HHS Public Access
Author manuscript
Health Phys. Author manuscript; available in PMC 2020 December 16.

Published in final edited form as:
Health Phys. 2015 December ; 109(6): 566–572. doi:10.1097/HP.0000000000000345.

A DOSIMETRY STUDY OF DEUTERIUM-DEUTERIUM NEUTRON GENERATOR-BASED IN VIVO NEUTRON ACTIVATION ANALYSIS

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Abstract

A neutron irradiation cavity for in vivo neutron activation analysis (IVNAA) to detect manganese, aluminum, and other potentially toxic elements in human hand bone has been designed and its dosimetric specifications measured. The neutron source is a customized deuterium-deuterium neutron generator that produces neutrons at 2.45 MeV by the fusion reaction $^{2}H(d, n)^{3}He$ at a calculated flux of $7 \times 10^{8} \pm 30\%\ s^{-1}$. A moderator/reflector/shielding [5 cm high density polyethylene (HDPE), 5.3 cm graphite and 5.7 cm borated (HDPE)] assembly has been designed and built to maximize the thermal neutron flux inside the hand irradiation cavity and to reduce the extremity dose and effective dose to the human subject. Lead sheets are used to attenuate bremsstrahlung x rays and activation gammas. A Monte Carlo simulation (MCNP6) was used to model the system and calculate extremity dose. The extremity dose was measured with neutron and photon sensitive film badges and Fuji electronic pocket dosimeters (EPD). The neutron ambient dose outside the shielding was measured by Fuji NSN3, and the photon dose was measured by a Bicron MicroREM scintillator. Neutron extremity dose was calculated to be 32.3 mSv using MCNP6 simulations given a 10-min IVNAA measurement of manganese. Measurements by EPD and film badge indicate hand dose to be 31.7 ± 0.8 mSv for neutrons and 4.2 ± 0.2 mSv for photons for 10 min; whole body effective dose was calculated conservatively to be 0.052 mSv. Experimental values closely match values obtained from MCNP6 simulations. These are acceptable doses to apply the technology for a manganese toxicity study in a human population.

Keywords

mixed field dosimetry; neutron activation; neutron detection; neutron dosimetry

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The authors declare no conflicts of interest.

INTRODUCTION

Neutron activation analysis (NAA) has been employed for many years in multiple disciplines including, but not limited to geology, chemistry, metallurgy, criminology, medicine, astronautics, and agriculture (Corliss 1968; Sedda and Rossi 2011; Kim et al. 2011; Latif et al. 2013). The NAA method is used at Purdue University to detect manganese in human hand bone in vivo and will eventually expand to detect other potentially toxic elements in bone (Liu et al. 2013, 2014). The neutron and photon doses to the extremity (hand and arm) as well as to the whole body need to be determined for radiation exposure risk communication as well as Institutional Review Board (IRB) permission.

Overexposure to manganese is associated with various neurological disorders and cognitive deficits (Bouchard et al. 2007; Roels et al. 2012). At high levels, manganese exposure can lead to manganism, with clinical symptoms including tremors and poor eye-hand coordination similar to those of Parkinson's disease (Lucchini et al. 2009; Martin 2006). Biomarkers of manganese exposure currently used include blood and urine, among others. The half-life of manganese in these biomarkers is on the order of a few days to weeks; however, recent research has indicated the average half-life of manganese in bones of rats is 143 d (O'Neal et al. 2014). This leads to the conclusion of Liu et al. (2014) that bone is a usable biomarker of manganese exposure. NAA is a desirable way of measuring bone manganese, as the method is noninvasive (compared to a bone biopsy). The hand was chosen as there is no active bone marrow (Cristy 1981), and the extremity can be extended away from the body. Following neutron activation, a 60-min count with two high purity germanium detectors is performed, and the peak of E_γ = 847 keV is measured from the β, γ decay of ^{56}Mn to ^{56}Fe. With this system, a detection limit of 0.5 ppm manganese can be achieved, where a normal concentration in adult male hands is approximately 1 ppm (Liu et al. 2014). The extremity dose to the hand and effective dose to the subject is of legit-imate concern and is the topic of this paper.

As the neutron source, a deuterium-deuterium (DD) neutron generator from Adelphi Technologies, Inc., (Redwood City, CA, USA) model DD-109 is used. The generator has a customized frame and structure assembly to support its weight and the weight of the customized neutron moderator and radiation shielding. This generator produces quasi-monoenergetic neutrons at 2.45 MeV by the fusion reaction ^{2}H(d, n)^{3}He. The Q-value for the reaction is 3.3 MeV, splitting kinetic energy and momentum between the ^{3}He and ^{1}n, which yields the aforementioned neutron energy almost consistently at 2.45 MeV (Vainionpaa et al. 2013; Bergaoui et al. 2014).

Neutron and photon extremity dose inside the small hand irradiation cavity were measured with an electronic pocket dosimeter (EPD) and film badges and compared with Monte Carlo simulation results. Traditional survey meters will not fit inside. Neutrons exist in a continuous spectrum from 0.025 eV to 2.45 MeV, with scattered and reflected neutrons entering the cavity from every direction. Bremsstrahlung x rays are created from back-streaming electrons impinging upon aluminum with average energy of 40 keV (maximum 120 keV). Activation gammas, primarily E_γ = 2.2 MeV from the capture reaction ^{1}H(n, γ) ^{2}H, are created throughout the high density polyethylene (HDPE) moderator. This mixed field of neutrons and photons makes deep dose neutron and photon calculation and measurement difficult but at the same time extremely important, since the application of this research is ultimately for human studies.

The goal of this study is to determine the neutron and photon extremity dose equivalent to the subject's hand and the whole body effective dose to the subject while undergoing a 10 min NAA measurement for manganese in vivo at a flux of $7 \times 10^8 \pm 30\%\ s^{-1}$.

MATERIALS AND METHODS

Neutron source, flux, and energy

Deuterium plasma is produced by radio frequency (RF) power and accelerated by high electric potential difference to impinge upon a V-shaped target of titanium-coated copper. Acceleration voltage can be set between 80–125 kV; current can be set up to 16 mA. Typical operating parameters are 120 kV and 10 mA. The target is actively cooled, allowing for sustained operation. Through the first 3 to 5 s of operation, deuterium becomes embedded in the target creating titanium hydride, allowing for constant replenishment of the deuterium on the target (IAEA 2012; Vainionpaa et al. 2013) and therefore a constant, sustained neutron flux. The target is kept under high vacuum by a roughing pump and turbo vacuum pump. This prevents a buildup of gas around the target causing a high voltage breakdown—resulting in a short to ground and likely equipment damage. Fig. 1 shows the system with no moderator, reflector, or shielding.

Unavoidably, tritium becomes present in the target through the $^2H(d, p)^3H$ reaction, contaminating the 2.45 MeV neutron flux with some 14 MeV neutrons through the $^2H(t, n)^4He$ reaction (Cecil and Nieschmidt 1986). Following the assumptions of Cecil and the total integrated operating time of approximately 100 h (typically no more operation than 1 h d^{-1}), at most 1% 14 MeV neutrons are expected to be generated.

To calculate the output flux of the DD neutron generator, the moderator was removed, and the NSN3 neutron survey meter (discussed further below) was exposed to a fast neutron flux. The dose displayed on the NSN3 was compared with MCNP simulation of an identical setup. The simulated neutron fluence rate incident upon the simulated detector was used to back-calculate the output flux using ICRP 74 fluence-to-dose conversion factors (ICRP 1996). From this method, the output flux of the DD neutron generator is calculated to be $7 \times 10^8\ s^{-1}$ (Liu et al. 2014). The error in this measurement is ±30%, where ±5% is the maximum error seen in the MCNP simulation output and ±25% error from the response of NSN3 to fast neutrons.

Moderator, reflector, and shielding

To lower the overall irradiation time and therefore neutron and photon extremity dose to the human subject, the system is designed to expose the hand to maximum thermal neutron flux. The subject places his/her hand and arm inside the irradiation cavity, which is shown in Fig. 2. The cavity measures 9.5 cm wide by 14 cm high by 44.5 cm deep. The centerline of the location of the generator source is 28 cm into the cavity. The straight-line thickness of HDPE between source and cavity is 5 cm; this represents the lower limit of moderator thickness at any point between source and cavity. Graphite with 5.3-cm thickness is chosen as a neutron reflector on the opposite side of the generator head from the cavity. HDPE with 6.5-cm thickness is added to reflect back into the cavity neutrons that have passed through it. Borated HDPE with 5.7-cm thickness is used as a neutron shield on the outside of the HDPE (Liu et al. 2014). Lead sheets with 3 mm and 7 mm thicknesses are placed strategically to attenuate bremsstrahlung x rays produced from back-streaming electrons as well as activation gammas. With this neutron and photon shielding, the rest of the body receives very little effective dose.

Monte Carlo simulation

Because the irradiation cavity is small, Monte Carlo N-particle transport (MCNP) version 6, developed by Los Alamos National Laboratory, was used to simulate neutron transport and extremity dose (http://mcnp.lanl.gov/; accessed 29 July 2014). This simulated extremity dose was compared with Fuji model NRY 21 (EPD) and personal dosimetric film badges—both sensitive to neutrons and photons.

Using schematics from this lab's customized moderator/reflector/shielding design, a MCNP6 simulation input deck was written. Much attention was paid to ensure that the geometry most closely matched the true DD neutron generator. From this code, the neutron source and activation gammas were simulated. The number of tracked particles was always set at 1×10^8, with the neutron source modeled as an isotropic point source. A simulation model is shown in Fig. 3.

A 15-cm-long hand (712 g soft tissue, 70 g bone) and 19-cm-long arm (872 g soft tissue, 92 g bone) were modeled inside the irradiation cavity, with the centerline of the hand at 28 cm deep and the arm extending back to the lip of the cavity. Bone mass was determined based on ICRP 89 reference male values (ICRP 2002). Atomic composition for tissue follows the ICRU 44 four-component model; bone follows the ICRU 44 compact bone model (www.physics.nist.gov; accessed 3 March 2015). Using the F6 card, which reports the track length estimate of heating in MeV g^{-1}, extremity dose was computed for the hand tissue, hand bone, arm tissue, and arm bone.

The simulation was also run with an empty hand cavity using the point detector function of MCNP. Extremity dose was calculated with the F5 card on a small volume point detector with radius = 0.5 mean free paths of corresponding neutron energy. Its center location was simulated as 17 cm from the center of the source. This placed the simulated detector at the same physical location inside the cavity as the EPD discussed below.

Radiation detection

Electronic pocket dosimeter (EPD).—The Fuji model NRY 21 EPD, which contains four sensitive p-type silicon semiconductors to detect neutron and photon radiation, has been used. It is small enough to fit inside the irradiation cavity. Its design response specifications were found to be ±20% photon, and the under-response and over-response range was −60% + 200% neutron, respectively, from energies of 0.025 eV to 15 MeV (Sasaki et al. 1998). Inside the hand cavity, it was placed at a depth of 28 cm (where the subject's fist would be located), aligning the sensitive volume to centerline with the source. A 10.2 cm × 20.3 cm × 5.1 cm HDPE brick was placed behind the EPD, and another brick was placed at the opening of the cavity, as shown in Fig. 4. Considering neutron scattering and reflecting and similar density of HDPE and soft tissue, this better simulates an arm in the cavity vs. simply air.

Dosimetric film badges

Two photon and neutron sensitive personal dosimetric film badges were used in the same setup as Fig. 4. They were processed by Purdue University's National Institute of Standards and Technology (NIST) traceable and National Voluntary Laboratory Accreditation Program (NVLAP) accredited dosimetry processors (Mirion Technologies, Inc., Smyrna, GA, USA) in accordance with their standard operating procedures.

Neutron survey meter NSN3

The Fuji NSN3 is a proportional counter sensitive to neutrons from 0.025 eV to 15 MeV. Above about $E_n = 1\ eV$, elastic scattering of hydrogen in methane gas is used to detect fast neutrons. Below $E_n = 1\ eV$, the $^{14}N(n, p)^{14}C$ reaction (proton energy of 626 keV) is used to detect slow and thermal neutrons. Its design gamma rejection is such that at a gamma dose rate of 0.1 Sv h^{-1}, the NSN3 displays an apparent neutron dose rate of 0.01 mSv h^{-1}. Its design responses for both dose and angular response (all design neutron energies) were found to be ±25% (Nunomiya et al. 2011). To measure ambient neutron dose equivalent, the NSN3 is placed outside the entirety of the moderator and shielding and used as a survey meter to obtain dose rates where the subject's trunk would be located during an IVNAA measurement.

The responses of NSN3 and NRY 21 EPD to neutrons at varying neutron energies were compared. Given the design large response range of the NRY 21, it was desirable to compare its response with the NSN3. A modification of the shielding was arranged such that the two radiation detection devices could be placed in the same geometry, with the sensitive volumes exposed to the same neutron flux. In three trials, fast neutron response (no moderator), mixed flux response (5-cm HDPE moderator), and primarily thermal response (10-cm HDPE moderator) were tested. They were placed on a 5-cm HDPE sheet with a 5-cm HDPE sheet 30.5 cm behind the device. Fig. 5 shows the setup with 5-cm HDPE and the NSN3 meter. The NRY 21 EPD was placed on a small HDPE brick to align the sensitive volume in the Z-axis to that of where the NSN3 was located. Each device was irradiated in each configuration for 3 min.

RESULTS

MC simulated hand dose

In MCNP6, the F6 card reports the track length estimate of heating results in MeV g^{-1} over the specified cell. Because of energy dependent radiation weighting factors (W_R), many energy bins were created in the MCNP output file. Neutron energy, E_n, was used as the highest cutoff energy for that respective bin, and for each bin a separate W_R calculated the following eqns (1) and (2) from ICRP 103 (ICRP 2007):

$$2.5 + 18.2e^{-\left[ln(E_n)^2\right]/6}, E_n < 1\ MeV \quad (1)$$

$$5.0 + 17.0e^{-\left[ln(2E_n)^2\right]/6}, 1 \le E_n \le 50\ MeV\ . \quad (2)$$

Output was in MeV g^{-1}, corrected for the neutron flux of $7 \times 10^8\ s^{-1}$ and time of 600 s, then converted to J kg^{-1}, an appropriate W_R assigned for dose equivalent, and converted to mSv to simulate the extremity dose. Table 1 displays simulated activation gamma and neutron extremity dose per 10 min to tissue and bone of the hand and tissue and bone of the arm.

Simulation values for the hand tissue are very closely matched to the experimental values, which are discussed below. It is expected that significantly lower extremity dose will be received by the arm as it extends to the opening of the cavity and, furthermore, that the rest of the body receives even less effective dose.

The F5 card simulates a point detector and reports results in neutrons cm^{-2}. Energy bins were modified to match those of ICRP 74 (ICRP 1996). This allows conversion in pSv cm^{-2} and then multiplication by the generator's flux of $7 \times 10^8\ s^{-1}$ and by 600 s to obtain neutron extremity dose. The point detector simulation was designed to match the experiment of the NRY 21 EPD, which was shown in Fig. 4, with results presented in Table 2. That is, the detector was simulated at the same geometric location as the EPD's physical location, and results are demonstrated in Table 3.

Slight deviation of EPD vs. simulated values with increasing distance from the source was observed. This is likely caused by the EPD always facing the wall of the cavity; with increasing distance, the angle from source to detector becomes more extreme and thus the EPD has a lower response.

Measured hand dose by NRY 21 EPD

Table 2 shows five measurements by EPD at varying depths inside the cavity, always facing toward the neutron source. Photon extremity dose is included, but this was performed prior to adding a lead wrap to the generator head to attenuate bremsstrahlung x rays.

For a manganese IVNAA measurement, a 10-min irradiation would provide an extremity dose to the hand of approximately 31.4 mSv (corresponding to the 27.9-cm-depth measurement). Photon extremity dose was further reduced after this experiment.

After adding lead to the generator head, in an identical setup as Table 2 at 27.9 cm depth, the NRY 21 EPD recorded 4.1 mSv DDE photon.

Measured hand dose by film badge

Mirion Technologies, Inc., processed the film badges according to their standard procedures, and results for extremity dose were reported in deep dose equivalent (DDE). Photon DDE was taken directly from the film. Neutron DDE was corrected using the reading from the CR39 polymer due to a large signal from neutrons >200 keV, as opposed to the film.

Extremity dose results displayed on Table 4 are found to be in an excellent agreement with those of NRY 21 EPD in the same setup.

Comparison of NSN3 and NRY 21 EPD response

Design response associated with the NSN3 is ±25%, and the NRY 21 is much wider at −60% to +200%. However during this 3-min response testing, the two detectors responded very closely to each other in all three situations. Table 5 shows the response of each device with differing thicknesses of HDPE moderator.

It can be seen that the response of the two devices agree within their margins of error. Indeed, they agree with the more restrictive margin of ±25% from NSN3. An excellent agreement at the 5-cm moderation, which is the thickness of the moderator between neutron source and the cavity during normal operation, has been found.

Measured trunk dose by NSN3 and MicroREM

Using the NSN3 and MicroREM as survey instruments, the neutron and photon ambient dose equivalent rates around the shielding assembly where a subject's trunk would be positioned were measured to be 0.03–0.05 mSv h^{-1} neutron and 0.03–0.05 mSv h^{-1} photon. Assuming 0.10 mSv h^{-1} total ambient dose equivalent rate including both neutron and photon radiation, and assuming ambient dose equivalent is equal to effective dose in this case, an effective dose of 0.017 mSv was recorded for a 10 min IVNAA measurement at a flux of $7 \times 10^8 \pm 30\%$ s^{-1}.

Whole body effective dose

To calculate the whole body effective dose, the tissue weighting factors and fractions of irradiated tissues need to be considered. The mass of one hand and lower arm is about 5% of total body mass, and assigned tissue weighting factors per ICRP 103 are 0.01 for skin and 0.01 for bone surface. The bone marrow is not considered in this case, as there is no active bone marrow in the adult hand, wrist, ulna, or radius (Cristy 1981). Neutron and photon extremity dose to the hand were averaged at 31.7 ± 0.8 mSv and 4.2 ± 0.2 mSv, respectively, from the NRY 21 reading and film badges. The whole body effective dose from hand irradiation was calculated as follows:

$$E_{Hand+arm} = \Sigma_T W_T \times H_T \quad (3)$$

$$E_{Hand+arm} = (.05) \times [(0.01 \times 35.94) + (0.01 \times 35.94)] = 0.036\ mSv. \quad (4)$$

This is a conservative estimation in that the equivalent dose to the arm is lower than the equivalent dose to the hand as it extends away from the source; however, the equivalent dose to the hand is used for the calculation.

To complete the calculation, the effective dose to the rest of the body (calculated at 0.017 mSv) must be corrected to the remaining 95% of body mass. Since the same tissues are considered in the whole body, no correction for tissue weighting factors is required (that is:

$W_T = 1.00$). This is calculated to be 0.016 mSv. The total whole body effective dose then sums to:

$$E = 0.036 + 0.016 = 0.052\ mSv\,.$$

DISCUSSION

From a localized extremity dose, the calculation of effective dose displays a weak relationship at best (Drexler et al. 1993). The emphasis of this research, therefore, is that the subject's extremity dose is low, less than 10% of the Federal occupational limit of 500 mSv (USNRC 2015). The subject's effective dose is minimal whether or not the extremity irradiation is factored into the effective dose determination: at either 0.017 mSv or 0.052 mSv, either value is less than 1% of the Federal occupational limit of 50 mSv. Neither stochastic nor deterministic radiation effects from this extremity dose or effective dose would be expected.

A complex neutron scatter environment is present in this setup. Different energy neutrons, from 0.025 eV to 2.45 MeV incident on the arm from every direction in a mixed flux with x rays and activation gammas, create a situation that makes exact dosimetric calculations and experiments difficult. An excellent agreement between the EPD, film badges, and MCNP simulations for neutron extremity dose was demonstrated. Photon extremity dose also was found to be in an acceptable agreement between the EPD and film badges.

Bremsstrahlung x rays are produced in high intensity and scattered throughout the moderator. A 3-mm-thick lead wrap was added to the DD generator head to attenuate these x rays, and an extremity dose reduction of nearly 1.5 mSv (approximately 20%) was observed. Activation gammas are not as easily shielded because they are created throughout the moderator and at a much higher energy (primarily 2.2 MeV vs. 40 keV).

Fast neutrons in the hand cavity contribute little to the measurement but contribute significantly to extremity dose. Therefore, future work includes a review of the moderator design to maximize the thermal neutron component in the irradiation cavity and minimize the fast neutron flux but still keep the overall irradiation time as low as reasonably achievable. To attenuate more of the bremsstrahlung x rays, adding more lead sheets to the setup will be investigated. Also, a lead lining inside the irradiation cavity to further attenuate any photons before reaching the subject's hand and arm will be evaluated. Through balancing these parameters more efficiently, the goal is to reduce the subject's extremity dose and effective dose to an even lower value.

CONCLUSION

Detailed measurements of extremity dose of 31.7 ± 0.8 mSv neutron and 4.2 ± 0.2 mSv photon (mean of three doses from the NRY 21 and film badges) from a 10 min IVNAA measurement of manganese at a neutron flux of 7×10^8 (±30%) s^{-1} have been completed. The whole body effective dose was found to be minimal at about 0.052 mSv. The obtained results represent the best estimate of extremity and effective dose during a 10-min IVNAA measurement of manganese. These are acceptable doses to apply the technology for a manganese toxicity study in a human population.

Acknowledgments—

This work was supported by the Navy Medical Service Corps Duty Under Instruction Program for D. Sowers; the National Institute for Occupational Safety and Health (NIOSH) grants 1R21OH010044 and 1R21OH010700; the Purdue University Nuclear Regulatory Commission (NRC) Faculty Development Grant NRC-HQ-11-G-38-0006; and Purdue Bilsland Fellowship.

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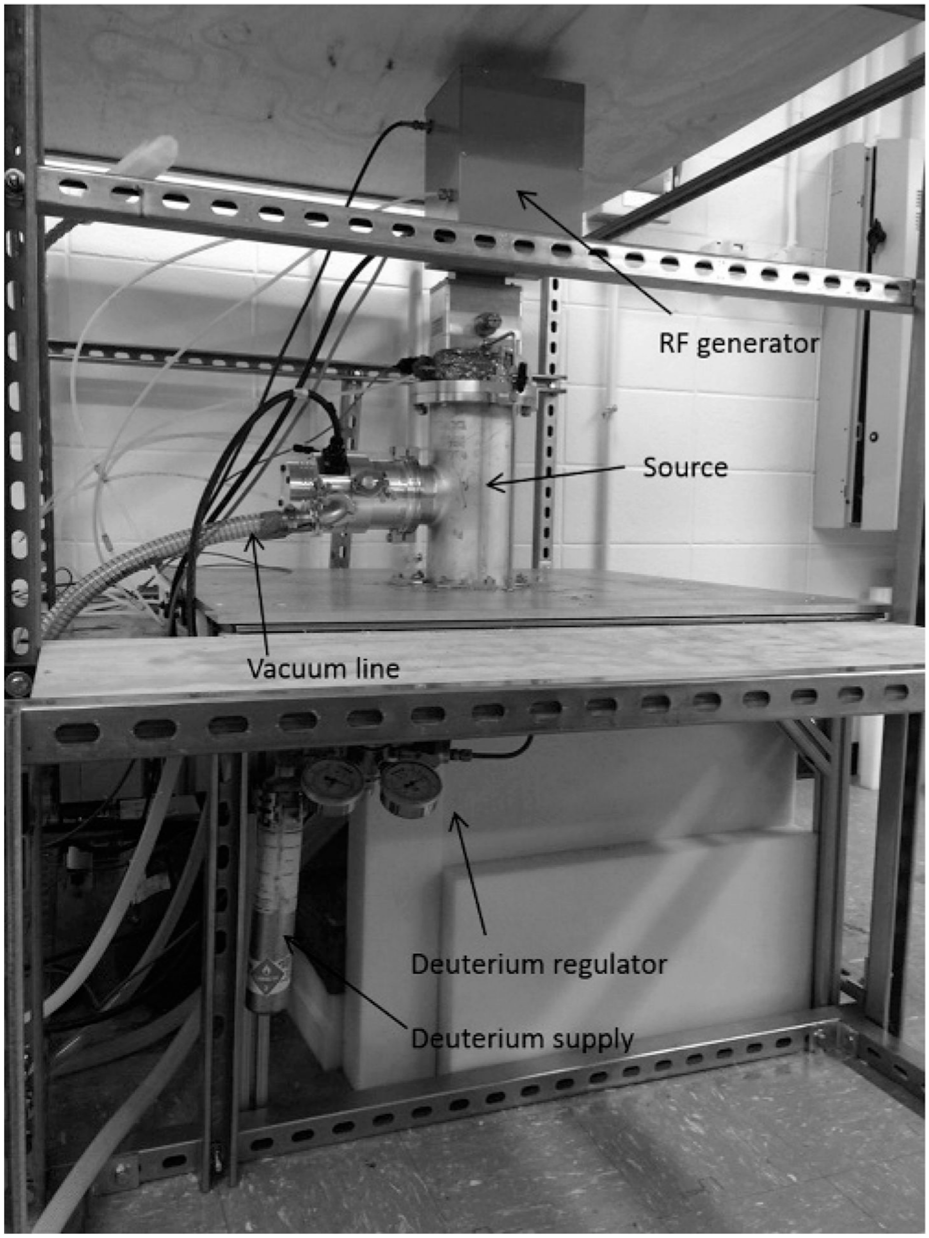

Fig. 1.
Open setup of DD neutron generator setup at Purdue University.

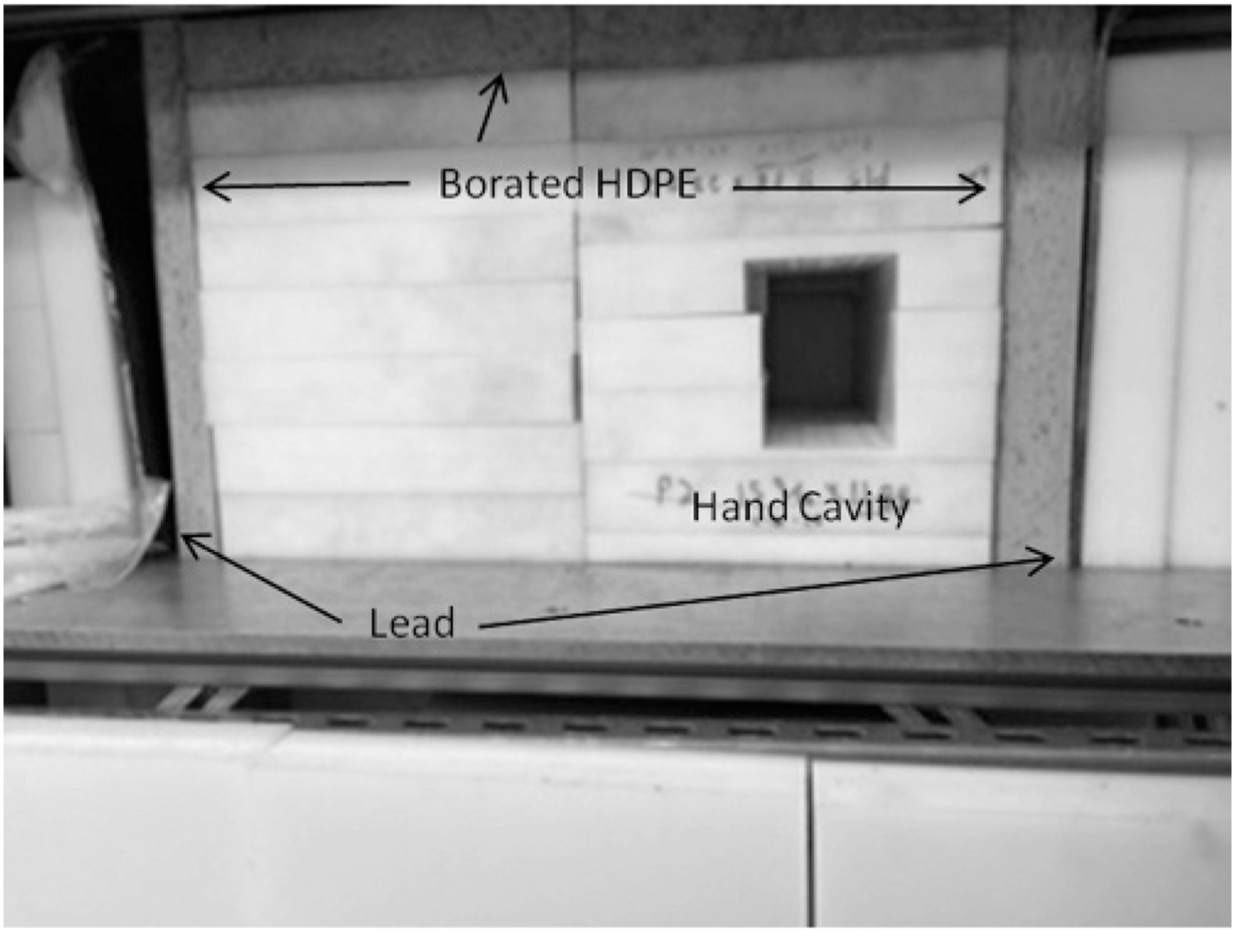

Fig. 2.
Setup showing hand cavity. Source location is 28 cm deep at center of cavity.

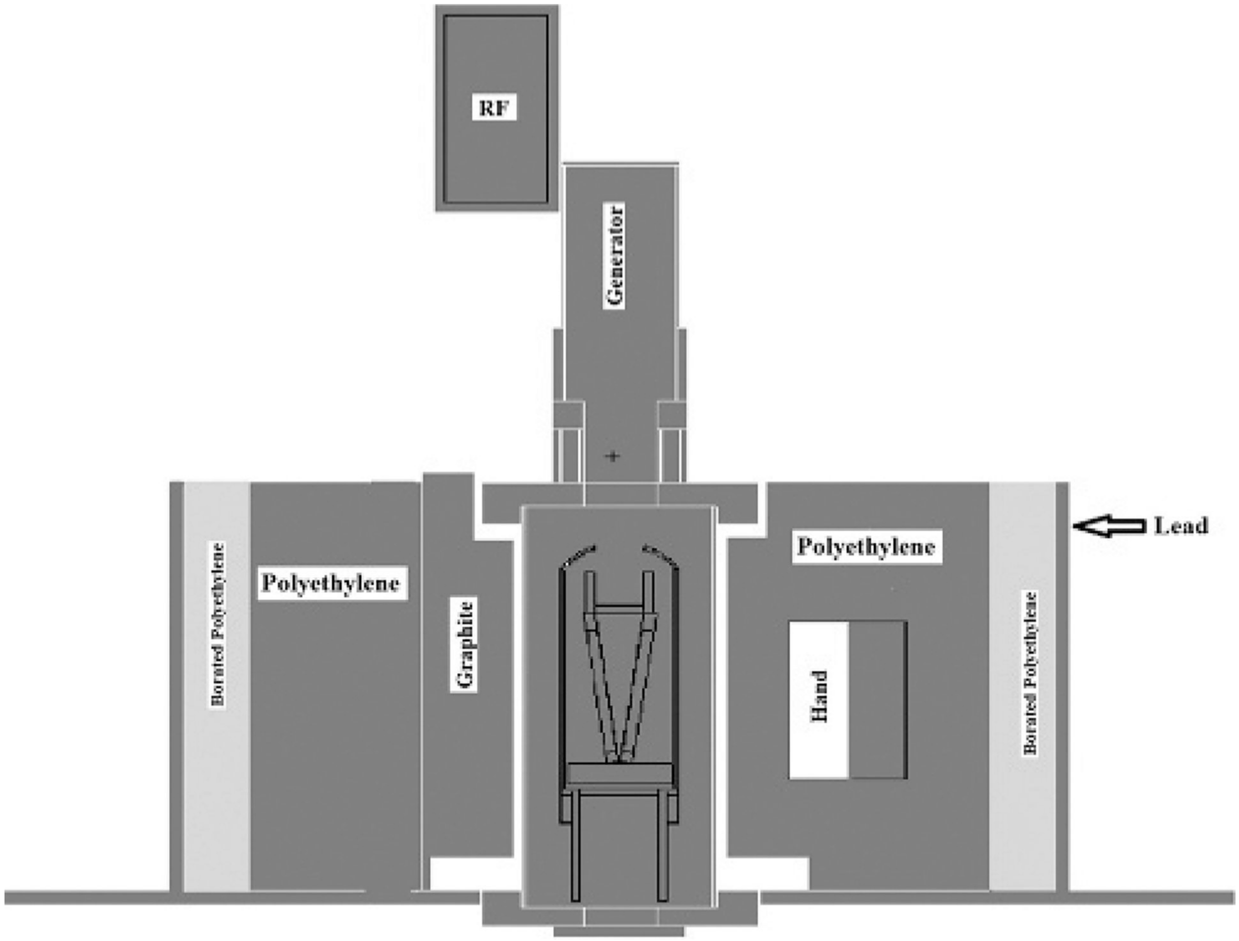

Fig. 3.
MCNP simulation schematic.

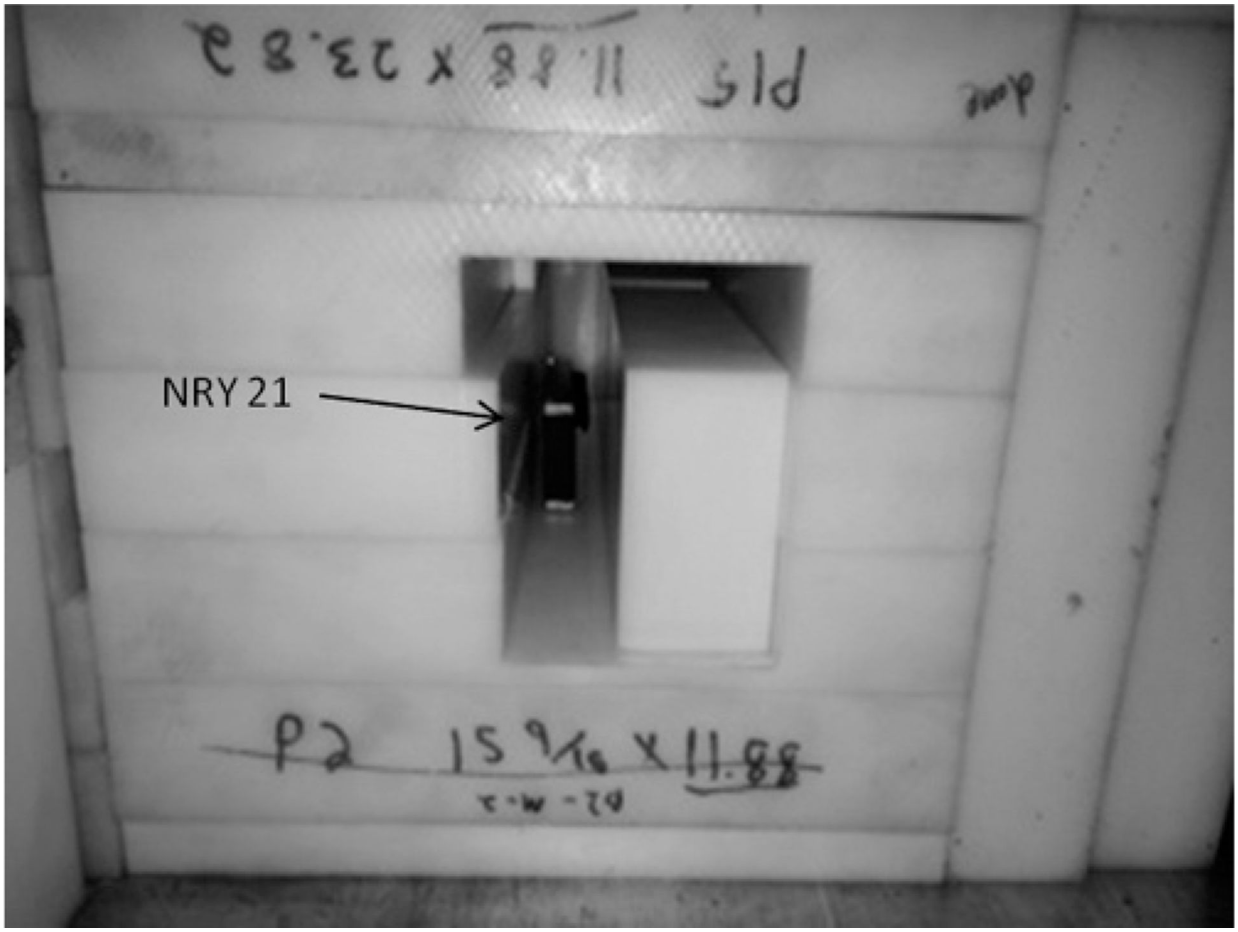

Fig. 4.
NRY 21 EPD placed inside irradiation cavity.

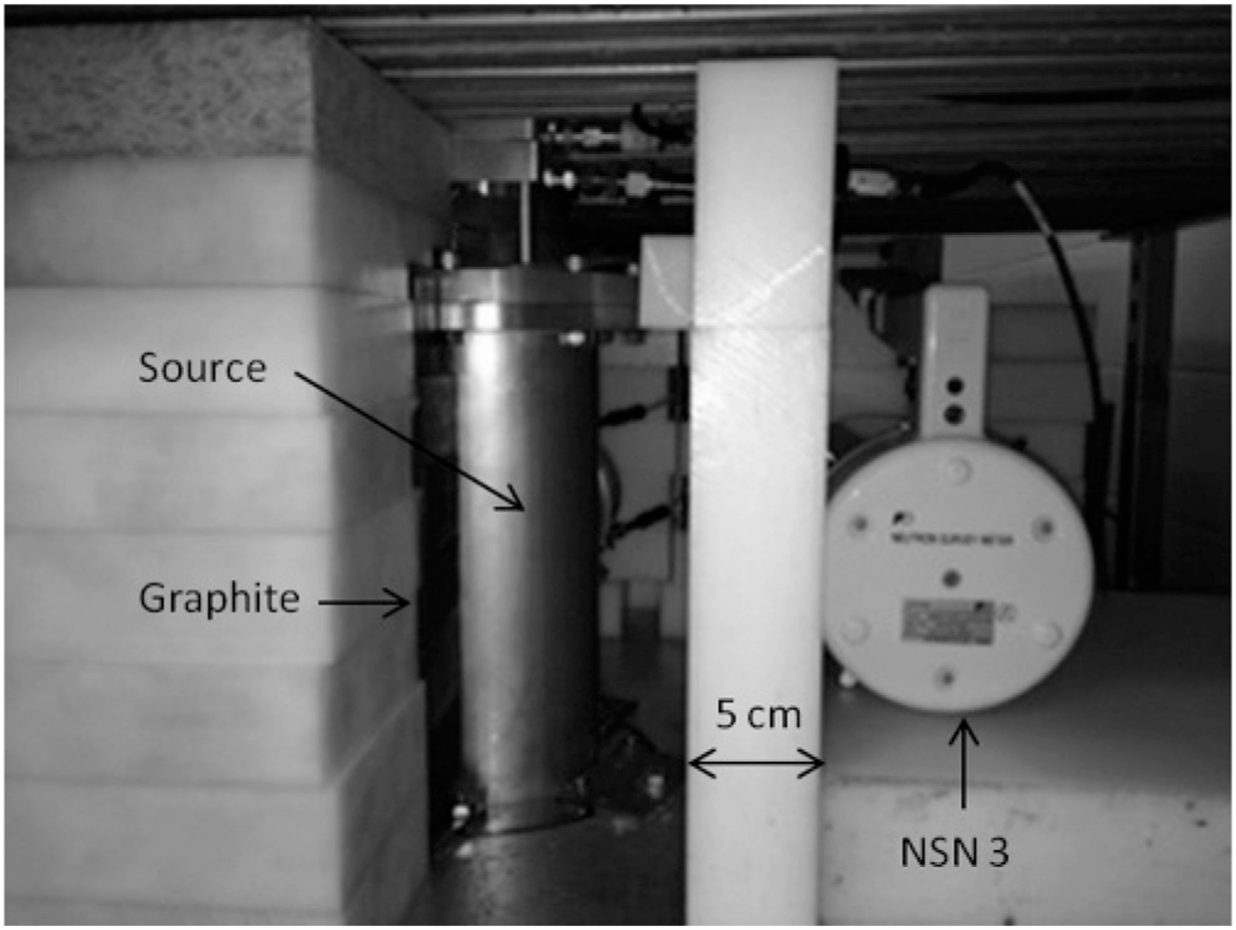

Fig. 5.
Moderator configuration to compare response of NSN3 and NRY 21 EPD.

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Table 1.

MCNP simulation results, extremity dose in tissue and bone.

	Act gamma	Neutron
Hand tissue	0.7	32.3
Hand bone	0.7	11.2
Arm tissue	0.4	4.1
Arm bone	0.4	1.6

(Unit: mSv).

Table 2.

MCNP simulation results, point detector.

Depth in cavity (cm)	Neutron dose (mSv)
27.9	31.9
25.4	30.3
22.9	26.6
20.3	22.3
17.8	17.4

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Table 3.

Neutron and photon extremity dose for 10 min irradiation, measured by NRY 21 EPD facing toward neutron source.

Depth in cavity (cm)	Photon dose (mSv)	Neutron dose (mSv)
27.9	5.6	31.4
25.4	5.7	31.1
22.9	5.2	24.1
20.3	5.1	14.9
17.8	4.2	10.3

Table 4.

Results from film badges irradiated in hand cavity.

	DDE Photon	DDE Neutron
Badge 299	4.4	31.2
Badge 300	4.2	32.6

(Unit: mSv).

Table 5.

Response of NRY 21 and NSN3 to fast, 5 cm HDPE moderated, and 10 cm HDPE moderated neutrons during 3 min irradiation.

	NRY 21	**NSN3**
0 cm (Fast)	17.4	18.7
5 cm	3.6	3.6
10 cm (Thermal)	0.8	0.9

(Unit: mSv).