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AHWR Simulator Facility in Integrated Test Station in BARC Trombay

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Engineering Simulator for Advanced Heavy Water Reactor

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Abstract

An engineering simulator for the Advanced Heavy Water Reactor (AHWR) [1] has been developed, installed and commissioned at the AHWR Integrated Test Station (ITS), RCnD, BARC, Trombay. This is a computer-based real time simulator employing mathematical models of coupled core neutronics-thermal hydraulics and all major plant systems of AHWR. The developed system is capable of simulating the dynamic response of the plant under both normal and off-normal operating conditions, and is being used in verification and validation of plant control algorithms and safety interlocks, hardware in the loop testing of prototype Control and Instrumentation (C&I) systems of AHWR, design of control room interfaces etc. Important features of this engineering simulator and results of some of the transient simulation studies.

Keywords: AHWR, Modeling, Simulation, Simulator, HIL, V&V

Introduction

Traditionally, Nuclear Power Plant simulators are widely used for operator training and licensing. With the requirements of physical and functional fidelity [2], these Full Scope Simulators (FSS) are dependent on the process design and operational data of reference plant, which is generally made available during the final stages of plant development cycle. Another category of simulators known as Nuclear Power Plant Analyzers (NPAs), typically consisting of detailed models of only nuclear and steam supply systems are used for design and safety studies. However, in the past few decades, availability of inexpensive hardware with advanced computational capabilities and improved knowledge on process modeling has led to a paradigm shift in the way simulators are built and used. Especially, use of another category of simulators called Engineering Simulators in various design phases of the plant proved to have a huge positive impact on the quality of C&I design and plant commissioning time. Unlike FSS, these simulators employ models of only major processes, instrumentation, control and protection systems, with no stringent requirement on the control room exactness. With the inclusion of auxiliary plant models and their integration with the replica control room, engineering simulators can further be extended to FSS for training and licensing purposes.

The Advanced Heavy Water Reactor (AHWR) engineering simulator in BARC would cater to the following key objectives: verification and validation (V&V) of control algorithms and safety interlocks, hardware in loop (HIL) response testing of prototype C&I systems, validation of performance and interfaces of control room and human factor engineering (HFE), virtual plant commissioning etc.

This is a computer based real time simulator capable of simulating dynamic response of the plant under both normal and off-normal operating conditions. It employs a coupled multipoint kinetics - thermal hydraulics model of the core, along with models of all major process, instrumentation, electrical, control and protection systems. It also has a hardware input-output (IO) system for interfacing with prototype controller hardware, so as to facilitate HIL response testing. **Fig.1** shows the AHWR Simulator facility installed in Integrated Test Station (ITS) in Reactor Control Division, BARC, Trombay. This simulator has been developed using an indigenous simulation tool (ECTools), which is an integrated platform for development, configuration and runtime control of simulators.

Simulator Architecture

The AHWR engineering simulator follows an architecture consisting of a simulation host (server), supervisory station, operator stations and IO system, as depicted in **Fig. 2**. Plant models are executed on real time basis in the simulation host. Provision to switch off or adjust the cycle time of individual system models is available in the simulation server. The Supervisory station serves the purpose of runtime control of

Fig.1. AHWR Simulator Facility in Integrated Test Station at RCnD, BARC, Trombay

simulation execution, including start/stop of the simulation, selection of initial conditions, introducing equipment/sensor/system malfunctions during runtime etc., as specified in [2]. Operator Stations consist of soft consoles used for operator interface and display of various simulated parameters. The Hardware IO system consists of configurable IO boards which are controlled by a local computer that communicates with the simulation server over the Ethernet link. It serves as an interface between simulator and external control system hardware besides providing a platform for HIL testing. The Control algorithms executed in simulation server during standalone simulations are replaced by those residing in external hardware during HIL testing, as shown in **Fig. 3**.

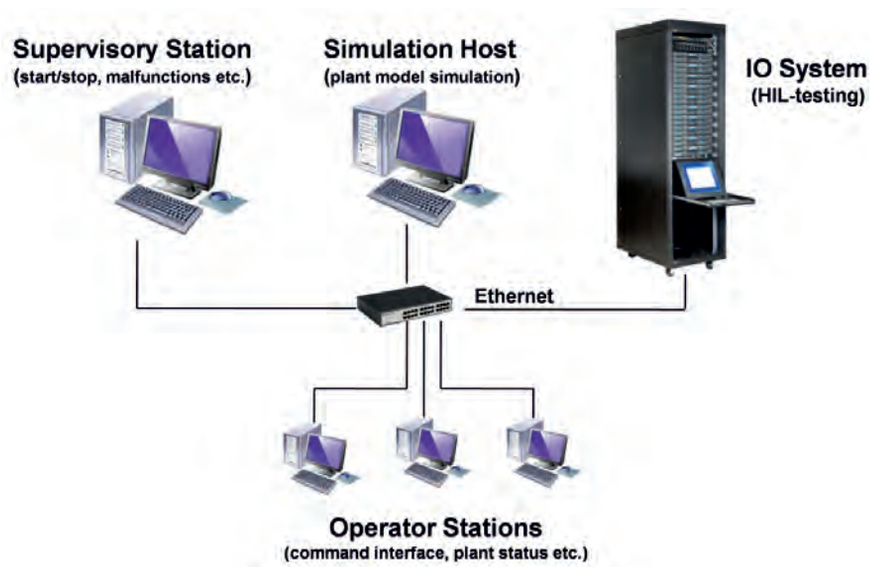

Fig.2. AHWR Simulator Architecture

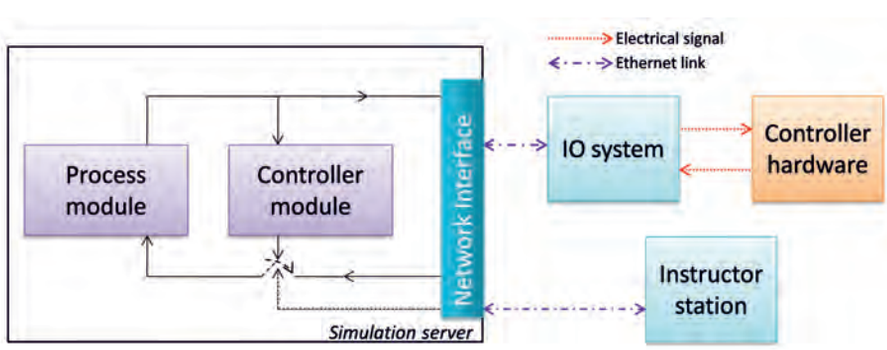

Fig.3. Interfacing external hardware with Simulator for HIL testing

Mathematical Models

The AHWR simulator employs integrated models of all major

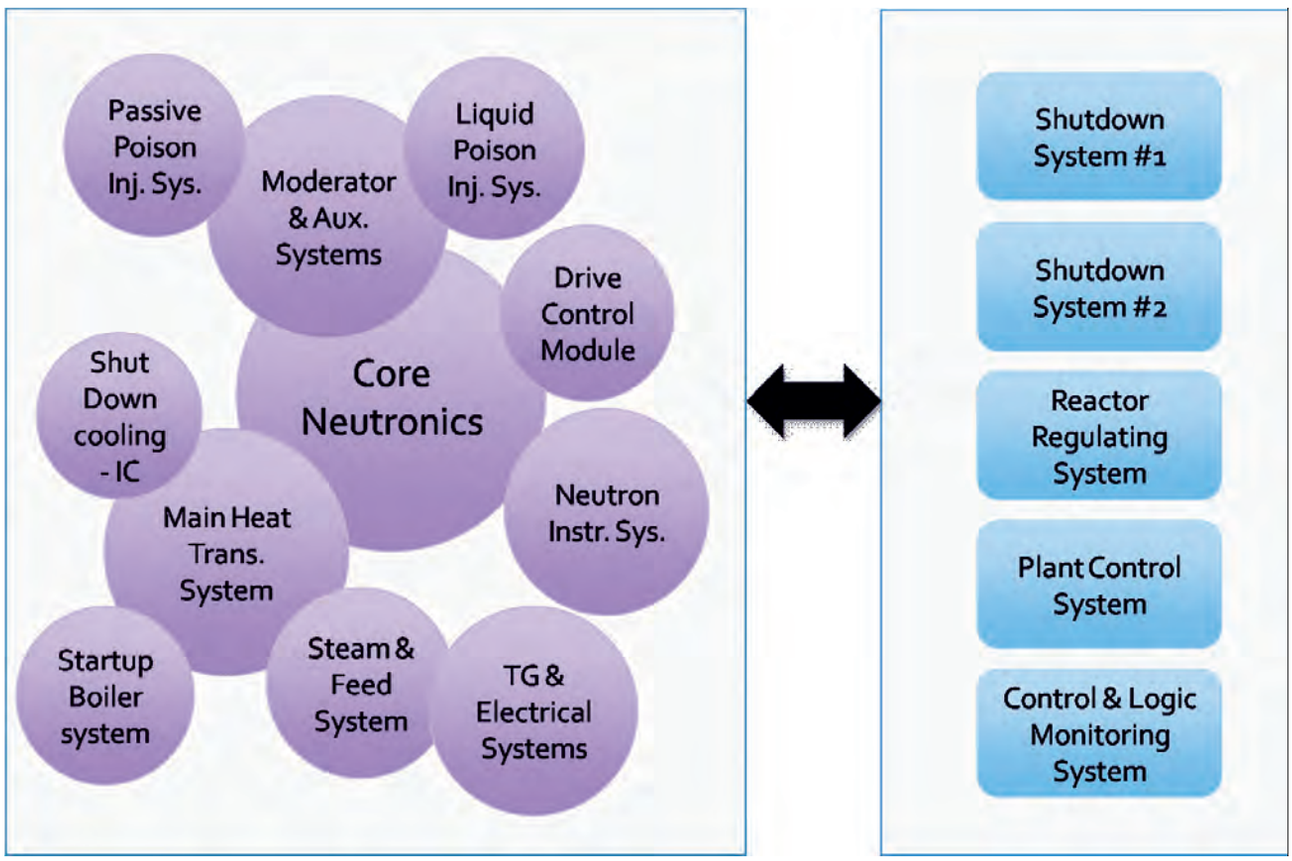

Fig.4. Process, control and protection system models in the simulator

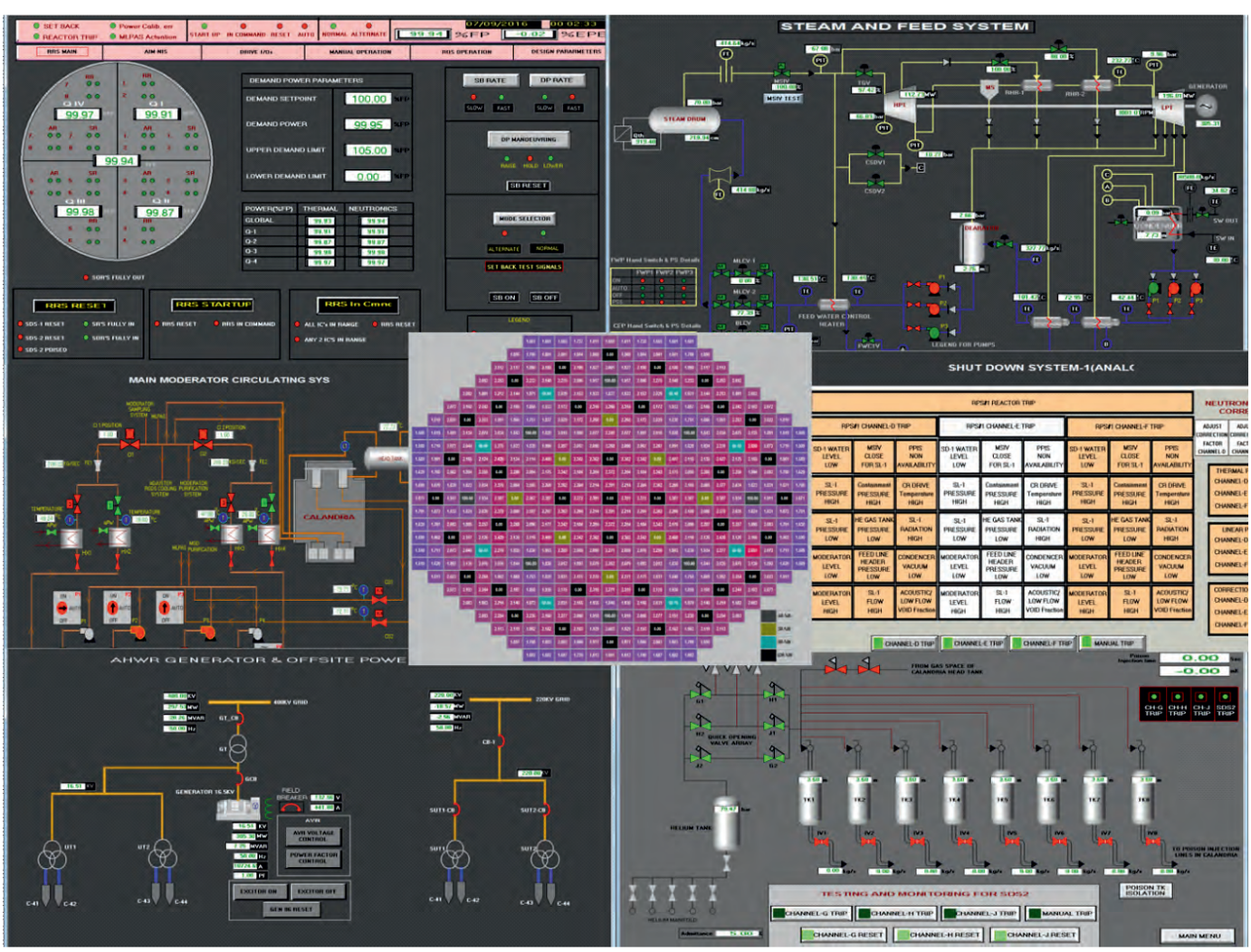

Fig.5. HMI screens of AHWR Engineering Simulator

processes, control and protection systems, as depicted in Fig.4. Models for specialized systems like core neutronics [3], thermal hydraulics [4]-[5] etc., are developed from first principles and validated using benchmark data from detailed design codes. On the other hand, process systems with generic components (like steam and feed system, moderator circulation system, electrical systems etc.,) are modeled using the simulation tool which offers a graphical editor with validated component libraries for model development and automatic code generation [6]. Complete algorithms and logics of control systems like Reactor Regulating System (RRS), Overall Plant Control System (OPCS), Control and Logic Monitoring System (CLMS), and protection systems including Shutdown Systems 1and 2 are also coded in the simulator [7].

Operator Interface

The Simulator provides a number of operator stations consisting of soft consoles for operator interface and display of various simulated parameters. Fig.5 shows a collage of some of the HMI screens developed for RRS, Steam and Feed system, 2-D Core map, Electrical System, Moderator Circulating System, Liquid Poison Injection System (LPIS), SDS#1 etc. of simulator.

Capabilities of Simulator

The AHWR Simulator is capable of simulating transients related to normal and off-normal operating conditions that are postulated for AHWR to enable validation of control logics and interlocks of safety and safety related systems of AHWR. It covers Category-I and Category-II Design Basis Events (DBEs) as per SG D-5 [8], pertaining to power transient as listed in 'Preliminary Safety Analysis Report–Part B' of Advanced Heavy Water Reactor [7]. Simulation studies of various normal operating transients as well as off-normal transients involving Postulated Initiating Events (PIEs) have already been carried out [6]. The Simulator has also been used to generate transient scenarios for dynamic probabilistic safety analysis studies. **Fig.6** shows simulated response of the plant parameters to an

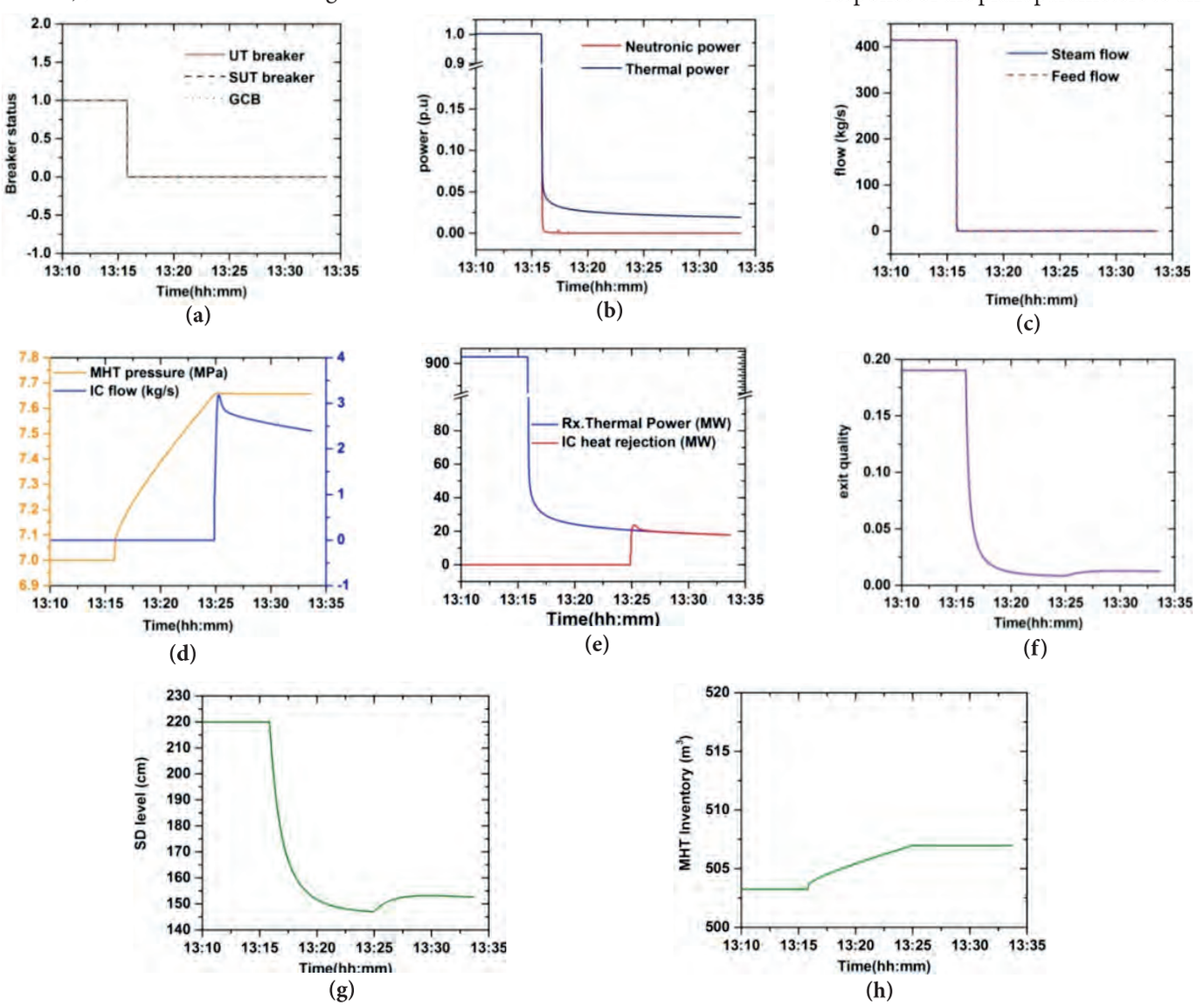

Fig.6. Variation in plant parameters for Class-IV power supply failure with availability of passive Isolation Condenser (IC) valve

off-normal operational transient involving class-IV power supply failure. Under class-IV failure, secondary system becomes unavailable for decay heat removal, and decay heat is passively removed through Isolation Condensers submerged in a Gravity Driven Water Pool. To investigate this scenario, simulator is initialized at 100% FP with all its modules running. After steady state operation of around 5 minutes, malfunction is invoked from supervisory station to simultaneously trip Unit Transformer (UT) breaker, Start-Up Transformer (SUT) breaker and Generator Circuit Breaker (GCB) breaker.

Opening of the circuit breakers (**Fig.6 (a)**) resulted in loss of voltage to the buses feeding feed water pumps and other class-IV loads. On sensing simultaneous unavailability of all the three feed water pumps, SDS#1 initiates reactor trip (**Fig.6 (b)**) and MSIVs are closed to prevent loss of inventory. This results in complete loss of steam and feed flows (**Fig.6 (c)**) and unavailability of heat removal path. The MHT pressure (**Fig.6 (d)**) starts increasing gradually due to the decay heat present in the core. After 8 minutes, MHT pressure reaches the set point for opening of passive IC valve. Once the passive valve opens partially, natural circulation flow (**Fig.6 (d)**) is established in IC system, which removes the decay heat generated from the system (**Fig.6 (e)**). The MHT pressure remains slightly above 7.65 MPa and valve opening is regulated automatically based on the MHT pressure. Exit quality (**Fig. 6(f)**) reduces continuously after reactor trip resulting in decrease in steam drum level (**Fig.6 (g)**) due to void collapse. Main Heat Transport (MHT) inventory (**Fig.6 (h)**) increases till opening of IC valve due to increased MHT pressure which converts steam into water. Once IC flow is established, MHT inventory remains steady.

Conclusion

An Engineering Simulator for AHWR has been developed to facilitate simulation based verification and validation of plant and C&I design during the development cycle. It is a computer based real time simulator consisting of all the major plant systems and is capable of simulating normal and off-normal operating transients. Simulation of a large number of normal operating transients and off-normal transients involving Postulated Initiating Events (PIEs) has already been carried out using this facility.

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Development of Active Neutron Interrogation Technique for Non-Destructive Assay of Hull Waste

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Abstract

Detection and quantitative estimation of nuclear materials is one of the most vital issues in management and safeguards in nuclear fuel cycle. Improving the sensitivity of detection is vital for accurate accounting and management of special nuclear materials (SNM) as well as to minimize loss of nuclear materials by monitoring residual quantities of fissile material in leached hull. Because of the limitations of passive gamma and neutron based methods for such an accurate accounting applications, active neutron interrogation technique has been developed to improve sensitivity and minimum detection limit. In this paper, we discuss one particular form of active neutron interrogation namely Differential Die Away (DDA) technique which is one of the most sensitive methods for detection of fissile material not only in hull but also in cargo containers for security application. We present experimental result of detection of plutonium in a simulated hull. The sensitivity of detection with the current experimental set-up used by us is about 300 mg of Pu in 150 kg of Zircaloy matrix in SS drum in the presence of neutron background of 10^4 n/s and an effective gamma dose of 30 mR/hr at the detector.

Keywords: Neutrons, Active interrogation, Differential Die Away (DDA)

Introduction

An early step in fuel reprocessing is the mechanical separation (shearing) of the incoming fuel assemblies and the dissolution of the spent fuel contained within the metal fuel assembly rod pieces. The shearing process is typically done with a large mechanical chopper which cuts the fuel assembly rods into short lengths of a few inches or less. The dissolution process is typically carried out using strong nitric acid (or electrochemically). When process is complete, the dissolved spent fuel/nitric acid solution proceeds to the radiochemical separation process. The metal fuel rod casings, or hulls, which do not dissolve in the acid are dried, melted and cast into waste ingots for permanent disposal. However, there is a possibility that not all of the spent fuel has been removed from these hulls and safeguards measurement must be made to quantify the residual Uranium/Plutonium in the hulls. Typically spent fuel hulls are assayed by sampling a small lot of the inventory of chopped hulls or by passive techniques. In contrast to this approach, active neutron interrogation techniques have been developed to perform complete assay of the entire inventory of leached hulls non-destructively. There are two primary approaches for detecting plutonium in hull matrix (a) passive detection and (b) active interrogation.

Passive techniques

Passive non-destructive detection is based on the detection of naturally emitted gamma rays and/or neutrons from fissile material. Because of the absence of any interrogating radiation source, the passive techniques are much simpler to deploy.

Passive Gamma Ray Detection

As a result of the spontaneous α or β decay in heavy nuclei, γ-rays are emitted during the de-excitation process. These γ-rays have energies that are characteristic of the nuclei from which they originate. However, the passive gamma techniques are often inaccurate as these measurements get contaminated due to interference from the intense gamma rays from other fission and activation products. Moreover, gamma radiation from the natural decay of ^{235}U and ^{239}Pu is relatively soft and effectively attenuated by a few centimeters of shielding such as lead or iron.

Passive Neutron Detection

Neutrons are emitted as a result of spontaneous fission or from the (α, n) reaction. These neutrons typically have high energies and can penetrate through thick shielding with a high probability. Passive neutron counting may be used for monitoring of wastes but this measurement can only quantify the total spontaneous fission neutron emission rate (due to

Fig.1. Experimental set-up of Active Neutron Interrogation Technique at Purnima Labs in BARC Trombay

emissions from even mass nuclides such as ^{238}U, ^{240}Pu and ^{244}Cm). To quantify specific transuranic nuclides or fissile isotopes (such as ^{235}U or ^{239}Pu) one needs to correlate it with fuel burn up and fuel composition. The assay is complicated by the fact that additional neutrons are generated due to (α, n) reaction resulting from the alpha activity of actinides present in the waste and their interaction with the light element such as fluorine and oxygen which are also present in the matrix. Moreover, for relatively low burn up fuel, spontaneous fission neutron emission of even isotopes of Pu is typically about 1000 fissions/sec/gram. With about 10^{-3} efficiency of fission to thermal neutron detection, isotopic abundance of such isotopes in the range of 8-20% and presence of large background neutrons from (α, n), simple gross counting methods is likely to give highly erroneous result unless the limit of detection is raised to quite high value. Though coincidence and/or multiplicity methods improve the signal to noise ratio, it results in either very large counting time or worsening of the detection limit. If advanced fuel based on LEU is used then presence of residual ^{235}U in hull will be substantial, and therefore accurate accounting of ^{235}U is not possible just by using passive neutron methods. Clearly, if ^{235}U or ^{239}Pu is to be reliably identified by non destructive assay (NDA) method, it must be induced to fission by external neutrons.

Active Neutron Interrogation Technique

Active neutron interrogation technique is based on differential die-away analysis (DDA)[1-3] and works by exposing a medium to pulses of neutrons. An external fast neutron detector measures the time-dependent decay of neutrons from the matrix. When no fissile material is present in the inspected medium (e.g. hull) the detector should only measure a signal representing the diffusion of the thermalized neutrons in the detector body (detector neutron "die-away" time). If fissile material is present, the detector will show, in addition to a signal decaying with the detector die-away time, an additional signal decaying with the die-away time of the inspected medium. If the latter is significantly longer than the former it will dominate the decay curve at later times and unequivocally establish the presence of fissile material in the inspected object. The advantage of active neutron interrogation technique is that it can detect very small quantities of fissile materials directly even in presence of high neutron and gamma background and is much more sensitive compared to passive techniques which are indirect measurement of fissile material with large error margins.

Experimental set-up

An experimental arrangement (**Fig.1**) to investigate the potential of this technique has been setup at Purnima Labs, BARC. Dry Zircaloy pieces weighing approximately 150 kg were placed in an SS drum of diameter 900 mm and height 1000 mm to simulate the cladding materials in hull. This assembly was surrounded by a bank of epithermal neutron detectors (^{3}He detector enclosed in cadmium). A 14 MeV neutron generator was used as external

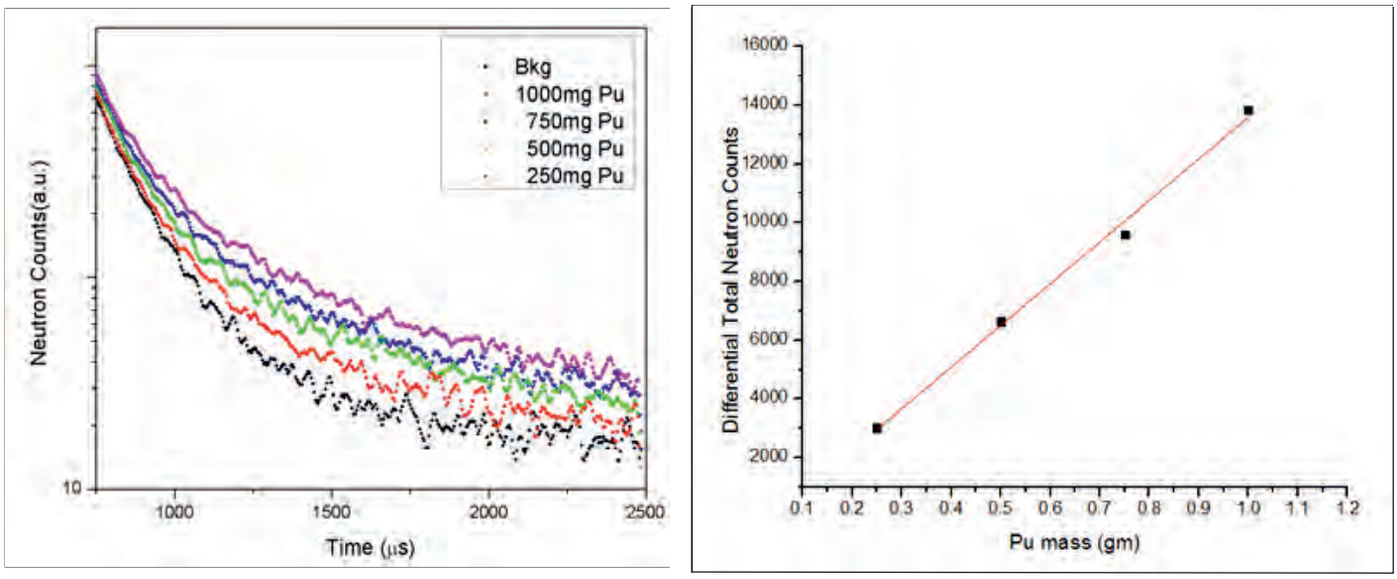

Fig.2. Plot of detector counts as a function of time and fissile material contents

neutron source for inducing fission. The drum assembly was surrounded by graphite to increase the die-away time of thermal neutron. Experiments to simulate various conditions as encountered in a hull matrix were carried out and are described in following sections.

Experimental Results

An experiment was conducted in clean environment and varying amount of Pu was kept within the hull matrix. The data was collected by operating the neutron generator in pulsed mode at a frequency of 400 Hz. The neutron counts detected were plotted as a function of time (**Fig.2**). The result clearly shows that up to 250 mg of Pu could be easily detected in 150kg hull under clean conditions. Figure 3 shows plot of total neutron counts (background corrected) as function of Pu mass. The plot shows that there exists a linear relationship between the two and hence this can be used as a calibration index for estimating precise quantity of leftover material in hull matrix. Moreover, as can be seen from the plot in **Fig.2** that even for 250 mg of Pu, integral neutron counts is well above the background levels thereby indicating that even smaller quantities of Pu could be distinguished by properly choosing the counting gate. The counting gate interval may be varied to get improved signal to noise ratio depending upon the matrix and other conditions.

Effect of Passive Background

The average burn-up of spent fuel from PHWR is typically around 6000-7000MWd/T. During the process of separation of fissile material from rest of actinides, around 1% to 4% of material may be left depending upon the process conditions. Assuming a cooling period of about 3-5 years and 1% loss, the neutron background of the hull matrix could be of the order of 10^4 n/s. Apart from these neutron signal, the background gamma radiation at the detector (after few HVL of lead/concrete shielding) may be around 20-30mR/hr. Experiments were carried out to find out minimum detectable quantity of fissile material under these conditions which may typically be encountered in actual hull monitoring conditions in nuclear plants. Experiments were carried by placing 0.30g to 1g of ^{239}Pu at the centre of the matrix. Active neutron (10^4n/s), gamma background (30mR/hr) and moisture conditions were simulated by keeping 5mCi Am-Be neutron source, 100mCi Cs-137 gamma source and water (60g), which represent the background radiation and moisture levels typically encountered in hulls. **Fig. 3** shows the plot of neutron counts under passive gamma and neutron background. It can be seen that we are able to detect up to 300mg of Pu under these stringent conditions, thereby making it very robust technique for applications involving monitoring of spent fuels, high activity level waste or even monitoring of small quantities of Pu in low activity waste assay.

Compact Neutron generator development

A compact multipurpose table top neutron generator (**Fig.4**) has been developed as source driver for DDA. It has been designed using self-focusing triode electrode geometry. As shown in **Fig. 4,** the generator consists of a RF ion source operated ~200 watts at 13.56 MHz operating frequency, a suppressor metallic shroud and an oil cooled target electrode that can accommodate solid deuterium/ tritium neutron producing targets. These targets are fabricated from titanium-coated copper loaded with either deuterium or tritium. The RF power is inductively coupled to excite the deuterium gas inside the ceramic housing of the ion source. Deuterium gas stored in a gas cylinder is inserted in the ion chamber through precision

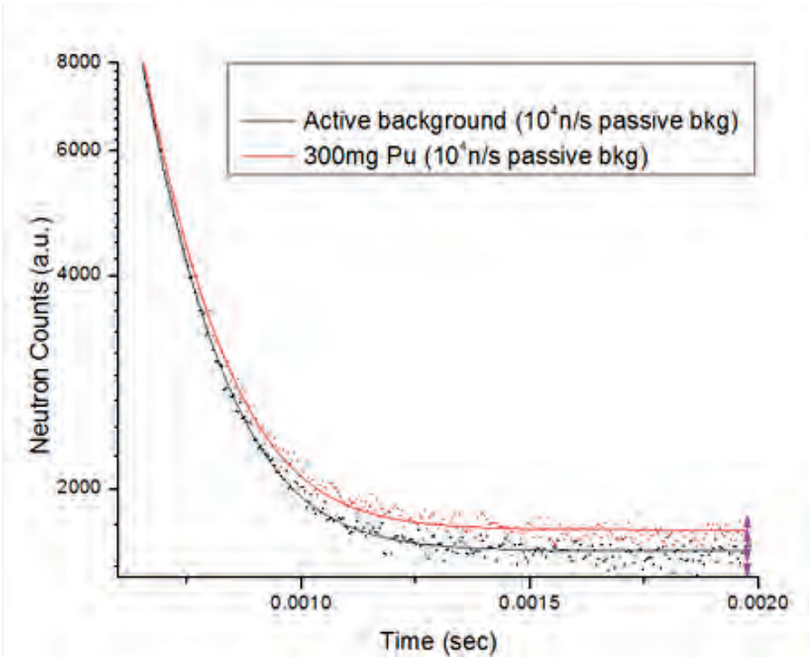

Fig.3. Neutron counts in presence of gamma and neutron background

gas dosi-valve. The deuterium ion beam is extracted through an iris at ground potential. The beam is extracted by applying high voltage at the extraction electrode at the rear side of the ion source. In this generator the target electrode is at a high negative potential (–100 kV), while the ion source is at ground. The deuterium ions (D+) are accelerated from the ion source's extraction iris to the target where the fusion reaction occurs. The shroud electrode is biased at slightly more negative potential (-102kV) than target electrode. Thus shroud electrode prevents the back-streaming electrons into the ion source. The generator is housed in a cylindrical metallic housing of 200 mm diameter with an overall length of 600 mm.

Conclusion

Active neutron interrogation technique using pulsed neutron generator has been developed at Purnima Labs. This technique is being used world-wide for improving sensitivity of fissile material detection for security as well as in nuclear industry for nuclear material accounting. The advantage of this method is that it can detect very small quantities of fissile materials even in presence of high neutron and gamma background and it is much more sensitive compared to passive techniques. An experimental facility comprising of pulsed neutron source and ^{3}He detectors has been set-up to verify the detection limits using this approach especially for the case of leached hull-monitoring. It has been shown that even under stringent conditions of neutron and gamma background, upto 300 mg of Pu could be easily detected in a 150 kg hull.

The tools and technique developed for hull monitoring can be very easily adopted for detection of special nuclear material (SNM) in a cargo or container. The generator developed has the additional capability of neutron tagging, which can give 3D location of explosive and narcotics using DC mode operation besides being switched to pulsed mode operation for SNM detection.

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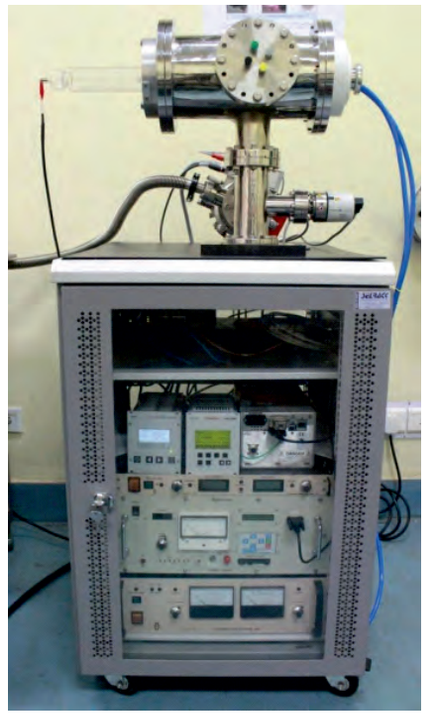

Fig.4. Trolley-mounted Neutron Generator

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A Brief Review on Identification of Irradiated Foods Using Physical Methods - Thermoluminescence and Electron Paramagnetic Resonance Spectroscopy

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Abstract

Food irradiation is now legally accepted in majority of the countries. The need for reliable and routine tests to determine whether or not food has been irradiated has arisen as a result of the progress made in commercialization of the food irradiation technology and increased international trade in irradiated foods. The availability of reliable identification methods would be of assistance in establishing a system of legislative control, and to help achieve acceptance of irradiated food by consumers. Identification of irradiated food is an extremely challenging task as negligible changes in irradiated food matrix are required to be detected. No single method has so far been established to deal with all types of foods. The present review discusses the efforts made in BARC to explore the use of electron paramagnetic resonance (EPR) spectroscopy and thermoluminescence (TL) measurements to identify several irradiated foods and allied products.

Keywords: Food Irradiation, Identification, EPR, TL

Introduction

Food irradiation is the treatment of food by ionizing radiation. The process involves exposing food, packaged or in bulk, to carefully controlled amounts of ionizing radiation for a specific time to achieve certain desirable objectives. In **1980**, a Joint Expert Committee of Food and Agriculture Organization/International Atomic Energy Agency/World Health Organization on Food Irradiation (JECFI)[1] concluded that foods treated with controlled quantity of radiation dose are toxicologically safe for consumption. Consequently, food irradiation is now legally accepted in more than 55 countries. The need for reliable and routine tests to determine whether or not food has been irradiated has arisen as a result of the progress made in commercialization of the food irradiation technology, increased international trade in irradiated foods, differing regulations relating to the use of the technology in many countries, and consumer demand for clear labelling of the treated food. The availability of reliable identification methods would be of assistance in establishing a system of legislative control, and help achieve acceptance of irradiated foods by consumers. Prior to **1980**s little progress had been made in the development of reliable detection methods for irradiated foods. In the last decade, significant progress was made in irradiation detection. In December **1996**, the European Committee for Standardization (CEN) adopted five European standards for detection of irradiation process in food commodities. The methods used in detection of irradiated foods are based on physical, chemical, biological and microbiological changes in food products during irradiation, although these changes are minimal. Ideally, detection methods should have the following major criteria to be accepted as a standard technique such as: 1. changes induced by radiation should be distinct and separable from non-irradiated food and specific to radiation processing, and 2. applicable through the dose range relevant to the food tested.

Identification of irradiated food is an extremely challenging task because negligible changes in irradiated food matrix are required to be detected. No single method has so far been established to take care all types of foods.

The present review discusses the efforts made in BARC to explore the use of electron paramagnetic resonance (EPR) spectroscopy and thermo luminescence (TL) measurements to identify several irradiated foods and allied products.

Identification of Irradiated Food Using EPR Spectroscopy

Application of EPR spectroscopy is based on the identification of a long-lived paramagnetic-centre produced by irradiation in the organic and inorganic component such as free radicals or inorganic complexes possessing a transition metal ion. The technique is non-destructive, specific, highly sensitive and rapid.

Free radicals trapped in a solid matrix of foods exhibit broad, poorly resolved, EPR signals which do not allow for chemical assignments. This analytical approach requires the

Fig. 1. a) Experimental set-up of the EPR spectrometer, (b) Experimental setup for TL measurements and the aluminium discs with isolated minerals from food samples for TL study

analysis of control(non-irradiated) samples because of variations in the background radical content. There are several reports in literature where spices, fruits and other food commodities have been identified after radiation treatment using EPR spectroscopy. EPR measurements were performed using X-band, BRUKER EMX spectrometer (BRUKER, Germany). All the spectra were recorded at ambient temperature (25±2°C) of EPR laboratory after 3 or 4 accumulated scans. Operating conditions of the EPR spectrometer in general were standardized as centre field 348 mT, scan range 20 mT for narrow scan and 100 mT for wide scan, microwave power 0.253 mW, microwave frequency 9.66 GHz, modulation frequency 100 kHz, receiver gain 4 X 10^4 and time constant 20.48 ms. The position of the radiation–induced EPR signal was compared with that of the standard 2, 2 diphenyl-1-picrylhydrazyl (DPPH) with g = 2.0032 (Sigma Chem. Co. USA). To determine the electron relaxation behaviour of radicals in food samples using EPR spectrometer, the microwave field strength was varied between 0.06 – 50mW to obtain progressive saturation behaviour (PSB). The experimental setup is shown in **Fig 1a**. Attempts have been made in BARC to identify irradiated food and allied products exposed to commercially relevant doses to assess the feasibility of this technique.

EPR Spectroscopy in Identification of Irradiated Meat and Mango

EPR technique has been used to detect radiation-induced species in bone since 1950[2] (Gordy et al, **1955**). Later, in BARC, Chawla et al, 1999[3] showed that the lamb meat with bone

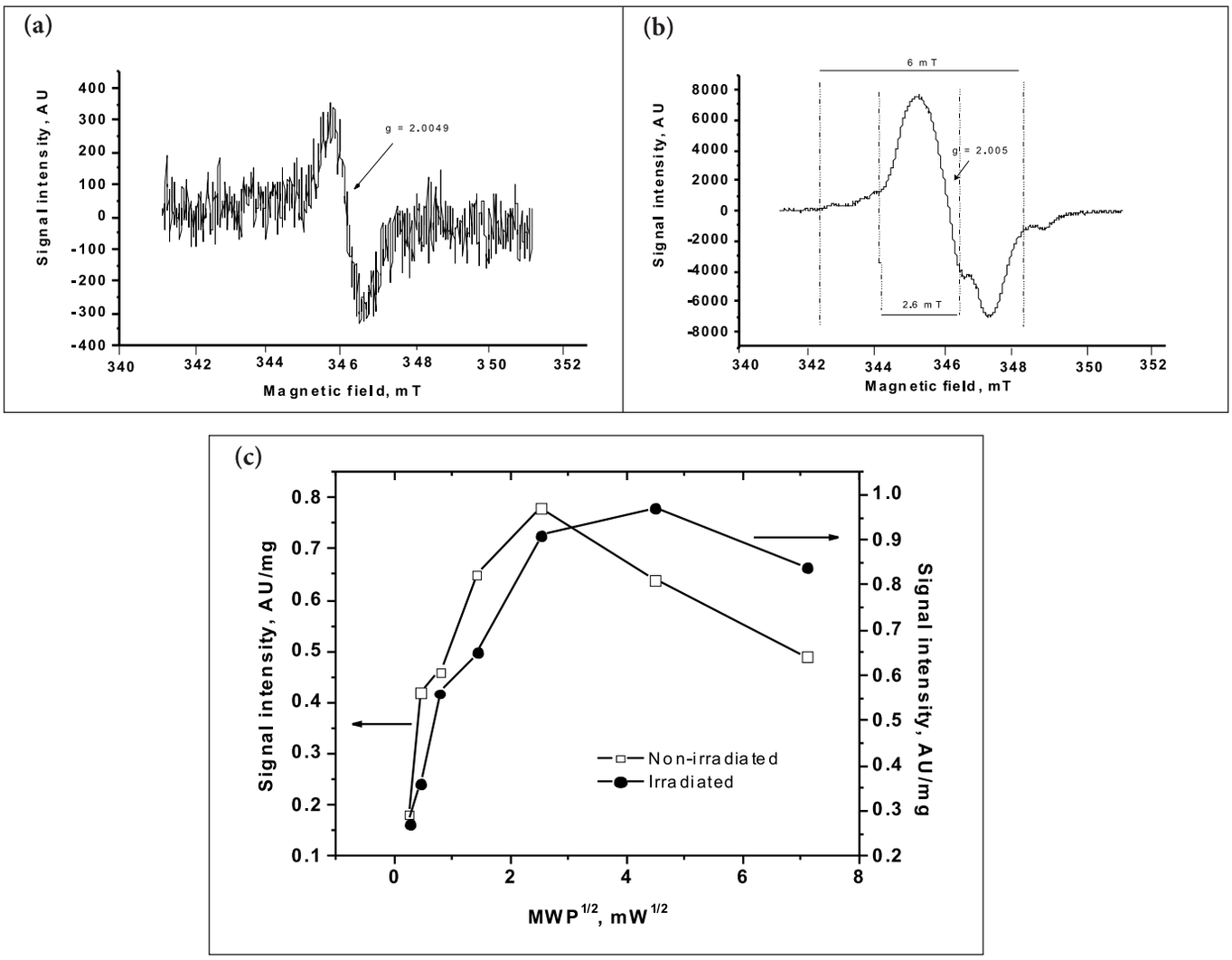


Fig. 2. (a) EPR spectra of the non-irradiated Basmati rice, (b) EPR spectrum of the irradiated (1 kGy) Basmati rice and c) Progressive saturation behaviour of the paramagnetic centres before and after irradiation as reported by Sanyal et al, 2009[6]

could be identified after irradiation. Irradiated bone of lamb meat exhibited anisotropic EPR signal with $g_{\perp}$ = 2.002 and $g_{\parallel}$= 1.998 originated from CO_2^- radical ion trapped in hydroxyapatite matrix of the bone. They also showed that the irradiated meat can be identified even after storage. In another study, Chawla et al, 2004[4] established that identification of irradiated meat was possible without knowing the processing status of the bone-in meat chunks.

Bhushan et al, 1995[5] has investigated the efficacy of EPR technique in detecting irradiated mango. A symmetric EPR signal at g = 1.988 was detected in the hard seed cover (endocarp), the dry epidermal layer (testa) surrounding the kernel, and the soft kernel portions of the seed from four mango cultivars. Qualitatively, however, no new signal was observed following irradiation, except that line width increased by 50%. The similarity of naturally occurring EPR signals to that induced by irradiation seems to restrict the practical utility of this method in irradiated mangoes.

Identification of Irradiated Basmati Rice by EPR Technique

Irradiated (0.25 – 1 kGy) Basmati rice, a high value fragrant rice of India, did not exhibit insect infestation during a storage period of 6 months, at room temperature, while the control (non-irradiated) samples were spoiled due to infestation. Sanyal et al, 2009[6] showed that the EPR signal of non-irradiated rice samples exhibiting a weak singlet characterized by g = 2.0049 ± 0.0004 and ΔB_{pp} = 1.3 mT, centered around 346.1 mT (**Fig 2a**). A complex spectrum was observed immediately after irradiation (1 kGy) of rice samples with an increase in signal intensity of the existing weak singlet (**Fig 2b**). Gamma radiation leads to change in rice matrix, producing two new types of paramagnetic species. One pair of intense satellite lines at a distance of 6 mT from each other and the other less intense pair of lines situated at a distance of 2.6 mT from each other.

These radiation specific signals observed in the rice matrix were not particularly stable and disappeared after 3-4 days. To address this problem, the electron relaxation behaviour of radicals was studied.

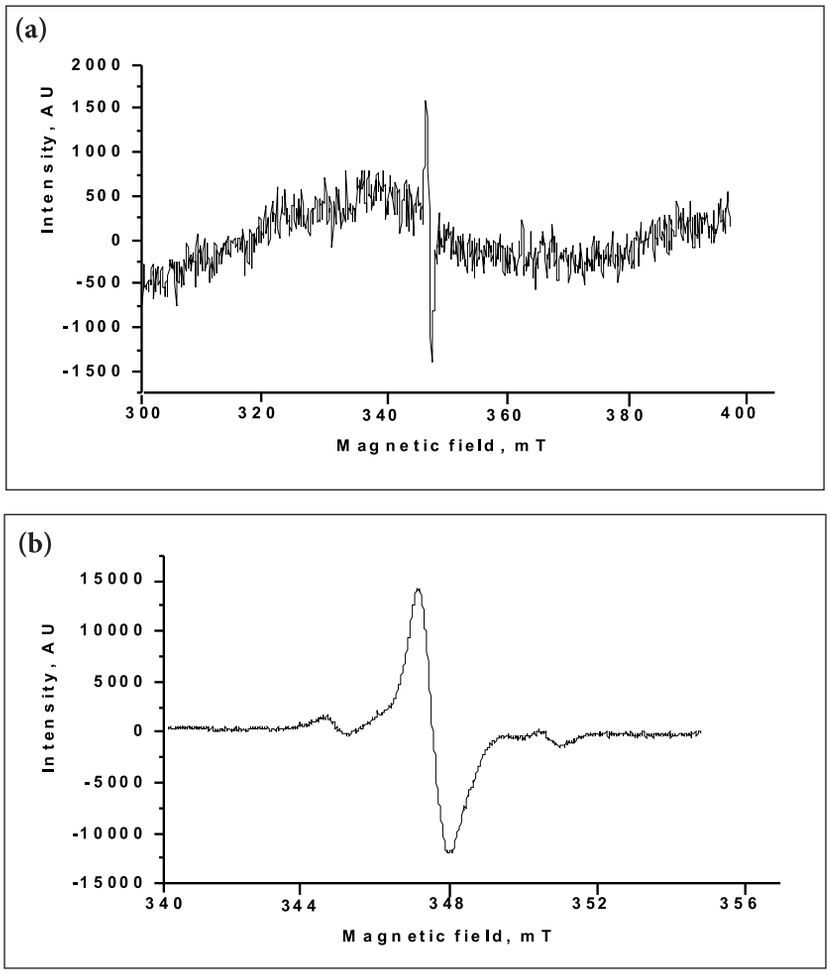


Fig. 3. EPR spectra of non-irradiated seed part a) and the irradiated seed part b) of soybean as reported by Sanyal et al, 2009[7]

The microwave field strength was varied from 0.063 to 50 mW to obtain progressive saturation behaviour (PSB). For the radicals of non-irradiated rice sample, a comparatively faster saturation at microwave power around 6 mW followed by a decrease in signal intensity in a monotonic fashion was observed. This saturation behaviour revealed the characteristics of organic radicals with a large relaxation time. Consequently, they were poor to maintain the spin-population difference between the ground and excited states manifesting early saturation. Whereas, the radicals of irradiated rice sample exhibited saturation at microwave power at around 20 mW even after a storage period of 90 days (**Fig 2c**).

Thus, this improved approach was successful to identify irradiated Basmati rice.

EPR Spectroscopy in Detection of Irradiated Soybean

The main objective of this study carried out by Sanyal & Sharma, 2009[7] was to identify irradiated soybean with commercially approved radiation dose (0.25 to 1 kGy) for insect disinfestation, particularly after prolonged storage. Skin and kernel parts of soybean were studied separately using EPR spectroscopy. The EPR spectra recorded using a broad magnetic field sweep width (100 mT) of skin part of soybean before radiation treatment is shown in **Fig. 3a**. A singlet EPR signal at g = 2.0046±0.0004 centered around 347.8 mT was observed. In **Fig. 3b**, the EPR spectrum one-day after the radiation treatment with dose 1 kGy of the skin part has been depicted which shows an enhancement in signal intensity of the existing weak singlet (g = 2.0046) by a factor of 7. Irradiation was explained to be responsible for the relatively high intensity increase. The exposure to gamma radiation leads to another paramagnetic species (triplet signal) at the same g value as that of non-irradiated samples with hyperfine coupling constant (hfcc) 3 mT. EPR spectra simulation studies were carried out in order to characterize radiation induced signals in the skin part of irradiated soybean. Simulation of EPR spectra obtained from the irradiated skin revealed the formation of two paramagnetic species, one was singlet attributed to phenoxyl radical ($C_6H_5O^-$) ion and the other was triplet signal of cellulose radicals. The signal attributed to cellulose radical was a marker of radiation treatment.

Identification of Irradiated Cashew Nut

The nut of the plant, Anacardium occidentale, commonly known as cashew is an important cash crop in many countries. Sanyal et al, 2008[8] carried out a detailed EPR study of the radical species produced by gamma irradiation of cashew. Cashew nut samples were irradiated at γ-radiation doses within the permissible dose range for insect disinfestation of food commodities (0.25- 1 kGy). A weak and short-lived triplet (g = 2.004 and hfcc = 30 G) along with an anisotropic signal ($g_\perp$ = 2.0069 and $g_{\varnothing}$= 2.000) were produced immediately after irradiation (**Fig 4a & b**). These signals were assigned to that of cellulose and CO_2^- radicals. The nature of the free radicals formed during conventional processing such as thermal treatment was also investigated and showed an increase in intensity of the central line (g = 2.0045) similar to that of irradiation. To address this problem, relaxation and thermal behaviours of the free radicals were studied. The study explored the possibility to

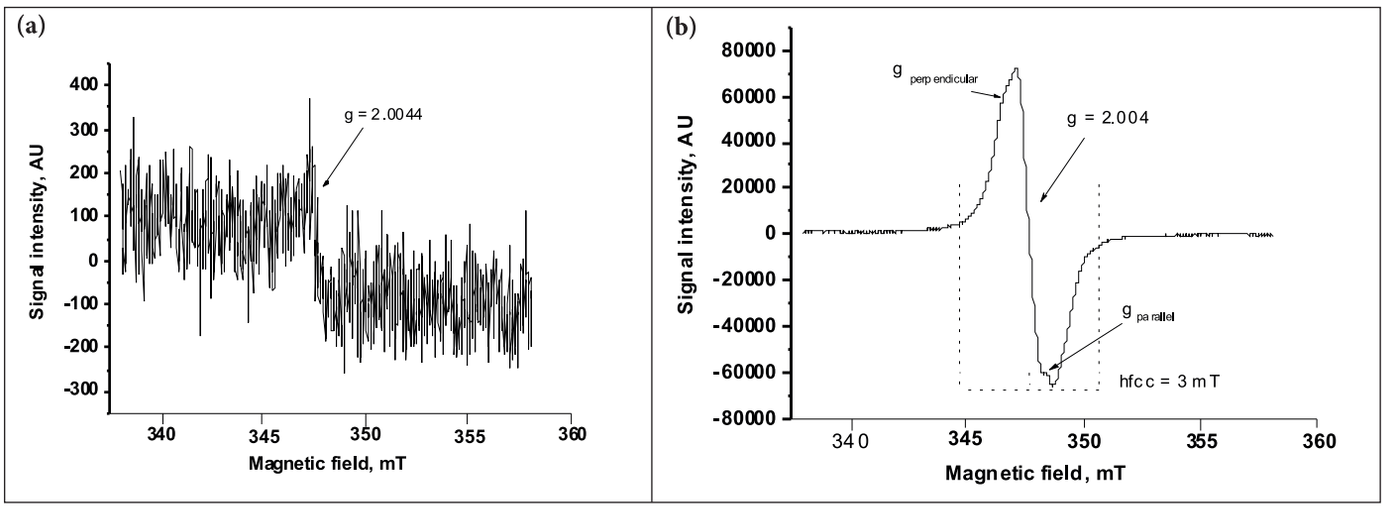


Fig. 4. EPR spectra (a) non-irradiated and (b) irradiated cashew nut and (c) the thermal behaviour of the EPR singlet of non-irradiated, irradiated and roasted samples as reported by Sanyal et al, 2008[8]

identify irradiated cashew nuts from non-irradiated ones by the thermal behaviours of the radicals beyond the period, when the characteristic electron paramagnetic resonance spectral lines of the cellulose free radicals have essentially disappeared (**Fig 4c**). In addition, this study for the first time reported that relaxation behaviour of the radicals could be a useful tool to distinguish between roasted and irradiated cashew nuts.

Identification of Indian Spices Using Various Analytical Approaches

The effect of flavour pattern of irradiated Indian spices, namely cumin and red chili powder was analyzed using electronic nose with a surface acoustic wave sensor in collaboration with Kyungpook National University, South Korea. The spices were irradiated with accelerated electron beam at doses 6, 10 and 14 kGy respectively. Both the non-irradiated and irradiated samples of weight 1 g were added to a 40-mL vial (Supelco, Bellefonte, PA, USA) fitted with a Teflon septum (PTFE/silicone, Supelco), sealed, and placed at room temperature for overnight to obtain the headspace equilibrium. The electronic nose registers the electrical signals generated due to the alteration of the state of the sensors after the chemical interaction with the odour compounds. The characterization of gaseous samples was conducted using the combined responses coupled with a suitable pattern-recognition system. Different electronic fingerprints were observed in irradiated spice samples compared to the non-irradiated ones[9]. In order to ensure the results obtained from electronic nose approach the irradiated spices were also attempted to detect using EPR spectroscopy. The e-beam induced signals were similar to that of the gamma irradiated samples. The non-irradiated spectra were characterized by a central line at g = 2.006 with a line width (ΔB_{pp}) of 0.54 mT. In case of cumin samples a sextet signal at g = 2.006 with a hyperfine interaction of hfcc (hyperfine coupling constant) = 9.6 mT was also observed showing a trace of Mn^{2+} ion. Existence of Mn^{2+} has also been reported for ground black pepper and wheat flour[10]. All the irradiated spice samples (6, 10 and 14 kGy) showed complex EPR spectra with an increase in signal intensity of the existing weak singlet. The exposure to electron beam leads to another triplet signal at g= 2.006 with a hyperfine coupling constant (hfcc) of 3 mT. This signal was attributed to cellulose radical due to the cleavage of cellulose polymer chain and exhibited detectable signature for the irradiated spices.

EPR Spectroscopy to Detect Irradiated Pet Feed and Medicinal Plants

In this study, probably for the first time, a detailed analysis of the radiation-induced radical species of irradiated dog feed were reported[11]. The irradiated samples exhibited a

complex EPR spectrum. During high power (50 mW) measurements, a visible change in the shape of the EPR spectrum was observed and characterized by EPR spectrum simulation technique. An axially symmetric anisotropic signal with $g_{\perp}$ =1.9976 and $g_{\parallel}$= 2.0028 were identified (**Fig 5a**). Another study on gamma-irradiated Indian medicinal plant products was carried out using EPR spectroscopy[12]. Improved approaches like high-power measurement, microwave saturation, and thermal behaviour of the radicals were explored for detection of irradiation. However, amongst all the three approaches, high-power measurement of EPR spectra emerged as a suitable technique in identification of the irradiated products (**Fig. 5b**).

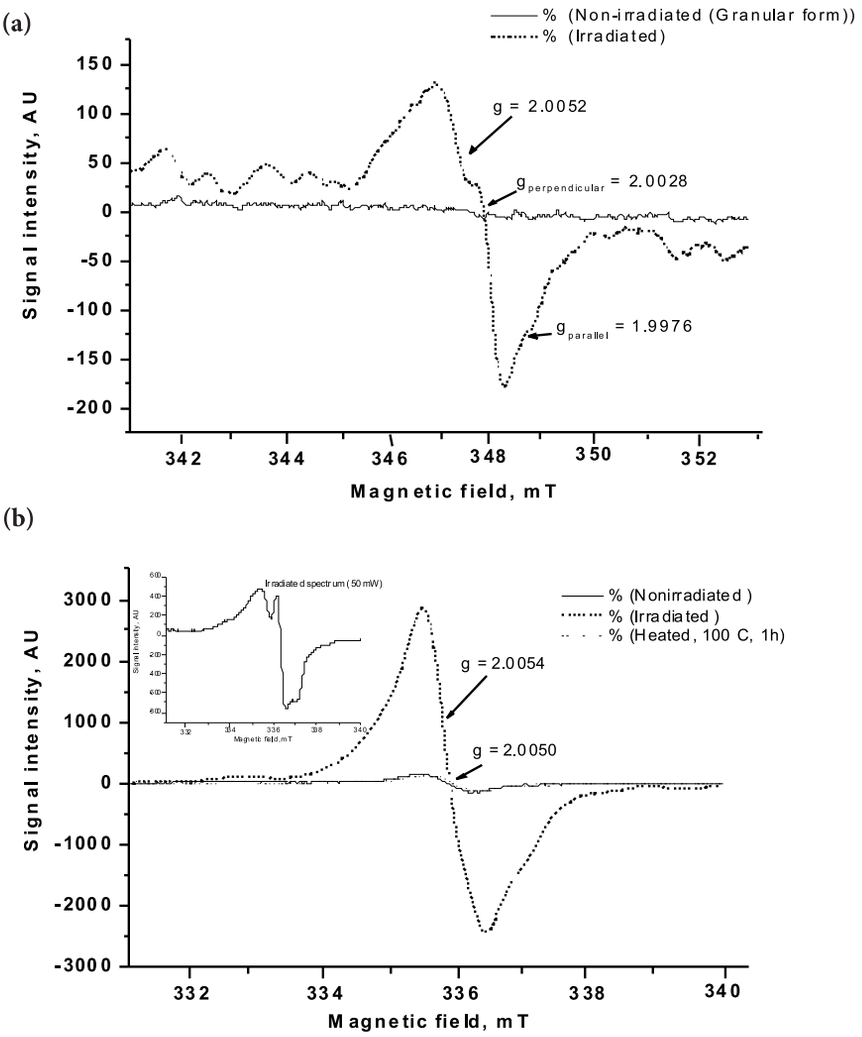
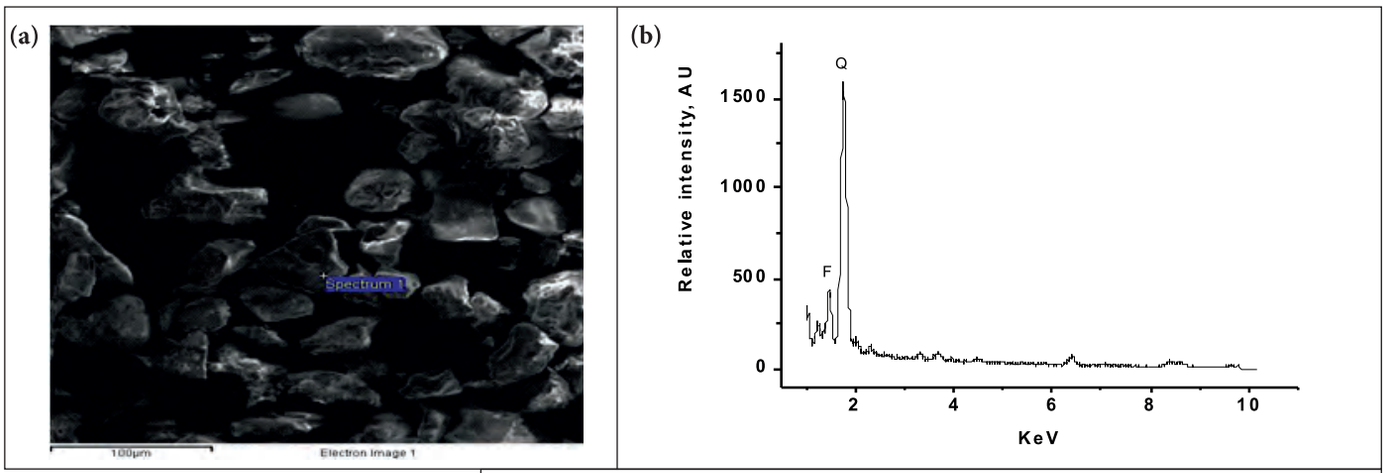


Fig. 5. EPR spectra of irradiated and non-irradiated (a) dog feed reported by Sanyal et al, 2011[10] and (b) non-irradiated, heat-treated (373 K, 1 h) and irradiated (7.5 kGy) Aswagandha. Inset shows irradiated spectrum at high power (6 dB)as reported by Sanyal et al, 2012[11].

Identification of Irradiated Food using Thermoluminescence (TL) Measurements

Thermoluminescence (TL) is a radiation specific phenomenon that arises due to energy stored by trapped charge carriers following irradiation[13]. By controlled heating of the isolated minerals from the irradiated foods, the charge carriers were stimulated to return back to the ground state thereby emitting some of the energy as light. The light emission which is dependent on the temperature is recorded as glow curve using a sensitive photon counter. An early application of thermo luminescence related to food was described by Chadwick and Oosterheert in 1967[14], who measured the thermoluminescence of tomato seeds irradiated at liquid nitrogen temperature with X rays at 0.05-1 kGy. To use thermoluminescence as detection method for irradiated food was first proposed by Heide and Bogl in 1984[15]. They suggested detecting the radiation treatment of spices and herbs by investigating whole samples. TL has been tested for detection of spices and herbs and was adopted as a standard method for detecting irradiated foods from which silicate minerals can be isolated[16]. For TL measurements, it is essential to isolate natural mineral contaminants of the various commodities. To make sure that the inorganics isolated from the food samples were free from any organic impurity, a solution of sodium polytungstate ($Na_6W_{12}O_{39} * H_2O$, density 2 g/ml) was used. The samples were subjected to ultrasonication in a bath sonicator for 5 min followed by centrifugation at 1000 g for 2 min. The organic material floating on the top of the polytungstate solution was removed. The bottom layer was washed three times with double distilled water. After total removal of water, 2 ml of acetone was added. The minerals in acetone were transferred to clean and weighed aluminium discs (diameter 9.0 mm; thickness 0.4 mm, fabricated at workshop, FTD, BARC) with the help of Pasteur pipette. The discs containing minerals were stored overnight at 50°C. Thermoluminescence analyses were carried using TL 1009I Reader (Nucleonix Systems Pvt. Ltd., India). The initial ambient temperature was increased to 300°C by a linear heating at a rate of 5°C/s. Nitrogen was flushed in the heating chamber to reduce spurious TL arising due to the presence of oxygen. The experimental setup is shown in **Fig. 1b**. Identification of irradiated food



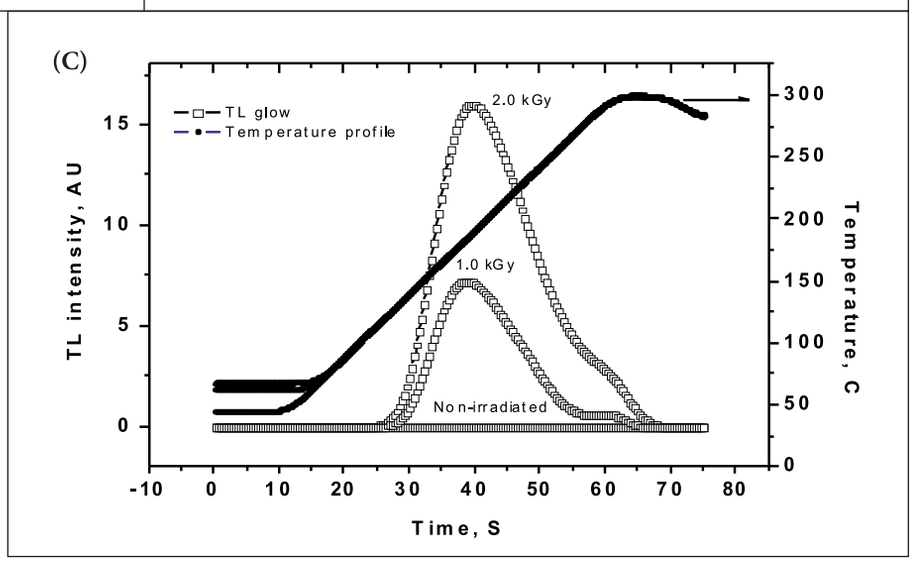


Fig. 6. (a) SEM image and (b) EDX spectrum of the isolated polyminerals, (c) TL intensities of glow curves for separated polyminerals from the non-irradiated and irradiated rice samples as reported by Sanyal et al, 2009[6].

samples namely rice, potato, ginger, fresh turmeric, shrimp, and dog feed were carried out in BARC using thermoluminescence (TL) technique.

Identification of Irradiated Basmati Rice Using TL Technique

Investigation on the composition of the isolated minerals from rice samples was of paramount importance to assess the possibility of employing TL for identification of irradiated rice[6]. In view of this, the composition of the separated polyminerals from rice sample was studied. SEM image of the isolated polyminerals from the rice sample revealing the morphology has been shown in **Fig. 6a** while **Fig. 6b** shows the EDX spectrum of polyminerals which mainly composed of quartz (SiO_2) and K-feldspars ($KAlSi_3O_8^-$) with a higher abundance of quartz (about 59.6%) than K-feldspars (20.7%).The TL intensities of glow curves for separated polyminerals from the non-irradiated and irradiated rice samples 10 d after radiation treatments presented in **Fig. 6c**. In case of irradiated sample, the glow curve was characterized by a low temperature peak at about 184±4°C and a high temperature peak at about 282±5°C. The position of glow peak for non-irradiated sample through all temperature ranges was not clear. However, low level natural radioactivity exhibited TL signal because of deep traps around 295°C. Therefore, discrimination between irradiated and non-irradiated rice samples was possible based on the shape of the first glow curve from the separated polyminerals. TL glow of the irradiated samples were recorded after 65 days of storage in dark with normal laboratory conditions and around 20% fading in TL glow intensity was observed with clear discrimination from the TL glow of the non-irradiated samples.

Identification of Irradiated Potato, Ginger and Fresh Turmeric Using TL Technique

The objective of the study was to identify potato of different geographical locations in India irradiated to sprout inhibiting dose, using TL characteristics of the isolated minerals. Potato (Chandramukhi) from two different districts, Bankura and Midnapore of West Bengal and markets of metro cities of Delhi and Mumbai were procured. The samples from each location were divided into three lots. One lot was kept as non-irradiated (control) and the remaining lots were irradiated with gamma radiation doses of 100 and 250 Gy. Minerals were isolated from both the non-irradiated and irradiated potatoes by density separation method. Characterization of the isolated minerals was carried out by X-Ray diffraction (XRD) technique to know the composition (**Fig. 7a**). The relative abundance of K-feldspars ($KAlSi_3O_8^-$) and quartz (SiO_2) were predominant. TL glows were recorded from room temperature to 350°C at a heating rate of 5°C/s (**Fig. 7b**). The intensities of the glow curves for both the non-irradiated

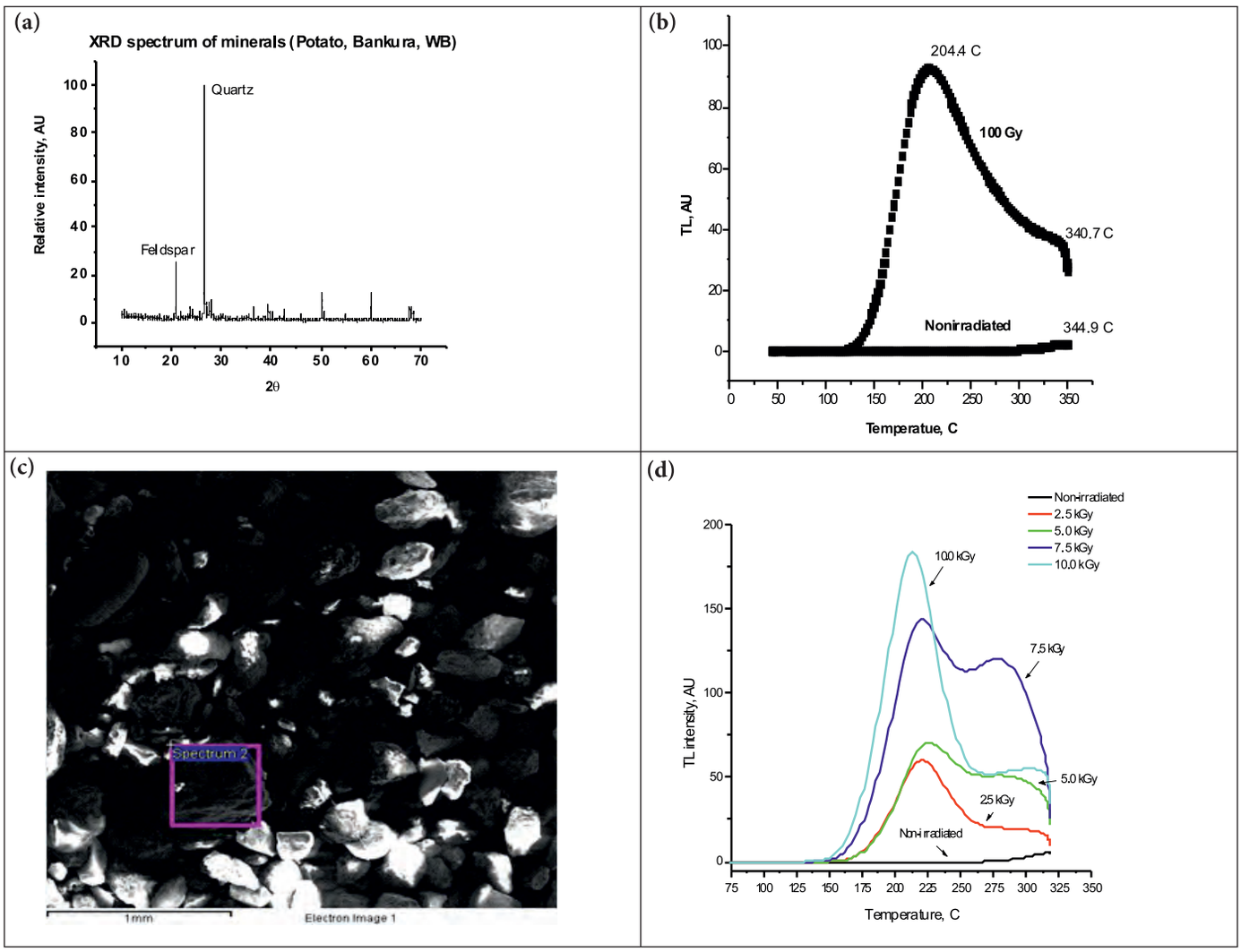


Fig. 7. (a) XRD spectrum and (b) TL glows of the isolated minerals from the potato (Bankura, WB),(c) SEM morphology and (d) TL glows of the minerals isolated from the dog chew samples reported by Sanyal et al, 2011[10].

and irradiated samples showed significant difference even after a storage period of one month. In all the cases, the glow curves of irradiated samples were characterized by a peak temperature at around 204°C, whereas, the position of glow peak for control sample through all temperature ranges was not clear. Identification of irradiated ginger (Zingiber officinale) and fresh turmeric (Curcuma longa) were also studied using TL technique. Minerals were isolated from both the non-irradiated and irradiated samples by density separation method. The integral TL and the intensities of the glow peaks for both the non-irradiated and irradiated samples showed significant difference even after a storage period of one month. Around 50-fold increase in integral TL glow in irradiated ginger and around 20-fold increase in glow in case of irradiated fresh turmeric were observed in comparison with their respective non-irradiated counter parts.

Thermoluminescence Measurements of Irradiated Edible Dog Chew

Thermoluminescence study of the non-irradiated and irradiated edible dog chew was carried out[11]. The composition of the isolated minerals and their morphology were found out using XRD and SEM analyses (**Fig. 7c**). In case of irradiated sample the glow curve was characterized by a low temperature peak at about 213±3°C and a high temperature peak at about 303±5°C. The areas under the glow curves for irradiated sample (10 kGy) were 55 times more than the areas under the non-irradiated sample (**Fig. 7d**), establishing a successful discrimination between irradiated and non-irradiated samples.

TL Characterization of Isolated Minerals from Irradiated Oranges to Detect Irradiation

The purpose of the study was to determine the TL glow curve structures of inorganic minerals isolated from irradiated oranges and to characterize the TL properties in comparison with that of the standard minerals. The efficacy of TL measurements in identifying imported oranges exposed to γ-ray, electron beam, and X-ray irradiations for quarantine applications was assessed even after a prolonged storage. The study was carried out in collaboration with Kyungpook National University, South Korea[17].

The TL characteristics of the inorganics separated from oranges before and after irradiation with γ- and X-rays and an electron beam were studied. No differences were observed among the TL glow curve structures obtained using different sources of ionizing radiation. The glow curves of the irradiated samples were characterized by a prominent glow peak in the range 158-163°C with considerable enhancement of the TL intensity. The area under the glow curve was around 100 to 600 times more than that of the curve for a non-irradiated sample, depending on the delivered dose. The non-irradiated sample exhibited negligible integral TL without any defined TL peak within the entire temperature range studied. Signal intensities of TL glow peaks of the isolated minerals from oranges irradiated at 2 kGy were measured during storage at two different temperatures of 4 and 20°C till 6 weeks. The irradiated samples exhibited considerable reduction in TL signal intensities. However, oranges stored at 4°C after irradiation with gamma, electron beam and X-ray showed more stable intensity in comparison with those stored at 20°C. Clear differences in TL glow curve structures between irradiated and non-irradiated samples were observed and a successful detection of oranges exposed to different types of radiations was possible even after a prolonged storage.

Conclusion

Identification of the irradiated food using EPR spectroscopy is a sensitive tool to detect radiation-specific paramagnetic centres. The difference in shape of the EPR signal due to radiation-induced radicals in food matrix are of extremely low concentration and comparable with other conventional food preservation technique making determination of radiation marker in food commodities a difficult task. In case of TL measurements, the inorganic contamination of the food items is studied. Isolation of the same is another challenging task. Biological samples are highly diversified and therefore detection of irradiation is normally product specific. The Government of India has gazette notified the class based approval of food and allied products for commercial radiation processing in 2016.Thus, a wide spectrum of food commodities now can be irradiated. Hence, there will be more scope for research in the field of identification of various types of irradiated food.

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Development of a Novel Process for Purification of Tummalapalle Milled SDU: Plant Scale Experience

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Abstract

Plant scale experience is reported on the purification and recovery of uranium from Tummalapalle Milled sodium di-uranate (SDU: yellow cake), the key raw material for the production of uranium metal ingot used in research reactor as fuel. The raw material consists of several impurities such as Zirconium, polyacrylamide, humic acid, silica and other organo-metallic compounds along with sulphates and carbonates which make the purification and recovery process complicated. The challenges was undertaken and successfully resolved by developing a new treatment methodology in Uranium Extraction Division (UED). The developed novel process has been adopted successfully in plant production activities. The effect of impurities is suppressed by addition of ethylene-di-amine tetra-acetic acid (EDTA) before acid dissolution and fluidity of the slurry generated after dissolution is enhanced by heating it upto 95°C. The slurry after dissolution is fed into a solvent extraction cycle for purification without filtering it. Uranium is extracted with tri-butyl phosphate dissolved in dodecane. The purified uranium solution is further processed to produce nuclear quality uranium metal ingot. In five batch campaigns, 5 tonnes of SDU have been processed and demonstrated successfully. The chemical yield of the dissolution and solvent extraction operations is > 99.5%. All purified materials have been used for Dhruva fuel fabrication after qualifying quality checks.

Keywords: Tummalapalle SDU, Purification, Recovery, EDTA, Dissolution, Solvent Extraction, U-metal, Yield, Dhruva, Fuel

Introduction

Tummalapalle ore is the single largest reserve of uranium in India having uranium concentration of ~0.042% on U_3O_8 basis[1] (Suri A.K. 2008). The ore is classified as phosphatic-siliceous-calcitic-dolostone (PSCD) having 60–65% dolomite and calcite for which alkaline leaching route has been chosen as milling process (**Fig. 1**) unlike the conventional sulphuric acid leaching technology used for the production of yellow cake[2] (Suri A.K. 2010) in the existing uranium ore processing mills at Jaduguda and Turamdih. The milled product of Tummalapalle ore is sodium diuranate (SDU), also known as yellow cake. The chemical composition of a typical mill batch SDU is given **Table 1**. The impure yellow cake is purified in various stages and converted to various forms as per the desired applications. During the process of purification, the yellow cake is dissolved in nitric acid at about 80°C. The resulting slurry (if required) is filtered to remove silica and acid-insoluble materials. The uranium solution obtained after filtration or the unfiltered slurry is taken for purification by solvent extraction (SX) using Tri-n-butyl phosphate (TBP). There are well identified problems associated with the processing and purification of SDU-mainly due to presence of acid insoluble materials, fine silica content, presence of excess carbonates and sulphates, higher level zirconium contamination and presence of different organic materials such as poly-acrylamide (PAM) derivative used as flocculent and ore-originated humic substances. Effervescence during acid dissolution process is obvious due to presence of carbonates in the SDU. Filtration rate of the resultant slurry after acid dissolution under vacuum is very poor because of sticky and gelatinous nature of the slurry-which may be due to presence of PAM, humic substances and other organo-metallic complexes. Fine silica passes through the filtering medium under forced filtration. Cake obtained after filtration carries significant uranium value which warrants re-dissolution to improve uranium recovery.

At SX stage by using slurry extractor and/or mixer settler, the process becomes complicated due to presence of fast settled silica, zirconium impurity and soluble organics[3] (Pradip A. and Biswas S. **2017**).

It is well-understood that Zr (IV) being tetravalent metal creates a third layer band in SX process using TBP as organic extractant. Additionally, degradation of TBP is also envisaged

due to contamination of process feed with sulphates, soluble silica and organic substances. Such experiences and understanding of processes clearly indicates that development of a new treatment method suitable for processing and purification of Tummalapalle SDU in plant scale is a worth challenging task for purposeful utilization of the material. The uranium metal production facility under Uranium Extraction Division (UED)/ Materials Group (MG) in Bhabha Atomic Research Centre (BARC), Trombay, produces nuclear grade uranium metal ingots to meet Dhruva reactor's fuel requirement. Impure uranium oxides, including yellow cakes (Diuranates etc.) are used as a starting material in the UED facility. Simplified flow sheet of dissolution and SX processes of the facility is given in **Fig. 2.** A novel method has been developed in UED for processing and purification of Tummalapalle SDU for the production of uranium metal. The developmental activities along with the experience in processing 5 tonnes of SDU in the facility are unique in nature which has been illustrated and shared here. **Table 1** shows the typical analysis of Tummalapalle SDU as received from Nuclear Fuel Complex (NFC).

TABLE-1

ELEMENT/ COMPOUND	QUANTITY
Acid Insoluble	3%
Ag	< 1 ppm
Al	< 0.03 %
CO_3	4.9 %
B	2 ppm
Ca	0.4 %
Cd	< 1 ppm
Ce	1 ppm
Cl	0.04 %
Co	39 ppm
Cr	15 ppm
Cu	41 ppm
Dy	< 1 ppm
Er	< 1 ppm
F	0.02 %
Fe	0.08 %
Gd	< 1 ppm
Ni	34 ppm
SO_4	2.6 %
SiO_2	2.6 % (as Silica)
Sm	< 1 ppm
Zn	9 ppm
Zr	~3%
U	58.95 %
As U_3O_8	69.51 %
Moisture	0.9 %

Materials and Instruments:

The source material - SDU (**Refer Table 1**) - was obtained from the Uranium Corporation of India Limited (UCIL)'s Tummalapalle Mill in Andhra Pradesh. However, uranyl nitrate standard was obtained from the Quality Control Laboratory (QCL) of UED, BARC. TBP (> 99% assay) used for the solvent extraction studies in the laboratory was obtained from Heavy Water Board (HWB), DAE. Ethylene-di-amine tetra-acetic acid (EDTA) and other reagents used were of Analytical Reagent (A.R) grade purity obtained from T.A. Corporation, Mumbai, and were used without any further purification/treatment. Distilled water as is being used in QCL was used for washing/dilution as required throughout this work. Poly-propylene 1µ filter cloth (Khosla) was used for filtration. Demineralised water (DMW) generated from the operating plant was used for stripping. A Resistance-type electrical heater was used for heating the solution. Thermo Fischer scientific make ICP-AES, Jobinyvon Emission Model JY 328 was used for determining elemental purity. Quantitative determination of U (VI) and Zr (IV) was done using EDXRF (Jordan Valley, EX-3600M). TBP analysis was done by nitric acid equilibration method, and Di-butyl phosphate (DBP) was analysed by following the method of separation and titration against standard alkali technique (established).

EXPERIMENTAL

Laboratory scale Experiment

Process development study was initiated with a target to improve filterability of acidic slurry after dissolution of SDU in nitric acid. Laboratory scale study was initiated with 5g SDU materials per batch of experiment. The SDU was directly dissolved in nitric acid at 80°C (Laboratory batch, LB – 1). Another method was tried following tri-sodium phosphate – phosphoric acid addition route prior to nitric acid dissolution (LB – 2). Further, experiments were initiated by treating 100g SDU of each batch with EDTA (LB – 3). Solvent extraction and stripping experiments using 32% TBP – Dodecane and DMW respectively in batch equilibrium mode were conducted with the solution obtained after dissolution & filtration.

Plant Scale Experiment

Based on process requirement and availability at the existing facility, five (05) plant scale trials were conducted. In all trials, a one (01) Ton SDU dissolution batch size was taken and 32% TBP diluted with dodecane was used as a solvent in the SX process. A 4000 litre capacity SS-304L tank along with NO_x scrubbing system was used for dissolution operation with commercial nitric acid (~60%). Also, solvent extraction unit of 50 kg-U/h capacity consisting of eight (8)-stage-slurry extractor for extraction, eight (8)-stage mixer settler for scrubbing and a ten (10)-stage mixer settler for stripping, were used. 0.5 (N) nitric acid solutions with O/A = 10 was used as scrub solution for final removal of contaminants and DMW was used as the stripping medium for recovery of uranium from the organic medium. The standard feed: solvent: DMW=1:2:3 flow rate was maintained for extraction and stripping processes. The details of the

conducted trials are briefed as below:

Trial – 1: One (1.0) MT SDU, 10% EDTA (w/w), 80°C for 3h, Filtration in centrifuge and vacuum pan filter, Feed U: 223 g/L & FA: 2.01 (N), previous batch used solvent.

Trial – 2: 1 MT SDU, 10% EDTA(w/w), 80°C for 4h, Filtration in centrifuge & 2 washes, + Feed U: 184 g/L & FA: 2.2 (N), used solvent after Trial – 1.

Trial – 3: 1 MT SDU, 10% EDTA

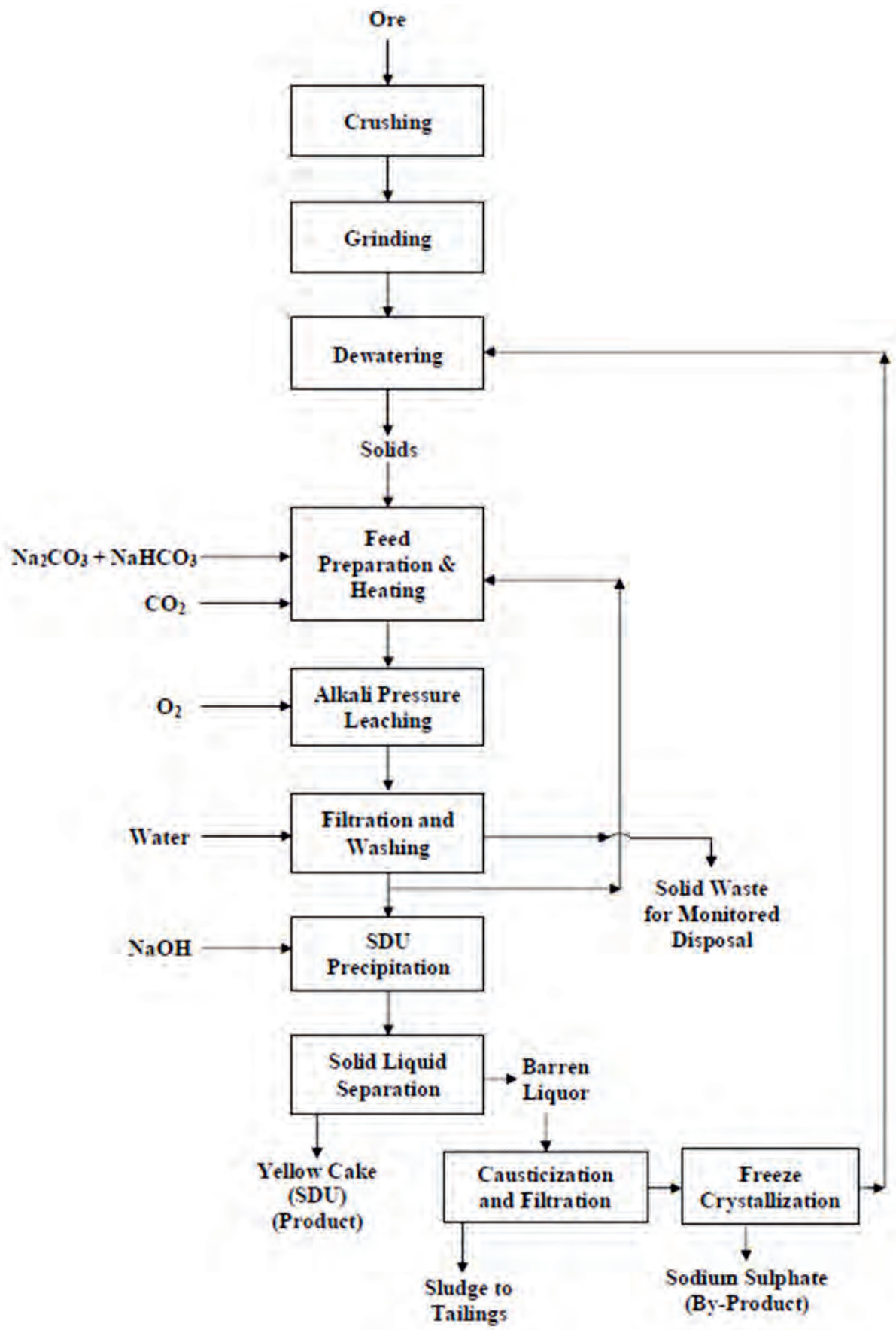


Fig. 1: Flow sheet of Tummalapalle ore milling process

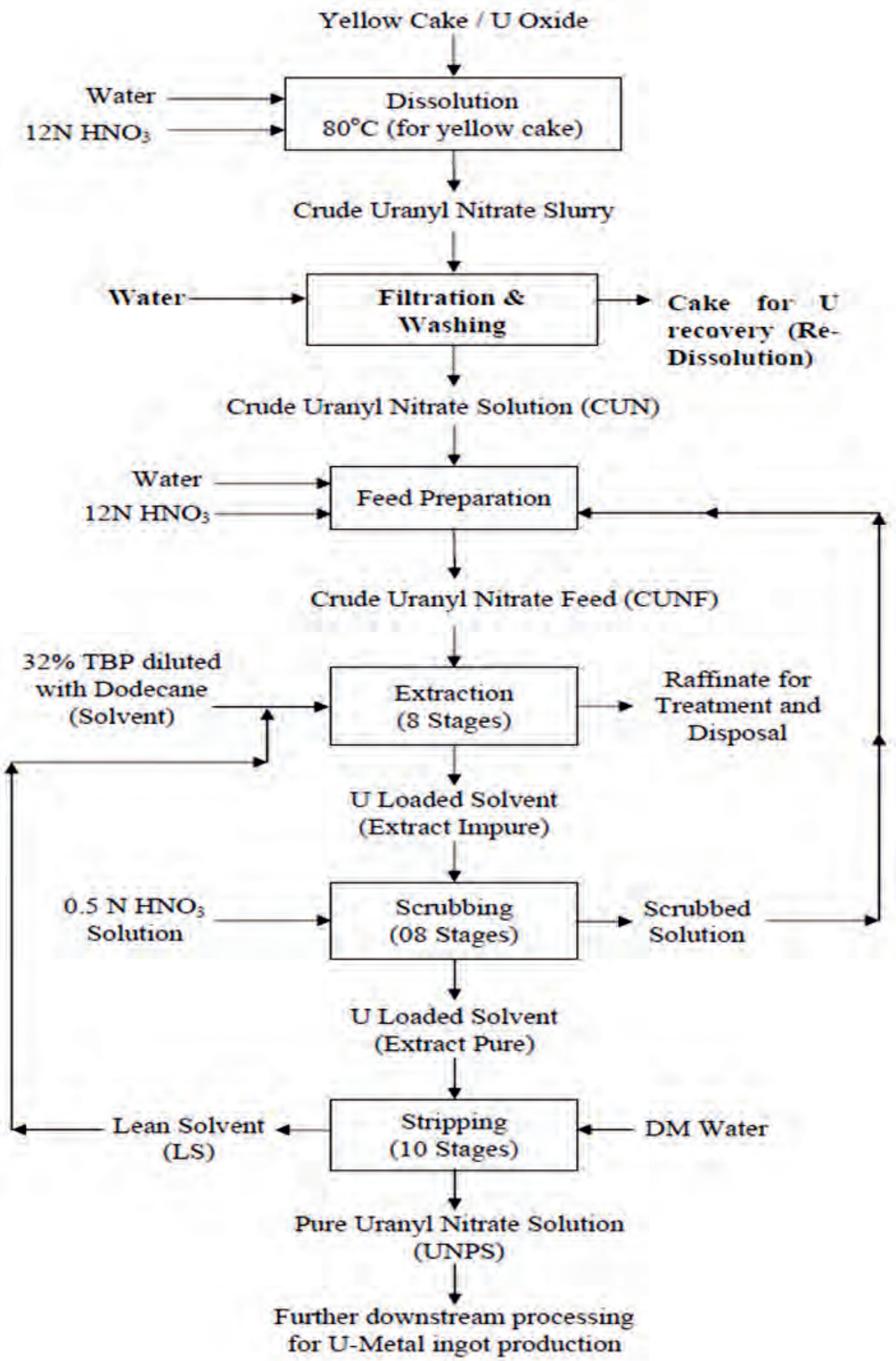


Fig. 2: Flow sheet of dissolution and SX processes of UED facility

(w/w), 95°C for 4h, without filtration, Feed U: 228 g/L & FA: 2.04(N), processed solvent.

Trial – 4: 1.0 MT SDU, 8% EDTA (w/w), 95°C for 4h, without filtration, Feed U: 223 g/L & FA: 2.5(N), solvent after Trial – 3.

Trial – 5: 1.0 MT SDU, 7% EDTA (w/w), 95°C for 4h, without filtration, Feed U: 236 g/L & FA: 2.07 (N), solvent after Trial – 4.

RESULTS AND DISCUSSION

Laboratory Scale

In the first laboratory study (LB – 1), it was found that the resultant slurry is poorly filterable and the generated cakes are sticky in nature having high levels of uranium, which is not recoverable even after repeated washing of the cake (**Fig. 3**).

Fig. 3: Gelatinous cake obtained after acid dissolution

The diluted uranium solution generated in the Laboratory Study-2 had sufficiently high levels of free acidity (FA) even as the filterability of resultant slurry is poor with reasonable quantities of uranium in the filtered cake. Importantly, the overall slurry is gelatinous in nature and is difficult to process in the existing SX system in UED's facility.

To get rid of the problem and to achieve a more acceptable process, several experiments like washing of the cake with water & repeated alcohols washing of the cake prior to acid dissolution; addition of Separan as a coagulant after acid dissolution for improving filterability; and carbonate leaching of uranium instead of the acid dissolution were carried out. In all the cases, recovery of uranium had been a considerable issue though improvement in filterability of acidic slurry was observed upon addition of coagulant Separan (Separan 2610, Dow Chemical Co.) to some extent. It was felt that presence of sufficient quantity of PAM and acid-insoluble organics in SDU may be playing a key role in the generation of gelatinous slurry in acid dissolution. PAM belongs to a class of responsive polymers that change their physical properties in response to a chemical or physical stimulus[4] (Hui F. et al 2010). In presence of acid, poly-acrylate (PAM) is neutralized and the non-ionized poly-acrylic acid is produced with extensive hydrogen bonding, which is insoluble in acidic aqueous medium. The rate of free radical crosslinking polymerization of acrylamide derivatives gets affected by addition of EDTA (a complexing agent)[5] (K. Byungsoo et al **2014**). EDTA having multiple chelating moieties, forms stronger complexes with heavy metals like uranium, zirconium etc. can be used as dislodging-assisted compound. EDTA cross-linked chitosan and polyacrylamide based double network hydrogel has been successfully synthesized and then employed for heavy metal ion adsorption[6] (M. Jianhong et al 2017). Being a mono-polar surface-nature, PAM dissolves in water and it is solvo-phobic in different solvents, including ethylene glycol (EG), acetone, ethanol, and dimethyl formamide (DMF)[7] (W. Shuhui and R.A. Shanks **2014**). This indicates the compatibility of PAM derivatives with the solvent used for extraction of heavy metal.

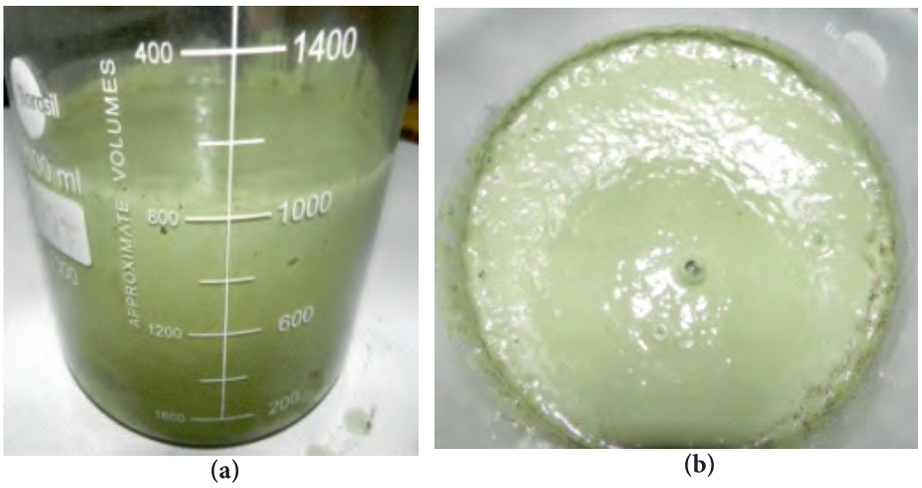
With this understanding, 100g SDU batch size treating with EDTA was carried out (LB – 3) and it was found that addition of EDTA (w.r.t. SDU, w/w) during nitric acid dissolution of SDU is useful with a particular sequence. Repeatability of the process along with addition of different quantity EDTA / U (w/w) was tested. Improved filtration rate is observed under vacuum pan as well as centrifuge filter and recovery of uranium after dissolution and filtration is ~98%. The generated slurry is appeared to be slippery in nature and the solid in the slurry is not separated /settled even after more than a month of storage. Most importantly, about 100% recovery of uranium is obtained in SX experiment. Both the experiments have given confidence to carry out plant scale trials.

Plant Scale

Processing of yellow cake having silica and zirconium (Zr) contamination is an unequalled experience in different purification facilities[8] (IAEA Proceedings **1986**).

During acid dissolution the resultant slurry is required to digest with 2M HNO_3 at 90–100°C for the complete flocculation of silica and if zirconium is present, the slurry is treated with controlled amount of phosphoric acid to separate both zirconium and silica. Process problems in SX and its process condition adjustments are regular features in any operating facility and these have been focussed as well as discussed with solutions in several literatures for different metal separation. Experience of processing of about 40 different sources of materials as yellow cake has been shared elaborately by J. W. Craig[9] (IAEA Proceedings **1979**). The cause of emulsion in extraction processes is for multiple reasons. The element silicon as a hydrated compound and zirconium are contributors to emulsions. Zr combines with di-butyl phosphate (DBP, degraded product of TBP) causes emulsion type problem. In nitrate medium solvent extraction, Zr as contaminant is generally picked up by unsaturated solvent which can be averted by keeping the solvent > 90% uranium saturation before entering to stripping unit. In presence of hydrated silica, the problem is corrected by increasing free acidity of feed. Tummalapalle SDU contains about 2-4% Zr, ~2.6% silica and about 3% insoluble acid, which indicates the necessity of preparedness for required parametric adjustment of the SX process in a similar way.

In Trial – 1, filtration rate in the pan filter is poor and filtrate is clear. In centrifuge filter, filtration is fast and slurry passes through the filter cloth. Generated cake is compact and hard, and the cake is having 12% U and ~50% moisture. Uranium concentration and free acidity (FA) in filtrate is 427 g/L and 0.64(N) respectively. Emulsion is observed in first two stages of stripping units which is removed by addition of nitric acid in line. Recovery in dissolution as well as SX is 98%. Uranium concentration in UNR and lean solvent (LS) is < 1 g/L and 6.4 g/L respectively, and DBP in LS is 0.76%.



(a) (b)

Fig. 4: Slippery slurry obtained after EDTA-acid dissolution: (a) side view in a beaker, (b) top view

In Trial – 2, filtration and washing both are fast; slurry passes through the 1µ filter cloth. Generated cake is appeared to be hard solid having 9% U and ~50% moisture. Uranium concentration and free acidity (FA) in filtrate is 372 g/L and 2.14(N) respectively. Emulsion was observed in mainly 1st stage of stripping unit which is removed by addition of nitric acid in line. Uranium concentration in UNR is < 1 g/L and LS is 13.1 g/L respectively, and DBP in LS is 0.34% which could not be correlated with U concentration in LS. Recovery in dissolution is 98.5% and 98% in solvent extraction.

The slurry generated in Trial – 3 is free flowing and slippery is nature (**Fig. 4**). Uranium concentration and free acidity (FA) in slurry is 416 g/L and 1.14(N) respectively. There is no chocking in any part of SX unit. Every day, after about 8 hours operation, complete unit is made complete shut-down and next day the full unit is started for operation without any operational difficulties. O/A volume ratio in different stages of slurry extractor as well as stripping unit and also densities of extract impure, extract pure, raffinate and lean solvent are major parameters which need close monitoring. There is no additional DBP generation after this trial (single pass of processed TBP). U concentration in UNR is 0.1 g/L, and in LS is 1.6 g/L. There is no dissolution loss and recovery in SX is >99.9%.

Observation and results of Trial – 4 & 5 are similar to Trial – 3. In Trial – 4, uranium concentration and free acidity (FA) in slurry is 467 g/L and 1.14(N) respectively. Increase of DBP generation after this trial is <0.1%. U concentration in UNR is 0.1 g/L and in LS is 4.7 g/L. There is no dissolution loss and recovery in SX is >99.9%. In Trial – 5, uranium concentration and free acidity (FA) in slurry is 476 g/L and 2.7(N) respectively. Increase of DBP generation after this trial is ~0.1%. The concentration of U in UNR is 0.1 g/L and in LS is 2.34 g/L. There is no dissolution loss, and recovery in SX is ~100%.

For last three batches (Trial – 3 to 5), it is observed that Zr comes out with raffinate which accumulates in cake after neutralisation treatment with magnesia as a regular practice before taking it for disposal monitoring. Based on the experience gained by processing 5 MT of the SDU in regular plant operation, processing flow sheet of dissolution and solvent

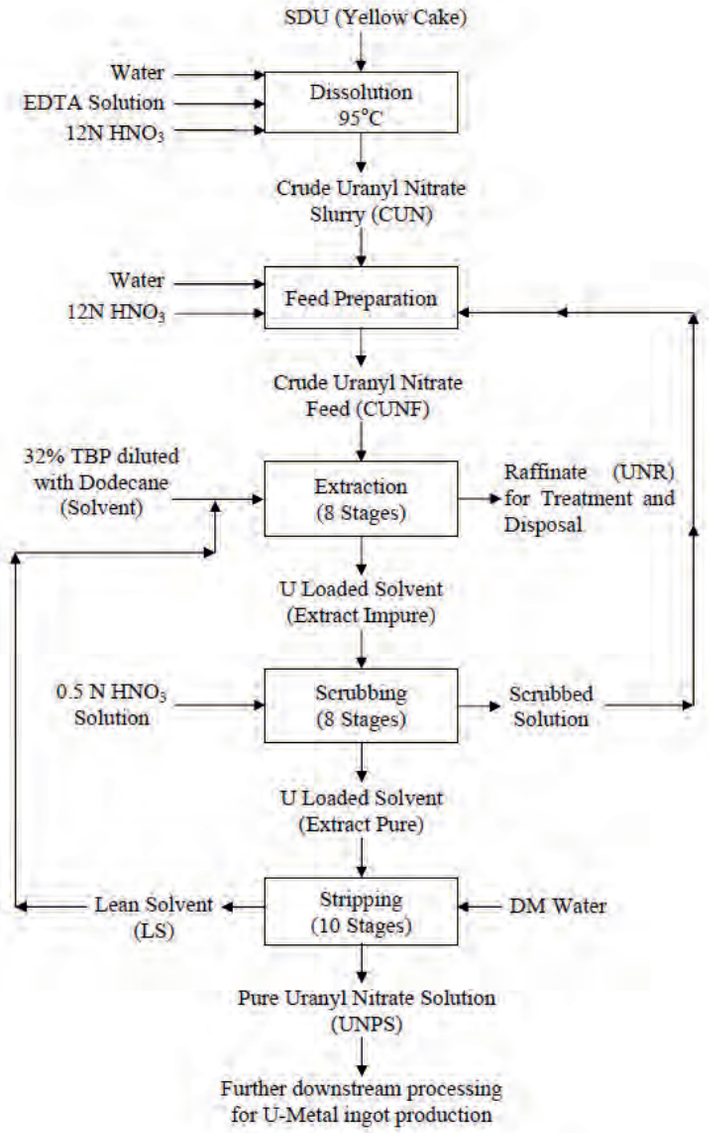


Fig. 5: New Flow sheet of hydro-processing of yellow cake in uranium metal production facility, UED for Tummalapalle SDU processing

extraction is developed for SDU processing as shown in **Fig. 5**. There are several benefits by adopting the new flow sheet:

· No additional equipment inclusion

· Filtration and washing operation is not required

· No additional manpower requirement

· No additional process footprint requirement and

· Process is easy and hassle free

Close monitoring of solvent extraction operation is essential and important which is anyway a requirement for processing of other yellow cake like magnesium di-uranate (MDU) etc. Comparing the experience of processing MDU by conventional method and SDU by newly developed method, latter is easier and simpler. All purified materials have been further processed successfully in downstream processes and equivalent quantities of uranium metal ingots have been produced for fabrication of Dhruva fuel (**Fig. 6**).

Fig. 6: NG-U metal ingot produced from SDU

Conclusion

The complexities involved in processing and purification of Tummalapalle SDU, generated by alkaline leached mill process, is well known to nuclear community mainly because of presence of adequate quantity of organics, silica and zirconium impurities. Gel and sticky cake formation in nitric acid dissolution process of the SDU and further micro emulsion and / or third phase formation in liquid – liquid extraction process using TBP – dodecane solvent are major challenges for processing the material. A new innovative and novel process has been developed for purification of the SDU to nuclear grade uranyl nitrate pure solution (Equivalent Boron Concentration (EBC) from 26.9 ppm to <1 ppm) avoiding all above said related problems during its processing. The slurry generated in newly developed EDTA – heating(>95°C) – nitric acid dissolution method is slippery in nature for which it can be processed as such in liquid – liquid extraction equipment avoiding the filtration and washing operation. In UED first time in about six decades of its operation, yellow cake has been processed without filtration. This has not only made the operation easy but also the overall recovery has improved to > 99.5%. Raffinate contents all in-soluble and zirconium has been treated and disposed following regular safe procedure. Generated pure uranyl nitrate solution has been processed successfully through regular downstream operation and equivalent uranium value has been converted to uranium metal following regular process. This method is highly useful and may be adopted in other Tummalapalle SDU purification facilities as per their operational needs.

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नवीनतम फिक्सेटिव कम्पोजीशन सैलाइन का आविष्कार तथा अनुप्रयोग

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सारांश:

श्वेत रक्त कोशिकाओं के अनुप्रयोग विविध हैं। इनकी प्राकृतिक/मूलभूत रूप में स्थिरण तथा पृथक्करण आवश्यक है। इस चुनौती को संबोधित करने हेतु हमारे शोध दल ने नवीनतम फिक्सेटिव कम्पोजीशन सैलाइन (एफ. सी. एस.) का आविष्कार किया है। एफ. सी. एस. विभिन्न प्रकार की श्वेत रक्त कोशिकाओं को उनके प्राकृतिक/मूलभूत रूप में स्थिर रखने मे सक्षम है। स्थिर किये गए रक्त नमूने को सामान्य तापमान पर कई महीनों तक संग्रहित एवं परिवहित किया जा सकता है। इन स्थरित श्वेत रक्त कोशिकाओं को जैवमात्रामिति सहित विविध अन्य नैदानिक तथा अनुसंधान कार्य हेतु प्रयोग में लाया जा सकता है।

प्रमुख शब्द: फिक्सेटिव कम्पोजीशन सैलाइन (एफ. सी. एस.), जैवमात्रामिती, श्वेत रक्त कोशिका, स्थिरण, पृथक्करण ।

प्रस्तावना

रक्त, मानव शरीर के स्वास्थ्य को प्रतिबिंबित करता है। संरचनात्मक रूप से रक्त विभिन्न प्रकार की कोशिकाओं और प्लाज्मा से निर्मित होता है। लाल रक्त कोशिकायें ऑक्सिजन वहन का कार्य करती हैं एवं श्वेत रक्त कोशिकायें शारिरिक प्रतिरोधक क्षमता के लिए समर्पित होती हैं। रक्त के नमूनों के साथ विभिन्न स्वास्थ्य संबंधी परीक्षण किए जाते हैं। विकिरण के प्रति विभिन्न रक्त कोशिकायों की संवेदनशीलता अलग - अलग होती है। विकिरण के प्रति अधिकतम संवेदनशील होने के कारण, श्वेत रक्त कोशिकायें (लिम्फोसाइट्स) जैवमात्रामिती (Biodosimetry) के लिए सबसे उपयुक्त मानी जाती हैं। किसी विकिरणीय दुर्घटना में उच्च स्तरीय विकिरण के संपर्क में आने वाले व्यक्तियों की प्रारंभिक देखभाल के लिये, रक्त नमूनों का शीघ्रतम संग्रहण, प्रारंभिक पहचान एवं चिकित्सीय वर्गीकरण आवश्यक होता है। विकिरण सम्बन्धी आपातकालीन परिस्थितियों में बहुत से व्यक्तियों के प्रभावित होने की संभावना होती है। विकिरण सम्बन्धी दुर्घटना-स्थल पर सूक्ष्मजीव मुक्त, रक्त नमूनो का संग्रहण एवं परिवहन अभी भी एक चुनौती है।

इन समस्याओं को दूर करने के लिए हमने एक नवीनतम फिक्सेटिव कम्पोजीशन सैलाइन (एफ. सी. एस.) विकसित किया है। यह सर्वविदित है कि शरीर के बाहर जीवित रक्त कोशिकाओं को अधिक समय तक नहीं रखा जा सकता है। इससे अवांछित परिवर्तन होने लगते हैं, जो रक्त के नमूनों की जाँच के वास्तविक परिणाम को प्रभावित कर सकते हैं। जैवमात्रामिती के अतिरिक्त कर्क रोग, अनुवांशिक परिवर्तन या अनुवांशिक अस्थिरता का अनुमान/पता लगाने के लिये भी सभी प्रकार की श्वेत रक्त कोशिकाओं को मूल रूप में बना रहना अत्यंत आवश्यक है। यह नवीनतम एफ. सी. एस. विभिन्न प्रकार की श्वेत रक्त कोशिकाओं को उनके प्राकृतिक/मूलभूत रूप में स्थिर रखने की क्षमता रखता है, तथा इस प्रक्रिया में सम्पूर्ण लाल रक्त कोशिकायें विखंडित हो जाती हैं। इस प्रकार स्थिर किये गए रक्त नमूने को सामान्य तापमान पर कई महीनों तक संग्रहित एवं परिवहित किया जा सकता है।

रक्त स्थिरीकरण एवं संग्रह की विधि :

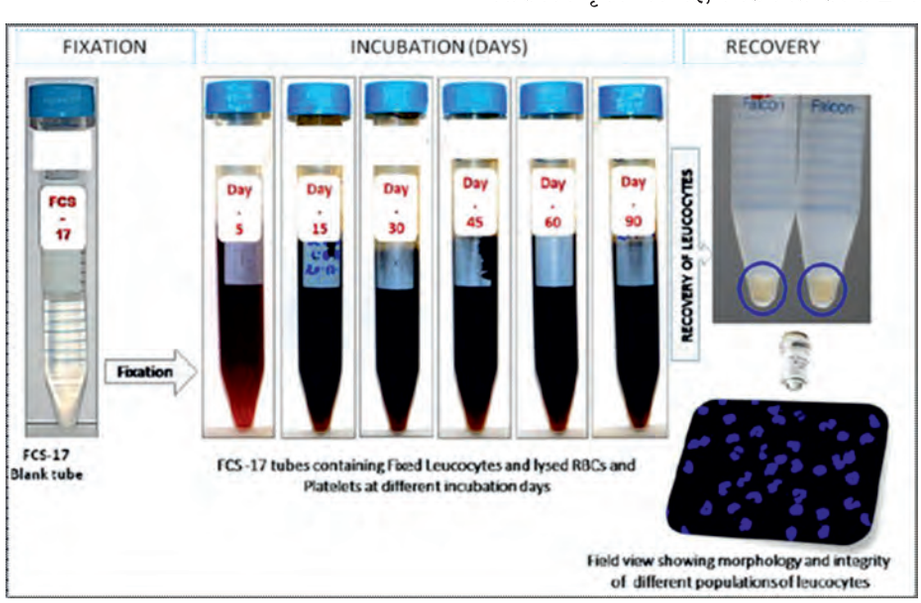
1. एक अन्तर्त्वचीय सुई की सहायता से 100 - 500 µl रक्त का नमूना प्राप्त किया जाता है।
2. रक्त के नमूने को 7.5 µl एफ. सी. एस. के साथ मिश्रित किया जाता है।
3. इस मिश्रण में श्वेत रक्त कोशिकायें स्थिर हो जाती हैं तथा लाल रक्त कोशिकायें विखंडित हो जाती हैं।
4. इसे सामान्य तापमान पर 6 महीने तक संचयित किया जा सकता है।

इस मिश्रण से श्वेत रक्त कोशिकाओं को बिना किसी अवांछनीय परिवर्तन के 6 महीने की अवधि तक पृथक किया जा सकता है। इस प्रक्रिया को चित्र क्रमांक-1 में दर्शित किया गया है।

परिणाम और विचार - विमर्श

पृथक्कृत श्वेत रक्त कोशिकाओं के भौतिक लक्षण

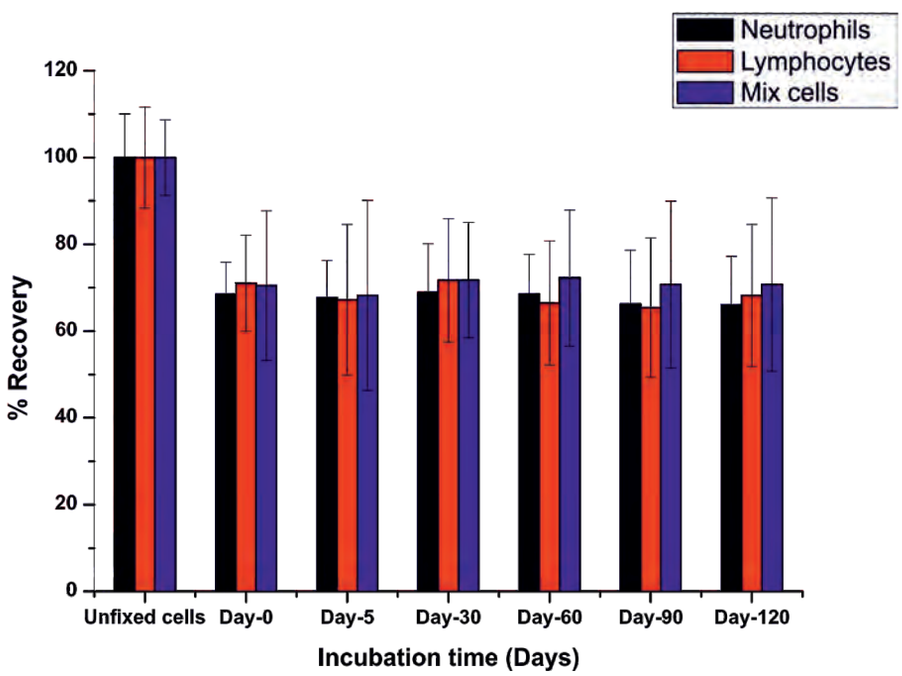
स्थिर किये गये रक्त से 120 दिनों तक, अलग - अलग समयांतराल पर विभिन्न प्रकार की श्वेत रक्त कोशिकायें पृथक की गईं, जिसे चित्र संख्या-2 में



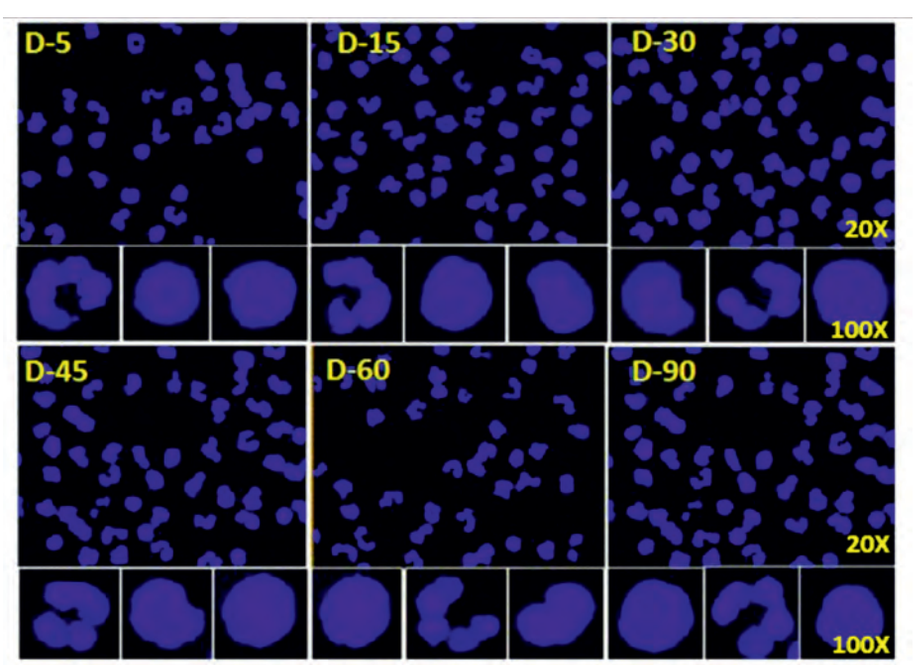
चित्र क्रमांक-1: रक्त संग्रह तथा संचयन की विधि का आरेखीय वर्णन

दर्शाया गया है। इस प्रक्रिया में 50 से 75 % तक श्वेत रक्त कोशिकायें पुनः प्राप्त हुईं, जो कि एक अच्छा अनुपात है। चित्र क्रमांक-3 से प्रमाणित होता है कि सूक्ष्मदर्शी द्वारा विभिन्न प्रकार की श्वेत रक्त कोशिकाओं को देखा और पहचाना जा सकता है। पृथक्कृत श्वेत रक्त कोशिकाओं के मूलभूत भौतिक लक्षण सामान्य पाए गए हैं तथा उनमें किसी प्रकार का परिवर्तन प्रदर्शित नहीं हुआ है।

न्यूट्रोफिल सबसे अधिक मात्रा में पाये जाने वाली श्वेत रक्त कोशिका होती है। संपूर्ण श्वेत रक्त कोशिकाओं में इनका अंश 40 - 70 % तक होता है। न्यूट्रोफिल के केन्द्रक में सामान्य रूप से 3 से 5 अनुभाग दिखे, जो कि इसकी पहचान का प्रमुख चिन्ह है। लिम्फोसाइट्स की पहचान उसके गोल केन्द्रक के आधार पर, सूक्ष्मदर्शी की सहायता से की गई और सामान्य पायी गयी, ये सभी श्वेत रक्त कोशिकाओं का 20 % से 40 % हिस्सा होते हैं। मोनोसाइट्स की पहचान उसके गुर्दे के आकार वाले केन्द्रक के आधार पर की गई, इनकी संख्या तुलनात्मक रूप से कम होती है (3 से 8 %)। इओसिनोफिल और बेसोफिल की संख्या न्यूनतम होती है तथा इसे सूक्ष्मदर्शिक परीक्षण से नहीं पहचाना जा सकता है। अन्ततः हम यह कह सकते हैं कि पुनर्प्राप्त श्वेत रक्त कोशिकाओं के मूलभूत भौतिक लक्षण में स्थिरन की प्रक्रिया से किसी प्रकार का कोई परिवर्तन नहीं पाया गया है।



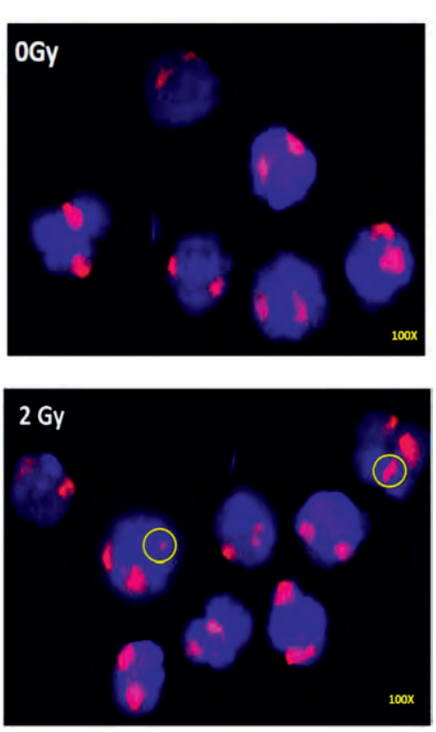
चित्र क्रमांक-2: विभिन्न समयांतराल पर पृथक्कृत श्वेत रक्त कोशिकाओं का अनुपात



चित्र क्रमांक-3: विभिन्न समयांतराल पर पृथक्कृत श्वेत रक्त कोशिकाओं का सूक्ष्मदर्शीय दृश्य

विकिरण प्रेरित गुणसूत्रीय जैवचिन्ह का निर्धारण - लिम्फोसाइट्स में गुणसूत्र स्थान-विनिमय

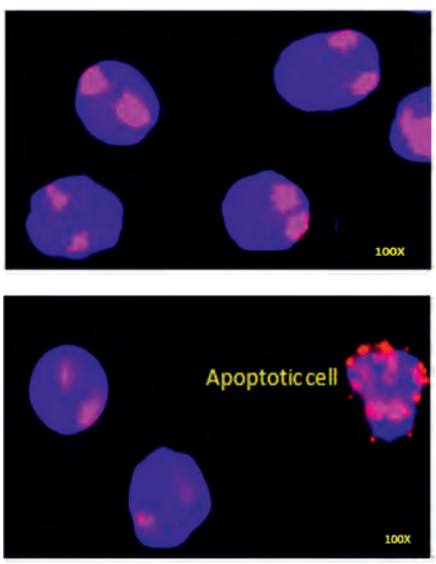
कई दशकों से कोशिका आनुवांशिकी पर आधारित तकनीकी जैवमात्रामिति के लिये सर्वोच्च विधि मानी जाती रही है। गुणसूत्र स्थान-विनिमय का इसमें एक समुचित स्थान है। यह तकनीकी दशकों पुरानी अवशोषित विकिरण मात्रा का मापन कर सकती है। अलग अलग समयांतराल पर पृथक कृत लिम्फोसाइट्स में, गुणसूत्र स्थान-विनिमय का पारीक्षण जी-0 फ्लूरोसेंस इनसिटू हाईब्रिडाइजेशन (G0-Fluorescence In-situ Hybridization, G0-FISH) द्वारा किया गया (चित्र क्रमांक-4), और यह कई महीनों तक संरक्षित पाए गये। एफ. सी. एस. का उपयोग कर विकिरण सम्बन्धी दुर्घटना - स्थल पर ही रक्त नमूनों का स्थिरन, एक व्यावहारिक दृष्टिकोण हो सकता है। चित्र क्रमांक-4 में बाईं ओर नियन्त्रित तथा दाईं ओर 2 Gy विकिरण डोज़ प्राप्त नमूनों को दर्शाया गया है। चित्र से दर्शित होता है कि दोनों प्रकार के नमूनों में गुणसूत्रों की स्थिति यथावत बनी रहती है।



चित्र क्रमांक-4: विकिरण प्रेरित लिम्फोसाइट्स में गुणसूत्रीय स्थान-विनिमय, जी-0 फ्लूरोसेंस इनसिटू हाईब्रिडाइजेशन (G0-FISH)

रेडिओधर्मी विकिरण- प्रेरित लिम्फोसाइट की कार्यक्रम कोशिका मृत्यु (एपापटोसिस) एवं जैवमात्रामिति

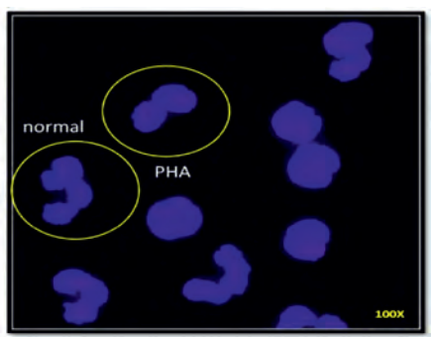
अंतर्राष्ट्रीय परमाणु ऊर्जा एजेंसी (आई.ए.इ.ए.) दिशानिर्देश के अनुसार, कार्यक्रम कोशिका मृत्यु को जैवमात्रामिति का चिन्ह माना जा सकता है। इस सन्दर्भ में हमारे शोध दल ने लिम्फोसाइट्स को उच्च रेडिओधर्मी विकिरण मात्रा से विकिरणित कर, एफ. सी. एस. में मिश्रित कर, सामान्य तापमान पर कुछ महीनों के लिए उदभवित (incubate) किया। फिर अलग अलग समयांतराल पर लिम्फोसाइट्स को पृथक किया गया। जैसा कि चित्र संख्या-5 से प्रतीत होता है, G0-FISH तकनीकी द्वारा कार्यक्रम कोशिका मृत्य का पता लगाया गया। इस प्रक्रिया द्वारा स्थिर की गई कोशिकाओं में किसी प्रकार का कोई परिवर्तन नहीं देखा गया। इस तकनीकी को अवशोषित विकिरण की मात्रा के मापन हेतु प्रयोग में लाया जा सकता है।



चित्र क्रमांक-5: विकिरण प्रेरित लिम्फोसाइट्स में कार्यक्रम कोशिका मृत्यु (एपोप्टोसिस) - जी-0 फ्लूरोसेंस इनसिटू हाईब्रिडाइजेशन (G0-FISH)

विकिरण-प्रेरित न्युट्रोफिल के केन्द्रक में आकृतीय परिवर्तन

एक सामान्य मानव परिपक्व न्यूट्रोफिल के केंद्रक में आमतौर पर तीन या चार खंड होते हैं जो कि एक पतले धागा नुमा डी. एन. ए. से जुडे होते हैं। एक प्रसिद्ध आनुवांशिक रोग (सूडो पेल्जर ह्यूट विसंगति) में, अस्थिमज्जा तीन या चार खंड के स्थान पर, दो-खंड वाले असामान्य न्यूट्रोफिल उत्पादित करता है। यह असामान्य न्यूट्रोफिल सूक्ष्मदर्शी की सहायता से आसानी से पहचाना जा सकता है। हाल ही में एक खोज में यह पता चला है कि, विकिरण भी दो-खंड वाले असामान्य न्यूट्रोफिल के उत्पादन को प्रेरित कर सकता है तथा इसे अवशोषित विकिरण की मात्रा के मापन हेतु प्रयोग में लाया जा सकता है। जैसा कि उपरोक्त जिक्र किया जा चुका है, एफ. सी. एस. कोशिका तथा केंद्रक को मूलभूत स्थिति में स्थिर कर देता है, हमने अपनी प्रयोगशाला में रक्त को एफ. सी. एस. में मिश्रित करके सामान्य तापमान पर कुछ महीनों तक उदभवित (incubate) किया, तथा अलग अलग समयांतराल पर श्वेत रक्त कोशिकाओं को पृथक कर, न्यूट्रोफिल के केन्द्रक की आकृति का अध्ययन किया। इसकी आकृति में किसी प्रकार का कोई परिवर्तन नहीं दिखा। ज्यादातर न्यूट्रोफिल्स के केन्द्रक में तीन या चार खंड दिखे, जो कि सामान्य है, तथा कुछ दो-खण्डीय केन्द्रक वाले न्यूट्रोफिल्स भी दिखे, जो की परिप्रेक्ष्य माने जा सकते हैं (चित्र क्रमांक-6)। अतः एफ. सी. एस. जैवमात्रामिती के साथ-साथ आनुवांशिक विकारों की जाँच करने में भी उपयोगी साबित हो सकता है।



चित्र क्रमांक-6: सामान्य तथा सूडो पेल्जर ह्यूट न्यूट्रोफिल्स, स्थिर रक्त से पृथक किया हुआ।

अन्य उपयोग

सैद्धांतिक रूप से एफ.सी.एस. का प्रयोग वाह्य कोशिकीय या प्लाज्मा में उपस्थित गुणसूत्रों / डी.एन.ए. के स्थिरन में किया जा सकता है जो कि कर्क रोग की जांच का एक अहम तरीका है। एफ.सी.एस. द्वारा स्थिर की गयी कोशिकाओं में क्षतिग्रस्त डी.एन.ए. का पता लगाने के लिए धूमकेतु आमापन विधि (Comet Assay) का प्रयोग भी किया जा सकता है। विभिन्न प्रकार के शोध कार्यों में जैविक नमूनों के स्थिरन, संचयन एवं परिवहन में भी एफ.सी.एस. उपयोगी हो सकता है।

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