DEVELOPING A RELIABLE APPROACH TO ESTIMATE THE STOICHIOMETRIC RATIO OF O/U IN UO_2 PELLETS USING MCNP-5 AND ARTIFICIAL INTELLIGENCE

by

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Uranium dioxied is used as a nuclear fuel. Depending on the temperature and oxygen partial pressure, it is incredibly versatile and can accept a wide variety of stoichiometry. Many methods are used to estimate the non-stoichiometric O/U ratio such as the coulometric titration, gravimetric and voltammetric methods. These methods have some disadvantages and may be time and cost-consuming. This work develops an approach to determine the stoichiometric ratio by using MCNP-5 code and hyper pure germanium detector to estimate the count rate at 185.7 keV for UO_2 pellets. The studied pellets are proposed to have ^{235}U mass content (3 %, 4 %, and 5 %) and 1 cm away from the detector. The mass of the oxide within the pellets is 7.8995 grams. The relation between volume and density has been studied during different steps in which temperature increases. Finally, a reliable model is established to describe the process of converting green pellets to sintered pellets. The model is supported by employing artificial intelligence to predict some features and the overall correlation equals 0.99929.

Key words: stoichiometry, UO_2 pellet, artificial intelligence, MCNP-5

INTRODUCTION

The primary nuclear reactor fuel, uranium, is widely distributed around the globe. It is mined, refined, and then enhanced before being fed into a nuclear reactor to create the fuel [1]. The enriched uranium is shipped to a facility that makes fuel, where it is transformed into powdered UO_2 [2]. The little fuel pellets made from this powder are then heated and pressed into hard ceramic material. The pellets are then placed into tubes called fuel rods, which are eventually gathered together to form fuel assemblies [3]. Depending on the type of reactor, each fuel assembly can contain anywhere from 90 to well over 200 fuel rods. The fuel often remains loaded in the reactor core for several years [4].

Nuclear fuel, which is produced in various forms based on the kind of reactor, is the source of energy in a nuclear reactor. The majority of operational commercial nuclear reactors use UO_2 pellets that are typically 1 cm in diameter and 1 cm in length as fuel, including both pressurized (PWR) and boiling (BWR) water reactors [5].

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Figure 1. Pellets of UO_2

This type of uranium fuel is manufactured by first processing mined uranium through conversions and enrichment operations and then transforming it into solid, compacted pellets as the final product. The UO_2 fuel is typically 35 % more abundant in the fissile isotope ^{235}U than in the fertile isotope ^{238}U (the ^{235}U proportion in natural uranium is roughly 0.7 %) [6].

The UO_2 pellets are made by sintering annealing powders at high temperatures (1650 °C to 1750 °C) for two or more hours in a hydrogen environment. An alternate process is sintering by oxidation, which is carried out in a CO_2 or CO/CO_2 atmosphere at lower temperatures (1000 °C to 1300 °C) [7].

One of the most important parameters to characterize the uranium oxide nuclear fuel: UO_{2+X} (where x is the stoichiometric deviation), is the relation between oxygen and metal, O/Me. During the production of the UO_2, used in nuclear reactors such as PWR, the relation O/Me should be 2.07 to 2.18, for the UO_{2+X} powder and 2.00 for the sintered pellets [8].

Many uranium compounds are used in the fuel cycle across a broad range of isotopic compositions so the isotopic ratio of a sample is often the target for assaying. This assaying is most often employed in samples containing uranium to get the ratio of fissile ^{235}U. The uranium isotopes emit neutrons, alpha and beta particles, and gamma-rays [9].

Gamma-rays, which are often dominated by emissions from ^{235}U decay, are the main radiation employed in passive non-destructive assay (NDA) of uranium samples. The X-ray, on the other hand, is the most potent part of the emission spectrum in low-content uranium samples. The most frequently employed signature to assess ^{235}U content is the 185.7 keV gamma-ray. It is the most noticeable single gamma-ray from any uranium sample that has been enhanced above the level of naturally occurring ^{235}U [10]. While conducting nuclear measurements on uranium and its compounds, some codes are used to conduct simulations, an example of these codes is the Monte Carlo code.

The Monte Carlo code is software that performs various nuclear simulations [11]. This code is extremely important in nuclear measurements as it gives us accurate results for nuclear tests, provided that the measured sample, the device used in the measurement process, and the conditions of the experiment are well described [12].

In this study, a proposed approach has been developed to estimate the stoichiometric ratio of O/U using Monte Carlo simulation and artificial neural network (ANN) modeling.

METHODOLOGY

The count rate was obtained according to eq. (1). The equation shows the formula used to get the count rate of a mass of ^{235}U in the UO_2 pellets at energy line 185.7 keV. It was altered to include the calculated absolute full-energy peak efficiency utilizing MCNP-5 modeling at the same energy as well as the measured gamma energy line's particular activity [13]

$$C_r \quad M_{235} \quad A_{\text{eff}} \quad S_a \tag{1}$$

where C_r is the count rate (counts per second – cps), M [g] – the ^{235}U mass, S_a [g^{-1}s^{-1}] – the specific activity at a single gamma line, and A_{eff} – the detector's absolute full-energy peak efficiency at 185.7 keV.

The results obtained from the Monte Carlo code and the previous equation are used to be the backbone on which the neural network will be built. The neural network is built according to a set of steps represented in fig. 2. The steps involve constructing an approximate solution, setting up the data, deciding on a network design, neural network training, boosting generalization abilities, testing outcomes, and finally deploying the model.

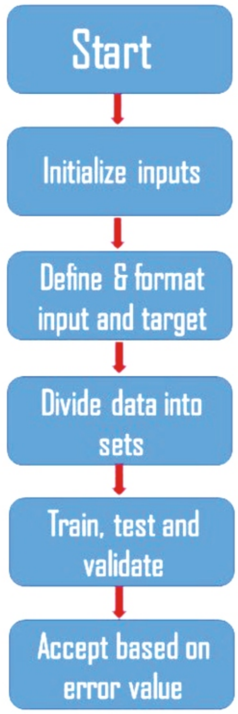

Figure 2. Basic steps for building ANN

EXPERIMENTAL SETUP AND TECHNIQUES

The proposed green and sintered pellets of UO_2 were modeled as in fig. 3 depending on certain specifications such as the height and radius of each pellet. The volume of each pellet decreases while the density increases. For this simulation, the used detector is a high-purity germanium detector (HPGe), GL0515R model Canberra which has the following specifications (active volume height and area are 1.5 cm and 540 mm^2, respectively, and FWHM 540 eV at 122 keV. The operating voltage of the multi-channel analyzer is (–2500 V) [14].

The Monte Carlo code was used to simulate the process of measuring the pellet at different temperatures by expressing the decrease in volume, which corresponds to an increase in density at different stages in

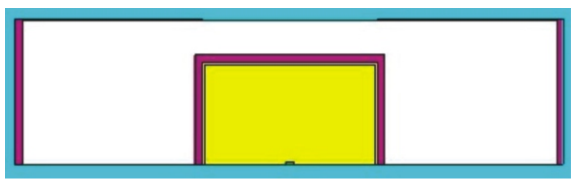

Figure 3. The HPGe detector appearance in MCNP-5

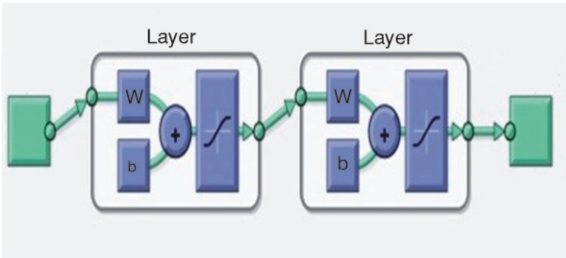

Figure 4. The structure of ANN

which the pellet moves from being green to being sintered. The simulation was done using a computer with a processor i5 core and the number of histories was 108. Figure 3 shows the detector view as it appeared in MCNP visual editor.

The ANN was built using the data obtained for volume, density, absolute efficiency, and account rate [15, 17]. The network in fig. 4 consists of two layers and the layer consists of 10 neurons. The number of epochs was selected to be high as possible to avoid any increase in error.

The training function is TRAINLM, the adaptation learning function is LEARNGDM, the performance function is MSE, and the transfer function is TANSIG for the feed-forward backprop network type.

RESULTS AND DISCUSSIONS

The results of volume and density for the UO_2 pellets

The density of the pellet changes with the change in volume and mass. Concerning volume, the factor affecting the change in volume is the height and radius of the pellet. The mass varies according to the mass of the ^{235}U isotope present in the sample being characterized. As shown in fig. 5, the greater the mass of the pellet the greater the density.

In fig. 5, as the temperature increases, the density of the pellet increases, and this is in line with what happens in practice, as the sintered pellet is denser than the green one. The starting point of the density has the value of 5.493 gcm^{-3} and this value increases till it becomes 10.063 gcm^{-3} in the final step.

It is clear from fig. 6 that as the temperature increases, the volume of the pellet decreases, and this

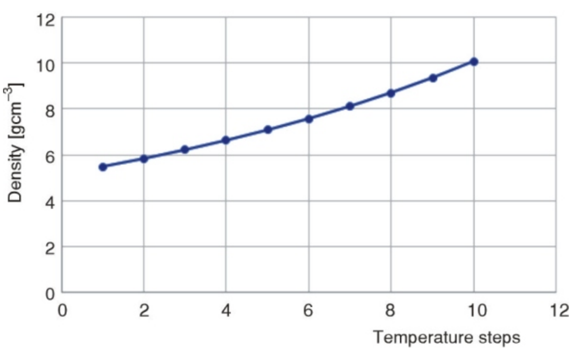

Figure 5. Density variation with temperature

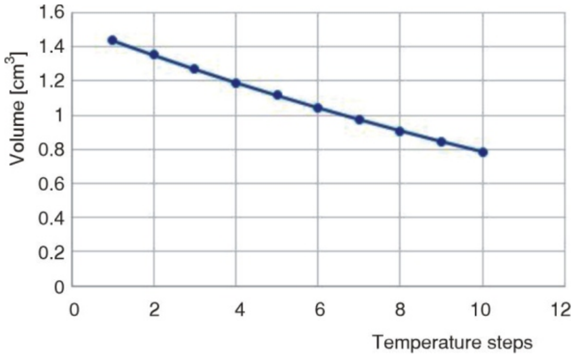

Figure 6. Volume variation with temperature

corresponds to the practical reality, as the sintered pellet is denser than the green one and so its volume decreases. The starting point of the volume has a value of 1.438 cm^3 and this value decreases till it becomes 0.785 cm^3 in the final step.

The results of the count rate at a certain mass ratio of ^{235}U

Figure 7 shows the effect of increasing temperature on the value of the count rate. It is clear as the temperature becomes greater the count rate decreases. The value of the count rate at the first step is 3.886 cps and the value of the final step is 2.230 cps. The values of the corresponding absolute efficiency and its related error are demonstrated in tab. 1 for the 3 % UO_2 sample.

Figure 8 shows the same trend between the count rate and temperature as the previous figure but the

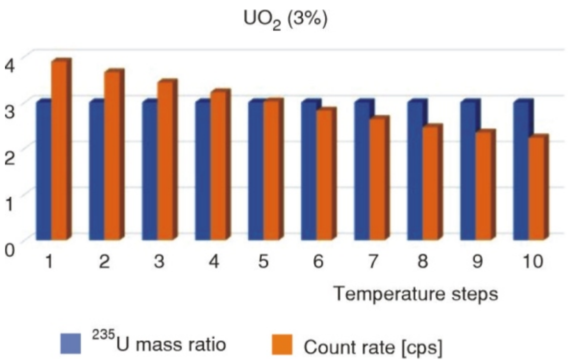

Figure 7. Count rate variation at 3 % UO_2

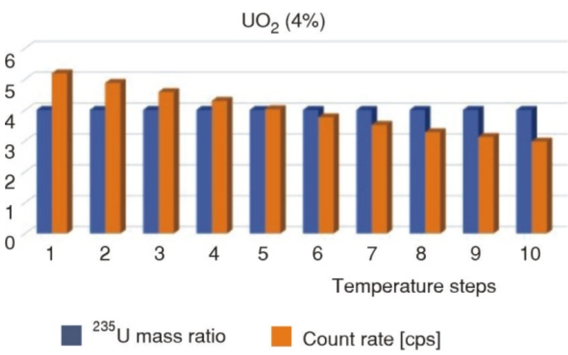

Figure 8. Count rate variation at 4% UO_2

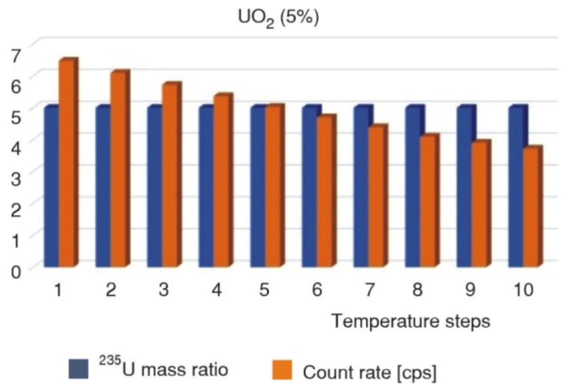

Figure 9. Count rate variation at 5 % UO_2

Table 1. The absolute efficiency values at different steps for the 3 % UO_2 sample

Step/ temperature	Absolute efficiency	Measurement uncertainty
1	$4.0352\ 10^{-4}$	0.0004
2	$3.7953\ 10^{-4}$	0.0004
3	$3.5645\ 10^{-4}$	0.0004
4	$3.3422\ 10^{-4}$	0.0004
5	$3.1311\ 10^{-4}$	0.0004
6	$2.9289\ 10^{-4}$	0.0005
7	$2.7355\ 10^{-4}$	0.0005
8	$2.5513\ 10^{-4}$	0.0005
9	$2.4320\ 10^{-4}$	0.0005
10	$2.3161\ 10^{-4}$	0.0005

Table 2. The absolute efficiency values at different steps for the 4 % UO_2 sample

Step/ temperature	Absolute efficiency	Measurement uncertainty
1	$4.0347\ 10^{-4}$	0.0004
2	$3.7948\ 10^{-4}$	0.0004
3	$3.5640\ 10^{-4}$	0.0004
4	$3.3418\ 10^{-4}$	0.0004
5	$3.1307\ 10^{-4}$	0.0004
6	$2.9286\ 10^{-4}$	0.0005
7	$2.7351\ 10^{-4}$	0.0005
8	$2.5510\ 10^{-4}$	0.0005
9	$2.4317\ 10^{-4}$	0.0005
10	$2.3158\ 10^{-4}$	0.0005

value of the count rate at the first step is 5.180 cps and the value of the final step is 2.973 cps. The values of the corresponding absolute efficiency and its related uncertainty are demonstrated in tab. 2 for the 4 % UO_2 sample.

In fig. 9, the same trend between count rate and temperature also exists as in the previous figures but the value of the count rate at the first step is 6.474 cps and the value of the final step is 3.716 cps. The values of the corresponding absolute efficiency and its related error are demonstrated in tab. 3 for the 5 % UO_2 sample.

It is well known there is no relationship between count rate and temperature, and the Monte Carlo program cannot perform a simulation process in which temperature or pressure is included as one of the conditions surrounding the experiment being simulated, but the proposed approach succeeded to a large extent in crystallizing all the different factors and conditions surrounding the special sintering process of the pellet to reach a clear concept through which the stoichiometric ratio can be deduced using different and innovative tools such as Monte Carlo methods and artificial intelligence. Part of the inputs was used to do the training process for the neural network. The network was also tested after low error rates were reached, which can judge the network as valid.

Table 3. The absolute efficiency values at different steps for the 5 % UO_2 sample

Step/ temperature	Absolute efficiency	Measurement uncertainty
1	$4.0342\ 10^{-4}$	0.0004
2	$3.7943\ 10^{-4}$	0.0004
3	$3.5635\ 10^{-4}$	0.0004
4	$3.3413\ 10^{-4}$	0.0004
5	$3.1304\ 10^{-4}$	0.0004
6	$2.9282\ 10^{-4}$	0.0005
7	$2.7347\ 10^{-4}$	0.0005
8	$2.5506\ 10^{-4}$	0.0005
9	$2.4314\ 10^{-4}$	0.0005
10	$2.3155\ 10^{-4}$	0.0005

Figure 10 shows the four graphs for training, testing, validation, and the overall coefficient. The results indicate that the model is valid for predicting values with good accuracy and it is recommended for dealing with such issues.

The four graphs of fig. 10 represent the regression plots using the ANN. Each graph is a relation between target and output data, however, the data values entered into the network differ according to the state so each state has its unique equation. Each plot expresses the performance of the studied state of the network. The higher the value of the coefficient the more the efficiency of the state being studied. The coefficients for the training, testing and validation states have the same value, and the overall coefficient is 0.99929. This value is good to accept the output from the network when using the samo conditions.

CONCLUSION

The process of converting green pellets of UO_2 to sintered pellets is vital in the life cycle of nuclear fuel. The quantitative measurement of the stochiometric ratio of metal to oxygen is of great importance and can be done through various methods. Neural networks are an effective way to deal with data that does not have a linear relationship, as they can predict results with high accuracy, especially if the neural network is well-built. The proposed method can predict the ratio depending

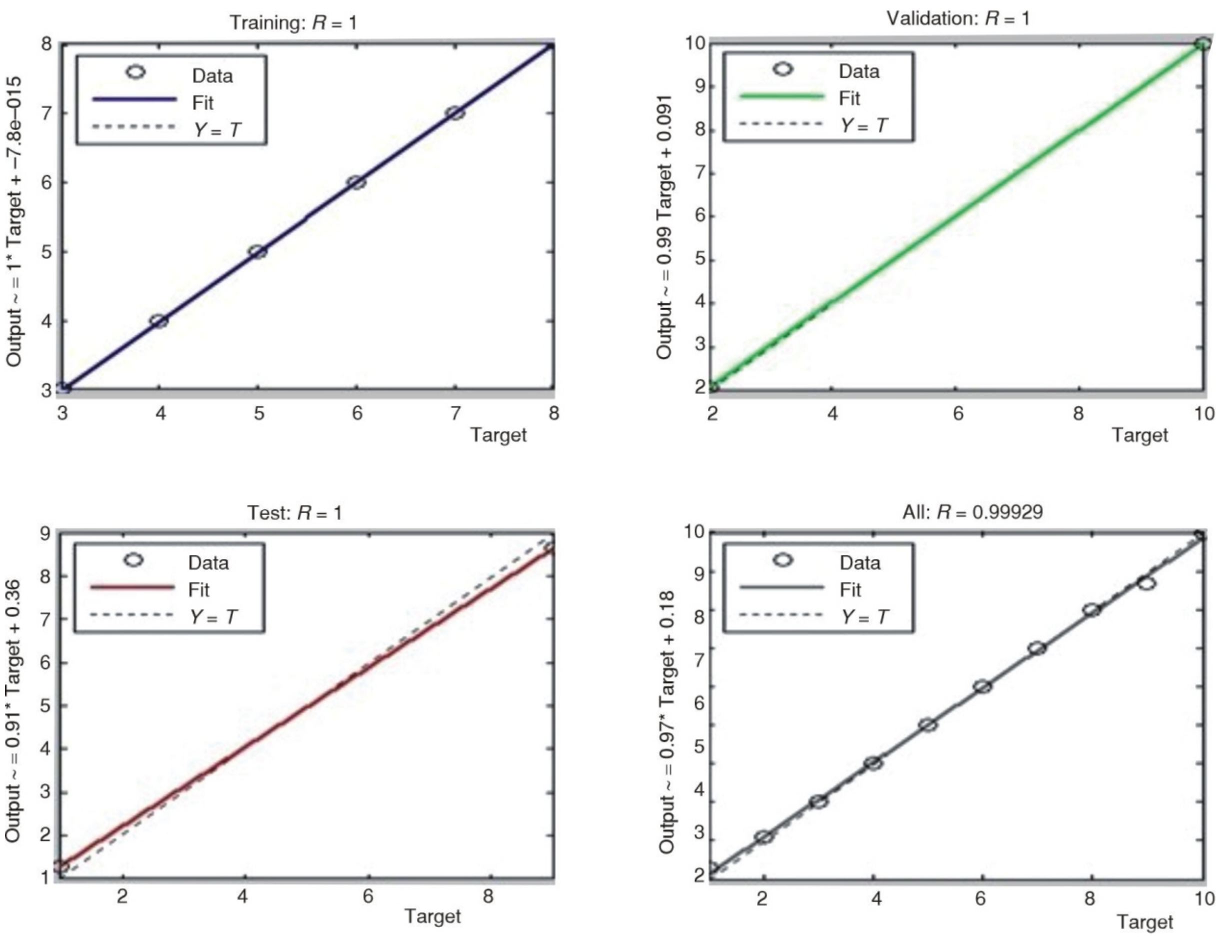

Figure 10. The ANN model of UO_2

on the data that comes from the Monte Carlo simulation. The developed model takes the variation in volume, density, and temperature into consideration. It proved to be valid for predicting the stochiometric ratio between uranium and oxygen. It gives accurate and precise results.

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AUTHORS' CONTRIBUTIONS

S. E. Shaban is the first and the corresponding author of the paper, and he is the main implementer and one of the thought contributors to the idea. A. R. Agha is one of the thought contributors and participates in accomplishing the research. The third author K. A. Aladham is the supporter and scientific advisor of the work.

REFERENCES

[1] Degueldre, C. A., *et al*., Nuclear Fuel Cycle, with a Liquid Ore and Fuel: Toward Renewable Energy, *Sustainable Energy & Fuels, 3* (2019), 7, pp. 1693-1700

[2] Larson, L. N., The Front end of the Nuclear Fuel Cycle: Current Issues, Congressional Research Service, USE, June 6, 2019

[3] Hung, N. T., *et al*., Modeling the UO_2 ex-AUC Pellet Process and Predicting the Fuel Rod Temperature Distribution Under Steady-State Operating Condition, *Journal of Nuclear Materials, 504* (2018), June, pp. 191-197

[4] Bodansky, D., *Nuclear Energy: Principles, Practices, and Prospects*, Springer Science & Business Media, New York, USA, 2007

[5] Kulyako, Y. M., *et al*., Preparation of Powdered Uranium Oxides by Microwave Heating of Substandard Ceramic Pellets of Oxide Nuclear Fuel, *Radiochemistry, 57* (2015), March, pp. 127-130

[6] Ge, L., *et al*., Influence of Processing Parameters on Thermal Conductivity of Uranium Dioxide Pellets Prepared by Spark Plasma Sintering, *Journal of the European Ceramic Society, 34* (2014), 7, pp. 1791-1801

[7] Yang, J. H., *et al*., Microwave Process for Sintering of Uranium Dioxide, *Journal of Nuclear Materials, 325* (2004), 2-3, pp. 210-216

[8] Moura, S. C., *et al*., Characterization of the Stoichiometric Ratio O/U in UO_2 Samples by Gravimetric and Voltametric Methods, 2001

[9] Abdalsamia, M. A., 2009, Thesis Mathematical Calibration of a Gamma-Ray Detector for Nuclear Material Measurement, Canberra, Cairo, 1996

[10] Dewji, S. A., *et al*., Validation of Gamma-Ray Detection Techniques for Safeguards Monitoring at Natural Uranium Conversion Facilities, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, *Detectors and Associated Equipment, 823* (2016), July, pp. 135-148

[11] ***, A guide to Monte Carlo Simulations in Statistical Physics (3rd ed., pp. xv, 471), New York: Cambridge University Press. ISBN: 978-0-521-76848

[12] Hou, D., *et al*., Monte Carlo Simulation of HPGe Gamma Spectrometry Systems, *The Journal of Engineering, 2019* (2019), 23, pp. 9064-9068

[13] Shaban, S. E., *et al*., Countering Distortion Scenarios Outcome from Shaping and the Chemical Mixing in U_3O_8-Al Compact Using Reliable Model, *Journal of Radiation Research and Applied Sciences, 13.1* (2020), pp. 88-93

[14] ***, Canberra, Detector Specification and Performance Data (HPGe Model GL0515R-7905SL-5). USA, Canberra Industries, 1995, Germanium Detectors, User's manual, USA. David, P. L., & Kurt, B. 2009

[15] Shaban, S. E., *et al*., Applying Monte Carlo and Artificial Intelligence Techniques for ^{235}U Mass Prediction in Samples with Different Enrichments, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, *Detectors and Associated Equipment, 916* (2019), Feb., pp. 322-326

[16] Kim, J., *et al*., Quantitative Analysis of NaI (Tl) Gamma-Ray Spectrometry Using an Artificial Neural Network, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, *Detectors and Associated Equipment, 944* (2019), Nov., 162549

[17] Tam, H. D., *et al*., Estimation of Liquid Density Using Artificial Neural Network in Gamma-Ray Scattering Measurement, *Nucl Technol Radiat, 37* (2022), 1, pp. 31-41

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РАЗВОЈ ПОУЗДАНОГ ПРИСТУПА ЗА ПРОЦЕНУ СТЕХИОМЕТРИЈСКОГ ОДНОСА O/U У ПЕЛЕТИМА UO_2, КОРИШЋЕЊЕМ ПРОГРАМА MCNP-5 И ВЕШТАЧКЕ ИНТЕЛИГЕНЦИЈЕ

Уранијум диоксид, који се користи као нуклеарно гориво, у зависности од температуре и парцијалног притиска кисеоника изузетно је свестран и може да прихвати широк спектар стехиометрије. Многе методе користе се за процену нестехиометријског односа O/U, као што су кулометријска титрација, гравиметријска и волтаметријска метода. Ове методе имају извесне недостатке и захтевају време и трошкове. У овом раду развијен је приступ за одређивање стехиометријског односа UO_2 пелета, коришћењем MCNP-5 програма и детектора хипер чистог германијума за процену брзине бројања на 185,7 keV. Предлажено је да проучаване пелете имају масени садржај ^{235}U од 3 %, 4 % и 5 %, и да су удаљене 1 cm од детектора. Маса оксида унутар пелета је 7.8995 грама. Однос између запремине и густине проучаван је током различитих корака у којима температура расте. Коначно, успостављен је поуздан модел за описивање процеса претварања зелених пелета у синтероване пелете. Модел је подржан коришћењем вештачке интелигенције за предвиђање неких карактеристика и укупна корелација је једнака 0,99929.

Кључне речи: стехиометрија, UO_2 пелет,вештачка интелигенција, MCNP-5 програм