Nuclear Instruments and Methods in Physics Research B 337 (2014) 62–67

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

Determination of boron in water using neutron scattering and transmission, and prompt gamma ray neutron activation analysis methods: A comparative study

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ARTICLE INFO

Article history:
Received 10 March 2014
Received in revised form 14 July 2014
Accepted 17 July 2014
Available online 12 August 2014

Keywords:
Neutrons
Boron
Prompt gamma-rays
Scattering
Transmission

ABSTRACT

The boron concentration in water was determined using neutron scattering and transmission, and prompt gamma ray neutron activation analysis methods. The experimental setup is based on a Pu-Be neutron source, ^{3}He neutron detectors and an NaI(Tl) gamma-ray detector. Water samples of different volumes and known amounts of boron were prepared. Transmitted and scattered neutrons and prompt gamma rays resulting from neutron capture reactions within the samples were simultaneously measured. The sensitivities of the neutron scattering and transmission measurements were determined to be better than those of the prompt gamma ray neutron activation analysis. The neutron scattering measurements were determined to be more sensitive than those of the neutron transmission measurements.

1. Introduction

Boron has two stable, naturally occurring isotopes, ^{10}B and ^{11}B, with natural abundances of approximately 19% and 81%, respectively [1]. Boron is mainly present as undissociated boric acid in water. It is essential to plant life (in minute quantities); however, larger amounts are toxic to plants and may cause soil sterilization [2]. In the nuclear industry, boric acid is mixed with moderators as a neutron poison to control excess neutron flux [2]. Boron-alloyed steel sheets are used in nuclear engineering as neutron shielding for radioactive waste disposal equipment, such as components for compact fuel storage racks and transportation baskets [3]. It is used in other important applications [3], such as some medical applications [4], including boron neutron capture therapy (BNCT) [1]. In addition, it may be a source of ground water contamination [2]. Thus, it is important to search for simple, fast, alternative methods for boron detection in aqueous solutions.

Prompt γ-ray neutron activation analysis (PGNAA) is the traditional nuclear method for boron detection in many sample types belonging to various fields. It is a well-known, rapid, nondestructive, instrumental technique that complements conventional neutron activation analysis (NAA). The PGNAA method is based on the detection of γ-rays from neutron capture emitted by the target material as it is irradiated with neutrons [5–8]. The greatest number of publications using this technique is concerned with light elements such as H or with large elements having a large cross section (Cd, Sm, B and Gd) [1]. Usually, PGNAA facilities rely on a neutron beam from a reactor that impinges on a sample to produce the neutron capture γ-ray emission reactions. However, other PGNAA facilities relying on radioisotope neutron sources (^{252}Cf, Pu-Be and ^{241}AmBe) and/or accelerator-based neutron generators have been widely used and tested for elemental analysis in various fields. These fields include the bulk detection of illicit drugs, minerals, oil, explosives and other contraband materials as well as the field of industrial exploration [9–21]. Boron is one of the most sensitive elements in PGNAA because the cross section for the ^{10}B(n,αγ)^{7}Li* reaction is large (σ_{th} = 3840 b) [22]. Boron is found via the γ-ray line at 478 keV, which is emitted by the recoiling ^{7}Li* nucleus.

Lower (better) detection limits for boron as well as other elements in the investigated samples by a PGNAA depend on several factors: the neutron flux, cross section of the investigated isotope, background resulting from the surrounding matrix of the investigated sample, recording time of the γ-ray spectra and the efficiency of the γ-ray detection system used. As the values of these parameters increase, better detection limits can be achieved [2,23–25]. In addition, samples with large volumes are needed when using beams of low thermal neutron fluxes [2,23–25].

Generally, the main drawbacks of the PGNAA method are the high acquisition time of the emitted γ-rays; the large volume of

http://dx.doi.org/10.1016/j.nimb.2014.07.017
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the samples analyzed, especially when using low neutron fluxes; the high background, especially in the low-energy range of the recorded γ-ray spectra; corrections for both neutron self-shielding and γ-ray attenuation; neutron moderation when using samples with high neutron scattering cross sections such as water; and γ-ray interference. In addition to these limitations, the boron peak (478 keV photopeak) has a broad line shape width (as a result of Doppler broadening) that is larger than that of normal γ-peaks [21,26]. The 472 keV line (resulting from the neutron capture reaction with Na) and the annihilation peak (511 keV) interfere with the boron peak.

Thus, an alternative method and/or experimental setup are needed for boron detection in water. This method should be quicker, simpler and cheaper than the PGNAA method. Moreover, the problems mentioned above should be avoided in this method.

Methods based on neutron transmission and the scattering of neutrons are well known and used in many applications [27,28]. For boron detection in water samples using neutron transmission, a neutron beam incident on the sample is mainly attenuated by scattering (a removal process). When an element with a large cross section (such as boron) exists in the water sample, the attenuation increases. Thus, the count rate of the transmitted neutrons can be used to monitor the boron content in the water sample. However, the mechanism is different when using neutron scattering. As the boron content in the water sample increases, the number of scattered neutrons decreases. This is due to some of these scattered neutrons being absorbed by the boron. Therefore, neutron scattering can be used for boron detection in water.

To the best of the author's knowledge, studies related to the determination of boron in water using neutron transmission and scattering are rare. However, there are some studies related to boron detection in other sample materials [3,29,30].

The aim of this study is to use neutron transmission and scattering as an alternative method for boron detection in water. In addition, the PGNAA method is used. The advantages and additional capabilities of the neutron transmission and scattering method compared to the PGNAA are presented.

2. Experimental details and measurements

The experimental setup used in this study is schematically illustrated in Fig. 1a. It consisted of a cylindrical polyethylene container (length = 70 cm and diameter = 50 cm). A tube (length = 70 cm and diameter = 5 cm) was inserted along the central axis of the container. The container was filled with borated paraffin wax for the purposes of neutron moderation and shielding. Two layers (length = 10 cm) made of paraffin wax mixed with boron carbide were used to block the two faces (exit faces of the neutrons) of the cylindrical container. The container had two vertical tubes; a 1 Ci Pu-Be neutron source (yielding 1.5×10^6 neutrons/s) was inserted through one of the tubes into the central tube. After inserting the source, these tubes were blocked by borated paraffin blocks. Blocks of borated paraffin ($10 \times 20 \times 30$ cm) surrounding the cylindrical container were used to further shield against fast neutron leakage. Lead bricks ($5 \times 10 \times 20$ cm) surrounding the borated paraffin blocks were used for gamma-ray shielding for all sides, except for the exit faces of the neutrons. To further reduce the number of gamma rays emitted at the exit faces, two lead rings (thickness = 7 cm, outer diameter = 30 cm, and inner diameter = 7 cm) were used.

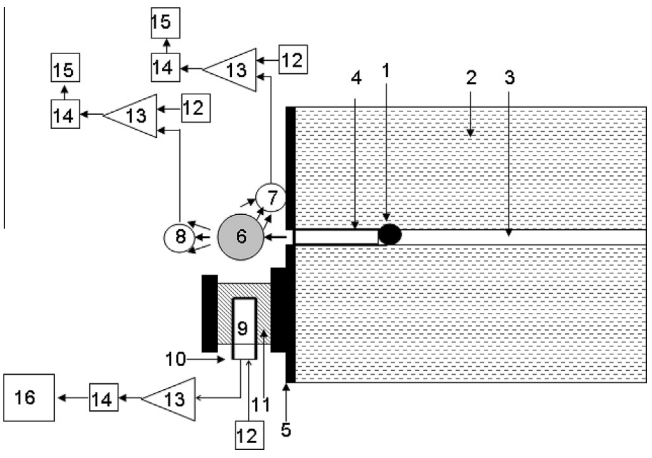

Fig. 1. Schematic diagram of the experimental set-up (not to scale): 1: neutron source, 2: borated paraffin, 3: central tube, 4: collimator, 5: Pb shield, 6: sample, 7,8: He-3 detectors, 9: NaI(Tl) detector, 10: Cd, 11: paraffin, 12 power supplies, 13: preamplifiers, 14: amplifiers, 15: counters, 16 MCA installed on PC.

The source container, borated paraffin blocks and lead shields were housed in a wooden box ($100 \times 100 \times 100$ cm). The box included two holes facing the exit faces of the neutrons. Thus, two beam exits from this setup could be simultaneously used as exits for the neutron beams for performing different experiments. In the current work, one exit face of the setup was used while the other was blocked by the borated paraffin tube. In such a setup, the neutrons travel from the source to the sample through collimators, and the collimators of the neutron beam ensure the appropriate geometry of the measurements. In the present work, a collimator made of two cylindrical polyethylene tubes (length = 19 cm, large diameter of the outer tube = 5 cm, and the diameter of the small tube = 2.5 cm) was used. The space between the two tubes was filled with wax mixed with lead powder. To prevent thermal neutron leakage, the exit face of the collimator providing the neutron beam to the samples investigated was covered with Cd, except at its opening. This collimator was inserted through the source container and along its central axis. Cadmium (Cd) sheets were used to cover the outer side of the wooden box to minimize the number of leaked thermal neutrons.

Two ^{3}He detectors (LND-252172) were used for counting backscattered and transmitted neutrons from the investigated samples. The ^{3}He detector, which registers backscattered neutrons, was placed adjacent to the neutron beam exit. The detector, which registers transmitted neutrons, was oriented along the same direction as the beam. The detector registers transmitted and forward-scattered neutrons. Power supplies (Canberra model 31060) providing high voltage to these detectors via preamplifiers (Ortec-142PC) were used. The output signals from the preamplifiers were fed to amplifiers (Canberra Amp/TSCA 2015A) and subsequently to counters (Ortec 772) for neutron counting.

An NaI(Tl) detector ($3'' \times 3''$) was used to count the prompt gamma rays. Power supplies (Canberra model 31060) providing high voltages to the detector were used. The output signal was fed to a preamplifier and then to an amplifier (Canberra Amp/TSCA 2015A). The output signal from the amplifier was fed to an MCA cart with 8000 channels (NDS). The MCA cart was installed on a PC and was controlled by the Genie 2000 software package. The MCA cart was used for acquiring and analyzing gamma-ray spectra. The NaI(Tl) crystal was shielded against scattered and leaked neutrons. It was wrapped with Cd and inserted into a cylindrical polyethylene tube filled with paraffin wax (outer diameter = 14 cm, inner diameter = 9 cm and length = 15 cm). Moreover, to reduce the background from the surroundings and the experiment, lead bricks were used to shield the NaI(Tl) detector. For further shielding against the fast neutrons of the incident beam, the NaI(Tl) detector was oriented perpendicular to the direction of the incident neutron beam.

The samples investigated in the present study were distilled water with different concentrations of boron. Boric acid, as a source of boron, was mixed with water to form solutions of different