Nuclear Instruments and Methods in Physics Research A 602 (2009) 306–310

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Pyroelectric crystal neutron production in a portable prototype vacuum system

Don Gillich, Andrew Kovanen, Bryan Herman, Travis Fullem, Yaron Danon *

Department of Mechanical, Aerospace and Nuclear Engineering, Rensselaer Polytechnic Institute, Troy, NY 12180, USA

ARTICLE INFO

Article history:
Received 13 October 2008
Received in revised form
3 December 2008
Accepted 8 December 2008
Available online 20 January 2009

Keywords:
Pyroelectric crystal
Nuclear fusion
Neutrons
Portable neutron generator

ABSTRACT

Pyroelectric crystals are being used to ionize and accelerate D_2 gas into a deuterated target to achieve D–D fusion at Rensselaer Polytechnic Institute (RPI). Previous results from RPI yielded up to 5.9×10^4 neutrons over one thermal cycle; however, these results were not reproducible from one experiment to another. A portable prototype vacuum system has been developed that enhances reproducibility of results. A thermal management system consisting of two thermoelectric module temperature controllers was developed to improve the reproducibility of the thermal cycle. Twenty-eight experiments were conducted over a period of three days to determine the optimum ambient gas operating pressure of the vacuum system. The optimum operating pressure was found to be 26 mTorr which is much higher than operating pressures that have been previously reported. Reproducibility of the results has been improved. At optimum pressure, five consecutive experiments yielded an average of 9×10^3 neutrons with a standard deviation of 14% for each thermal cycle. While the neutron yield is lower than previous results, increased reproducibility in a more stable vacuum environment may allow for an increase in neutron yield in the future.

1. Introduction

Pyroelectric crystals have been used to ionize deuterium gas and accelerate the ions into deuterated targets to achieve D–D fusion for the purpose of generating neutrons. Pyroelectric-generated fusion was first achieved in 2005 by Naranjo, et al. [1] at the University of California at Los Angeles (UCLA). In 2006, Geuther et al. [2] at Rensselaer Polytechnic Institute (RPI) used a dual pyroelectric crystal system to generate D–D fusion thereby confirming the phenomena. More recent work [3] reported doubling the neutron yield of Ref. [1] but also reported on the lack of reproducibility in results.

Commercial neutron generators generally require a high voltage power supply to achieve the electrostatic fields necessary to ionize and accelerate deuterium (or tritium) gas into a deuterated target. By heating (or cooling) a pyroelectric crystal using 10–20 W of power (considering thermal losses), a single crystal can generate an electrostatic potential on the order of 100 kV which is sufficient to achieve D–D fusion. The two crystal system at RPI typically doubles the accelerating potential and deuterium ions can be accelerated with a maximum measured energy of approximately 220 keV. Higher energy increases the likelihood of fusion because the D–D fusion cross-section is energy dependent. Because hundreds of keV can be generated by pyroelectric crystals, a neutron generator can be engineered to be small, battery operated and portable.

Before such a system can come to fruition, several challenges must be solved. One of the main challenges is reproducibility of results. One reason for this problem is that the temperature profile across the crystal from one experiment to another is not precisely reproducible due to the lack of a controlled thermal management system. Other reasons include high voltage discharges and erosion of the field emitting tip used to locally enhance the electric field [3].

One goal of the pyroelectric crystal fusion project at RPI is to develop a portable, battery operated neutron generator capable of providing $\sim 10^6$ D–D neutrons per 100 second thermal cycle. By introducing tritium into the system, the neutron yield will increase to $\sim 10^6$ neutrons per second which is suitable for some applications such as a calibration source, research and homeland security. In working towards this goal, RPI has constructed a portable prototype vacuum system that is capable of being sealed after it has been evacuated and filled with deuterium gas. Preliminary experiments using this portable system have been conducted and results are reported in this paper.

2. Experimental set-up

The portable prototype vacuum chamber consists of a 9.3 cm long, 7.62 cm diameter chamber with 3.3 cm reducer flanges to

*Corresponding author. Tel: +1 518 276 4008; fax: +1 518 276 4832.
E-mail address: danony@rpi.edu (Y. Danon).

0168-9002/$ - see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.nima.2008.12.239

minimize the overall vacuum volume. Two isolation valves are attached which can be closed after the system is pumped down and filled with deuterium gas. Fig. 1 is a picture of the system. It is important to note that during the experiments presented here, the lower valve was closed almost all the way to conserve deuterium gas and to ensure a more stable ambient pressure in the main chamber. A gas leak valve was used to introduce D_2 gas into the chamber.

Two 1 cm thick, 2 cm diameter lithium tantalate ($LiTaO_3$) crystals were used in these experiments. A ~8 mm diameter, ~0.8 mm thick copper disk with a standard DIP socket soldered in the middle was mounted on the +z face of one of the crystals (henceforth called the tip crystal). During cooling, the +z face of a pyroelectric crystal accelerates positive ions away from the crystal face. Mounted in the DIP socket was a 70 nm radius tungsten tip that was used to locally enhance the electric field in order to ionize the D_2 gas. The tip apex was approximately 1 cm from the face of the crystal. On the other crystal, deuterated polyethylene (CD_2) was melted onto the –z crystal face which attracts positive ions during cooling (henceforth called the target crystal).

The crystals were mounted onto the thermoelectric heater/coolers (TECs) using conductive epoxy (GC electronics part number 19-2092). The TECs were then mounted onto the aluminum vacuum flanges using silver paint (SPI High Purity Silver Paint). Type T (copper and constantan) thermocouples were attached to each TEC with JB Weld® Epoxy. CPU fans (MassCool 5R057B3) were attached to the outside face of the aluminum flanges to facilitate cooling. Fig. 2 graphically depicts the two crystal–flange system.

Heating and cooling of the crystals was done using a LabView program which provided input to a pair of thermoelectric module temperature controllers (Oven Industries Model 5R7-388). Several experiments were done to determine a thermal cycle for each crystal that would ensure both crystals were thermally in phase during cooling. The tip crystal was heated from ~27 to 130 °C over a period of 5 min, allowed to soak at 130 °C for a period of 3 min and then cooled to 15 °C over a period of 5 min. The target crystal was heated from ~27 to 145 °C over a period of 5 min, allowed to soak at 145 °C for a period of 3 min and then was allowed to cool naturally. Neutron emission occurred during the crystal cooling phase. Fig. 3 provides a typical thermal cycle for the two crystals during these experiments.

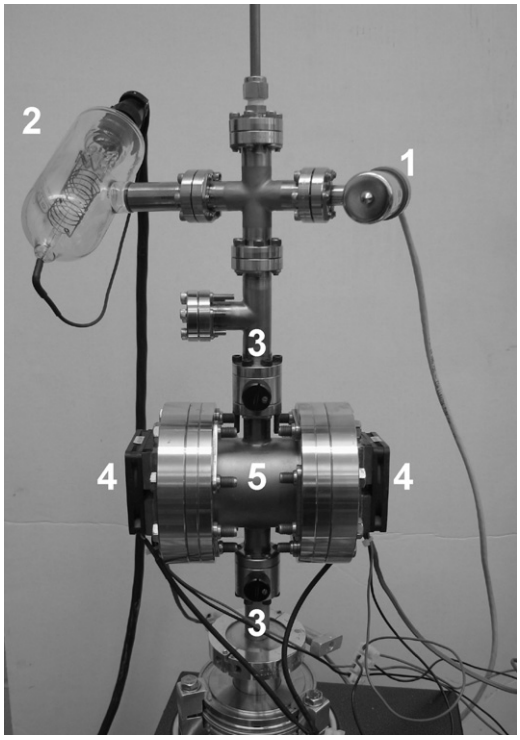

Fig. 1. Picture of the RPI portable prototype vacuum chamber: (1) Granville–Phillips Convectron® pressure gauge, (2) MDC ionization pressure gauge (part number 432023), (3) isolation valves, (4) CPU fans, and (5) vacuum chamber.

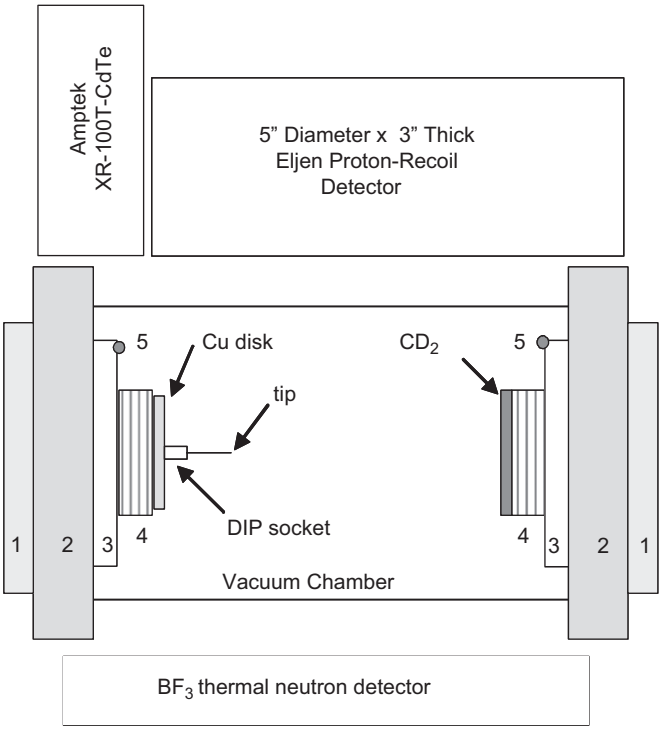

Fig. 2. Schematic of the two crystal system inside the vacuum chamber: (1) CPU cooling fans, (2) aluminum vacuum flanges, (3) TECs (Laird Technologies part number 430141–501), (4) lithium tantalate crystals and (5) type T thermocouples. The distance between the two crystals is ~7 cm. The distance from the target crystal to the face of the proton-recoil detector is ~6.55 cm and to the BF_3 detector is ~7.65 cm. The stainless steel chamber is ~0.17 cm thick.

An Amptek XR-100 T, cadmium telluride (CdTe) semiconductor X-ray detector was used to monitor X-ray emission. The signal was input to an Amptek Model PX2T-CdTe power supply and amplifier and output to a Canberra Multiport II multi-channel analyzer (MCA) and multi-channel scalar (MCS). GENIE 2000 Virtual Data Manager Software was used to collect the X-ray energy and the time data.

Neutrons were detected by using a 5 in. diameter , 3 in. thick proton-recoil detector (Eljen 510-50x30x-9/301) which was coupled to a Photonics photomultiplier tube (VD105K/01). In order to lower the X-ray flux incident on the proton-recoil detector, a ~6.5 mm thick lead sheet was placed on the detector face. This shield attenuates 180 keV X-rays by a factor of 1000 while transmitting 98.4% of the 2.45 MeV neutrons from D–D fusion. Data from the neutron detector were collected using an Acqiris AP240 data acquisition board. The data were then analyzed by software written at RPI [4]. This analysis included the use of pulse fall-time discrimination curves to discriminate the neutrons from gamma radiation [5].

Previous work called to question the number of neutrons being produced using pyroelectric crystals speculating that the results may be contaminated with X-ray pile-up due to insufficient shielding of the proton-recoil detector [6]. To address this issue, a BF_3 detector consisting of four electrically linked 2 in. diameter, 14 in. long BF_3 tubes (Reuter–Stokes Electronic components Inc. RSN-108-S) surrounded by polyethylene, was used to detect

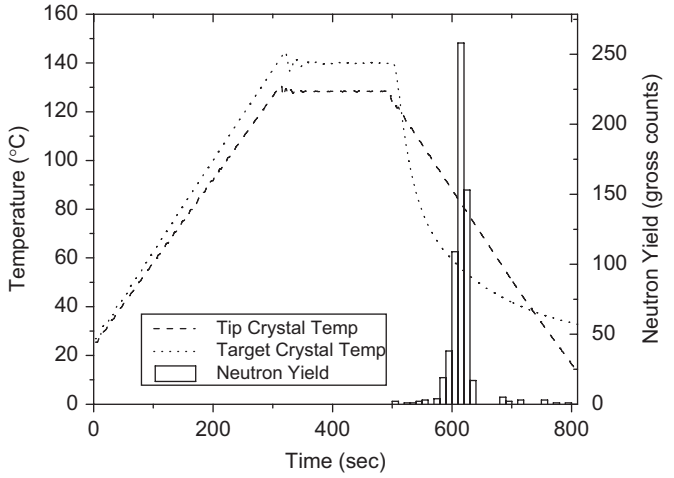

Fig. 3. Controlled thermal cycle and neutron production of a typical experiment for the two crystal system. The time bins for the neutron production is 10 seconds.

neutrons. This detector is insensitive to X-ray and gamma radiation and was used to verify neutron results. To verify that the detector was insensitive to gamma radiation, a 5.7 μCi sodium-22 source was placed directly in front of the detector with no significant change in counts compared to background. A lead shield was therefore not used with the BF_3 detector so as to ensure the maximum neutron flux would interact with the detector.

Ref. [7] has shown that the use of an optimum operating pressure in the vacuum chamber can more than double the maximum accelerating energy generated by pyroelectric crystals. The same work concluded that the optimum ambient gas pressure is chamber volume dependent. The portable system used in the experiments presented here with a volume of approximately 425 cm^3 is significantly smaller than the chambers previously used in pyroelectric crystal fusion experiments. Geuther et al. used a cylindrical chamber that is 14.5 cm in diameter and 15.7 cm tall [5] with an approximate volume of 2600 cm^3. Tang et al. reported the use of an 8.4 in. diameter spherical vacuum chamber [3] equating to an approximate volume of 5100 cm^3. Because the portable chamber is so small, a series of 28 experiments were conducted to determine the optimum operating pressure of RPI's portable prototype vacuum system.

3. Results

Fig. 4 presents the results of the 28 experiments and shows that the optimum pressure is approximately 26 mTorr. This pressure is significantly higher than that in the previous experiments in which the pressure ranged from 3 to 8 mTorr [1–3]. At pressures below optimum, there are fewer D_2 gas molecules in the chamber which results in lower D–D fusion probability and a reduced neutron yield, as shown in Fig. 4. The results also show that at pressures above the optimum, the X-ray energy and yield decline because the higher pressure allows the charge on the crystal face to dissipate more easily, thereby reducing the overall accelerating potential generated by the crystals.

Another factor that likely contributed to achieving stability in the system was the separation distance between the two crystals. Crystals that are farther apart are less likely to cause spontaneous discharge which results in a more stable electric field between the anode and cathode. In previous experiments, the crystals were ~2.1 cm apart whereas the crystals in these experiments were approximately 7 cm apart.

As shown in Fig. 4, the neutron yields measured by the two different detectors have the same trend in terms of relative counts. Data missing in the BF_3 detector results are due to a malfunction in the detector as a result of electronic noise during several experiments. The data presented in Fig. 4 are normalized to the maximum neutron and X-ray counts which, for both detectors, occurred at the optimum pressure. The data are presented in this way to show the relative neutron and X-ray yields when compared to those achieved at optimum pressure.

During one of the experiments the proton-recoil detector measured 627 gross neutron counts over the 300 second cooling cycle. The same experiment yielded end-point X-ray energy of

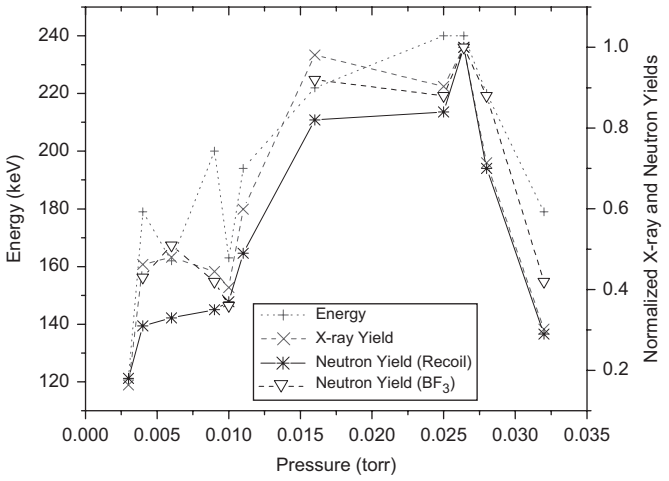

Fig. 4. X-ray energy and normalized X-ray and neutron yield results versus operating pressure [8]. Each data point represents two to four separate experiments. The scatter of the data points at each pressure is a measure of the reproducibility between experiments and is estimated to be ~14% at the optimum pressure. At optimum pressure the proton-recoil detector yielded an average of 507 (±11) net counts and the BF_3 detector yielded 366 (±12) net counts.

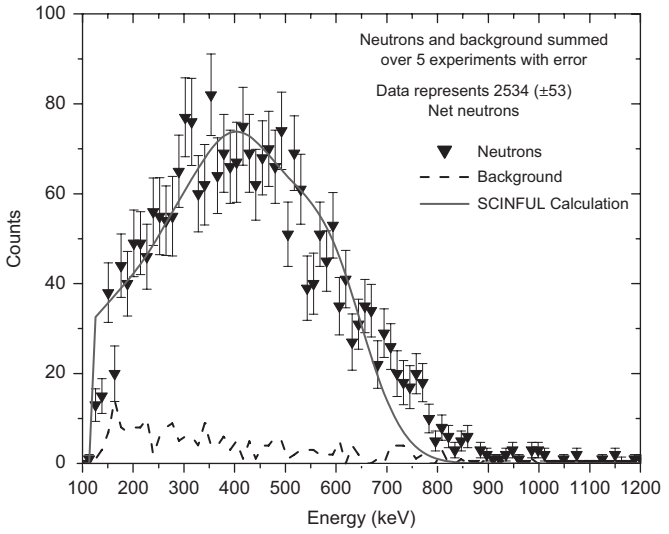

Fig. 5. Histogram of the neutron pulse height measured during five consecutive experiments. The energy axis was calibrated using the Compton edges resulting from electron recoil from a Na-22 gamma source. The error bars represent the counting statistic error (the square root of the counts). The SCINFUL calculation shows that measured and calculated detector responses for 2.45 MeV neutrons are in good agreement.

239 keV and 780,000 X-ray counts on the CdTe detector. RPI's fall-time discrimination software includes the ability to separate pulses into two windows: gamma/X-ray and neutron. The background for the neutron detector was measured over five experiments under operating conditions similar to the neutron production experiments (without deuterium gas) and an average of 47 counts was measured in the neutron window. The X-ray energy and yield produced in these background experiments are similar to those produced during neutron production experiments. The background count in the lab in the absence of an X-ray source in the neutron window is 40. Considering the statistical error of these measurements, X-ray pileup does not significantly change the background. A Monte Carlo calculation (MCNP) was used to calculate the effects on the neutron yield of the solid angle, the stainless steel chamber wall and lead shielding. Subtracting the background and accounting for the solid angle and efficiency of the proton-recoil detector (48% based on a SCINFUL calculation), this particular experiment resulted in emission of 10,720 ($\pm$430) neutrons.

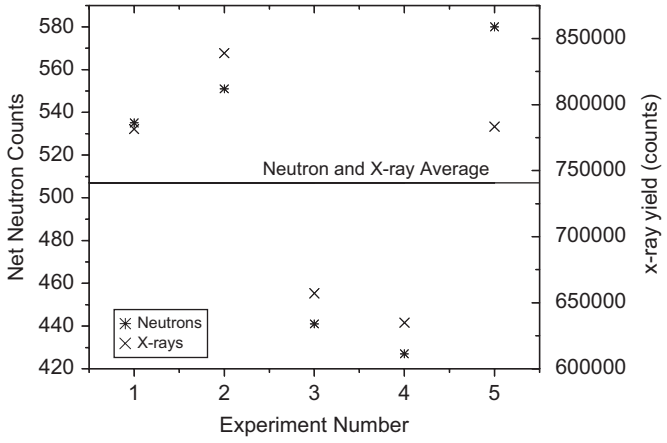

Fig. 6. Neutron and X-ray detected counts during five experiments conducted near the optimum deuterium gas pressure plotted with the average values demonstrate the reproducibility of the system (~14% standard deviation).

The average and standard deviation of the net neutrons detected by the proton-recoil detector for each experiment over five consecutive experiments at optimum pressure were 507 and 11, respectively (the BF_3 detector yielded 366 and 9, respectively). Fig. 5 presents the sum of these experiments with the sum of the five background experiments. Using Monte Carlo methods, the SCINFUL code calculates the anticipated proton-recoil detector response for 2.45 MeV neutron interactions [11]. Fig. 5 shows that the measured and calculated neutron responses are in good agreement. Fig. 6 is a scatter plot of the distribution of data from these five experiments around the average which demonstrates the reproducibility of results.

It is important to note that the results presented here were achieved with a single 70 nm tungsten tip. Previous results [3] have cited varying tip conditions due to erosion as a possible reason why reproducibility is a problem. Experiments were conducted at RPI to characterize tip erosion using a scanning electron microscope (SEM) before and after pyroelectric crystal experimentation. Fig. 7 shows these before and after pictures. It can be seen that there is significant tip erosion after only 10 experiments. Refs. [9, 10] provide similar results relating to tip erosion due to high current densities. For an eroded tip, the radius of gas ionization is smaller and therefore higher gas pressure is needed to provide more molecules locally to attain the same ionization as achieved using a smaller tip. For the smaller tip, the sphere of ionization around the tip would be larger which may lead to more frequent discharges given higher pressures and a smaller chamber size. The experiments reported here may have obtained the optimum pressure for an eroded tip in the RPI portable prototype system.

4. Conclusions

Because it operates at higher ambient D_2 pressures, the RPI portable prototype vacuum system establishes a more stable environment for pyrofusion experiments and reproducibility of experimental results is improved. An average of 507 (with standard deviation of 72) net counts which equates to 9200 (with a standard deviation of 1300) were achieved over five consecutive

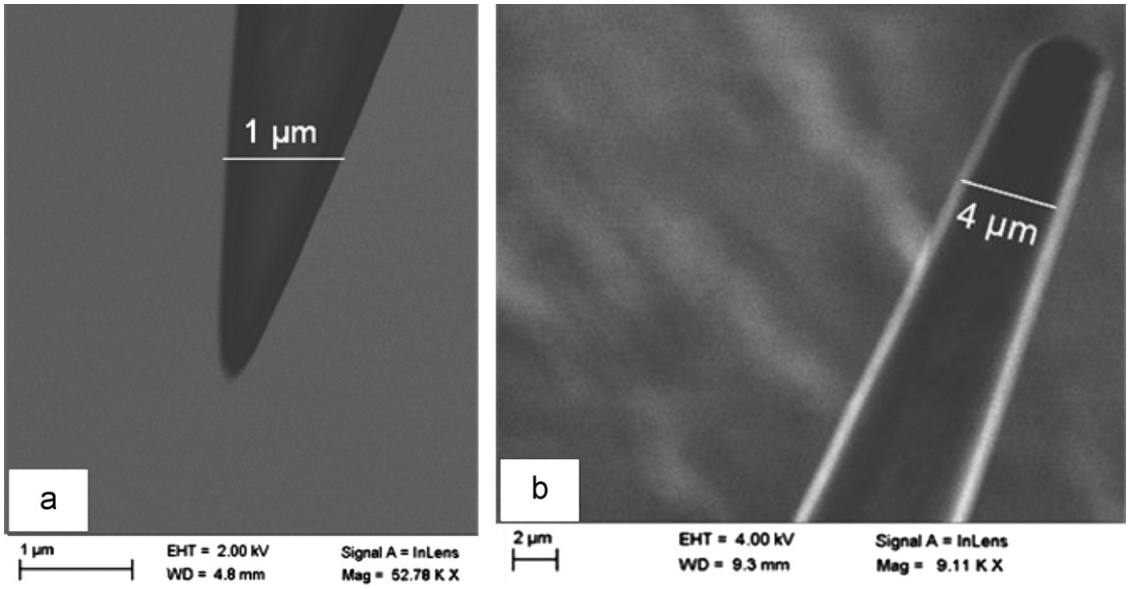

Fig. 7. SEM pictures of a 70 nm radius tungsten tip before (a) note the 1 μm scale and after (b) note the 2 μm scale; 10 two-crystal system experiments. The tip diameter changed from 140 nm to 2.5 μm.

thermal cycles. A more precise thermal management system also contributed to the reproducibility of results.

Acknowledgments

The authors acknowledge the help RPI students including Zuankai Wang, Ezekiel Blain, Frank Saglime, and Brian McDermott in the conduct of the experiments that provide the basis of this paper. This work was supported by DHS cooperative agreement number 2007-DN-077-ER0003.

References

[1] B. Naranjo, J.K. Gimzewski, S. Putterman, Nature (London) 434 (2005) 1115.
[2] J. Geuther, Y. Danon, F. Saglime, Phys. Rev. Lett. 96 (2006) 054803.
[3] V. Tang, G. Meyer, et al., Rev. Sci. Instrum. 78 (12) (2007) 123504-1-4.
[4] F. Saglime, Y. Danon, R. Block, Digital data acquisition system for time of flight neutron beam measurements, ANS 14th Biennial Topical Meeting of the Radiation Protection and Shielding Division, Carlsbad New Mexico, USA. April 3–6, 2006, p. 368.
[5] J.A. Geuther, Radiation generation with pyroelectric crystals, Ph.D. Dissertation, RPI, 2007.
[6] W. Tornow, S.M. Shafroth, J.D. Brownridge, J. Appl. Phys. 104 (2008) 34905.
[7] J. Brownridge, S. Shafroth, The effect of vacuum chamber size and maximum electron energy for pyroelectric crystal electron accelerators, CAARI 2004, Fort Worth, TX, 10–15 October 2004.
[8] D. Gillich, Y. Danon, et al., Pyroelectric crystal-generated neutron production: preliminary results using a portable vacuum system, ANS Transactions TBP, ANS Winter Meeting, Reno, NV, 9–13 November 2008.
[9] K.S. Yeong, J.T.L. Thong, J. Appl. Phys. 100 (2006) 114325.
[10] R. Tanabe, Y. Ito, H. Takezawa, N. Mohri, J. Appl. Phys. 97 (2005) 053301.
[11] J.K. Dickens, Computer code SCINFUL, ORNL-6462, 1988; NEA Data Bank Program no. PSR-0267, 1994.