

Influence of uncertainties of isotopic composition of the reprocessed uranium on effectiveness of its enrichment in gas centrifuge cascades

A Yu Smirnov¹, A R Mustafin¹, V A Nevitsa², G A Sulaberidze¹, A A Dudnikov² and V E Gusev¹

¹ National Research Nuclear University MEPHI (Moscow Engineering Physics Institute), Kashirskoe shosse 31, Moscow, 115409, Russia

² National Research Center “Kurchatov Institute”, Moscow, Russia

E-mail: AYSmirnov@mephi.ru

Abstract. The effect of the uncertainties of the isotopic composition of the reprocessed uranium on its enrichment process in gas centrifuge cascades while diluting it by adding low-enriched uranium (LEU) and waste uranium. It is shown that changing the content of ^{232}U and ^{236}U isotopes in the initial reprocessed uranium within 15% (rel.) can significantly change natural uranium consumption and separative work (up to 2-3%). However, even in case of increase of these parameters is possible to find the ratio of diluents, where the cascade with three feed flows (depleted uranium, LEU and reprocessed uranium) will be more effective than ordinary separation cascade with one feed point for producing LEU from natural uranium.

1. Introduction

In the context of increasing needs of the nuclear power in the fuel for thermal reactors and the gap between production and consumption of natural uranium [1], the reprocessed uranium and plutonium are considered as an alternative to natural raw materials, and partly involved in the light water reactors' fuel cycle. In addition to that there are schemes of using reprocessed uranium and plutonium together [2-5] and separately [6-9].

In the case of separate use of these materials for the production of nuclear fuel, reprocessed uranium must first be enriched in ^{235}U by means of gas centrifuge cascades [10-16]. Moreover, besides saving natural resources and reducing the volume of radioactive waste, reprocessed uranium fully meets the requirements for nuclear non-proliferation treaty (NPT) [17-18]. But enrichment of this material in separation cascades is impeded due to the presence of ^{232}U и ^{236}U [6], concentrations of which are strictly limited to fulfill the requirements of radiation safety in the manufacture of fuel elements and to preserve the neutron-physical characteristics of the fuel.

For today, a number of methods to enrich the reprocessed uranium in gas centrifuge cascades have been proposed [10-16]. They could be based on an ordinary three-flow centrifuge cascades (one feed point, one product out (enriched output), one waste out (depleted tailings outlet)) or on use of cascades with additional feed point of reprocessed uranium or two additional points (reprocessed and waste uranium) and in some cases, additional product out. At the same time the main advantage of centrifuge cascades with additional feed points is to minimize separative work, due to mixing of flows with



different contents of ^{235}U . However, the use of the ordinary centrifuge cascade is technologically easier.

Computational experiments show that cascade with three feed points (waste uranium, LEU, reprocessed uranium) is one of the most effective schemes due to the simultaneous input of LEU and reprocessed uranium which leads to significant decrease in concentrations of harmful isotopes (^{232}U , ^{236}U) while increasing the concentration of ^{235}U isotope [15]. In particular, it is shown that this scheme may be more efficient than cascade with two feed points (natural and reprocessed uranium) [11] or cascade with three feed points (waste uranium, natural uranium, reprocessed uranium) [13]. In addition, preliminary economic calculations have shown that this cascade can be more effective than ordinary separation cascade [19]. But it should be noted that in all calculations was considered spent fuel from Russian type of light water reactors, VVER-1000, and suggested that the concentration of isotopes in it are constant. At the same time, alterations in the concentrations of the unwanted isotopes (^{232}U , ^{236}U) in reprocessed fuel, which needs to be enriched, can have a significant impact on the key parameters of the separation cascade (the consumption of diluent per unit of product, separative work). That must be considered in the further elaboration of the possibility of involving reprocessed uranium in the production of fuel for light-water reactors.

This paper deals with the analysis of the impact of uncertainties of isotopic composition of the reprocessed uranium on efficiency of its further enrichment in gas centrifuge cascades with three feed points. Estimates of changes in consumption of natural uranium and in separative work are made by varying the ^{232}U and ^{236}U concentrations in the initial reprocessed uranium. As the object of study a mathematical model of the quasi-ideal cascade [20] with three feed points is considered [13].

2. Mathematical model

Let us consider cascade with three feed points (Fig. 1.). The cascade has product out (P) and waste out (W) with a concentration C_i^P, C_i^W (from now on, the concentration is expressed as a percent by mass; $i = \overline{1, m}$, i - index number of component of separated mixture; m - the total number of components of the separated mixture). F_1, F_2 and F_3 are flows with concentrations $C_i^{F_1}, C_i^{F_2}, C_i^{F_3}$ into feed points of waste uranium, reprocessed uranium and low-enriched uranium respectively. Detailed description of this mathematical model is given in [13].

The calculations were performed for the problem statement as follows: the concentrations C_n^P, C_n^W and relations $F_2/F_3, F_1/F_3$ are given (n – the index number of the target component). Through calculations is necessary to compute remaining parameters of the cascade, including the concentrations of other components in outlets and the number of stages in the cascade, etc. The problem is interesting from a practical point of view, because in this case clearly defined product quality that allows you to compare the effectiveness of different ways to produce commercial reactor-grade LEU.

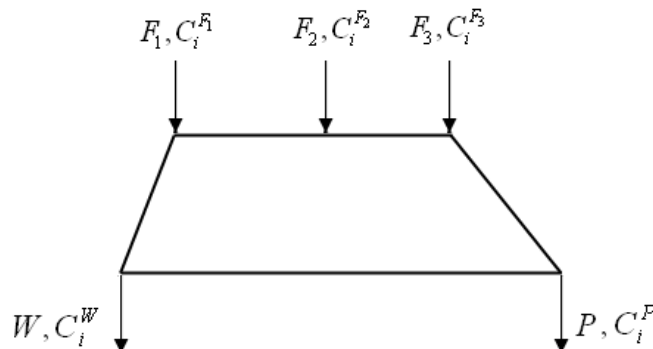


Figure 1. The schematic drawing of the cascade with three feed flows to enrich recycled uranium.

3. Results and discussion

As an example consider the enrichment of spent fuel from VVER-1000 with a burnup level of 40 GWd/t [15]. The isotopic composition of the material is shown in Table 1.

Table 1. The isotopic composition of spent fuel from Water-Water Energetic Reactor, VVER-1000.

Component	^{232}U	^{233}U	^{234}U	^{235}U	^{236}U	^{238}U
Concentration, %	$2.98 \cdot 10^{-7}$	$5.81 \cdot 10^{-7}$	$1.91 \cdot 10^{-2}$	0.901	0.573	The rest

As a calculation model is considered a particular case of "quasi-ideal" cascade - *R*-cascade [16] or cascade with non-mixing condition for chosen pair of components, in this case ^{235}U and ^{238}U isotopes. In the calculations the following parameters were given: concentration of ^{235}U in product was 4% (excluding additional enrichment in order to compensate the effect of ^{236}U), concentration of ^{235}U in waste was 0.1%, the reactivity compensation coefficient was 0.29. The concentration of ^{232}U in the product was limited by value $2 \cdot 10^{-7}\%$ [18]. As diluents were considered the waste uranium with 0.25% of ^{235}U and LEU with 1.5% of ^{235}U . As a comparison has been considered ordinary *R*-cascade to enrich natural uranium to the same concentration of ^{235}U .

For these conditions were calculated cascade parameters with different concentrations of ^{232}U and ^{236}U isotopes in reprocessed uranium, which varied within the range of 15% (relat.). Moreover, concentrations of ^{238}U were calculated by subtracting the sum of all concentrations from unity for each set of concentrations in order to satisfy the condition of equality of total concentration to unity.

Three-dimensional surfaces (Fig. 2-4) show the consumption of natural uranium, separative work and relative costs of Low-enriched uranium production versus concentrations of ^{232}U , ^{236}U isotopes in reprocessed uranium. The relative costs of Low-enriched uranium production was calculated as follows:

$$S = A_C \cdot Z + A_{F_1} \cdot F_1 + A_{F_2} \cdot F_2 + A_{F_0} \cdot F_0, \quad (1)$$

where Z is the total number of separation units, in this case centrifuges, necessary to enrich uranium to the given concentrations of ^{235}U in product and in waste. For cascades with three feed points, Z also takes into account the number of separation units needed for producing LEU diluent. A_C -factor which takes into account the costs associated with the work of gas centrifuges; F_0 - consumption of natural uranium for producing commercial reactor-grade LEU. A_{F_1} , A_{F_2} , A_{F_0} – prices of waste, reprocessed and natural uranium, respectively.

Properly speaking, the value S is not “the relative costs of Low-enriched uranium production” itself, but is a value determining relative costs to a large degree [21]. The first term of (1) is proportional to the cost of separative work, and the others represent prices of input material. The calculations assumed that the average price of natural uranium approximately 9 times higher than the average cost of waste uranium [22], and 3 times less than the cost of the uranium hexafluoride produced from reprocessed uranium. [8]. In the case of calculating the value of S for the ordinary cascade, values F_1 and F_2 is set to 0.

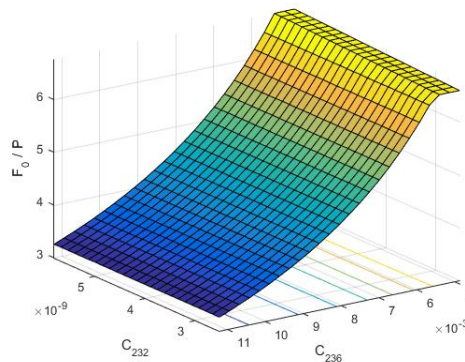


Figure 2. Consumption of natural uranium versus concentrations of ^{232}U , ^{236}U in reprocessed uranium.

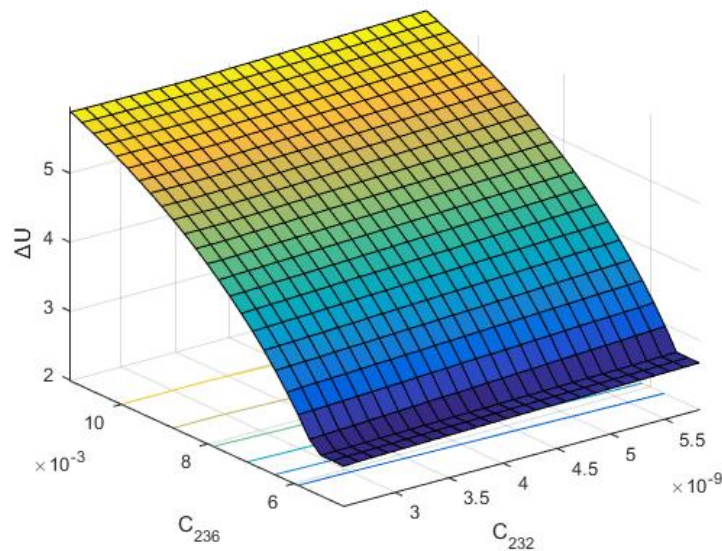


Figure 3. Separative work versus concentrations of ^{232}U , ^{236}U in reprocessed uranium.

Analysis of surfaces (Fig. 2-4) shows that uncertainties of isotopic composition of the reprocessed uranium can have a significant impact on consumption of natural uranium, on separative work and, as a result, on relative costs of Low-enriched uranium production. Therefore, this factor must be considered in a feasibility study of various options of reprocessed uranium enrichment in the gas centrifuge cascades. However, the relative costs of Low-enriched uranium production on the same or even lower level comparing to enrichment of natural uranium can be achieved by varying the cascade parameters.

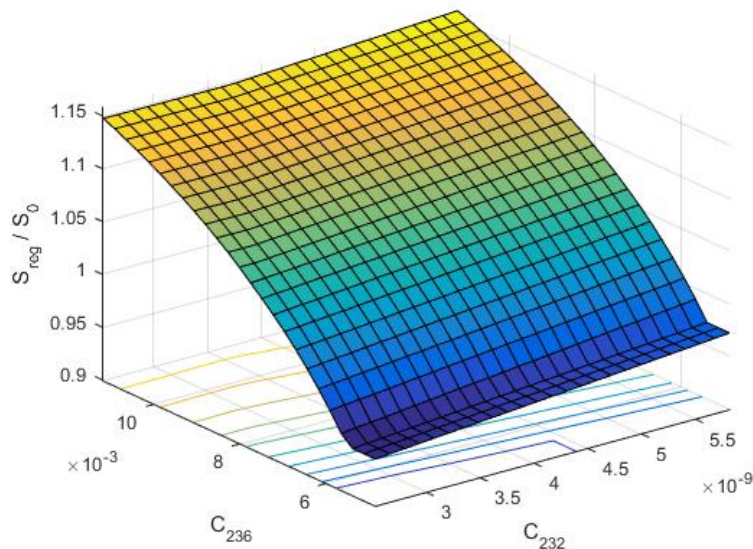


Figure 4. Relative costs of Low-enriched uranium production versus concentrations of ^{232}U , ^{236}U in reprocessed uranium.

4. Conclusion

The impact of uncertainties of isotopic composition of the reprocessed uranium on effectiveness of its enrichment in gas centrifuge cascades with simultaneous dilution with waste and low-enriched

uranium is considered. It is shown that changing the content of ^{232}U and ^{236}U isotopes in the initial reprocessed uranium within 15% (rel.) can significantly change natural uranium consumption and separative work (up to 2-3%). However, even in case of increase of these parameters is possible to find the ratio of diluents, where the cascade with three feeds (depleted uranium, LEU, reprocessed uranium) will be more effective than ordinary separation cascade with one feed flowing for producing LEU from natural raw material.

References

- [1] Andrianova E A, Davidenko V D, Tsibul'skii V F 2015 *At. Energy* **118** 301–6
- [2] Pavlovichev A M et al 2006 *At. Energy* **101** 407–413
- [3] Zilberman B Ya et al 2012 *At. Energy* **113** 383–391
- [4] Fedorov Yu S et al 2005 *At. Energy* **99** 136–141
- [5] Pavlovichev A M et al 2008 *At. Energy* **104** 195–8
- [6] Smirnov A Yu et al 2012 *Phys. At. Nuclei* **75** 1616-25
- [7] Coleman J.R., Knight T.W. 2010 *Nucl. Eng. Design* **240** 1028–1032
- [8] Butler G G, Wilcox P 1988 *Nucl. Engineer* **28** 186-90
- [9] Hida K, Kusuno S, Seino T 1986 *Nuclear Technology* **75** 148-159
- [10] Palkin V A 2013 *At. Energy* **115** 32–7
- [11] Sulaberidze G A, Borisevich V D, Xie Q X 2006 *Theor. Found. Chem. Eng.* **40** 5-14
- [12] Palkin V.A. 2010 *Prosp. Mater.* **8** 11–4
- [13] Smirnov A Yu, Sulaberidze G A 2014 *At. Energy* **117** 44–51
- [14] Sulaberidze G A, Borisevich V D, Xie Q X 2004 In: *Proc. of IX All-Russia (International) Scientific Conference “Physical and Chemical Processes on Selection of Atoms and Molecules”* (Russia, October 4-8, 2004) p 70
- [15] Smirnov A Yu, Sulaberidze G A 2015 *Phys. Proc.* **72** 132-5
- [16] De la Garza A, Garrett G A and Murphy J E 1961 *Chem. Eng. Sci.* **15** 188–209
- [17] Abbas K et al 2013 *Esarda Bulletin* **49** 75-81
- [18] Alekseev P N et al 2012 *Phys. At. Nuclei* **73** 2264-2270
- [19] Smirnov A Yu et al 2016 *IOP Conf. Ser.* **751** 012005
- [20] Sazykin A A 2000 In the book: ISOTOPES. Properties. Production. Application. Ed. V.Yu. Baranov Moscow. IzdAt. 72-108 (in Russian)
- [21] Smirnov A Yu, Borisevich V D, Sulaberidze G A 2012 *Theor. Found. Chem. Eng.* **46** 373-8
- [22] Varela C, Akapo D, Schneider E A et al. 2009 In: *Proc. of GLOBAL-2009* 9463

Acknowledgements

This research is supported by the grant No.14.Y30.16.6284-MK of the President of the Russian Federation for young scientists.