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Application of medium energy plasma focus device in study of radioisotopes

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ABSTRACT

Pulsed neutrons generated in a plasma focus device are used for the thermal neutron activation analysis (TNAA) of selected three elements having widely different half-lives varying from a few seconds to a few days [Dysprosium (Dy), Manganese (Mn) and Gold (Au)]. Neutron pulse having strength of $(1.2 \pm 0.3) \times 10^9$ neutrons/pulse with a pulse width of 46 ± 5 ns is produced by "MEPF-12" device operated at a filling gas (deuterium + 0.5% krypton) pressure of 3 mbar. The fast 2.45 MeV D–D neutrons are thermalized before irradiating the sample. The decay gammas from the radioisotopes ^{165m}Dy ($T_{1/2} = 1.26 \text{ min.}$), ⁵⁶Mn ($T_{1/2} = 2.58 \text{ hrs.}$), and ¹⁹⁸Au ($T_{1/2} = 2.69 \text{ days}$) produced via reactions, ¹⁶⁴Dy(n, γ) ^{165m}Dy, ⁵⁵Mn(n, γ) ⁵⁶Mn, and ¹⁹⁷Au(n, γ) ¹⁹⁸Au respectively are counted off-line in a lead shielded well type 76 × 76 mm² Nal(Tl) detector coupled to a calibrated 2048 channel analyzer. The values of half-lives evaluated from the measured decay gammas, 1.43 ± 0.3 min., 2.56 ± 0.5 hrs. and 2.84 ± 0.6 days respectively for the radioisotopes of Dy, Mn and Au, are seen to be close to the values reported in literature.

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1. Introduction

Neutron activation analysis (NAA) is one of the popular nuclear analytical techniques for identification and guantification of different elements in any sample. Further, thermal neutrons are mostly used for NAA owing to the high capture cross section for many elements and the technique is referred as thermal neutron activation analysis (TNAA). TNAA is based on the activation of stable isotopes through capture of thermal neutrons followed by the determination of temporal profile of the activity of the radioisotopes so produced. Some of the commonly used neutron sources for TNAA are ²⁵²Cf, D-T LINAC and nuclear reactors. A major disadvantage of using these neutron sources is high source background itself and hence the high minimum detection limit (MDL) for sample of a given nuclear material. This necessitates putting additional shielding around the sample to reduce background which in turn makes the system bulky. Also, operation and maintenance of these neutron sources are costlier and somewhat complex.

Plasma focus (PF) is a well-established pulsed neutron source known for decades [1,2]. It is simple in operation and a relatively

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https://doi.org/10.1016/j.physleta.2018.09.015 0375-9601/© 2018 Elsevier B.V. All rights reserved. low cost device which produces a wide range of radiations [microwaves to hard X-rays, energetic charged particles] and neutrons if operated with deuterium (D₂) or deuterium-tritium (D-T) gas mixture. The duration of neutron pulse being of a few 10 s of ns only, ensures negligible background before and after experiments. It is based on a kind of Z-pinch discharge that uses self-generated magnetic field to accelerate the plasma axially within the coaxial region and then compress it to form a short lived (\sim 10 to 100 ns), high density $(\approx 10^{25}\ m^{-3})$ and high temperature (1 to 2 keV) plasma pinch at the open end of the anode. The disruption of the pinched plasma column due to various plasma instabilities results in creation of high electric and magnetic fields which accelerate the ions and electrons to high energies. It is unambiguously demonstrated that using deuterium (at times with tritium) as a filling gas, neutrons are produced due to fusion reactions. The energy of neutrons as expected is close to 2.45 MeV (for D-D) and 14.1 MeV (for D-T). The neutron emission from a PF device is anisotropic in nature with more numbers in axial than that in radial direction. The anisotropy factor i.e. ratio of neutron flux in axial to that in the radial direction, is typically observed [3] as 1.2 to 3. The mechanism of neutron production points to their origin being thermal as well as non-thermal [4] with a major $(\approx 90\%)$ contribution being attributed to non-thermal. An order of magnitude enhancement in neutron yield has been reported in a







Moderator

Sample

Evacuation

&

Gas filling

Anode

Cathode

Quartz

Triggered spark gap

(HDPE)



vield. PF device has found a number of applications [6] including as an ion source which are widely reported. However, as a neutron source even feasibility studies are still limited. A PF device generating $(3-4) \times 10^8$ neutrons/pulse has been reported [7] to be used in fast neutron activation analysis of gold by inelastic scattering ¹⁹⁷Au $(n, n'\gamma)^{197m}$ Au. A compact PF device producing 2×10^8 neutrons/pulse has been used [8] for detecting water contents of a few percent in volume placed about 8.5 cm away from the PF chamber by neutron scattering. A recent report [9] also shows its use for a single-shot detection of illicit materials and explosives. The main principle of the technique is based on measurement of time of flight of neutrons scattered from nuclei of various elements. An interesting application of a medium energy plasma focus "MEPF-12" operating at 11.5 kJ was demonstrated for non-destructive assay of fissile materials (²³⁵U) where using delayed neutrons [10] as well as delayed gamma counting [11], minimum detection limit (MDL) of 18 mg and 14 mg of ²³⁵U mass respectively was observed. Here we report on using the MEPF-12 device as a pulse neutron source for thermal neutron activation analysis of Dysprosium (Dy), Manganese (Mn) and Gold (Au), chosen due to their widely differing half-lives from a few tens of seconds to a few days. The energy spectrum of decay gammas from radioisotopes and their respective half-lives have been measured. The next section describes the method for experiment followed by results with discussions in the subsequent section after that. The work is summarized and concluded at the end.

2. Experimental method

A series of experiments have been carried out for activation of Dy, Mn and Au, using MEPF-12 device and measurement of their half-lives. The schematic of the setup along with various diagnostics are displayed in Fig. 1. The PF set up comprises of a capacitor bank and a matching PF device. The capacitor bank consists of four energy storage capacitors (each 10 μ F, 25 kV). The PF device is of coaxial squirrel-cage geometry. The PF electrodes are separated at one (closed) end by a quartz tube of 2 mm thickness whereas top end is kept open. The dimension and other details of PF device are described elsewhere [12]. The experimental chamber has four

side ports. One of the ports is used for evacuation of experimental chamber and filling of operation gas. The filling gas pressure is measured using a capsule dial gauge (0–25 mbar).

The diagnostics for characterizing PF device include a miniature pickup coil for monitoring current derivative, Geiger–Muller (GM) counter based silver activation detector (SAD) for measurements of neutron yield and plastic scintillator detector (PSD) coupled to a photomultiplier tube for recording time resolved neutron and X-ray emissions. For neutron yield anisotropy (ratio of neutron yield in the axial to that of in the radial direction) measurement, two identical silver activation detectors were placed at 1 m distance from anode tip in the radial and axial direction w.r.t. PF electrode axis. The PSD detector was positioned at 3 m in the radial direction. The separation time between the X-ray pulse and the neutron pulse is used for the evaluation of neutron energy using time of flight method. All the signals from the detectors are recorded in digital storage oscilloscope (1 GHz, 5 GS/s) kept inside an electromagnetic noise shielded enclosure.

The decay gamma counts from thermal neutron activated samples were measured using a well type $76 \times 76 \text{ mm}^2 \text{ Nal(Tl)}$ detector coupled to a 2048 channel analyzer. The multichannel analyzer (MCA) has been calibrated using gammas of varying energies emanating from different sources viz. ²⁴¹Am (59.5 keV), ¹³⁷Cs (661.5 keV), and ⁶⁰Co (1173 and 1332 keV).

3. Results and discussion

For plasma focus operation, experimental chamber is initially evacuated to a base pressure of less than 10^{-5} mbar by a rotarydiffstac pump. It is then filled with deuterium mixed with krypton gas (0.5%) at low pressure (1 mbar). Initially five to six shots are taken at 20 kV charging voltage for conditioning of insulator surface. These shots are required to form uniform plasma sheath over insulator surface and termed as "insulator-conditioning shots". For each shot, current derivative signal is recorded. In these shots, either mild or no dip, indicative of plasma focus formation, in the current derivative signal is observed. A minimum time gap of 30 min. between two shots was maintained for cooling of insulator material to avoid its breakdown due to rise in temperature during plasma focus operations. On conditioning of insulator surface, sharp and strong dip near zero cross-over (maximum peak current) of current derivative signal was observed. The form of dip is signature of plasma focus characteristics. The sharp and large dip is an indication of strong focus formation and hence high neutron yield. The filling gas pressure of deuterium and krypton admixture was optimized at a charging voltage of 24 kV. The sharp and strong dip at maximum peak current was observed at 3 mbar. A typical current derivative signal indicating plasma focus formation is shown in Fig. 2.

The measurement of the neutron yield, its anisotropy and the energy evaluation of neutrons through the time-of-flight were carried out at optimized pressure. The maximum neutron yield of $(1.2 \pm 0.3) \times 10^9$ neutrons/pulse in the radial direction was observed. The neutron pulse duration (FWHM) was measured to be 46 ± 5 ns. The anisotropy factor of (1.3 ± 0.2) was estimated at 3 mbar. The observed maximum neutron yield is found to be nearly same as the neutron yield of 1.03×10^9 neutrons/pulse estimated from scaling law $(Y_n = 1.7 \times 10^{-10} I_0^{3.3}; I_0 = 491 \text{ kA})$ for neutron emissions in PF devices [13]. It is also found to be close to neutron yield at same capacitor bank energy/current reported by others [14,15]. The two pulses observed in the PSD (Fig. 2) are of hard X-rays (> 10 keV) and neutrons. They are separated due to difference in their time of flights which is also used to infer the neutron energy (assuming both the hard X-rays and neutrons are produced at the same time) estimated to be 2.45 ± 0.2 MeV.



PSD

SAD

Pickup coil-

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