

# Estimation of Argon-41 concentrations in the vicinity of a high-energy medical accelerator

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## Abstract

This study presents the estimation of <sup>41</sup>Ar concentrations using the neutron activation method. The distribution of thermal neutron flux in a 15 MV medical accelerator (linac) treatment room was determined and contoured by measuring the radioactivities of indium foils irradiated by thermal neutrons. The <sup>41</sup>Ar concentrations were calculated based on the spatial distribution of thermal neutrons. The evolution of <sup>41</sup>Ar concentration with time in the treatment room was predicted and the corresponding radiation dose associated with <sup>41</sup>Ar was derived and shown to be insignificant for both patients and workers, being below the regulatory level. Indium foil activation method showed high detection sensitivity for estimating the low-level <sup>41</sup>Ar in the vicinity of medical accelerators, yielding a minimum detectable concentration of less than 10 Bq m<sup>-3</sup>. © 2007 Elsevier Ltd. All rights reserved.

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## 1. Introduction

High-energy electron accelerators including the electron linear accelerator and the betatron are routinely used to produce high-energy electrons and X rays for cancer therapy. Accelerators operated at above 10 MeV can produce neutrons through photonuclear reactions in the target, field-flattening filters, beam-defining collimators and other accelerator components, resulting in a mixed radiation field in the beam and the treatment room. The calculation of photoneutron yields and the subsequently induced photons in different components of medical accelerators and barriers has been extensively investigated to design shielding to protect personnel outside treatment rooms (Kase et al., 1998; Mao et al., 1996, 1997; McGinley, 1992). The contribution of neutrons and photons can be estimated for therapeutic and radiation safety purposes. Neutron fluences and the corresponding dose can also be measured experimentally

and compared to the calculated results (Lin et al., 2001; Paredes et al., 1999; Palta et al., 1984; Gur et al., 1978; McGinley et al., 1976; Uwamino et al., 1986). Although the dose of photoneutrons is less than 0.5% of that of photons on the beam central axis at the depth of dose maximum, and less than 1% in treatment rooms (Paredes et al., 1999), the photoneutrons can also produce activation of materials in treatment rooms to generate radioactive substances, which raise a concern about radiation safety. To date, little attention has been paid to the gaseous radionuclide <sup>41</sup>Ar, which can be generated by thermal neutron activation of stable <sup>40</sup>Ar in air, although its contribution to the radiation dose of both patients and workers may be negligible.

Measurement of <sup>41</sup>Ar concentration using a gamma-ray spectrometer following the collection of an air sample in a sealed container is convenient but impractical because the <sup>41</sup>Ar concentration is generally lower than the detection limit of the counting system. In situ monitoring of gaseous radionuclides using a gamma-ray spectrometric system may be effective for an air source of infinite space (Chung et al., 1998; Chung and Tsai, 1996) but is not applicable to a treatment room with an air volume of specific geometry. Accordingly, almost no data on <sup>41</sup>Ar concentration near a medical accelerator has been

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reported. In this work, the concentration of  $^{41}\text{Ar}$  in a treatment room equipped with a 15 MV accelerator was determined. Initially, the thermal neutron flux, or thermal neutron fluence rate, was estimated with the activation method using indium foils. Therefore, the variation of the concentration of  $^{41}\text{Ar}$  with time and the corresponding dose were calculated for radiation safety assessment.

## 2. Materials and methods

### 2.1. Neutron activation of indium foils

The activation technique has been widely used for measurement of neutron fluxes and the corresponding doses (Knoll, 1989). Fast and thermal neutrons can be discriminated by irradiating appropriate foil materials and measuring the induced radioactivities (Lin et al., 2001; Paredes et al., 1999; Palta et al., 1984; Deye and Young, 1977; Price and Holeman, 1978; Gur et al., 1978; McGinley et al., 1976; Uwamino et al., 1986). In the measurement of thermal neutrons, indium foils are commonly used due primarily to indium's high cross-section and suitable half life ( $t_{1/2} = 54.1$  m) (Reus and Westmeier, 1983). In this study, indium foils were used to contour the distribution of thermal neutrons around a medical accelerator. The thermal neutron flux is defined and determined by measuring the induced radioactivities of indium foils, as follows:

$$A = \phi_{\text{th}} \times \sigma_{\text{In}} \times N_{\text{A}} \times \frac{m \times a}{M} \times (1 - e^{-\lambda t_i}) \times e^{-\lambda t_c}, \quad (1)$$

where  $\phi_{\text{th}}$  is the thermal neutron flux ( $\text{cm}^{-2} \text{s}^{-1}$ );  $\sigma_{\text{In}}$  is the cross-section of the activation reaction (161 barns);  $N_{\text{A}}$  is the Avogadro's number ( $6.02 \times 10^{23}$  in atoms per  $g$ -atom);  $m$  is the mass of indium foil ( $g$ );  $a$  is the isotopic abundance of  $^{115}\text{In}$  (95.7%);  $M$  is the atomic weight of indium (114.82);  $\lambda$  is the disintegration rate of  $^{116\text{m}}\text{In}$  ( $2.135 \times 10^{-4} \text{ s}^{-1}$ );  $t_i$  is the irradiation time, and  $t_c$  is the time duration between irradiation and measurement.

### 2.2. Neutron irradiation

Twenty indium foils (purity > 99.9%; 25 mm  $L \times 25$  mm  $W \times 1$  mm  $H$ ) with an averaged mass of ( $4.86 \pm 0.07$ ) g were used in the experiment. The foils were placed and distributed evenly in the vicinity of a medical accelerator (Clinac 21EX, Varian, Palo Alto, CA) for neutron irradiation. The electron accelerator provides dual photoenergies with accelerating voltages of 6 and 15 MV. The beam intensity was controlled by changing the pulse interval. Fig. 1 displays a floor plan of the radiotherapy facility. The accelerator was operated at 15 MV for 2.5 min, delivering a dose of 1000 cGy at depth of dose maximum in a water-equivalent phantom with source-surface distance of 100 cm and the collimator open to a field size of  $20 \times 20 \text{ cm}^2$ . For batch irradiation, all measured values were normalized to a reference foil, which was placed at the isocenter (0,0,0), which is exactly 100 cm below the X-ray target, such that the relative intensity of the thermal neutron in the treatment room

can be simply described. The thermal neutron flux can be averaged by integrating thermal neutron flux with distance from the reference point.

### 2.3. Radioactivity measurement

The irradiated foils were immediately transferred to a gamma-ray spectrometric system, which consisted of a 30% high-purity germanium detector (GC3520, Canberra Industries, Meriden, CT, USA). The measured gamma-ray spectra were collected with a multichannel analyzer (35-Plus, Canberra Industries, Meriden, CT, USA) and were further analyzed by gamma-ray spectrum software. The foils were placed immediately on the face of the detector for counting; the efficiency was determined to be 4.0% at the characteristic gamma-ray energy of 417 keV emitted from the activated nuclide  $^{116\text{m}}\text{In}$ .

### 2.4. Calculation of $^{41}\text{Ar}$ concentrations

Argon-41 is produced through the neutron capture reaction  $^{40}\text{Ar} (n, \gamma) ^{41}\text{Ar}$  in air. The argon-40 concentration in air is derived as

$$N_{\text{Ar}} (\text{m}^{-3}) = \frac{P}{RT} \times f_{\text{Ar}} \times N_{\text{A}}, \quad (2)$$

where  $P$  is the atmospheric pressure (1 atm);  $f_{\text{Ar}}$  is the fraction of Ar molecules (1.28%);  $R$  is the universal gas constant ( $8.2 \times 10^{-5} \text{ atm m}^3 \text{ mol}^{-1} \text{ K}^{-1}$ ), and  $T$  is the temperature in the treatment room (295 K). Therefore,  $N_{\text{Ar}}$  was calculated to be  $3.18 \times 10^{23} \text{ m}^{-3}$ .

The accumulation of  $^{41}\text{Ar}$  concentration ( $C_{\text{Ar}}$ ) by neutron irradiation in the treatment room with irradiation time  $t_i$  is given by

$$C_{\text{Ar}} (\text{Bq m}^{-3}) = F \times \bar{\phi}_{\text{th}} \times \sigma_{\text{Ar}} \times N_{\text{Ar}} \times (1 - e^{-\lambda_{\text{Ar}} t_i}), \quad (3)$$

where  $\bar{\phi}_{\text{th}}$  is the averaged thermal neutron flux in the room;  $\sigma_{\text{Ar}}$  is the neutron capture cross-section of argon (0.64 barns); and  $\lambda_{\text{Ar}}$  is the disintegration rate of  $^{41}\text{Ar}$  ( $1.05 \times 10^{-4} \text{ s}^{-1}$ ) (Reus and Westmeier, 1983). The neutron capture cross-sections of  $\sigma_{\text{In}}$  in Eq. (1) and  $\sigma_{\text{Ar}}$  in Eq. (3) were obtained from the published data (Lederer and Shirley, 1977). A correction factor  $F$  is introduced in Eq. (3) to accommodate the neutron energy distribution of the medical accelerator. The  $F$  can be estimated by relating the thermal neutron flux, determined by Eq. (1), to the  $^{41}\text{Ar}$  concentration, measured at the same irradiation positions (as described in detail in the following section).

### 2.5. Estimation of the $^{41}\text{Ar}$ concentration and corresponding dose

An air-filled Marinelli cylindrical container ( $\phi = 25$  cm;  $H = 20$  cm) with a volume of 9.2 L was irradiated together with an indium foil in the treatment room, but placed outside the direct radiation beam to prevent fast neutron interference, to determine the correction factor  $F$  in Eq. (3). Prolonged irradiation (10 min) was conducted to ensure that the  $^{41}\text{Ar}$  concentration in the container was measurable. After irradiation, the foil and

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