AIP | Review of Scientific Instruments

Small plasma focus as neutron pulsed source for nuclides identification

M. Milanese, J. Niedbalski, R. Moroso, M. Barbaglia, R. Mayer et al.

Citation: Rev. Sci. Instrum. **84**, 103501 (2013); doi: 10.1063/1.4823522
View online: http://dx.doi.org/10.1063/1.4823522
View Table of Contents: http://rsi.aip.org/resource/1/RSINAK/v84/i10
Published by the AIP Publishing LLC.

Small plasma focus as neutron pulsed source for nuclides identification

M. Milanese,[1,2,a] J. Niedbalski,[3] R. Moroso,[1,2] M. Barbaglia,[1,2] R. Mayer,[4] F. Castillo,[5] and S. Guichón[2]

[1]*Centro de Investigaciones en Física e Ingeniería del Centro de la Provincia de Buenos Aires (CONICET-UNCPBA), Pinto 399, Tandil 7000, Buenos Aires, Argentina*
[2]*Universidad del Centro de la Provincia de Buenos Aires (CONICET-UNCPBA), Pinto 399, Tandil 7000, Buenos Aires, Argentina*
[3]*CONICET(Consejo Nacional de Investigaciones Científicas y Técnicas), Rivadavia 1917, Buenos Aires, Argentina*
[4]*CNEA (Comisión Nacional de Energía Atómica), Av. Bustillo 9500, San Carlos de Bariloche, Rio Negro, Argentina*
[5]*UNAM (Universidad Nacional Autónoma de México)–Circuito Exterior s/n, Ciudad Universitaria, Delg. Coyoacán, P.O. Box 70-543, México DF, Mexico*

(Received 7 August 2013; accepted 12 September 2013; published online 2 October 2013)

In this paper, we present preliminary results on the feasibility of employing a low energy (2 kJ, 31 kV) plasma focus device as a portable source of pulsed neutron beams (2.45 MeV) generated by nuclear fusion reactions D-D, for the "*in situ*" analysis of substances by nuclear activation. This source has the relevant advantage of being pulsed at requirement, transportable, not permanently radioactive, without radioactive waste, cheap, among others. We prove the feasibility of using this source showing several spectra of the characteristic emission line for manganese, gold, lead, and silver. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4823522]

I. INTRODUCTION

Neutron beams have considerable scope in several application fields, especially in non-invasive research, because of their possibility of deeply penetrating the structure of condensed matter. The identification of chemical elements in the composition of molecular structures using neutron beams has immense utility in research and industrial laboratories. This could be done by neutron activation of nuclei and the subsequent registration of characteristic emitted gamma radiation (*Neutron capture Gamma-ray Activation Analysis*, NGAA), by instantaneous *Prompt Gamma Neutron Activation Analysis* (PGNAA) and/or by Delayed decay *Gamma Neutron Activation Analysis* (DGNAA).[1–4] The practical implementation of this analysis method requires, mainly, the use of neutron beams in the thermal (E = 0.025 eV, about) and epithermal (0.025 eV < E < 100 eV, approximately) energy range. That is because numerous nuclides have considerable neutron capture cross section for the thermal and epithermal ranges. In general, the cross section of any nuclide has an "inverse power" law on the neutron energy in the range from very low (some meV) to hundreds of keV. Moreover, many nuclides present resonance peaks at high energies.

Once the substance is irradiated, the gamma decay spectrum of photons emitted by each component nuclear species is unique; it is the characteristic "signature" that allows identification and eventually quantification.

Neutron sources for irradiation normally are constituted by radioisotopes of relatively high activity (A) and medium and long decay lifetime (τ), such as the actinide ^{252}Cf (A = 170 × 10^6 (n/min)/μg and τ = 2.64 yr), or mixtures of α particles' emitter such as ^{241}Am or ^{226}Ra with ^{9}Be powder. These radioisotopes are commonly obtained from waste resulting from the "burning" of fissile material typically used in nuclear reactors. These are specimens of difficult affordability, are expensive, and are dangerous for manipulation. Other kind of sources are nuclear fission reactors or particle accelerators (cyclotrons or LINACs); they are expensive and very impractical to get "*in situ*" measurements compared with the machine used here.

This paper informs an assessment on the feasibility of a dense plasma focus (DPF) device of relatively low operating power (2 kJ, 31 kV) as clean source (non-radioactive), operable requirement, high security in handling, and safe transportation, of pulsed beam D-D fusion neutrons (2.45 MeV), for use in the analysis "*in situ*" of substances by nuclear activation. One of the aims of the present work is the future use of a plasma focus for detecting specific chemical elements in molecular structures' constituents of hazardous substances such as explosives, toxins, fissile material, etc.,[5,6] potentially to be deployed in high-volume port terminals' containers. This research includes the results of experiments to get a reduction of the energy of neutron beams provided by the DPF device to thermal and epithermal levels required to optimize the activation process. Results of the application of this technique to commercially available Ag, Mn, and Au are also given. Besides using energetic fusion neutrons (2.45 MeV), the activation of Au and Pb samples has been detected.

II. EXPERIMENT AND RESULTS

A. The source of energetic neutrons

The source of energetic neutrons used for irradiation experiments is a Mather type dense plasma focus device named PACO[7] with 2 kJ of energy stored in a capacitor bank and

[a]Author to whom correspondence should be addressed. Electronic mail: malenamilanese@yahoo.com.ar

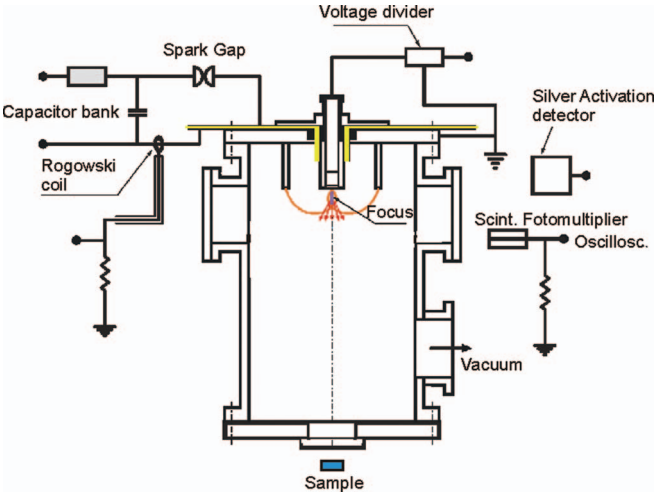

FIG. 1. Schematic of the PACO plasma focus device cross section and some diagnostics.

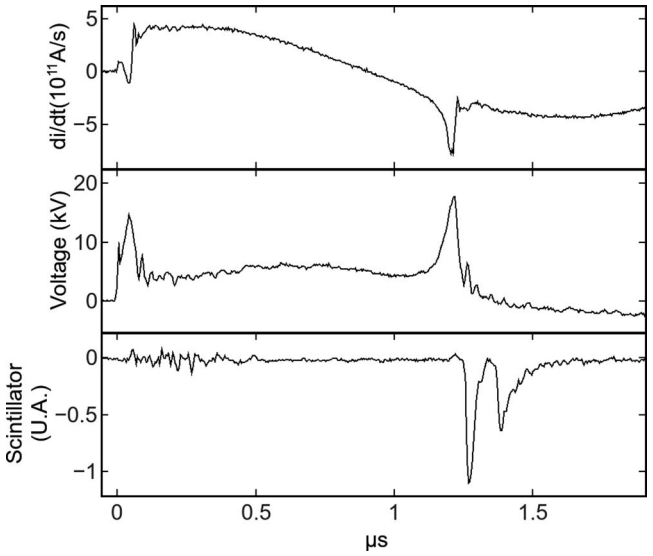

FIG. 2. Waveforms corresponding to time resolved discharge current derivative (top), anode-cathode voltage taken at the top of the coaxial gun (middle), and hard X-rays and fusion neutrons (bottom). Temporal swept: 200 ns/div. The hard X-ray and neutron pulses (lower trace) are separated by time of flight.

relatively small dimensions, placed in the National University of the Center of the Buenos Aires Province, Argentina, and operated within the limits of deuterium pressure range with neutron production.[8] A schematic of the DPF PACO is shown in Fig. 1.

This device has the feasibility of a simple mechanical modification of the support structure to allow its easy transportation. It normally operates as follows: a relatively fast discharge of high voltage (31 kV) and current (250 kA) is realized on a pair of coaxial electrodes under an atmosphere of D_2 at low pressure (1.2–1.8 mbar). As a result of self-compression of the deuterium plasma, a zone of high density and temperature (focus) is formed where very energetic deuteron and electron beams, hard and soft x-ray pulses,[9] and nuclear fusion reactions are generated. Therefore, a neutron flux of 2.45 MeV is produced in pulsed form, with variable duration (shot to shot) between about 80 and 150 ns (FWHM) and a total average irradiance of about 2×10^8 neutrons per pulse distributed in 4π sr.

Fig. 2 shows typical waveforms, simultaneously recorded, of the temporal evolution of hard X-rays and fusion neutrons (lower trace), anode-cathode voltage at the top of the coaxial gun (middle trace), and discharge current derivative (upper trace). The diagnostics used for the registration of the current derivative and voltage are a calibrated Rogowski coil and a calibrated fast resistive voltage divider, respectively. The time integrated, in each pulse, neutron yield is registered by a calibrated Silver Activation Counter (SAC). The time resolved X-ray pulses and neutrons are recorded by a plastic scintillator NE102A type, optically coupled to a photomultiplier tube (S-PMT) located approximately 2 m from the emission source (focus). This distance can accurately separate both photon and neutron pulses by a difference of time-of-flight. X-ray emission is a secondary process that occurs almost simultaneously with neutron emission,[10] as a result of the formation of a beam of very energetic (relativistic) electrons in the region of the dense and high temperature plasma during the final pinch stage. Experimental evidence suggests that its origin is associated with the bremsstrahlung beam of electrons interacting with the back of the hollow central electrode (anode) in the PACO device. Due to the large flux of electrons and their high energy, an insert of a high atomic number material (tungsten) is placed inside to optimize the Bremsstrahlung conversion efficiency. The PACO device has a non-uniform angular distribution of the emission of neutrons. This was determined previously[11] using solid state nuclear track detectors CR-39.

B. Moderation of pulsed beams of energetic neutrons

High density polyethylene (HDPE) was used to moderate the neutron flux up to the thermal and epithermal ranges proper to maximize the material activation response. The molecular structure of HDPE is constituted by a straight chain (unbranched) obtained from ethylene (C_2H_4). According to data provided by the manufacturer, this polymer has a density of 0.957 g/cm^3 and a molecular weight of 2.5×10^5 g/mol. HDPE was chosen, among other potential moderator materials, because of its present good capacity of moderation taking into account numerical simulations performed using the Monte Carlo method. Then, when energetic neutron beams irradiate a piece of HDPE, the low mean free path of neutrons (due to the high density of this type of polythene) and the huge amount of energy lost in the collisions (due to the high concentration of hydrogen atoms) allow us to obtain strongly moderated neutrons emerging from the material. The easy handling of this material is another advantage with respect to others such as wax or water. HDPE plates (45×45 cm^2 and 8 mm thickness) were arranged in parallel for making possible to modify gradually the moderator thickness; this allows increasing or decreasing the number of collisions of neutrons with hydrogen nuclei of the material; then, it is possible to achieve different levels of moderation.

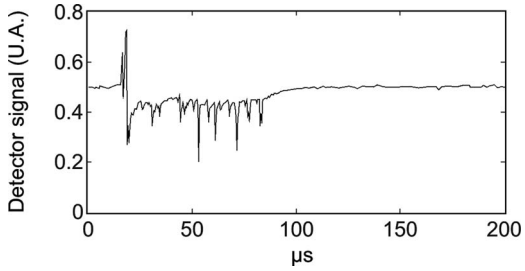

FIG. 3. A typical oscillogram of slow neutrons emerging from the moderator. Energetic range: 0.01–1 eV. The detector is a Bicron BC 702 Thermal Neutron scintillator coupled to Electron Tubes photomultiplier.

A series of experiments have been carried out in the DPF PACO to test the capacity of obtaining thermal and epithermal neutrons from a collimated fast neutron beam. For these experiments, the entire PACO was shielded with a series of wide paraffin plates allowing the fast neutrons to pass through a small orifice through a PVC tube to the collection zone. The emerging slow neutrons were recorded with a time-resolved detection system based on a Bicron BC 702 Thermal Neutron scintillator that detects neutrons only in the range of 0.01 to 1 eV. The detection material basically consists of a matrix formed by a compound of Li 95% enriched in the isotope ^{6}Li, which is homogeneously dispersed into a fine powder of ZnS (Ag). This scintillator is optically coupled to an Electron Tubes photomultiplier tube of high gain (10^5 for 1 kV bias voltage) containing a bi-alkaline photocathode material with spectral response extending from 300 to 650 nm (maximum at 420 nm). The operating mechanism is based on the high capture cross section of ^{6}Li in the energy range previously mentioned. The illumination of photocathode occurs via the de-excitation of the ^{6}Li produced by α emission described for the nuclear reaction ^{6}Li(n,α)^{3}H. The signal from this detector is recorded in an oscilloscope; time sweeps (usually between 10 and 100 μs) were selected in agreement to the flight times expected for neutrons emerging from the moderator (speeds previously estimated of the order of 10^3 m/s). For the control of fast neutron production, we used the other two detectors mentioned above (S-PMT and a SAC). A typical oscillogram of slow neutrons can be observed in Fig. 3. The distribution of peaks associated with slow neutrons can be appreciated in the form of a "comb" in the pictures, the peak at ~18 μs corresponds to the hard x-ray produced by the discharge. A series of DPF discharges were made to determine the best moderator thickness according to the experimental needs. The number of peaks of slow neutrons obtained from each measurement was registered and an average was obtained from series corresponding to each moderator thickness $\boldsymbol{d}$. This average number of peaks varies with $\boldsymbol{d}$; a maximum value is observed (in our case) for $\boldsymbol{d} = 24$ mm. Then, we used this optimum thickness (24 mm) in our experiments.

C. Application of a DPF to the nuclear activation method

We made the first feasibility test for using the NGAA method for substances identification with plasma focus as primary neutrons source, using some samples of different materials. The DPF PACO was used like a neutron source. The device used for acquiring the γ decay spectrum was a well-shielded ORTEC digiBASE Multichannel Analyzer (MCA) coupled to a 2 in. ORTEC NaI(Tl)-PMT detector. The current pulses from the PMT are amplified, digitized, and processed by the MCA device and associated software. Basically, the analyzer separates, collects, and takes into account the different energy levels present in a definite time. We have analyzed several materials by the present method. First, a sample of natural Mn has been used.

The natural Mn has the only isotope ^{55}Mn (stable). The reaction by neutron capture is: ^{55}Mn (n, γ) ^{56}Mn, and cross section $\sigma_n\gamma$15 b approximately for thermal and epithermal neutrons. The τ of ^{56}Mn that decays (β^-) to ^{56}Fe is 2.57 h. A sample of pure Mn pellets, 100 g total weight, was subjected to moderated neutron pulses placed at 0.38 m from the focus. Each slow neutron pulse lasts about 2 ms. Several series of PACO shots (10 to 50 shots, frequency 10^{-1} s^{-1}, each series) have been made. After the irradiation with moderate neutrons, the gamma radiation resulting from the ^{56}Mn decay was measured for each series. The gamma spectrum for a series of 40 DPF shots is shown in Fig. 4. A peak can be observed at the energy of 850 keV. This corresponds to a characteristic line in the spectrum of the mentioned ^{56}Mn decay.[12] It is convenient to point out that the slow neutron irradiation time is much smaller than the decay time of Mn.

Au and Pb have been used searching for identification through NGAA with PACO DPF neutrons. Au has one stable isotope, ^{197}Au, the only natural one. A nuclide ^{197}Au transforms into ^{198}Au through the neutron capture reaction ^{197}Au (n, γ) ^{198}Au that has a mean lifetime of 2.7 days. It, via β decay, passes to an excited state of ^{198}Hg and then passes to the ground state emitting gamma radiation (411.8 keV for the more probable ^{198}Hg excited state).[13]

On the other hand, the nuclear reaction ^{197}Au (n,n$'$,γ) ^{197m}Au presents an activation cross section of about 800 mb for fast neutrons (around 2.5 MeV) and a half-life of 7.8 s for ^{197m}Au. The very short half-life of ^{197m}Au and a good activation cross section make the measurement, using fast neutron activation, quick and accurate; it is an important consideration for diagnosis of a large sample, where it is important to reduce the trial time, for commercial viability of this technique. Although activation with thermal neutrons would yield a better sensitivity, because of its large activation cross section, a comparatively long half-life of ^{198}Au would entail a long analysis

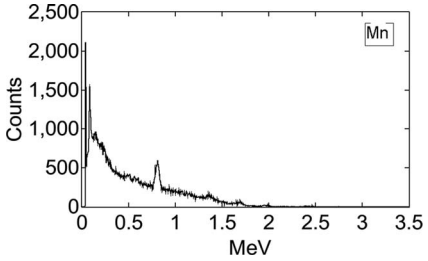

FIG. 4. Gamma spectrum of a natural Mn sample for a series of 40 DPF shots. A peak can be observed at the energy of 850 keV. This corresponds to a characteristic line in the spectrum of the mentioned ^{56}Mn decay.

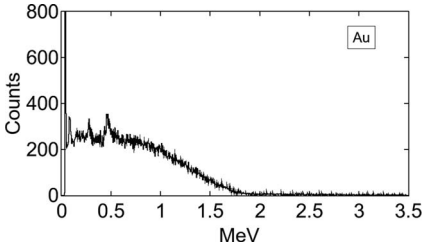

FIG. 5. A spectrum of 18 carat gold sample after irradiation with neutrons generated from D-D fusion reactions at the DPF PACO and then moderated. The result of this figure corresponds to 100 DPF shots (10 ms of irradiation) and 1000 s of MCA measurement. A peak around 400 keV can be observed, corresponding to the reaction ^{197}Au (n,gamma) ^{198}Au, half life of 2.7 days, cross section for slow neutrons: 98 b, and gamma-ray of interest: 412 keV (95.5%). Other peaks observed here could correspond to different components of the annealing.

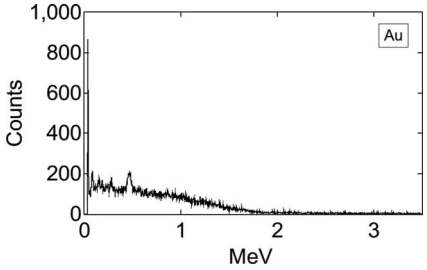

FIG. 6. Fast neutron Activation Analysis: Gamma spectrum of a piece of 18 carat gold subjected to 2.45 MeV neutron flux (70 DPF shots). A peak is observed at about 280 keV corresponding to the nuclear reaction ^{197}Au (n, n′ γ) ^{197m}Au. Other intense peak around 500 keV, which may be due to reaction ^{207}Pb (n, n′γ) ^{207}Pb^{m} or to a reaction with other components of the gold annealing.

time. An antecedent of this part of the present work is that of Tartari *et al.*[14] which employed fast neutrons from a plasma focus device for Au detection.

Activation in Au was carried out through the following procedure: 100 g of 18 carat-gold (80% gold; the rest copper and other components) encapsulated in high density polythene box (150 × 150 × 40 mm^3) were placed at 0.36 m from the source of fusion neutrons (the focus). The solid angle subtended by the sample surface is then 0.1736 sr. One hundred shots of DPF PACO were performed (total duration of the series: 1000 s).

The decay spectrum is shown in Fig. 5. A peak around 400 keV can be observed, corresponding to the reaction ^{197}Au (n, γ) ^{198}Au with half life of 2.7 days and a cross section for slow neutrons of 98 b. The gamma-ray of interest is 412 keV (95.5%). Other peaks observed here could correspond to different components of the annealing.

Later on, the same piece of gold was located about 2 m from the neutron emission zone. Fast neutrons (2.45 MeV) were used in this experiment because no moderator was used. The expected reaction is ^{197}Au *(n, n′γ)* ^{197}Au^{m}, related photon energy: 279.0 keV, τ = 7.8 s, and 800 mb cross section.[15] A series of 70 shots of plasma focus were made with about 1.8 × 10^8 neutrons per pulse in 4π sr. The solid angle subtended by the gold sample is $\Delta\Omega = 0.25 \times 10^{-3}$ sr. The neutrons that arrive to the sample are 3.6 × 10^5 per pulse.

The procedure was as follows: the registration of γ radiation with the NaI detector began immediately (one tenth of a second) after every shot, and was maintained during 10 s. The detector was put off and the following plasma focus discharge was performed. The accumulation of radiation emitted as a consequence of the activation reached in these 70 shots was then registered by the MCA system (Fig. 6). A peak is observed at about 280 keV corresponding to the nuclear reaction ^{197}Au (n, n′ γ) ^{197m}Au. The other intense peak is around 500 keV, which may be due to the reaction ^{207}Pb (n, n′γ) ^{207}Pb^{m} or to a reaction with other components of the gold annealing.

An attempt to identify commercially available Ag was made too. Some samples were irradiated with neutrons generated in the DPF PACO and then moderated. The natural Ag is a mixture of the isotopes ^{107}Ag and ^{109}Ag (51.8% and 48.2%, respectively). The corresponding nuclear reactions by neutron capture are ^{107}Ag *(n,eνγ)* ^{108}Ag and ^{109}Ag *(n,eνγ)* ^{110}Ag; its cross sections for thermal neutrons $\sigma_{n}\gamma$ are 37 and 89 b, respectively.[16]

Experiments were carried out using a sheet 10 × 10 cm^2 and 1 mm thick, disposed perpendicular to the DPF axis (see Fig. 1) in order to maximize the neutron flux irradiation energy; this sample was placed outside the discharge chamber in the direction of maximum emission. It was fixed behind HDPE plates 24 mm thick that, according to the results previously shown, yield a maximum of the moderated neutron flux.

The radiation emitted by the irradiated Ag sample was recorded with a 2 in. NaI(Tl)-MCA Canberra. A photon spectrum recorded with 11 shots from the PF device is shown in Fig. 7.

Due to the statistical nature of the energetic neutron yield typically generated by PACO DPF, the neutron yield was significant in these seven shots corresponding to a single charge of gas D_2. In the picture, a peak around 450 keV can be clearly observed, also other peaks at higher energies (there is one around 650 keV) can be observed. This could correspond to the ^{110}Ag decay (τ = 24 s, β decay) that led to intense gamma radiation at 374 keV and 657 keV. Also, a contribution could be done by the peak of 447 keV of the decaying of ^{110}Ag (τ = 250 d, β decay). With respect to the ^{108}Ag decay (τ = 24 s, β decay) the most intense peak of gamma radiation is at 633 keV. In our experiment, this could contribute to the observed peak around 650 keV. It is necessary to remember that our detector does

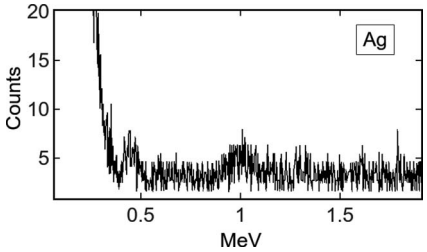

FIG. 7. Spectrum obtained by irradiating a sample of natural Ag with neutrons from 11 (7 with high neutron yield) DPF shots. The arrow marks a peak of 446.8 keV, characteristic of the ^{110} Ag decay spectrum. Other peaks of less intensity can be also observed.

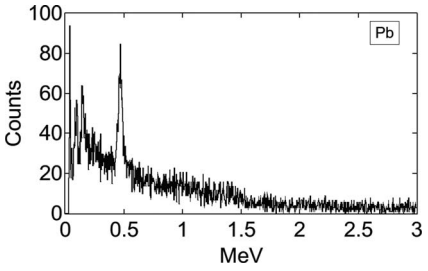

FIG. 8. Fast neutron Activation Analysis: Gamma spectrum of a piece of Pb subjected to 2.45 MeV neutron flux (20 DPF shots).

not have a very high resolution (it is a NaI detector) and the collection solid angle is narrow.

An identification of a sample of lead by NGAA was also accomplished by using 2.45 MeV neutrons. A plate of Pb, $100 \times 100 \times 3$ mm^3, was placed at the ORTEC NaI-digital MCA detector inlet, located at a distance of 2 m from the fast neutron source zone. Several series of shots were made. The gamma spectrum was recorded for each one. The nuclear reaction of interest for 2.5 MeV neutron energy is ^{207}Pb $(n, n'\gamma)$ ^{207}Pb^m, whose spectrum presents a peak at 569.7 keV ($\tau = 0.81$ s). In Fig. 8 a Gamma spectrum obtained for a series of 20 DPF shots can be observed. An intense peak around 500 keV can be observed, which may be due to reaction ^{207}Pb $(n, n'\gamma)$ ^{207}Pb^m.

Moreover, preliminary experiments have been made to evaluate the feasibility of DPFs application in the activation of substances for its detection and also activation to obtain short life isotopes (the last for eventual applications in industry or medicine). This point is in development at present.

III. CONCLUSIONS

This work must be understood as a first attempt to test the feasibility of using a plasma focus as primary neutron source for NGAA applications. Preliminary results obtained with the small 2 kJ DPF PACO suggest promising prospects for the use of DPF devices as pulsed neutron sources for the determination "*in situ*" of the chemical composition of substances by a nuclear activation technique. From a comparative perspective with other neutron sources currently in use, it is a tool with significant advantages, particularly as a clean source (free of radioactive waste), with easy transportability and high operational safety. The possibility to make it functional in a repetitive mode significantly reduces the time taken for irradiation and subsequent acquisition of the gamma spectrum emitted by samples for quick and more selective identification. The decay γ detector system can be modified in order to reach a qualitative improvement of the detection. It must be taken into account that the only detector used here was a 2 in. diameter NaI(Tl); instead of this, a detector of greater diameter or an array of several detectors in parallel could be set without losing practical advantages.

ACKNOWLEDGMENTS

The authors wish to thank Mr. Claudio Santiago, Mr. Lucas Conde, Mr. Marcelo Rodriguez, and Mr. Osvaldo Toscano for technical assistance and researchers of IFIMAT-UNICEN Institute for providing some material for measurement. This work was supported in part by the Science and Technology Promotion National Agency of Argentina under Grant No. PICT 718-06.

[1]E. S. Gladney, D. B. Curtis, and E. T. Jurney, J. Radioanal. Chem. **46**, 299 (1978).
[2]E. S. Gladney, E. T. Jurney, and D. B. Curtis, Anal. Chem. **48**, 2139 (1976).
[3]E. T. Jurney, D. B. Curtis, and E. S. Gladney, Anal. Chem. **49**, 1741 (1977).
[4]S. M. Lombard, T. L. Isenhour, P. H. Heintz, G. L. Woodruff, and W. E. Wilson, Int. J. Appl. Radiat. Isot. **19**, 15 (1968).
[5]E. M. A. Hussein and E. J. Waller, Appl. Radiat. Isot. **53**, 557 (2000).
[6]Y. J. Park and B. J. Song, Anal. Sci. Technol. **16**, 426 (2003).
[7]J. W. Mather, Phys. Fluids **8**, 366 (1965).
[8]J. Pouzo, D. Cortázar, M. Milanese, R. Moroso, and A. Píriz, *Small Plasma Physics Experiments* (World Scientific, London, 1988), p. 80.
[9]P. Choi, C. S. Wong, and H. Herold, Laser Part. Beams **7**, 763 (1989).
[10]H. Bruzzone, M. M. Milanese, J. J. Niedbalski, H. N. Acuna, R. L. Moroso, J. L. Supán, S. Guichón, and F. Malamud, IEEE Trans. Plasma Sci. **38**, 1592 (2010).
[11]F. Castillo Mejía, M. Milanese, R. Moroso, and J. Pouzo, J. Phys. D: Appl. Phys. **30**, 1499 (1997).
[12]S. H. Byun, A. Pejović-Milić, W. V. Prestwich, D. R. Chettle, and F. E. McNeill, J. Radioanal. Nucl. Chem. **269**, 615 (2006).
[13]K. H. Lieser, *Nuclear and Radiochemistry: Fundamentals and Applications* (Wiley-VCH Verlag GmbH, 2001), p. 61, Fig. 5.12.
[14]A. Tartari, G. Verri, A. D. Re, F. Mezzetti, C. Bonifazzi, and L. Rapezzi, Meas. Sci. Technol. **13**, 939 (2002).
[15]A. A. Ebel and C. Goodman, Phys. Rev. **93**, 197 (1954).
[16]R. Sher, *Handbook on Nuclear Activation Cross-Sections* (IAEA, Vienna, 1974).