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Cascade design for isotopically modified molybdenum as an alternative to zirconium alloys

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ABSTRACT

Search of the optimum design used in practical squared-off cascades is usually started with optimization of various model cascades with the specified concentrations of a target component in their outgoing flows. After that, the approximation of the found model cascade is carried out. These two stages of research have been executed in solution of the problem of manufacturing the isotopically modified molybdenum (IMM), which is considered as an alternative construction material for nuclear power plants. A quasi-ideal cascade is chosen as a model cascade. Based on the results obtained for model cascades, the performance of single-cascade and double-cascade schemes has been analyzed under various requirements for thermal neutron capture cross section of the produced isotope product, and design of the squared-off cascades is completed. It is demonstrated that the most efficient separation scheme could be realized by means of either a single-cascade or double-cascade scheme, depending on the specific requirement. The 4-section squared-off cascades provide the highest metrics for cascade efficiency. The results of this research could provide necessary guidance for further design and construction of a separation plant for large-scale production of IMM.

1. Introduction

At the moment zirconium alloys are the basic material for claddings of fuel elements in the most commercial nuclear power plants (NPPs), primarily because of its low capture cross section for thermal neutrons and good corrosion resistance in water. To improve the corrosion resistance of zirconium under reactor conditions in water and steam at a working temperature of 300–400 °C, such chemical elements as tin, niobium, iron, chromium and others are used as the alloying elements (Kubaschewski and Alcock, 1976).

In Russia, the preference for fuel cladding was given to binary alloys of zirconium with niobium, in particular with a content of niobium equal to 1% (the E110 alloy). Niobium has also a small neutron capture cross section. In addition, it reduces effectively zirconium's absorption of hydrogen, forming only solid solutions with zirconium, which provides high plasticity to the alloy (Blumenthal, 1958).

Although zirconium alloys possess significant physical properties mentioned above, they have some disadvantages too. One of them consists in the negative impact on spent fuel reprocessing. The Zr-92 isotope under the influence of radiation inside reactors converts into the radioactive Zr-93 isotope, whose half-life is 1.53 million years. As a result, the extracting spent assemblies made from the E110 alloy have the level of beta radiation 200–300 times higher than the maximum permissible one (Clark et al., 1973). This leads to the necessity of their long exposure before reprocessing.

Another one drawback is that zirconium reacts actively with water vapor, generating heat, producing hydrogen and accelerating degradation of a fuel rod cladding by means of the vapor-zirconium chemical

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Table 1 – Nuclear-physical and thermo-physical properties of Zr and Mo (Shmelev et al., 2016).

Material	Density, g/cm^3, 20 °C	T_{melt}, °C	Thermal capacity, 10^7 J/(m^3 K)	Thermal conductivity, W/(m K), 1000 K	Neutron capture cross-section, σ_c, barn (E_n = 0.025 eV)
Zr	6.5	1855	0.23	21.5	0.196
Mo	10.22	2623	0.30	112	2.570

Table 2 – Isotopic composition and neutron capture cross section for natural Mo.

Isotope	Mo-92	Mo-94	Mo-95	Mo-96	Mo-97	Mo-98	Mo-100
Natural content, %	14.84	9.25	15.92	16.68	9.55	24.13	9.63
σ_c, barn (E_n = 0.025 eV)	0.061	0.339	13.602	0.447	2.488	0.132	0.194

reaction at temperatures higher than 700 °C (Foreman, 2015):

$$Zr + 2H_2O \rightarrow ZrO_2 + 2H_2\uparrow. \quad (1)$$

This reaction is extremely hazardous during accidents at NPPs with a water coolant or moderator. In particular, it was the main reason for series of explosions occurred in the Fukushima nuclear accident (Foreman, 2015). Besides, note that uranium dioxide in nuclear fuel being heated up also reacts with zirconium by the following reaction:

$$Zr + UO_2 \rightarrow ZrO_2 + U, \quad (2)$$

which generates further heat and also reduces fuels' melting point, contributing to the overheating and melting of active zone of NPPs (Foreman, 2015; Burakov et al., 1997).

One of possible substitute for zirconium alloys could be molybdenum. This idea is discussed for a long time (Ki et al., 1997; Goldstein et al., 1988; Shmelev et al., 2016; Youinou and Sen, 2014). Its thermal conductivity is higher than that of zirconium. Its low coefficient of expansion and good corrosion resistance make it as an attractive option for construction materials of fuel cladding and/or a part of nuclear fuel at NPPs (Youinou and Sen, 2014). The data of Table 1 demonstrate the primary physical properties of zirconium and molybdenum, including their capture cross section for thermal neutrons whose typical kinetic energy E_n is about 0.025 eV.

However, certain difficulties exist for the application of natural molybdenum. The primary one is its considerable absorption of thermal neutrons, which, if applied at NPPs, would increase reactors' requirements for uranium enrichment significantly. For example, compared with the standard Zr-UO_2 fuel system, the Mo-UO_2 system producing the same energy will need about 30%–45% more natural uranium and 45%–60% more SWU (Youinou and Sen, 2014).

Since the isotope Mo-95 defines the biggest thermal neutron absorption (see the data in Table 2), the idea to modify the isotopic composition of natural molybdenum by reducing its concentration and approaching a new neutron capture cross section close to that of zirconium has been presented in Shmelev et al. (2016). The molybdenum hexafluoride (MoF_6) is used as a processing gas for this purpose.

This idea looks quite realistic, since the technology to separate molybdenum isotopes by a gas centrifuge method has been developed in Russia in 90s based on the experimental and calculation results of multicomponent isotope separation in a single centrifuge and in cascades (Cheltsov and Sosnin, 2006; Aisen et al., 1996).

2. Problem statement

A double-cascade separation scheme (see Fig. 1b) has been introduced to manufacture IMM with a content of the Mo-92 or Mo-100 isotopes up to 95% (Aisen et al., 1996). It is also demonstrated that, for production of the IMM with a thermal neutron capture cross section as in natural zirconium, a double-cascade scheme provides a noticeable advantage over a single-cascade scheme (see Fig. 1a) both in terms of the total flow (by 14%) and raw material consumption (by 63%) per the product unit (Shmelev et al., 2016).

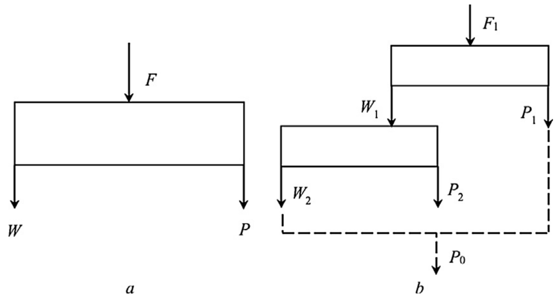

Fig. 1 – Sketch of a single-cascade scheme (a) and a double-cascade scheme (b) under investigation.

It seems that a double-cascade scheme could be regarded as an "universal" solution for the production of IMM. Nevertheless, in the future application of IMM, the actual requirement for thermal neutron absorption may not be fixed at a definite value. If the requirement is changed, whether a double-cascade scheme is still more efficient or not needs careful investigation.

Model cascades, because of their convenience to calculate, are employed for the performance comparison of separation schemes. Although optimization results based on characteristics of model cascades could not be directly applied into practice, they could be used as a template for design of squared-off cascades (SOCs), which are usually adopted for large-scale isotope separation. The major idea of this study is to take a next step (following the first one presented in Shmelev et al. (2016)) to designing a separation plant for large-scale production of IMM. For this purpose, we will examine the efficiency of different separation schemes under various requirements for thermal neutron capture cross section, and showing the design of the SOCs for production of IMM.

However, in the previous research (Shmelev et al., 2016), concentrations of target components in outgoing flows are chosen as optimization variables, which are not directly controlled parameters of cascades, making the optimization procedure inconvenient to use. Besides, the Mo-98 isotope should also be considered as one of the target components, due to its small thermal neutron absorption. Therefore, a new optimization procedure should be developed, employing different optimization variables as well as a new set of components as the target ones.

Based on the results of optimization, performance of single-cascade and double-cascade schemes under various requirements will be compared, and the design of SOCs to produce IMM with specified characteristics on a thermal neutron capture cross section will be carried out.