Abstract:
Smuggling, Trafficking, Illegal possession and Theft of nuclear or radioactive material have increased and have become a worldwide problem. This has result in development of new discipline “Nuclear Forensics”. The rate of illegal trafficking and smuggling of the nuclear or radioactive material has increased, so the proper investigation and identification of nuclear material to prevent illegal trafficking of nuclear or radioactive substance has become necessary. Once the nuclear or radioactive material is seized, the identification of the sample and determination of its source is of prime importance. The nuclear trafficking or smuggling creates the potential threat due to its hazards. In this paper an overview is given regarding the samples and the techniques used for the investigation purpose.

Key words: Uranium, Plutonium, Proliferation, Nuclear Material

Introduction:
The element is made up of basic building blocks known as atom. Each atom has centrally located nucleus. The nucleus has protons and neutrons. The stability of nucleus depends on the protons and neutrons. The nucleus having excess of neutron gets stable by converting into proton while the nucleus having the excess of protons gets converted to neutron. The conversion of protons to neutrons or neutrons to protons emits the certain radiations or particles. This phenomenon of spontaneous disintegration of the nucleus is called the radioactivity and the nucleus that emits the radiation is called the radionuclide. The rate at which the radionuclide decays in the given amount of radioactive material is called its activity and is measured in becquerel (Bq). [2]

Illicit trafficking of the nuclear material leads to the nuclear proliferation and the construction of the nuclear of radioactive explosive devices. The involvement of unauthorized group creates the potential threats. [1]. The term nuclear forensics was first coined in context of combating nuclear smuggling and trafficking. According to the International Atomic Energy Agency, the nuclear forensics can be defined as: “the analysis of intercepted illicit nuclear or radioactive material and any associated material to provide evidence for nuclear attribution” while attribution refers to “the process of identifying the source of nuclear or radioactive material used in illegal activities, to determine its point of origin and routes of trafficking and ultimately the prosecution of those responsible.’[3] The nuclear forensic also includes analysis of materials obtained from the unused nuclear materials, or from the debris of nuclear explosion. Nuclear forensic works best with law enforcement agencies, traditional forensics, radiological protection dosimetry etc. It is based on the physical and chemical analysis of the radioactive materials and the analysis of impurities.[4]

In nuclear forensics, the samples are analysed to the detection of isotope of uranium and plutonium isotopes. The uranium-235, uranium-238, and plutonium -239 are used as nuclear fuels in the nuclear the nuclear explosive devices. This is known as nuclear characterization. It is the study of the elemental composition of the seized material for the detection of the presence of any radioactive isotopes. It includes study of the dimensions such as particle size, and distribution of shape.
The nuclear explosive devices produce a large amount of energy from small quantity of radioactive material. The primary advantage of nuclear weapon is orders of magnitude of energy obtainable than the chemical explosives. They constitute the significant long lasting hazard to the population over a significance distance [2].

**Samples and Specimens:**

The handling, collection, preservation and storage of the sample are important to maintain the quality and for the interpretation of results and data. In forensic investigation, the specimen must be obtained that are representative of questioned material. The sampling protocols and storage conditions of the specimens are more important for the valid data and interpretation of results from the analysis of those specimens. The specimen includes the various matrices such as soil, flora and fauna, air and water. [2]

**Matrices:**

Soil and sediments are good source of undeclared nuclear material and serve as good sorbents of actinide, lanthanide and transition metal analytics. These chemical classes correspond to nuclear fuels, fission products and activation products respectively. Nuclear indicators can be isolated from soil using techniques like ion exchange or by extraction. The terrestrial vegetation can collect several orders of magnitude of the nuclear material released in environment. Plants with large surface areas such as pine needles, grasses etc. are good source of nuclear material. The exposed portion can serve as an air collector while root system ambient ground water and soil. The rooting strength and growing cycle are main factors that need to be studied during the ground water sampling. Many radioactive traces can be found in the aqueous effluents. Also the traces of the material can be found in the aquatic animals. The water pump is used for the collection of nuclear material from the aqueous medium. The aqueous medium is pumped through the filter cartridge consist of mix-bed ion exchange resin for dissolved species. However air sampling has been most effectively employed in study of nuclear proliferation for many years. The air sampling is performed with the high volume air collectors through the rates of tens of cubic meters per minute. The tools that are used for the collection of the specimens may vary but most commonly used are disposable gloves, spatulas, forceps, sticky tapes, razor blades, transfer pipette, swipe papers etc. The specimen of radioactive material includes the traces of radioactive devices along with the transport material such as hair, soil, dust, paper pollen etc. The material collected for the analysis contains the traces of radioactive material so containers made of proper material should be used for the packaging purpose such as Teflon or can be packaged in a multilayer zip lock polythene bags.[2]

**Analytical Techniques in Nuclear Forensics:**

Investigation in forensic science starts after the material is seized. The aim of analysis of material is to determine its source, its mode of production, the batch of production, the plant of production, route of trafficking and owner of the material. The composition of nuclear material depends on the nature of its production. The nature of production of material constitutes its elemental and isotopic composition. The determination of origin or mode of production from the composition of nuclear material is called “Nuclear fingerprinting”. Nuclear material is either obtained from uranium or from the nuclear reactors. In the nuclear reactors, the uranium is subjected to the neutrons due which it gets convert in to trans-uranium elements. The analytical techniques in nuclear forensics are applied to gain the maximum information regarding the nuclear material. [5]

The analytical techniques employed in forensic investigation of nuclear material are radio analytical techniques along with physical and chemical analytical methods. The most important step is the proper preservation of
samples to prevent hazard. Before subjecting the samples to the analytical techniques the physical inspection is done which may reveal the useful information such as physical form, its shape and geometry etc. If the fuel pellets are seized, its macroscopic quantities should be measured which is helpful in the determination of the reactor where it was used. Seizing of nuclear materials may take place under any circumstances.

**Chronometry:**
The determination of age of nuclear sample seized is of prime importance in nuclear forensics. The chronometry is the dating technique used for the identification of the sample source as well the procedures that have been carried out on the material. Many age dating relationships can be used such as $^{234}\text{U}/^{238}\text{Th}$, $^{241}\text{Pu}/^{241}\text{Am}$ etc. Nuclides related to the radioactive decay process will have relative sample concentration that may be predicted using parent-daughter growth relations and relevant half-lives. The radioactive element decays at the rate determined by the amount of isotope in a sample and half life of parent isotope, the relative amount of decay products is compared with parent isotope to calculate the age of the sample. The chronometry is very useful in the archaeology and geology where the age of organic material is to be determined.[6]

**Alpha Spectrometry:**
The alpha particles are the helium nucleus. The helium nucleus has mass number equals to 4 and atomic number equals two. When any radioactive substance emits the alpha particles, its mass number gets decreased by 4 units and atomic number by 2 units.

$$mX \rightarrow m-4X + \text{He}$$

The alpha radiations are emitted by the combination of nucleons in the nucleus. During each decay of nucleus, only one alpha particle is emitted, but each decay may result in the emission of alpha particles of different energy because any two protons and neutrons in the nucleus may combine to form the alpha particle that is emitted. The alpha spectroscopy involves the detection of the material which emits the alpha radiations. The alpha particles have characteristic energy. Alpha spectrometry is a destructive technique because it requires the preparation of the sample preparation prior to analysis[5]. The alpha emitters have energy levels between 3 MeV to 10 MeV. Alpha particle energies for different nuclei can also be similar (e.g., $^{238}\text{Pu}$ & $^{241}\text{Am}$). The spectrum of this to nuclei overlaps which cannot be resolved and thus can be measured only as sum of activities of two nuclei. The principle of distinguishing the energies of alpha particle is that it forms an ion pair with the electron. Higher is the energy of the alpha particle, more the number of ion pair will be formed with electron. The electron density is more in solids, therefore the path length of alpha particles get decreased in solids. The formation of each ion pair will results in signal called pulse. The pulse size varies with energy of alpha particles [13].

**Gamma Spectroscopy:**
Gamma spectroscopy involves the identification of the samples that emits the gamma particles or gamma radiations. The gamma spectroscopy is non destructive in technique because it does not require any nuclear
standards or calibrated standards, so it is preferred for the investigation of the seized samples. Gamma rays are emitted when the nuclei of higher energy decays to the nuclei of lower energy. The radioactive material decays in this way emit the gamma radiation of specific energy which is unique to that element.

\[\frac{m_X}{n} \rightarrow \frac{m}{n}Y + \gamma\]

Codes like FRAM (Fixed-Energy, Response Function Analysis with Multiple Efficiency), MGA (Multi-Group Analysis for Uranium) are used for the detection of gamma particle. The FRAM is developed by Los Alamos, used to study the pulse height spectra by the gamma ray detectors. FRAM is primarily used to determine the isotopic composition of plutonium in the seized nuclear material. The MGA is a code used for the determination of the uranium content in the nuclear material. The information is derived from the \(^{235}\text{U}\) and \(^{238}\text{U}\) emission in 90-94 KeV energy range. The measured peak intensity is directly related to the number of atoms.[5,11, 12]

**Thermal Ionisation Mass Spectroscopy**

Thermal Ionisation Mass spectroscopy is a single elemental analysis technique which requires the sample to be present in the purest form. The technique is very useful for the determination of isotopic composition of uranium, plutonium and thorium. The technique is gives the results with high precision and accuracy. The disadvantage of this technique is that it requires laborious sample preparation.[5, 8]

**Inductively Coupled Plasma-Mass Spectroscopy (ICP-MS)**

ICP-MS is a very useful technique used for the detection of the radioactive element traces in the sample of nuclear fuel. The technique is used for the determination of the neutron absorbing nuclei. The nuclear fuel is a material that can be burned by nuclear fission or fusion to gain nuclear energy. The uranium-235 and plutonium-239 are most commonly used as nuclear fuels in the reactor. The fuel can be obtained in various forms such as oxides, nitrides, carbides or molten salts. Uranium oxide from dust and environmental particles can be analysed using this technique. [5, 7, 10]

**Secondary Ion Mass Spectroscopy (SIMS):**

SIMS is useful technique for the analysis of powder material or micro particles. With the help of SIMS, elemental and isotopic information can be gained. SIMS is applicable when very less amount of sample is seized or the sample is powder mixture and the each constituent is to be investigated individually. The sample in SIMS is characterized by isotope ratio.[5]

**Profilometry:**

Profilometry is the study of surface characteristics. The technique is applied to study the surface characteristics like roughness on the surface of the nuclear fuel pellets. The surface characteristic acts as “fingerprint” of the sample which is very useful the purpose of identification. The pellets are prepared either by wet grinding or dry grinding. Depending upon the grinding methods used by the plant, the surface characteristics of the fuel pellets will vary from each other which help in identification.[9]

**Scanning Electron Microscopy:**

SEM is used to study the morphology of the nuclear samples. In SEM, the electron beam (primary electron) is used for the sample study. Depending upon the sample, the electrons are scattered back from the sample which are known as secondary
electrons. The SEM is used when the sample seized is in powder form. Under the high magnification, the morphology of the sample is studied. Different components can be studied using the particle morphology and the composition. The study of particles by the SEM gives the information regarding the production process of the samples.[5]

Discussion:
The analytical techniques discussed above are helpful in the identification of the radioactive samples. The analytical techniques in nuclear forensics are radiometric and micro structural in nature. The techniques like Chronometry, Alpha Spectroscopy, Gamma Spectroscopy, Secondary Ion Mass Spectroscopy, Inductively Coupled Plasma Mass Spectroscopy and Thermal Ion Mass Spectroscopy are radiometric techniques, while Profilometry and SEM are the micro structural techniques. Radiometric techniques detect the type of radiation emitted by the radioactive substances. The analytical techniques should be applied depending upon the quantity of sample. The Gamma spectroscopy, SEM etc. are non destructive techniques so it should be preferred first for the analysis. The TIMS is very useful when the sample is pure and it gives results with very high accuracy. The powder samples or heterogeneous samples can be analysed by SEM to study the morphology of each grain and SIMS. The Profilometry is very useful technique for the identification of the plant where the fuel pellets are used. The identification is made possible using the study of the surface characteristics. All the analytical techniques discussed are employed in the investigation and identification of the radioactive samples which are trafficked, smuggled or obtained from post nuclear explosion scene.

References: