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MEASUREMENT OF D_2O IN WATER USING $^2H(\gamma,n)^1H$ REACTION

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Abstract

Utilization of Photodisintegration reaction of Deuterium, $^2H(\gamma,n)^1H$, is a precise technique for determining the percentage of ^2H in tritiated and natural water. The $^{23}Na(n,\gamma)^{24}Na$ reaction is a suitable γ-source for producing the $^2H(\gamma,n)^1H$ reaction. Tehran Research Reactor was used as a neutron source for producing ^{24}Na isotopes. The BF_3 detectors were used to detect the neutron emission of H_2 content of water. We obtained a linear calibration plot by repeating the experiment with many samples of different concentrations of tritiated water. With this simple portable apparatus which have been used for this purpose for the first time, and corresponding linear plot, we can monitor and determine easily and accurately the ^2H concentration of an unknown sample.

Keywords: Gamma source (Na^{24}); Tehran research reactor; Thermal neutron; BF_3 detectors; D_2O water; Linear plot

Introduction

Advancements in nuclear physics have provided powerful means for chemical analysis. Bombardment of selected nuclei with particles or gamma rays, to produce certain nuclear reaction, and detecting the emitted radiation is a well-established analytical process in nuclear technology. Thermal neutrons have been extensively used in neutron activation analysis. Although gamma irradiation did not find wide application, it has been utilized in Deuterium Photodisintegration [1-3].

When gamma rays of sufficient energy, (greater than the neutron binding energy) hit a light element, a probability exists that a neutron will be emitted by the energized nuclei. The majority of nuclei have a neutron binding energy of 5 MeV or more, except $^9_4$Be (1.67 MeV) or $^2_1$H (2.23 MeV). Gamma rays from artificially produced radio-nuclei can serve in Photodisintegration reactions. A standard ^{228}Th or ^{24}Na gamma source is placed at the center of a vessel filled with tritiated water (D_2O). The yield, the number of neutrons produced per second by 1 g of Be or D_2O at a distance of 1 cm derived from 1 curie of any of various gamma sources is given in Table 1.

The advantages of Na or Th sources are:

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(a) Reproducible neutron yield.

(b) The choice of E_n by choosing a suitable value of E_γ.

(c) The wide variety of artificial gamma sources which are available from atomic reactors.

(d) The neutrons produced are more mono-energetic than those obtained from (α,n) sources.

Wattenberg [4,5] gives the equation of the emitted neutrons for the energy (E_γ):

$$E_n = \frac{A-1}{A}\left[E_\gamma - Q - \frac{E_\gamma^2}{1862(A-1)}\right] + E_\gamma \cos\theta\left[\frac{2(A-1)\cdot(E_\gamma - Q)}{931\cdot A^3}\right]^{\frac{1}{2}}$$

where, for A(γ,n)A-1 reactions:

A: The mass of target nucleus

E_γ: The energy of the incident gamma rays in MeV [the threshold energy, in MeV, for the (γ,n) reaction] on target nucleus

|Q|: 2.2246 MeV for $H^2(\gamma,n)^1H$

θ: The angle between the path of gamma rays and the M direction of the emitted neutrons.

When deuterium nuclei undergo Photodisintegration, they emit neutrons with relatively high energies. These neutrons must be slowed down to improve their detection efficiency. The availability of efficient neutron detectors and the electronics necessary to obtain a count will provide a nondestructive means for determining deuterium oxide in water.

$^2H(\gamma,n)^1H$ Reaction

Bombarding a water sample with a flux of gamma rays produces this reaction. Under the effect of these rays, the present deuterium (^2H or D) nuclei will disintegrate to produce neutrons with an energy of about 0.2 MeV (Table 1). After being slowed down and detected, the monitored neutrons can be associated to the ^2H concentration in the water sample.

Safety considerations limit the intensity of gamma rays that can be utilized in this monitoring technique. The main disadvantage of Na^{24} is that it has a short life and must be reactivated as needed.

Gamma Source

The principal means for producing ^{24}Na is from $^{23}Na(n,\gamma)^{24}Na$ reaction. Either sodium carbonate or lime glass (10-wt %sodium) can prepare this gamma source by means of neutron activation. The activation can easily be accomplished in a reactor with a thermal neutron flux of about 3×10^{12} $cm^{-2}s^{-2}$.

Overman *et al.* [2] prepared a fairly strong gamma source by electro-depositioning. They imbibed 4 Ci of ^{228}Th into a Bismuth matrix to reduce the emission from (α,n) reactions (Table 2). This source was so effective that they were able to determine D_2O, even in depleted H_2O. Calibration with standard mixtures of D_2O over concentrations of 120 to 190 ppm gave a linear response with an average standard deviation of 0.6%.

Neutron Detection

Fast neutrons are of little interest to us, but after being moderated, the resulting thermal neutrons are very efficient to produce nuclear reactions. This not only provides a source of artificially produced isotopes, but also gives a means of detecting neutrons.

The monitored neutrons can be related to the D_2O concentrations in water sample. An equation for D_2O detection sensitivity is derived elsewhere [3]. This equation is expressed as:

$$S = \frac{R}{N_D G} = \frac{\sigma A \varepsilon}{4\pi r_\gamma^2} \quad (1)$$

where

S: Detection sensitivity

R: Moderated neutron count rate (n/s)

N_D: Number of Deuterium nuclei

G: Gamma source strength (γ/s)

σ: $^2H(\gamma,n)^1H$ Cross-section (cm^2)

A: Correction factor for gamma ray attenuation

ε: Efficiency of neutron detector(s):

$$\varepsilon = \varepsilon(r) = \sum_{i=1}^{N} \varepsilon(r_i),$$

where

$\varepsilon(r_n)$: The sum of the efficiencies for all neutron detectors.

r_i: Source to Deuterium distance (cm). A few remarks concerning the variables that determine S are in order:

σ: The 2754-KeV gamma of ^{24}Na (15 h) has a $\sigma=1.5\times10^{-27}$ cm^2. This σ is 23% larger than that of the 2615-KeV gamma of ^{228Th} (1.9 y), which has been used in some $^2H(\gamma,n)^1H$ monitors [3].

A: The attenuation for gamma rays with energies greater than 2000-KeV is close to unity for typical

probe moderators [3] (A ≈ 1 in Equation 1).

Table 1. Characteristics of some radioactive (γ,n) sources [4]

γ- Source	Target	Half-life	E_γ (MeV)	E_n (MeV)	Standard yield
^{24}Na	D_2O	14.8 h	2.76	0.22	27×10^4
^{24}Na	Be	14.8 h	2.76	0.83	13×10^4
^{88}Y	Be	87 d	1.9, 2.8	0.16	10^5
^{88}Y	D_2O	87 d	2.8	0.31	0.3×10^4
^{124}Sb	Be	60 d	1.7	0.025	19×10^4
Rd-Th*	D_2O	1.9 y	2.26	0.197 ± 0.010	9.5×10^4

*Rd-Th source emits some neutrons through (α,n) reactions with light elements around it, yielding an undesirable background.

Table 2. Basic radiations of ^{24}Na and ^{228}Th

Nuclide	Radiations			Produced by
	α	β^-	γ	
^{24}Na		4.17 max (0.003%) 1.389 max (100%)	1.369 (100%) 2.754 (100%)	^{23}Na(n,γ)^{24}Na
^{228}Th	5.43 (71%) 5.34 (28%)		0.084 (1.6%) 0.132 (0.2%) 0.167 (0.1%) 0.214 (0.3%)	$Ra^{226}(n,\gamma)Ra^{227}(\beta^-)$ $Ac^{227}(n,\gamma)Ac^{228}(\beta^-)$

ε: Neutron detector efficiency depends on the neutron moderator and the geometry. Winn *et al.* used Helium (^{3}H) detectors in their work [3].

At low neutron energies, the total cross section for Boron (B) is higher than that of Hydrogen [6]. Therefore, the use of B detectors will probably improve the detection efficiency.

r_γ: Minimizing Source to Deuterium distance, maximize neutron production for a given gamma intensity [3].

Statistical Considerations

Using statistical considerations, we can express the count rate limit ΔR, corresponding to just-detected D_2O, as follows [3]:

$$\Delta R = 2\left(\frac{R_b}{t}\right)^{\frac{1}{2}} + \frac{1}{t} \qquad (2)$$

Determining the R_b (Count/min) in a particular interval of time t enables us to find ΔR according to Equation 2. For any particular detecting system, S can be easily calculated. Hence, by rearranging Equation 1, the detection limit ΔN_D can be expressed by:

$$\Delta N_D = \frac{\Delta R}{SG} \qquad (3)$$

For a 1- mCi source, and for various pipe geometries, the minimum detection limit $(\Delta N_D)_{min}$ varies 5 to about 100 μl/cm for D_2O streams. In other words, for the best pipe geometry $(\Delta N_D)_{min}$ is:

$$\Delta N_D = 5\ \mu l\ D_2O \equiv 1.5 \times 10^{20}\ D_2O\ Molecules$$

Since natural water contains about one D_2O molecule in every 7000 water molecules, then it is expressed that 7000 × 0.005 = 35 ml of natural water is required to avail 1.5×10^{20} D_2O molecules. This volume is fairly small and does not present practical difficulties.

Instrumentation

All the instruments needed are usually available in any nuclear research center. This apparatus is portable, which is very important factor for monitoring any

residual heavy water in pipes during the maintenance of power reactors.

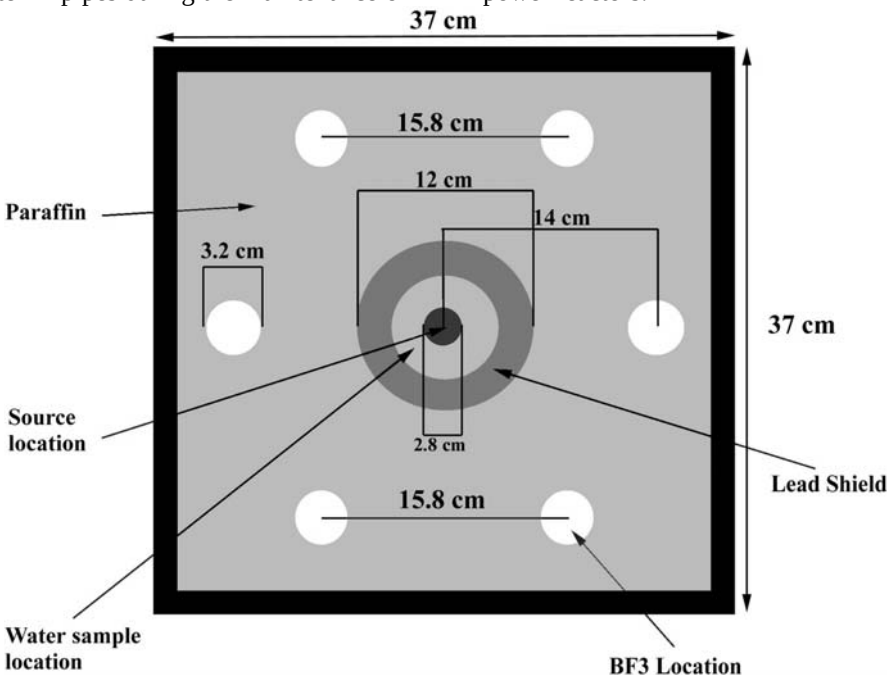

Figure 1. Top view of apparatus.

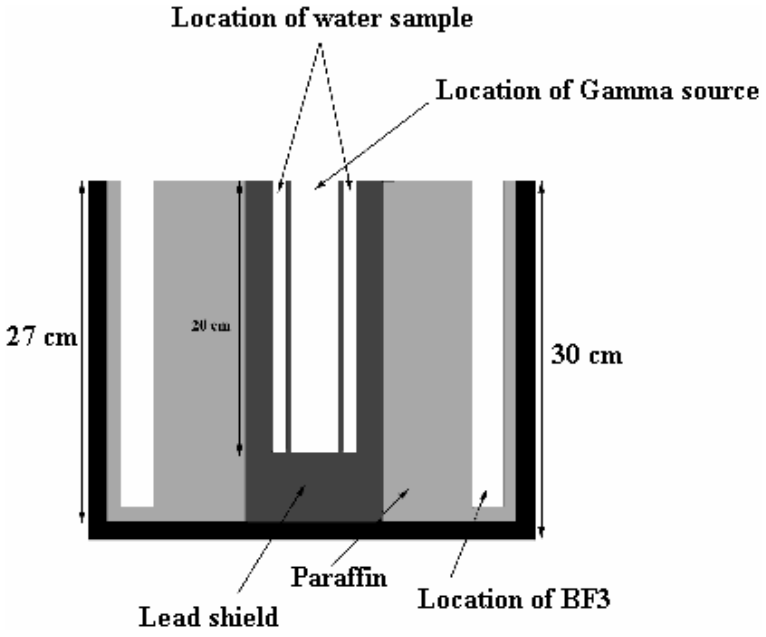

Figure 2. Side view of apparatus.

Design of Experiment

The BF_3 detectors are surrounded by attenuators and the whole apparatus in confined within a 37.0 × 37.0 × 30 cm^3 cubic box. Inside of it, six BF_3 detectors are placed for collecting neutrons. The detectors are located on the circumference of a circle that has a 14 cm diameter. The center of the circle coincides with the center of the box and six BF_3 detectors are located along vertexes of a hexagon. The specific locations for the gamma source and the water sample are at the center of box. As the gamma source is from ^{24}Na, it must be separated from the water sample. For this purpose, the

source container and water sample are designed in the form of coaxial cylinder (Figs. 1 and 2). The outer cylinder is made of lead to prevent the gamma rays from interacting with D_2O. Solid paraffin is used for attenuating the fast neutrons produced from the $^2H(\gamma,n)^1H$ reactions.

Source Preparation

In preparing the gamma source, 0.7 grams of Na_2CO_3 (99.9 percent pure) were poured into a polyethylene capsule, measuring 5-mm in diameter and 28 mm in height. This capsule is placed inside a second container made of aluminum. This ensemble was irradiated for 4 min with a flux of 10^{12} n/sec cm^2 inside the core of the research reactor at the Atomic-Energy Organization of Iran (AEOI). The activity of the source, after interrupting bombardment, was 40.35 mCi.

Electronic Circuit

The electronic circuit used (Fig. 3), includes the probes and the electronic units. All of the BF_3 detectors in this experiment are connected to a preamplifier. The bias of BF_3-detectors operating voltage for detection system was 1250 V. A Multi Channel Analyzer (MCA) collects the amplified pulses. To eliminate the gamma rays from the background, the lower limit voltage of the SCA is adjusted to 3 V. This voltage is less than any typical pulse produced by neutrons (4.5 V), and more than any gamma rays from the background. Integrating the surface under a spectrum curve, at 400-second intervals does the count (Fig. 4). After determining the background, the experiment is repeated using 200 cm^3 water samples, containing different D_2O percentages.

The heavy water (tritiated) samples used are 99.8% pure. For calibrating this system, the experiment was repeated with 21 different concentrations of heavy water. The concentrations ranged from 0.5% to 25.0%. We neglected the error in the D_2O concentration (this concentration is less than 150 ppm or 0.15% in ordinary water). The results, after adjusting for errors, are given in Figure 5. The corrections we have performed were background counts, coefficient reduction of source activity, and adjusting the statistics for errors in counting. The count rate during 400 sec, ranged from 280 ± 20 for 0.5% D_2O to 8050 ± 90 for 25% D_2O. A linear calibration plot was obtained. We tested this plot for a few determined mixtures. For example, using a 16% of D_2O, we should have a count rate equal to 4860. The experimental result gave 4760, which is in good agreement with the calibrated plot.

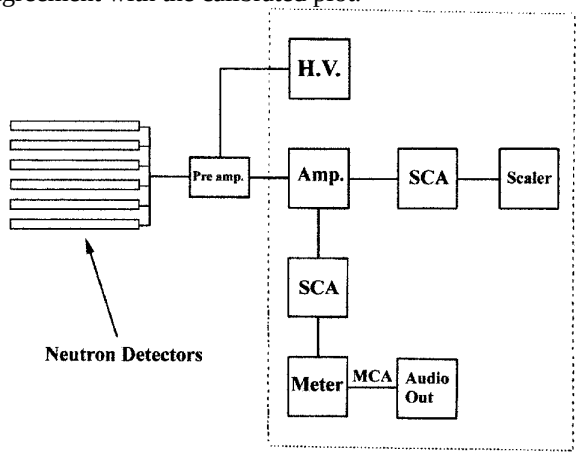

Figure 3. Layout of circuit used with main elements.

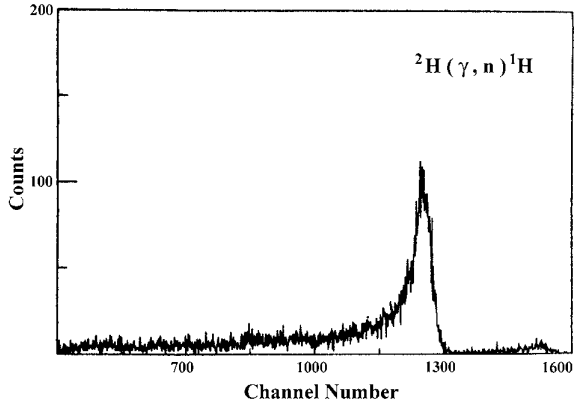

Figure 4. A typical spectrum of $^2H(\gamma,n)^1H$ reactions collected in MCA.

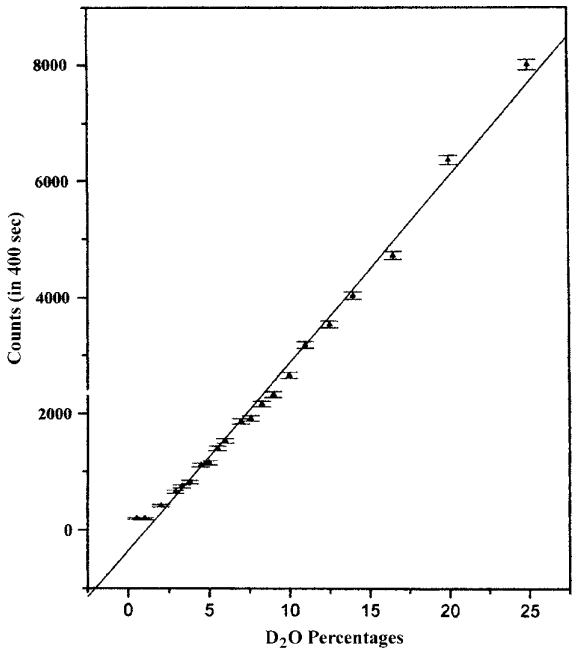

Figure 5. Calibrated plot representing the count rate as a function of ^{2}H percentage.

Conclusions

(1) Utilization of the simple reaction $^{2}H(\gamma,n)^{1}H$, can provide a very powerful analytical technique for determination of Deuterium in water. The technique is so efficient that it can even detect D_2O in depleted water.

(2) For preliminary experiments, if ^{24}Na (15 h) is not obtainable, one can utilize ^{228}Th (1.9 y) as the gamma source. As noted previously, the latter nuclide can provide more-lasting gamma source that is necessary for any practical works.

(3) Parameters like the volume of sample, the detector type, and different Gamma source, examined in order to reach the most efficient detection capacity. The recommended BF_3-detectors will possibly serve the purpose.

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