromagnetic steel as the material. D.C saturation eddy current testing method has been developed in-house for the quality control of 9Cr-1Mo ferromagnetic steel tubes. Micro-focal radiography and replication techniques have been applied for the inspection of



TTS weld joints of the evaporator assembly during manufacturing. Videoimagescopy and digital radiography have been used for the assessment of concavity and convexity of TTS welds. Few of the important NDT techniques for the inspection of steam generator have been described in detail here.

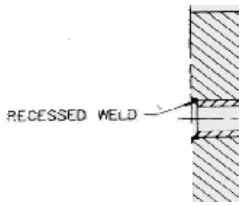
Microfocal Radiography of Tube to Tube Sheet Welds

Typical configurations of the TTS welds are shown in Fig. 2. The first three configurations (a, b, c) involve a fillet weld to the face side which have the following disadvantages and are not desirable.

(i) The existence of crevice between the tube and tubesheet (TS) may lead to failures by crevice corrosion and stress corrosion cracking

(ii) The weld configuration is not amenable for NDT inspection

Due to the typical joint configuration, conventional ultrasonic testing or X-radiography are ruled out due to problems of access. Hence, gamma radiography using a very small thulium source (size 0.1 mm) or micro-focal radiography using a rod anode tube is the only other alternative. Theoretical analysis revealed that gamma radiography is not an appropriate choice leaving only micro-focal



radiography as the possible technique. Special wire-type penetrameters with wire diameters ranging from 120 microns down to 14 microns were used. An ultra-fine-grain, slow-speed film (Kodak DR5 / Agfa D2) was used.

Procedures [1,2] have been developed for the radiography of these welds to achieve a defect detectability level of 50 micron size (porosity) and an overall sensitivity level of 1.4% of the wall thickness. The technique has been successfully applied on the shop floor for the radiography of re-heater TTS weld joints (50 nos), TTS weld joints of SG of SGTF and 590 TTS weld joints of evaporator.

Remote field Eddy Current Technique (RFECT) for ISI of SG Tubes

Conventional saturation method used during fabrication of SG tubes cannot be implemented for ISI due to severe limitations of the geometry for magnetizing and demagnetising. In view of this difficulty, remote field eddy current technique has been developed in-house for ISI of studies. In this technique an exciter and receiver coils are kept separated at a distance in the ID of the tube to be inspected. The electromagnetic fields linking with the receiver coil after diffusing twice through the tube wall is detected. The receiver coil voltage is expected to be proportional to wall loss and less sensitive to lift-off variations. Any wall thinning variations especially uniform wall thinning variations can be efficiently detected with this method. The principle of remote field eddy current and the instrumentation flow chart has been shown in Fig.3. The operating parameters such as number of coil turns, frequency of operation and exciter receiver separation etc are optimised for better defect detection and signal sensitivity using 2D axi-symmetric finite element model and compared with the experimental results. Typical RFECT signals from a calibration a tube consisting of artificial uniform wall thinning grooves has been shown in Fig. 4. Flexible RFEC probe has been developed for negotiation of bend regions of the SG tubes. Advanced signal processing methods such as

wavelet transform method has been developed and applied for suppressing large amplitude signals from the bend regions of the SG tubes due to misalignment in the exciter and receiver locations during probe passage in the bend regions. High density tungsten carbide material has been used for manufacturing probes for easy sliding of the probe during in-service inspection. The permeability influence of tungsten carbide on RFEC signals has been studied and it has been found that addition of tungsten carbide rings in the probe assembly has increased the signal amplitude by 20%. Effect of sodium deposits on the outer wall of tubes has also been studied.

Ultrasonic Technique for ISI of SG Tubes

An ultrasonic based Internal Rotary Inspection System (IRIS), which is available commercially, is considered as a complimentary to RFECT for inspection of SG tubes for measurement of wall thinning and pitting due to corrosion. Working on the principle of ultrasonics, the instrument displays circumferential cross-section (wall thickness) of the tube with an accuracy of 50μm thickness [3]. The advantages of this equipment includes better detection sensitivity of up to 500μm, equal sensitivity to ID and OD defects, circumferential positioning of the defects and detection of defects under support plate.

Ultrasonic Inspection of Weld Joints in sodium circuits and SG units

The coolant sodium is contained in austenitic steel material and most of the system operates at high temperature where high fracture toughness has to be ensured. The pre-service inspection of sodium circuit components and pipe joints for the baseline for the in services inspection to be carried out when the plant is in operation.

ECT of Cladding Tubes

This method has been developed for the inspection of cladding tubes of PFBR having different dimensions. The test frequency and differential probe design parameters have been optimised to detect and evaluate the defects in cladding tubes. An on-line test procedure using artificial neural network has been developed.

Ultrasonic methods have been developed for measurement of grain size in austenitic stainless steels and ferritic steels by judicious selection of ultrasonic transducers. The parameters derived from the frequency spectra, which are relatively insensitive to couplant conditions, are used for grain size measurements [4]. The recrystallisation behaviour in D9 alloy has been characterized using ultrasonic velocity measurements [5]. The influence of Ti/C ratio on recrystallisation kinetics could also be well characterized. Ultrasonic measurements carried out at elevated temperatures (up to 500K) in AISI 316LN stainless steel indicate better reliability for detection of creep-fatigue damage, in comparison to room temperature ultrasonic measurements.

By identifying the small variations in the ultrasonic velocity in the weld, heat affected zone and parent metal regions of ferritic steels, a procedure based on precise ultrasonic velocity measurements has been developed for imaging of 9Cr-1Mo ferritic steel welds [6]. The progress of damage due to thermal ageing and creep in 9Cr-1Mo ferritic steel has been characterized using ultrasonic velocity measurements [7].

Magnetic Barkhausen Emission (MBE) technique has been found to be useful to characterize different microstructures and also to assess the tensile strength in 9Cr-1Mo ferritic steels [8]. It is demonstrated that different zones in the 9Cr-1Mo ferritic steel welds can be identified using MBE parameters [9]. By taking advantage of the dependence of MBE behaviour on microstructures and residual stresses, a methodology has been developed to assess the adequacy of post weld heat treatment for the TTS weld joints of ferritic steels [10]. The ageing induced degradation of 9Cr-1Mo steel leads to change in the size of ferrite and precipitates. Studies have been made to establish correlations between the MBE parameters and size of ferrite and precipitates [11]. The different stages of progress of fatigue damage in 9Cr-1Mo steel could be characterized using MBE technique [12, 13].

Low frequency ultrasonic and impact echo techniques show promise for the assessment of initial quality and in-service degradation of the concrete structures. Ultrasonic velocity

measurements using low frequency ultrasonics / impact echo techniques can be used for the assessment of compressive strength. Further, these techniques can also be used for the detection of any delaminations and crack in the concrete and corrosion and delamination of reinforced rods. Thermography technique also shows promise for studying the degradation in concrete structures, such as change in moisture content, particularly in the near surface regimes.

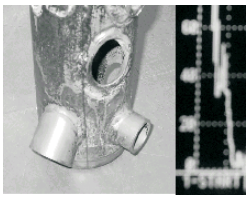
Gamma Radiography for NDT of Pipes

All reprocessing unit operations generally involve nitric acid as the process medium and AISI type 304L stainless steel (SS) as the main material of construction. In these steels, formation of a stable, adherent and self-healing passive chromium oxide (Cr_2O_3) film ensures excellent corrosion resistance. The reliability of the material with respect to corrosion resistance is the key factor, which decides the plant availability with minimum failure and shut down. Still, several incidences of failures of components made of this material have been reported in spent nuclear fuel reprocessing plants. The failure in general, is reported due to (i) intergranular corrosion due to sensitization, (ii) intergranular corrosion due to impurity segregation at grain boundaries, (iii) transpassive dissolution of passive films, and (iv) selective corrosion of welds.

X-ray and gamma-ray techniques are widely used for NDT of components in reprocessing plants. Accuracy in establishing the corrosion rate and its location in the critical areas of pipe has been a problem area in most of the chemical plants including reprocessing units. The wall thickness measurements can be done by tangential radiography without removing the pipe insulation. Many statistical evaluations about the condition of the whole pipe system may be easily performed after some initial measuring periods. Another method is based on radiometric measurement of the density profile of distillation columns and similar components. By this method too the inspection can be done without removal of the outer insulation and even without stopping the process. If process irregularities are observed, the internal structure of the components can be inspected and damages in the internal structure as well as the area of damage can be recognised and located so that a rapid repair is possible. An improved radiographic technique that requires accurately calibrated reference block has been developed. The technique consists of (a) comparing the film densities in the radiographic images of known step wedge and the corroded wall of the pipe and (b) the popularly known "Canadian" technique that uses a lead ball of known diameter as the reference standard. The combination of these techniques modified by application of the calibrated reference blocks is new. It reduces or eliminates "undercutting" and permits obtaining measurements with accuracy between 0.005 and 0.0075 mm. In practical field applications, this method has proved to be effective on pipes of 200 mm outer diameter and with a wall thickness upto 18 mm. For these applications commercially available high strength gamma ray sources of Ir-192 and Co-60 can be used. The method provides the ability to view the results of internal corrosion and to measure wall thickness in pipes accurately [14].

Electrochemical Noise Method for Corrosion Monitoring

An Electrochemical noise analysis method is used to detect localized corrosion such as pitting or SCC, exfoliation, erosion corrosion etc, in storage tanks at an early stage. This method is non-contact and remote and hence can be applied in reprocessing plants with ease. Electrochemical Noise (EN) is the low frequency random fluctuations in current or voltage of electrodes representing the corroding metallic material. During localized corrosion, EN is generated by a combination of stochastic processes, such as passivation breakdown and repassivation events, and deterministic processes which can be caused by film formation or pit/crack propagation processes. The inspection system measures electrochemical noise generated by corrosion in the component by means of electrodes. The time domain EN data is analysed in frequency domain using Maximum Entropy method or statistical methods. It has already been established that different types of corrosion create different patterns of electrochemical noise. Detailed analysis of the EN patterns can identify the mode and the rate of corrosion in a component. This method has been successfully applied to monitor the corrosion damage in the under ground storage tanks of nuclear waste at Hanford site, USA. Acoustic emission



technique can be applied to assess the corrosion damage as a complementary method. Capability for continuous monitoring and high sensitivity makes the EN method a preferred choice over the other methods.

In-service Inspection Techniques for Reprocessing Plant at IGCAR

Typical components used extensively in fast reactor fuel reprocessing plant (FRFRP) at IGCAR include, piping systems, fuel dissolvers, various types of heat exchangers like pot type and thermo siphon type and high active liquid waste storage vaults. The NDT methods being developed for the ISI of a few critical components are presented.

Fuel Dissolver Vessel Inspection

Immersion Ultrasonic Testing

The fuel dissolver vessel is a complex and intricate geometry with many inlets and outlets for the process fluid and the spent fuel as can be seen from Fig. 5. Highly corrosive nitric acid is used as solvent in such dissolvers. In view of this it is important to assess the wall thinning due to corrosion. Immersion ultrasonic testing is a suitable technique for the assessment of wall thinning. During testing the vessel is filled with water and this water column is used as the coupling medium between wall and transducer. Normal beam ultrasonic testing is performed with a waterproof probe operated at 20 MHz to evaluate the wall thinning. In this method the distance between the echo signals from inner and outer walls of the vessel is measured. The time difference between these echoes reduces with wall thinning. Fig. 5 also shows typical ultrasonic signals obtained during examination of the mock-up dissolver vessel. The accuracy obtained in measuring the wall thickness by this method is 0.1mm. The entire inspection has to be performed remotely with the help of a 2-axis manipulator. Manipulator should be centered first and is to be provided with special drive mechanism to carry the probe head to different elevations and scan circumferentially to obtain data.

Laser triangulation method for surface profiling of the Dissolver

Laser triangulation technique can be used as a complementary NDT technique for immersion ultrasonic testing. By this method corrosion damages inside the dissolver vessel can be detected. A schematic diagram of the laser triangulation method being developed is shown in Fig.6. In this method, a diode laser is used to generate a collimated beam of light to be projected on to the testing surface. The reflected light will be focussed to a photodetector with the help of an imaging lens. The photodetector generates a signal that is proportional to the spot position on its image plane. As the target surface height changes this imaged spot shifts due to parallax. A two dimensional scanning of the surface with this setup will give data, which represents the surface topography of the scanned part. With specialised software it is possible to construct a 3D image of the surface. The surface profiling accuracy

with this method has been found to be 100 microns. However the instrument should withstand radiation levels as high as 50 Sv/h.

Ultrasonic Guided Wave Inspection of the Tube Wall Thinning in Evaporator Tubes

NDT methods such as ECT and IRIS cannot be used for the inspection of evaporators due to higher radiation levels present and restricted access. To overcome the difficulty of going near the instrument, a guided ultrasonic wave inspection can be used. Wall thinning of the evaporator tubes of the order of 10 microns can be determined with this method and also fast and reliable inspection is possible. Guided wave technique is a remote and non-contact technique and hence suitable to this particular component. In this method, cylindrical guided ultrasonic waves are generated with Electro Magnetic Accoustic Transducer (EMATs) and passed through the tube wall. Various models of guided Lamb waves will be generated in the tube wall. The reflected wave amplitude and the information of the wave mode due to wall thinning and defects will give information on the extent and location of the defects.

Waste Vault Inspection

Various radioactive wastes originating from irradiated fuel reprocessing operation are collected in underground waste storage tanks from where it will be taken to waste immobilisation plant. It is necessary to use radiation resistant camera and fibre optic visual inspection systems. A multilink manipulator with pan and tilt mechanism for positioning and orienting the camera in space is being developed for the inspection of waste vaults of the fast reactor fuel reprocessing plants at IGCAR. The camera will provide the global and zoomed local view of the tanks and associated piping. Online corrosion monitoring probes based on measurements of electrical resistance, linear polarization resistance and electrochemical noise are also available for application in nitric acid service.

Tomography for Nuclear Waste Containers

Conventional radiograph is a 2-D image. Defects that are situated behind each other are superimposed in the image so that a three-dimensional volume is projected in two dimensions. To obtain a 3-D image, a number of images from different angles have to be integrated and this is termed as tomography. An online tomography system has been developed elsewhere employing two types of detectors [16] for the detection of voids, cracks and to locate the actual position of waste in concrete containers. The main limitation of this technique is the detection of liquids and the chemical characterisation of materials. However, by using low and high energy tomographic acquisition, objects of low attenuation i.e. organic materials, mineral and metals can be imaged and differentiated. This aspect has been used in the case of light containers containing low activity nuclear waste, to estimate precisely the actual activity contained in the container for identifying its processing. Similarly, it is required to ensure that there is no massive metallic part or forbidden items such as aerosol bombs inside the container.

Conclusion

A variety of advanced NDE techniques and their applications have been developed indigenously to meet the challenges not only in the PSI stage of critical nuclear components / structures, but also during ISI stage, to ensure safe and reliable operation. Judicious selection of a combination of NDT&E techniques is essential to ensure the ultimate objective of high level of plant availability and safety required in this crucial advanced technology. Robotics and automation also play a crucial role in usage of NDE for nuclear technology not only from the point of view of limited accessibility, speed and accuracy but also due to the hazardous nuclear radiation environments necessitating remote inspection.

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IANCAS Roundup

Shri Surendra Kumar *obtained his post graduate degree in Physical Chemistry from Meerut University, U.P. He joined Radiations and Isotopic Tracers Laboratory (R.I.T.L) of G.B.Pant University of Ag. & Technology, Pant Nagar 263145 (Uttaranchal) in Dec.1976. He has been associated with the development and planning of R.I.T.L since his joining. R.I.T.L is a duly approved B-class Radiochemistry laboratory and central facility of the University for radiotracers & radiations teaching and research. He has also been associated with the multidisciplinary radioisotopic research works carried out in Pantnagar. He has been teaching different P.G. courses related to radioisotopes, radiations & nuclear techniques etc for several years. His research interests are NAA, adsorption, remediation & kinetics studies. He has worked as convener & co-convener in the organization of a number of training workshops, summer schools & symposia.*

REPORT ABOUT THE USE OF IANCAS DONATED INSTRUMENTS

The nuclear instruments namely the G.M. counter & Gamma Ray spectrometer donated to R.I.T.L. in April 2000 have widely & regularly used in our laboratory (Radiations & Isotopic Tracers Laboratory), which functions as a central research facility for the whole university. These are being used here in the following activities;

(a) Teaching

In four P.G. courses namely (i) Radiochemistry (ii) Use of radioisotopes in research (iii) Radio-tracer techniques in Mol. Biology (iv) Radio-Isotopic techniques in Mechanical Engineering . These are offered by the laboratory to the P. G. (M.Sc. & Ph.D.) students of various departments whose research problems require use of radioisotopes / radiations. Besides, these are also used in other several inter-departmental collaborative courses of the university.

The List of experiments in which the instruments have used:

1. Determination of counter plateau, operating voltage, counter efficiency, step-factor, G.M. counter.
2. Study of statistics of counting and determination of counter dead time.
3. Determination of half-life of Phosphorus-32.
4. Study of the absorption characteristics of beta and gamma radiation by Aluminium and HVL determination.
5. Study of the back-scattering effect of beta rays.
6. Study of the self-absorption effect of beta rays.
7. Counting of ^{32}P-labelled plant samples by GM counting.
8. Counting of gamma emitters Zn-65, Cs-137 by NaI (T1) solid crystal scintillation counting assembly.
9. Gamma ray spectrometry of Cs-137 and Fe-59.
10. Study of the incorporation of Zn-65 and P-32 in plants.

(b) Research

The P.G. research scholars & scientists of different disciplines namely Agriculture, Veterinary, Basic Sciences & Biotechnology have been using these instruments for their research works. The research works carried out in last two years are;

1. Elucidation of causes and mechanism of Iron chlorosis in sugarcane –a NATP project. In this project radio- tracer work done using Fe-55 and Fe-59 isotopes.
2. Effect of weed's extracts on the uptake and transport of Zn-65 and P-32 in Wheat and Paddy.
3. Studies on feeding efficacy of coccinella septempunctata on lipaphis erysimi and their prey-predator relationship using radiotracer technique.
4. Effects of Nickel levels on phosphorylation and photosynthetic rate in rapeseed Brassica campestris var. Toria (PT-303)

(c) Training

The instruments have also used in the national training course on "Use of nuclear techniques in increasing agricultural productivity" in July-August2001 financed by B.R.N.S. for scientists/research scholars of agricultural universities in collaboration of Nuclear Agriculture and Biotechnology Division of B.A.R.C.

(d) Extension

The instruments have been used in scientific exhibitions /demonstrations to U.G. and School level students during Science Week Celebrations in February-2001,2002 &2003 in the college to familiarize them with Radioactivity Measurement And Radioisotopes Applications.

NUCLEUS

Prussian Blue: A counter measure to terrorist attacks involving a dirty bomb

Cesium-137 is one of the major fission products present in various liquid waste streams of nuclear facilities. It is also used as a source of radiation in blood irradiators and for cancer treatment. It is found in the fall out from the detonation of nuclear weapons. In addition to concerns about accidental industrial and medical exposure, Cs-137 is of particular concern because it is a potential component of a conventional explosive device (a dirty bomb) containing radioactive material. Although this radiological dispersal device is not a nuclear bomb, it is detonated as a means to spread radioactive material. Salient radiological features of Cs-137 are:

1. *Easy absorption by the body through different routes (ingestion, inhalation and skin penetration).*
2. *Relatively long biological half-life in humans of about 100 – 110 days.*
3. *Relatively long physical half-life is about 30 years.*
4. *Emission of beta and gamma radiations (660 keV).*
5. *(E_{mea} = 0.5/1.2 MeV) uniform distribution throughout the body due to its proximity to Potassium (K).*

As a part of the efforts of U.S. Food and Drug Administration (FDA) to foster the development and availability of drug products for treatment of people accidentally exposed to radioactive materials, it is encouraging the pharmaceutical companies to submit New Drug Applications for Prussian Blue (PB), which has been used as a pigment in industry since 1704 and it's chemical name is Ferric hexa cyano ferrate. FDA wants doctors to know that Prussian Blue is the first therapy that would be available to immediately help reduce the body's burden of exposure to Cesium-137. Long range objective is to ensure that PB consistently meets high standards of quality and proper instructions for use are available with the product.

The principle pathway of Cs excretion is through glomerular filtration in kidneys. Due to it's reabsorption in the intestine, Cs also gets cleared partly through faecal excretion and it is facilitated after it's chelation by PB. However, this compound is known to induce gastro-intestinal and cardio toxicity. There is global interest in developing alternate efficient chelating agents for Cs-137 which are free of these side effects.