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Accidental neutron dosimetry with human hair

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HIGHLIGHTS

- Human hair contains sulfur.
- Reaction $^{32}S(n,p)^{32}P$ can be used for dosimetry of fast neutrons.
- Relation between ^{32}P activity and neutron dose can be derived for a specific neutron spectrum.

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ABSTRACT

Human hair contains sulfur, which can be activated by fast neutrons. The $^{32}S(n,p)^{32}P$ reaction with a threshold of 2.5 MeV was used for fast neutron dose estimation. It is a very important parameter for individual dose reconstruction with regards to the heterogeneity of the neutron transfer to the human body. Samples of human hair were irradiated in a radial channel of a training reactor VR-1. ^{32}P activity in hair was measured both, directly by means of a proportional counter, and as ash dispersed in a liquid scintillator. Based on neutron spectrum estimation, a relationship between the neutron dose and induced activity was derived. The experiment verified the practical feasibility of this dosimetry method in cases of criticality accidents or malevolent acts with nuclear materials.

1. Introduction

The unique feature of radiologic accidents with neutron exposure is the induced radioactivity in body tissues, metal objects, and clothing. Human body contains stable elements which can be activated by neutrons. The nuclear reactions $^{23}Na(n,\gamma)^{24}Na$ and $^{32}S(n,p)^{32}P$ are of the greatest practical importance (IAEA, 1982; Feng et al., 1993; Miele and Lebaron-Jacobs, 2008) in this regard. The $^{32}S(n,p)^{32}P$ reaction with a threshold neutron energy of 2.5 MeV enables to estimate fast neutron dose based on the measurement of ^{32}P activity (Lebaron-Jacobs et al., 2007). The activation technique can complement standard personal dosimetry.

Human hair consists of proteins, lipids, water, trace elements and pigments (Robbins, 2012). In terms of raw elements, on average, hair is composed of 45% carbon, 28% oxygen, 15% nitrogen, 7% hydrogen, and 5% sulfur. However, the natural content of sulfur exhibits little variation from individual to individual or with anatomical distribution (Rutherford and Hawk, 1907). For dosimetry purposes, it usually is assumed that the content of sulfur in human hair is 45 mg g^{-1} (Lebaron-Jacobs et al., 2007). ^{32}S, when irradiated, gives ^{32}P following the $^{32}S(n,p)^{32}P$ reaction. ^{32}P is a pure beta emitter with a maximum energy of 1.7 MeV and a 14.3 days half-life. Analysis of hair samples taken from the head and front, left, right and back areas of the body can provide important information about the person's orientation at the time of an accident, based on the relative specific activity of each sample (Takeda et al., 2001). When hair is not contaminated by fission products, direct beta counting of ^{32}P is viable. Even in this case, hair should be carefully rinsed in order to remove ^{31}Si ($T_{1/2}=2.6$ h) and ^{24}Na ($T_{1/2}=15$ h) produced by activation of silicium from dust or peeling skin and of sodium from perspiration, respectively (Hankins, 1969; Feng et al., 1993). The activation occurs by means of reactions $^{30}Si(n,\gamma)^{31}Si$ and $^{23}Na(n,\gamma)^{24}Na$.

Severe neutron exposures are usually connected with criticality accidents (McLaughlin et al., 2000; Gusev et al., 2001). However, during the last decade not only nuclear accidents, but also eventualities of radiologic terrorism have been considered as a potential risk (González, 2007; Hall and Giaccia, 2012; Petrenko et al., 2005). Possible scenarios as the detonation of a radiological

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dispersal device or improvised nuclear device have been mentioned. Numerous institutions and expert groups have pointed out the need for dose assessment techniques that can be used in the aftermath of serious nuclear accident or terrorist event (González, 2007; Alexander et al., 2006; Klemic et al., 2006). The purpose of this experimental work was to establish a neutron dosimetry method based on utilization of human hair.

2. Materials and methods

2.1. Hair samples

Human hair from the head of one donor was divided into 9 portions of approximately 0.5 g. Each portion was weighed by a high-precision scale (ED 224S-OCE, Sartorius AG, Germany). The portions were packed in using a thin polyethylene film and sealed. These samples were inserted into a polyethylene vial. The used hair was natural. It was neither colored nor permed.

2.2. Irradiation

VR-1 "Sparrow" training nuclear reactor at the Czech Technical University in Prague, the Faculty of Nuclear Sciences and Physical Engineering (http://www.reaktorvr1.eu, last accessed on 13 November 2013), was used as a source of neutrons. It is a pool-type, light water reactor based on enriched uranium. For experimental purposes, the reactor is equipped with a radial horizontal channel (diameter of 200 mm) and vertical channels of different diameters. These channels can be used for samples irradiation.

The vial with the hair samples was inserted into a chosen position of the radial horizontal channel by means of a special holder. The position was located 3 cm from the channel axis at a distance of 22 cm from the active zone border. Neutron spectrum in this position is illustrated in Fig. 1. It was calculated using the MCNP code and corresponded to the maximum power of the reactor. The samples were irradiated at one third of the maximum reactor power. The irradiation time was 1423 s, which is negligible compared with the ^{32}P half-life.

2.3. Radiochemical preparation

The hair samples were washed with detergent, demineralized water and ethanol and dried for 30 min. The efficiency of washing procedure was checked by gamma spectrometry counting in a sample originally containing maximum activity of ^{24}Na. Washed and dried hair was either placed directly on the counting planchette and covered with plastic tape or carefully (slow ramp) ashed at 800 °C for 30 min (Feng et al., 1993).

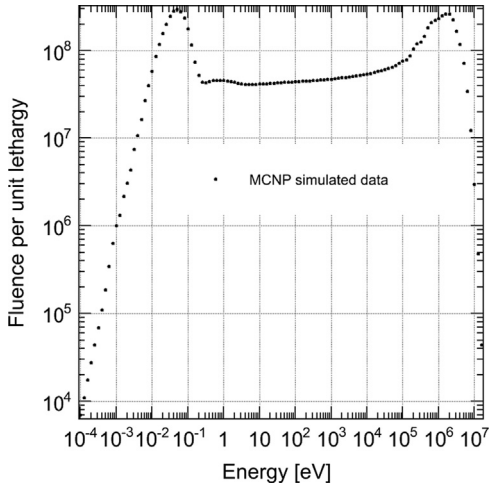

Fig. 1. Calculated neutron spectrum.

2.4. Activity measurement

Two measurement systems were used in the experiment. Both instruments were calibrated with ^{32}P standard solution beforehand. A low level gas flow proportional counter (PC) was used for both direct counting and ashed hair counting. Because the used hair weight on planchettes was lower than 14 mg cm^{-2}, no self-absorption corrections were necessary (Hankins, 1969). Samples of hair or hair ash were secured with plastic adhesive tape in the same configuration, which was used during the calibration of the instrument. Low level Tricarb 2770 liquid scintillation counter (LSC) was used for ashed hair counting. The counting window (counting energy range) was set at 6–1700 keV. Instagel liquid scintillator was used and the ratio of aqueous/LS was 3/17. The addition of the hair ash did not influence quenching as preliminary measurements with the standard solution of ^{32}P had shown. A possibility of direct beta counting of hair submersed in the liquid scintillator cocktail was tested with either 0.2326 g of irradiated hair dispersed in the scintillator or the whole pack immersed in the cocktail.

With the proportional counter the counting efficiency was 0.45 s^{-1}/Bq, with liquid scintillation it was 0.98 s^{-1}/Bq.

The actual counting with the proportional counter started on the next day after the irradiation and repeated on ten following days. The measurement with the LS counter started on the third day and was repeated 5 times.

All the activities measured were normalized to the time of irradiation and 1 g hair weight.

2.5. Relation between induced activity and neutron dose

Derivation of the relationship between the induced activity and neutron dose comes from the activation equation (IAEA, 1982):

$$A = \lambda N \int \sigma(E)\Phi(E)dE$$

where A is the ^{32}P activity in the time of irradiation, λ is the decay constant, N is the number of ^{32}S atoms in the sample, $\sigma(E)$ is the cross section as a function of energy E for the reaction considered and $\Phi(E)$ is the fluence of neutrons per unit energy interval. N can be expressed as $6.02 \times 10^{23}(m/M)p$, where m is the mass of sulfur in the sample, M is the molar mass of sulfur and p is the isotopic abundance of ^{32}S. For a hair sample of 1 g, the values of these quantities and parameters are: $m=45$ mg, $M=32$ g/mol, $p=0.95$. Consequently, the number N of ^{32}S atoms in 1 g hair is 8.042×10^{20}. The ^{32}P decay constant $\lambda=5.613 \times 10^{-7}$ s^{-1}. The cross section $\sigma(E)$ can be applied in form of a mean value $\bar{\sigma}$ calculated specifically for the present neutron spectrum. The cross section data for the ^{32}S(n,p)^{32}P reaction can be found in the ENDF library (Chadwick et al., 2011).

The dose that is of primary concern is the surface-absorbed dose, because the maximum absorbed dose from fast neutrons occurs close to the surface of the body where the neutrons are incident. The dose is defined as dose averaged over a surface element, 2 cm thick, in the middle of a cylindrical tissue-equivalent phantom (IAEA, 1982). The cylinder (30 cm in diameter and 60 cm high) is irradiated by a broad, parallel beam of neutrons incident normal to the axis if the cylinder. The surface-absorbed dose has two components – the surface absorbed dose from heavy charged particles (recoils and ^{14}N(n,p)^{14}C) and the surface absorbed dose from gamma radiation produced in the body via ^{1}H(n,γ)^{2}H reaction. At energies above 0.1 MeV, the surface-absorbed dose comes mainly from recoils and heavy charged particles, while below 0.01 MeV