Accounting for Uranium-234 During the Analysis of Experiments at the Research Fast Reactor with High Enrichment

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Abstract. Paper is an attempt to consider the influence of U-234 presence through the comparison of the calculated assessments of the temperature reactivity effect (TRE) and the sodium void reactivity effect (SVRE) of the fast research reactor CEFR with 64.4% enrichment on U-235 isotope and measured values of the temperature reactivity effect and sodium void reactivity effect. Additional investigation was performed to account for U-234 in the first criticality experiment. Experimental data are provided in frames of IAEA Coordinated Research Project “Neutronics Benchmark of CEFR Start-Up Tests”. The content of U-234 was calculated by us using mathematical model of the quasi-ideal enrichment cascade. Due to the high enrichment, the content of U-234, although still small, has ceased to be vanishingly small. The effect caused by this isotope is appeared not only through the presence of new isotope in the fuel load, but also in a decrease in the mass of the U-238 isotope in the fuel load. It is shown, that accounting for U-234 slightly increases the agreement of the calculated assessments of TRE among themselves (values of deviation shifted toward each other)/ Also accounting for the U-234 isotope leads to a small but systematic increase in the values of the effective multiplication factor by 120-150 pcm.

1. Introduction
During the calculations of fast neutron reactors with uranium fuel, with rare exceptions, it is not accepted to take into account the presence of the U-234 isotope in the fuel. Its presence in natural uranium is not great, approximately 0.0055 %, and despite a significant increase in its share during enrichment, due to the small starting value, it remains insignificant even after enrichment. Therefore, its influence on the neutron-physical characteristics of the reactor is negligible, the only characteristic that it has a noticeable effect on is the concentration of U-232 in the spent fuel [1-3], which is really the most important characteristic, because it has a strong influence on the radiation and economic characteristics of fresh fuel, which is made from regenerated uranium extracted from spent fuel [2,4]. However, in the case of high fuel enrichment, it is possible to try to detect its influence on other reactor characteristics, such as the effective multiplication factor, sodium void reactivity effect (SVRE) and temperature reactivity effect (TRE).

We have attempted to consider the influence of U-234 to computational analysis of the experimental measurements of the temperature coefficient of reactivity and sodium void reactivity effects of the research fast reactor CEFR [5 - 7] with the fuel enrichment 64.4% on U-235. In addition,
calculations have been made of the experiments on the release of the first critical state. The reactor core layout is presented on figure 1.

Figure 1. CEFR reactor core layout [5].

2. Computational Analysis
We have attempted to consider the impact of U-234 to computational analysis of the experimental measurements of the temperature coefficient of reactivity and sodium void reactivity effects of the research fast reactor CEFR [5 - 7] with the fuel enrichment 64.4% on U-235. In addition, calculations have been made of the experiments on the release of the first critical state. The reactor core layout is shown on figure 1.

In the description of the reactor loading, the U-234 isotope is missing, so its content was calculated by us using mathematical model of the quasi-ideal enrichment cascade [8]. Due to the high enrichment, the content of U-234, although still small, has ceased to be vanishingly small. The effect caused by this isotope is appeared not only through the presence of new isotope in the fuel load, but also in a decrease in the mass of the U-238 isotope in the fuel load.

In our calculations SERPENT (Monte-Carlo code with neutron cross sections from Jeff-3.3 library) and design diffusion code JARFR with ABBN-93 library were used [9,10].

A computational analysis of three series of experiments was performed: experiments on obtaining the first critical state of the reactor, on measuring the temperature effect of reactivity, and on measuring the sodium void effect of reactivity. Measurements of the temperature effect of reactivity, in turn, included two sequences of measurements, one series of measurements was performed when the temperature of the coolant increased, the second - when it decreased.
2.1. Results of calculations of the first criticality experiment

Results of calculations of the first criticality experiment are given in Table 1.

| Configuration | Number of FA | Loading depth of the RE rod, mm | Serpent Without U-234 | Serpent With U-234 | JARFR Without U-234 | JARFR With U-234 |
|---------------|--------------|---------------------------------|------------------------|---------------------|---------------------|------------------|
| 1             | 70           | 500                             | 0.99445                | 0.99585             | 0.98404             | 0.98520          |
| 2             | 71           | 500                             | 0.99857                | 0.99991             | 0.98842             | 0.98958          |
| 3             | 72           | 190                             | 1.00217                | 1.00334             | 0.99299             | 0.99415          |
| 4             | 72           | 170                             | 1.00203                | 1.00330             | 0.99288             | 0.99404          |
| 5             | 72           | 151                             | 1.00206                | 1.00327             | 0.99292             | 0.99407          |
| 6             | 72           | 70                              | 1.00164                | 1.00304             | 0.99276             | 0.99392          |

Results of calculation shows, that accounting for the U-234 isotope leads to a small but systematic increase in the values of the effective multiplication factor by 120-150 pcm.

2.1.1. Temperature Reactivity Effect Calculations. Each measurement of the temperature reactivity effect (TRE) was carried out as follows: temperature change at the reactor in a critical state, reactivity compensation by means of moving the control rods (CR), which was accompanied by a measurement using a reactivity meter of the part of reactivity (in fact, determining the uncertainty of fixing a new critical state) that could not be compensated by the CR, a new temperature change, etc. The total temperature reactivity effect was defined as the sum of the partial temperature reactivity effects, and the partial temperature reactivity effect, in turn, was determined using the control rods calibration curves with correction for the reactivity meter readings.

During the experiment, the calibration curves of the single CR were measured in a critical state, for which all other CRs, with the exception of emergency protection rods, were partially inserted into the core. The height position of these CRs is not given in the description of the experiment. Therefore, the calibration curves obtained by calculation may not coincide with the experimental ones, which will introduce an additional error in the analysis of measurement results.

However, we can eliminate the changes of rod worth in perturbed core and all following result deviations by performing an additional calculation per experimental step. After modeling of initial configuration, first we make a change of temperatures with the same positions of control rods. A difference between these 2 results are actual temperature reactivity effect, unaffected by other changes. Then a second step with a new CR positions are calculated, and its reactivity can be used to find control rod worth difference in perturbed core and are initial configuration for next temperature change. This approach allows to separate two physical effects and it was used in refined calculations phase. At the same time, “two step calculations” maintain all the experimental conditions and can be directly compared with it, while some intermediate steps can be used for cross verification between participants.

During the calculation analysis of the experiments three calculations were performed for each measurement:
1. Calculation of the effective multiplication factor in the initial state.
2. Calculation of the effective multiplication factor in the heated or cooled state, depending on which series of experiments was considered. The position of the control rods in this case corresponded to the previous state.
3. Calculation of the effective multiplication factor in the heated state with the new position of the control rods, which corresponded to the new initial state for the next temperature change.
The results of the first two calculations were used to calculate the partial temperature effect of reactivity a given. Emergency protection CR were removed from the core during calculations as well as during measurements.

In Table 2 the calculated with diffusion JARFR code TRE relative deviations from experimental data.

**Table 2.** Relative deviations of calculated TRE value with diffusion JARFR code from experimental data, %.

| T, °K | Without U-234 | With U-234 |
|------|---------------|------------|
|      | Increasing temperature from 250 °K |          |
| 275  | 17.52         | 16.76      |
| 283  | 20.55         | 19.86      |
| 293  | 19.94         | 19.30      |
| 302  | 9.50          | 8.79       |
|      | Decreasing temperature from 300 °K |          |
| 290  | -5.96         | -6.62      |
| 281  | 0.00          | -0.50      |
| 270  | -1.62         | -2.16      |
| 250  | -4.12         | -4.67      |

As can be seen from the results of calculations, accounting for U-234 slightly increases the agreement of the calculated assessments of TRE among themselves (values of deviation shifted toward each other).

### 2.1.2. Sodium void reactivity effect calculations.

The measurements of the Sodium Void Reactivity Effect (SVRE) were performed as follows: in a reactor in critical condition, the fuel assembly filled with sodium was replaced with a fuel assembly without sodium, then the reactivity was compensated by the CR moving.

When performing the calculation analysis of experiments on the measurement of SVRE two calculations corresponded to each measurement:

1. Calculation of the effective multiplication factor in the initial state.
2. Calculation of the effective multiplication factor after installing the emptied fuel assembly in the measurement position. The position of the control rods in this case corresponded to the previous state.

Deviations of calculated SVRE value with diffusion JARFR code from experimental data, pcm are shown in table 3.

**Table 3.** Deviations of calculated SVRE value with diffusion JARFR code from experimental data, pcm.

| Position of voided FA [6] | Without U-234, pcm | With U-234, pcm |
|--------------------------|-------------------|---------------|
| 2-4                      | -20.08            | -19.51        |
| 3-7                      | -15.81            | -15.28        |
| 4-9                      | -16.39            | -15.91        |
| 5-11                     | -12.77            | -12.34        |
| 6-13                     | -16.16            | -15.83        |
As follows from table 3, accounting for U-234 leads to a slight but systematic reduction in the error of calculating the SVRE. Since JARFR is a diffusion code that uses a simple homogenization method for preparing constants, and it is known from calculation practice that this method, combined with the diffusion approximation, makes the main error in the calculation of NPS, it can be assumed that the contribution of U-234 to reducing the error in the calculation of NPS may become greater when using kinetic codes.

Thus, it was shown that taking U-234 into account when analysing experiments on a fast reactor with high enriched uranium fuel leads either to a reduction in deviations from the experimental values (SVRE), or to an improvement in the agreement between the calculated estimates (TRE). In absolute terms, the improvements achieved are small. Accounting for the U-234 isotope leads to a small but systematic increase in the values of the effective multiplication factor by 120-150 pcm.

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References
[1] Zaritskaya T.S., Matveev L.V., Rudik A.P., Tsenter E.M. 1983 Accumulation of $^{232}$U in cyclic utilization of the fuel in the VVER-440. Sov. At. Energy, 54, 332.
[2] Nevinitsa V.A., Kolupaev D.N. 2020 U-232 Content Determination in Spent FA from Fast Reactor with a Uranium Load. Atomic Energy, 27, issue 3, pp.: 149-152.
[3] Yamamoto K., Okumura K. 2014 A Study of the Generation of $^{232}$U in UO$_2$ and MOX fuels. J.of Nucl. Sci. and Techn., 51, issue 4, pp. 568–573.
[4] Kusuno S., Watanabe S. 2009, Recycling of Reprocessed Uranium from High Burnup Fuel. Proc. of Int. Conf. Global 2009 Paris, France, September 6-11. Paper 9221.
[5] Yang X., YuH., Zhou K., Zhao J., Chen Y., Hu Y., Tian H., Li Z., Gang Z. 2013 The first Criticality of CEFR. Proc. of the 2013 21st Int. Conf. on Nuclear Engineering, ICONE21, July 29 - August 2, Chengdu, China. ICONE21-16187.
[6] Zhou K., Yu H., Zhao J., Chen X., Hu Y., Tian H., Li Z., Gang Z., Wang S. 2013 The Measurement and Analysis of CEFR Sodium Reactivity Effect. In Proc. of the 2013 21st Int. Conf. on Nuclear Engineering ICONE21 July 29 - August 2, Chengdu, China. ICONE21-15799.
[7] Yu H., Zhao J., Chen Y., Hu Y., Zhan Q., Fan Z., Li Z. 2013 Measurements and Analysis of Temperature Reactivity Coefficient of CEFR. Atomic Science and Techn. 47, p.89-91. [In Chinese].
[8] Borisevich V.D., Borshchevskiy M.A., Zeng S., Jiang D. On Ideal and Optimum Cascades of Gas Centrifuges with Variable Overall Separation Factors. — Chem. Eng. Sci., 2014, 116, p. 465–472.
[9] Leppanen J., Pusa M., Viitanen T., Valtavirta V., and Kaltiaisenaho T. 2015 The Serpent Monte Carlo code: Status, development and applications in 2013. Ann. Nucl. Energy, 82 142-150.
[10] Fomichenko P., et al. 2014 Results of Verification of Computer Codes Used for Used for Analysis BN-1200 Reactor Core Neutronics. Proc. of int. Conf. PHYSOR 2014 – The Role of Reactor Physics Toward a Sustainable Future. The Westin Miyako, Kyoto, Japan, September 28 – October 3, 2014, on CD-ROM.