Nuclear Engineering and Technology 54 (2022) 4231–4235

Contents lists available at ScienceDirect

Nuclear Engineering and Technology

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

Original Article

Measurement of deuterium concentration in heavy water utilizing prompt gamma neutron activation analysis (PGNAA) in comparison with MCNPX simulation results

Saeed Salahi [a], Mahdieh Mokhtari Dorostkar [b], Akbar Abdi Saray [b, *]

[a] Reactor and Nuclear Safety Research School, Nuclear Science and Technology Research Institute, AEOI, Iran
[b] Department of Physics, Urmia University, Urmia, Iran

ARTICLE INFO

Article history:
Received 24 April 2022
Received in revised form
15 June 2022
Accepted 21 June 2022
Available online 5 July 2022

Keywords:
Heavy water
PGNAA
MCNPX
Deuterium
Concentration

ABSTRACT

Considering the importance of deuterium in nuclear science including medical and industrial researches such as (BNCT) and nuclear reactors respectively, it is important to study various possible ways in addition to common methods for measuring its concentration. This study is an effort to measure deuterium concentration using PGNAA. The main idea is to calculate the area under 2.23 MeV gamma-rays photo peak resulting from neutron collision with Hydrogen atoms which are in mix with deuterium in samples. The study carried out by both simulation and experiment. Monte Carlo MCNPX2.6 code has been used for simulation and based on its acceptable results an experimental setup has been arranged. The coordination of results was in the range of $R = 0.99$ and $R = 0.98$ in simulation and experiment respectively. The accuracy of the study has been investigated by measuring the concentration of an unknown sample by both PGNAA and Fourier transform infrared spectroscopy (FT-IR) methods in which there were acceptable correlation between these two methods.

1. Introduction

Heavy water (D_2O) contains deuterium which is one of the hydrogen isotopes with one more neutrons which comprises about 1 in 6400 hydrogen atoms available in nature.

In addition to all applications of deuterium such as in neutron and energy production in accelerators and nuclear fusion reactors respectively [1,2] etc, it shows amazing results in boron neutron capture therapy (BNCT) [1]. The macroscopic absorption cross-section for heavy water is smaller than water, whilst the diffusion length is greater for heavy water [9]. Despite all positive benefits of heavy water, it can be toxic in medical applications and should be applied to the body in accurate doses. Heavy-water replacement of light water was practiced in an endeavor to enhance the maximum therapeutic depth for thermal BNCT [4,8]. It is proved that inclusion of heavy water in tissues significantly increases thermal-neutron transmission with reduced capture gamma ray absorbed dose rates.

Heavy water is important in reactors like HWZPR or CANDU and it is essential to determine the concentration of it to achieve better performance [10,11]. For example HWZPR is initiated by increasing the height of heavy water (water level) in the core, it is vital to calculate the water level after any change made to the core configuration [3]. The fuel rods and heavy water are kept under low pressure nitrogen gas to prevent humidity entering the core and to avoid heavy water degradation [6,7].

Calculating heavy water concentration may be a great challenge in this path. There are various ways to measure it but they can be divided into two main groups including: Analysis based on physical properties such use of a pycnometer or the falling drop method for determination of specific gravity, etc. The second group will be instrumental analysis including Infrared spectroscopy and Mass spectroscopy. There are benefits and difficulties in each solution. In first method a small amount of sample and fast timing process with simple and affordable instruments has been utilized but many errors can occur by human, conditions and system and lead to possible high uncertainties. In second method, fast and accurate measurements are available but instruments are expensive and have high sensitivity in different conditions.

This is where PGNAA is considered as a novel method for heavy water concentration measurements. In this paper, we focused on

* Corresponding author.
E-mail addresses: Saeedsalahi065@gmail.com (S. Salahi), m.mokhtaridorostkar@urmia.ac.ir (M. Mokhtari Dorostkar), ak.abdi@urmia.ac.ir (A. Abdi Saray).

https://doi.org/10.1016/j.net.2022.06.021

finding deuterium concentration using PGNAA by utilizing MCNPX2.6 code for simulation and validate the performance by experiment.

2. Material and method

Despite the similarities, there could be great difference in different applications as discussed before. Hence, there are great requirements for accurate calculations of deuterium's concentration in different applications.

NAA or Neutron Activation Analysis is a way to find a highly radioactive element within a sample after exposure to neutron source. There could be different nuclear reactions during neutron activation which is reported in Fig. 1 [4].

When a neutron interacts with the target nucleus via a non-elastic collision, a compound nucleus forms in an excited state. The excitation energy of the compound nucleus is due to the binding energy of the neutron with the nucleus. The compound nucleus will almost instantaneously de-excite into a more stable configuration through emission of one or more characteristic prompt gamma rays. In many cases, this new configuration yields a radioactive nucleus which also de-excites (or decays) by emission of one or more characteristic delayed gamma rays, but at a much slower rate according to the unique half-life of the radioactive nucleus [5]. Prompt gamma neutron activation has instant response and is not delayed by radioactive decay. This method has three main steps. First, the target is irradiated with neutrons. Then, each sample emits prompt gamma rays. Finally, emitted gamma rays are measured with a detector.

PGNAA has some remarkable advantages that can be useful and lead to novel ways of measurements. By utilizing this method low atomic mass elements can be detected. This method depends on cross section, sample mass and incident neutron flux; and as a result, it is independent of the amount and rate of decay. In addition, a gamma ray with higher energy has a lower possibility of absorption. Single excited states can be increased by adjusting beam energies. The sample has low residual activity after irradiation. Large samples can be analyzed by using neutron and photon beams.

This research is an effort to measure heavy water concentration using prompt gamma neutron activation analysis, PGNAA has some advantages comparing to delay neutron activation Analysis or NAA including the fact that it is not possible to place sample in front of a high neutron flux beam in NAA. In addition, NAA is dependent on radioactive decay and as a result it will be time consuming. It can also have some unwanted radioactive products. Finally, some elements with low atomic mass have less sensitivity in this method. In addition, PGNAA can measure elements that do not form radioactive nuclei upon irradiation (e.g. H, B), while NAA with delay gammas cannot.

2.1. Simulation

Fig. 2 illustrates the considered geometry for Monte Carlo simulation. An Am–Be neutron source provided the neutron beam. By focusing on measurement of prompt gamma, an NaI(Tl) detector was simulated with radius of 1 inch and length of 2 inches.

The neutron source is located in the beam tube and the sample is next to beam port. A cylinder of 1 cm radius and 2 cm height is considered as sample. The detector is located in 15 cm from the sample. Gamma-ray count on entrance surface of detector, volumetric flux and dose in detector volume was calculated through this geometry.

2.2. Experiment

After passing through simulation validation, the experimental set up was arranged as shown in Fig. 3.

Energy resolution is not the most important factor in this study; hence, NaI (Tl) detector is a great selection due to its lower price, higher efficiency and construction ability in various shapes and sizes. ^{241}Am-Be or ^{252}Cf are candidates as neutron sources. An Am–Be source with the power of 3 ci was our selection for this study according to its accessibility for us. Before counting, the detector must be calibrated with standard laboratory sources. Nine samples were measured as shown in Fig. 4, with different weight fractions of heavy and normal water as shown in Table 1.

The detection setup is shown in Fig. 5.

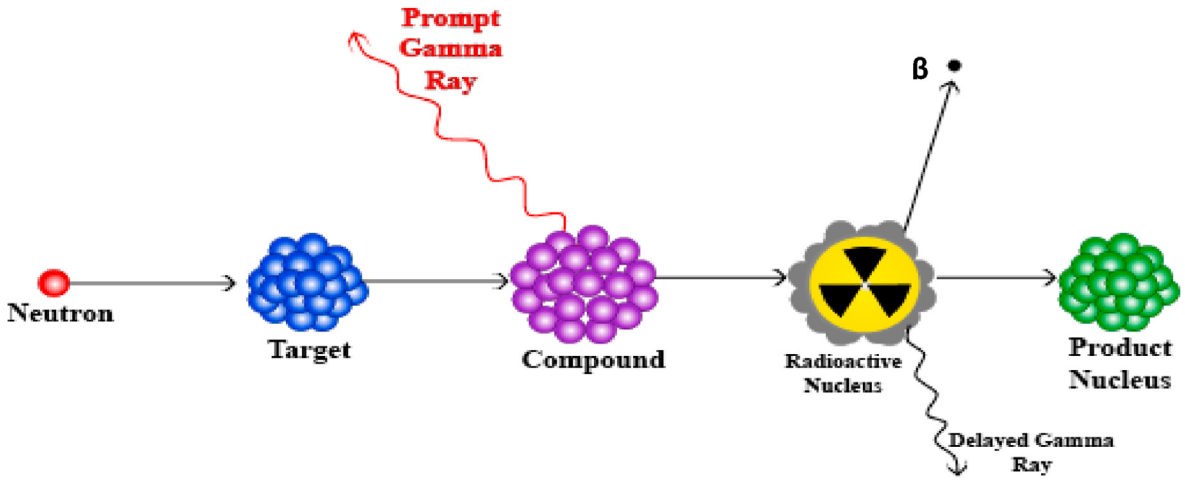

Fig. 1. Neutron activation process.

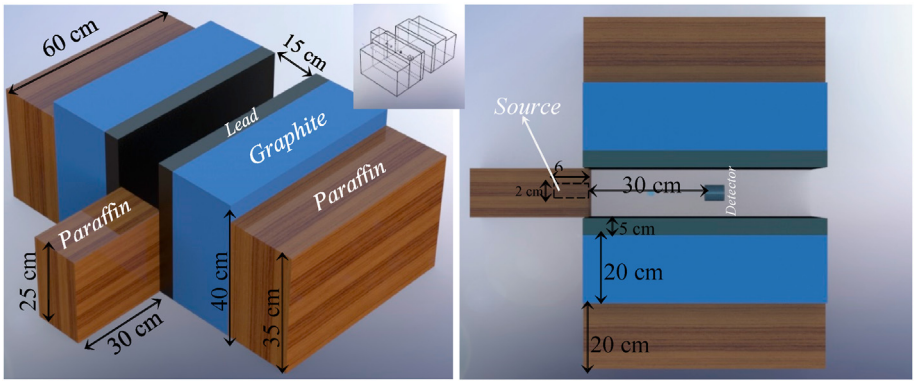

Fig. 2. 3D view of considered geometry.

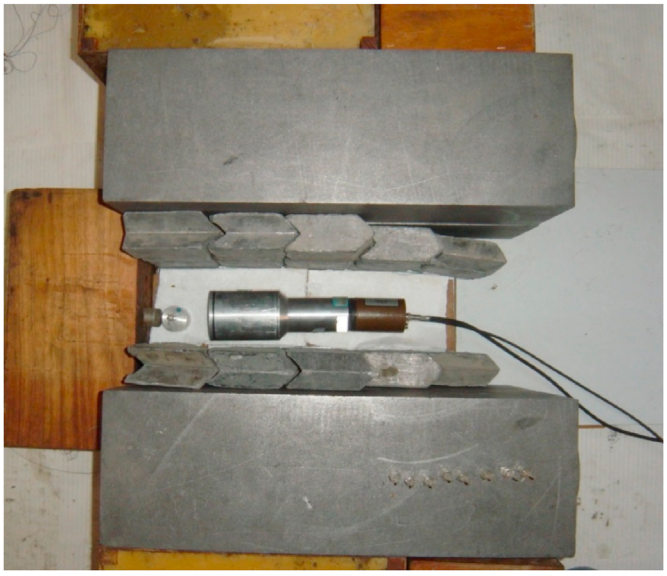

Fig. 3. Considered geometry for experimental calculations.

2.3. Measurement of heavy water concentration

The detector was located in front of the neutron source after calibrating it in terms of energy. The sample was placed in a glass container for analysis to prevent air and humidity penetration. According to equation 1, if we locate hydrogen isotopes in front of the neutron beam, a gamma-ray with energy of 2.23 MeV will be emitted.

$$^{1}_{1}H + n \rightarrow {}^{2}_{1}H + \gamma \quad (1)$$

Therefore, if we have more $^{1}_{1}H$ atoms and less $^{2}_{1}H$ atoms, more gamma rays of this energy will be emitted. By detecting these emitted gamma rays as shown in eq.1 with NaI (Tl) detector, $^{2}_{1}H$ concentration can be measured. The area under 2.23 MeV of gamma-rays photo peak area was calculated with MCA system and Maestro-Pro Advanced Spectroscopy application with an analysis time of 5400 s for each sample.

3. Results

3.1. Simulation results

By considering afore mentioned conditions, gamma-ray activity in detector volume was calculated. Fig. 6 reports gamma counts per second in detector in which the correlation coefficient between the concentration of heavy water and number of gamma-ray per second is equal to $R^2 = 0.99888$. Relative error of simulation results were under 0.1%.

3.2. Experimental results

Fig. 7 reports an example for gamma spectrum for a sample collected using NaI(Tl) detector. Because it wasn't possible to see energy directly with available instruments, the NaI(Tl) detector was calibrated with several check sources including ^{137}Sc, ^{60}Co and ^{22}Na. Hence, appropriate energy was assigned to each channel through proportion. After this collaboration and proportion, the detector was used for the experiment.

And according to Fig. 8, the correlation coefficient between the concentration of heavy water and number of gamma counts per unit time is equal to $R^2 = 0.98691$. In addition, standard error of obtained results is less than 1%.

Due to the results, by knowing parameters "a" and "b" of a linear equation 2,

$$y = (a + b * x) \pm error \quad (2)$$

In where y = counts and x = weight fraction sample. It is possible to determine the concentration of an unknown sample.

3.3. Validation of experiment

A sample with unknown concentration was used for validation. Utilizing the PGNAA method, there were 127249 counts under the 2.23 MeV photo peak area. According to calibration equation, heavy water concentration will be 36.1.

To validate the accuracy of measurements, the concentration was obtained by Fourier transform infrared spectroscopy (FT-IR) either and its result was equal to 36.824.

4. Discussion

In addition to many methods investigated and known as accurate ways to measure the concentration of heavy water, PGNAA can be a novel candidate for this purpose. In this study, Monte Carlo MCNPX2.6 code was used for simulation. Considering acceptable results of simulation due to their great correlation coefficient between the concentration of heavy water and obtained results in count and flux, experimental set up has been arranged as similar as possible to simulation set up.

Utilizing Am–Be neutron source and NaI (Tl) detector, number

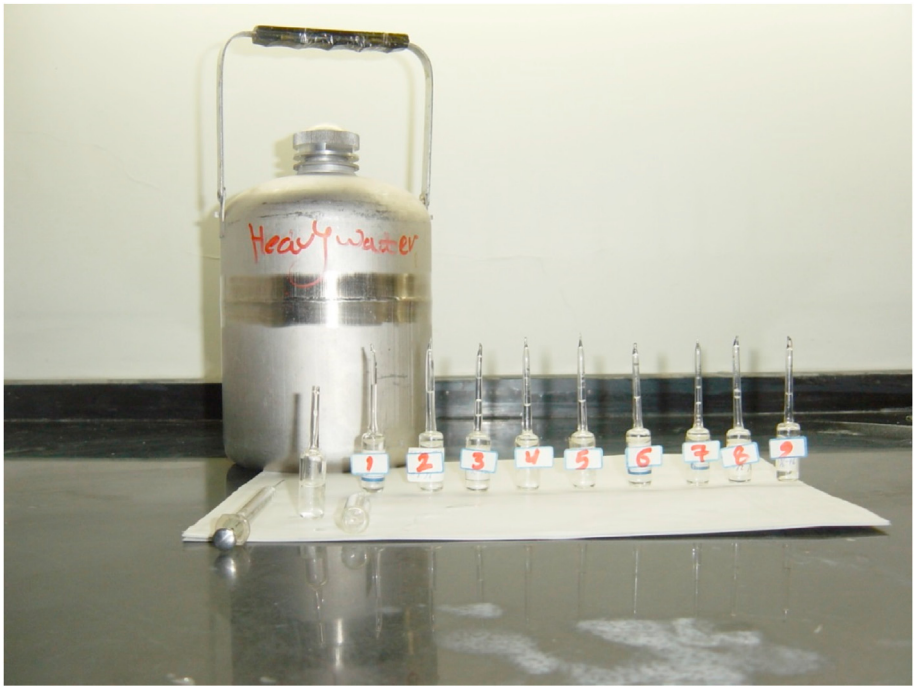

Fig. 4. Samples.

Table 1
considered weight fractions for samples.

No.	Heavy water weight fractions (%)	Normal water weight fractions (%)
1	8.01	91.99
2	24	76
3	29.94	70.06
4	35.98	64.02
5	41.97	58.03
6	58.01	41.99
7	61.99	38.01
8	79.86	20.14
9	97.01	2.99

of counts for gamma-ray have been measured after passing through each sample. The area under the photo peak of 2.23 MeV was calculated in both simulation and experiment. Accurate correlation coefficient was concluded from the experimental results.

Due to the utilized NaI(Tl) detector and comparing this detectors resolution to HPGe, detection of each energy with sharp energy peak was not possible. Hence, gamma counts in other energies like

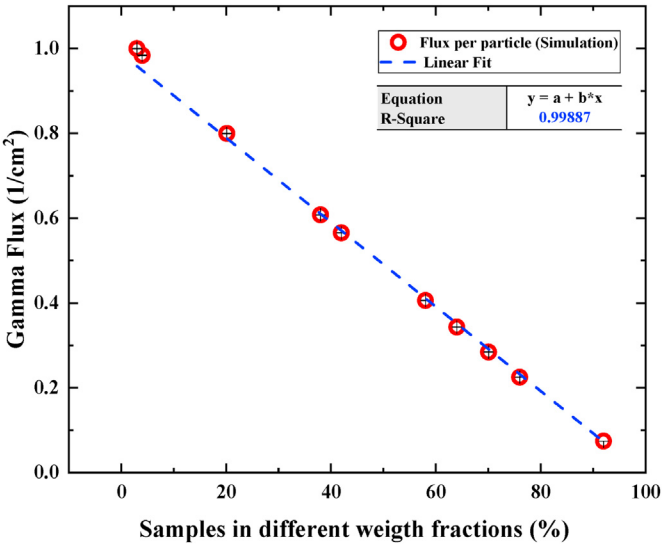

Fig. 6. Gamma-ray per second under 2.23 MeV photo peak for each sample.

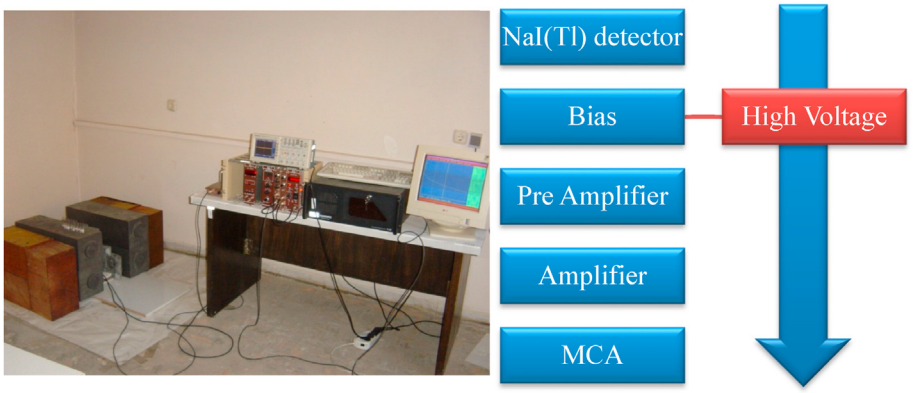

Fig. 5. Detection set up.

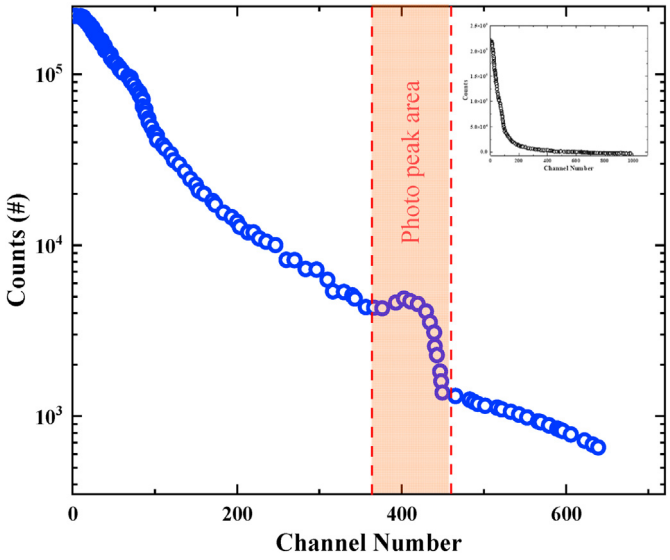

Fig. 7. Gamma spectrum of source and of neutron collisions with heavy water collected by the NaI detector using a multi-channel analyzer (MCA) system.

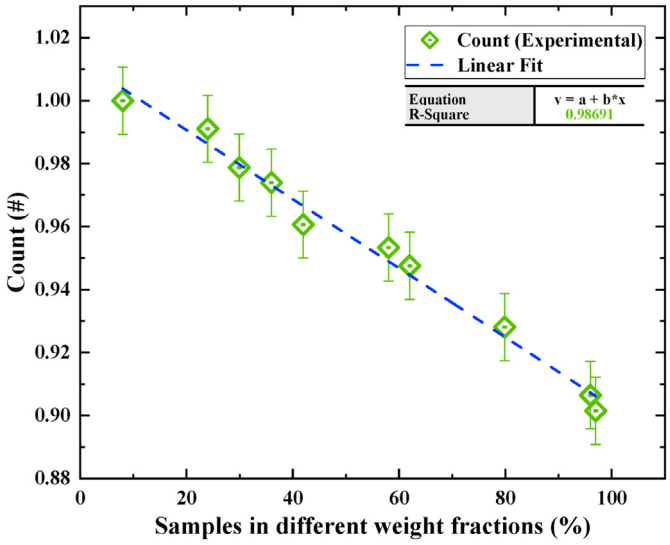

Fig. 8. Heavy water concentration calibrated curve in terms of counting gamma resulting from the reaction.$^1_1\boldsymbol{H}(\boldsymbol{n},\gamma)^2_1\boldsymbol{H}$

6250 keV was not calculated in our measurements. Although the energy peak exists but is not measured or simulated in this study.

The validation of this method was not only investigated by simulation, but also checked by measuring the concentration of heavy water in an unknown sample and the result was compared with FT-IR results of mentioned sample. According to acceptable error between these two methods, it can be concluded that PGNAA is an accurate, simple and affordable method for measurement of heavy water concentration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

[1] D.N. Slatkin, M.M. Levin, A. Aronson, The use of heavy water in boron neutron capture therapy of brain tumours, Physics in Medicine & Biology 28 (12) (1983) 1447.
[2] S.A. Wallace, B.J. Allen, J.N. Mathur, Monte Carlo calculations of epithermal boron neutron capture therapy with heavy water, Physics in Medicine & Biology 40 (10) (1995) 1599.
[3] M.N. Nasrabadi, B. Teimouri, A. Moosakhani, Critical heavy water level and some parameters for the HWZPR reactor core using MCNP4c code, Annals of Nuclear Energy 73 (2014) 496–499.
[4] D.L. Chichester, E. Empey, Measurement of nitrogen in the body using a commercial PGNAA system—phantom experiments, Applied Radiation and Isotopes 60 (1) (2004) 55–61.
[5] M.D. Glascock, An Overview of Neutron Activation Analysis, 2004. World Wide Web. [Online].
[6] S.F. Masoudi, F.S. Rasouli, M. Ghasemi, BNCT of skin tumors using the high-energy DT neutrons, Applied Radiation and Isotopes 122 (2017) 158–163.
[7] K. Bergaoui, N. Reguigui, C. Gary, C. Brown, M. Piestrup, Development of a BNCT Facility Based on Axial Deuterium–Deuterium (D–D) Neutron Generator Using MCNP Code, IAEA, 2017. CN–250.
[8] S.A. Wallace, J.N. Mathur, B.J. Allen, The influence of heavy water on boron requirements for neutron capture therapy, Medical Physics 22 (5) (1995) 585–590.
[9] J.R. Lamarsh, A.J. Baratta, Introduction to nuclear engineering. Upper saddle river, NJ: Prentice hall 3 (783) (2001).
[10] L. Taghizadeh, A. Zolfaghari, M. Zangian, J. Mokhtari, Y. Sardjono, Comparison of probabilistic and deterministic methods for calculation of kinetic parameters of HWZPR, Annals of Nuclear Energy 165 (2022), 108633.
[11] Zahra Nasrazadani, Manochehr Behfarnia, Jamshid Khorsandi, Mohammad Mirvakili, Investigating dynamic parameters in HWZPR based on the experimental and calculated results, Nuclear Engineering and Technology 48 (5) (2016) 1120–1125.