# Use of molybdenum as a structural material of fuel elements for improving the safety of nuclear reactors

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**Abstract**. Main purpose of the study is justifying the use of molybdenum as a structural material of fuel elements for improving the safety of nuclear reactors. Particularity of used molybdenum is that its isotopic composition corresponds to molybdenum, which is obtained as the tailing during operation of the separation cascade for producing a material for medical diagnostics of cancer.
When performing the study the neutron-physical properties of isotopes of natural molybdenum (nuclear data library JENDL-4.0) and thermal properties of metallic molybdenum were used.
The following results were obtained:
1. A method for reducing the thermal constant of fuel elements for light water and fast reactors by using dispersion fuel in cylindrical fuel rods containing, for example, granules of metallic U-Mo-alloy into Mo-matrix was proposed.
2. The necessity of molybdenum enrichment by weakly absorbing isotopes was shown.
3. Total use of isotopic molybdenum will be more than 50%.
A method for reducing the thermal constant of the fuel elements, allowing us to increase the safety of light water and fast nuclear reactors by using dispersion fuel in cylindrical fuel rods containing, for example, granules of metallic U-Mo-alloy into Mo-matrix with enrichment by weakly absorbing isotopes of molybdenum is proposed.

## 1. Introduction

It is well known fact that the safety of nuclear reactor in the case of introduction of reactivity comparable with delayed neutrons fraction depends largely on the properties of fuel and materials constituting fuel element [1]. Sharp reactivity increase initiates neutron flash, which is suppressed by means of feedback due to fuel heating and increase of neutron absorption by fertile nuclide ($^{238}U$ or $^{232}Th$) thanks to the Doppler effect. If there is enough time for heat transfer from fuel to coolant then there would be feedback caused by heating of the coolant. This second feedback depends largely on thermal-physical characteristics of the fuel element and materials constituting it.

As is known, a refractory material based on molybdenum is characterized by good thermal-physical properties [2]. Therefore, such a material appears to be attractive when using dispersion fuel elements with good heat-conducting molybdenum matrix. However, there are some difficulties to use such a material in the reactor core. Firstly, molybdenum of a natural isotopic composition is quite a strong absorber of neutrons. Secondly, it is necessary to take into account its compatibility with fuel material on the one hand and with coolant on the other hand. Resolving these issues is the subject of the present paper.

## 2. Natural isotopic composition of molybdenum and some peculiarities of its nuclear-physical properties

Zirconium alloys are used as a basic structural material in the core of thermal reactors. The advantage of zirconium alloys is small neutron capture cross-section in the thermal energy range. At the same time molybdenum of a natural isotopic composition is characterized by a significantly larger neutron capture cross-section in this energy range (see table 1). It can be seen that neutron capture cross-section at thermal point ($E_n$ = 0.025 eV) of molybdenum of a natural isotopic composition is 13 times larger than that of zirconium [3].

**Table 1.** Isotopic composition and radiative neutron capture cross-section of natural zirconium and molybdenum, as well as of different isotopes of molybdenum at thermal point ($E_n$ = 0.025 eV).

| Nuclide, atomic number of molybdenum isotope | Natural composition [%] | $\sigma_c$ [barn] ($E_n$ = 0.025 eV) |
|---|---|---|
| $Zr_{nat}$ | – | 0.196 |
| $Mo_{nat}$ | – | 2.57 |
| 92 | 14.8 | 0.061 |
| 94 | 9.25 | 0.339 |
| 95 | 15.9 | 13.6 |
| 96 | 16.7 | 0.447 |
| 97 | 9.55 | 2.49 |
| 98 | 24.1 | 0.132 |
| 100 | 9.63 | 0.194 |

It means that technology of isotopic enrichment should be applied in order to use molybdenum as a construction material in thermal reactors. It can be seen from the table that isotopes $^{92}$Mo and $^{94}$Mo account for almost a quarter of the natural material and are located on the “light” end of isotopes natural mixture. Their mixture provides about the same neutron capture cross-section as natural zirconium does. Isotope $^{95}$Mo provides a dominant contribution to the total capture cross-section, the atomic weight of which is intermediate in molybdenum isotopic composition. Use of $^{98}$Mo and $^{100}$Mo is not excluded, of course. Dependence of capture cross-section of these light and heavy isotopes of molybdenum on neutron energy is shown in figures 1 and 2 (here reactor energy range is considered). One can see that capture cross-section is generally inversely proportional to neutron velocity, and, generally speaking, the resonance integral is more than an order of magnitude smaller than that of $^{238}$U [3].

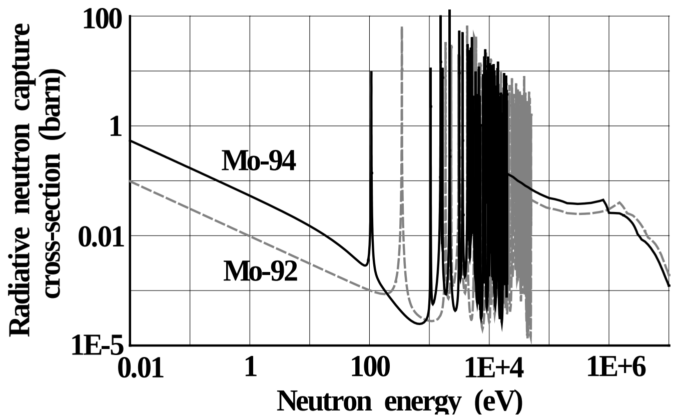


**Figure 1.** Radiative neutron capture cross-section of molybdenum isotopes $^{92}$Mo and $^{94}$Mo on neutron energy (nuclear data library JENDL-4.0).

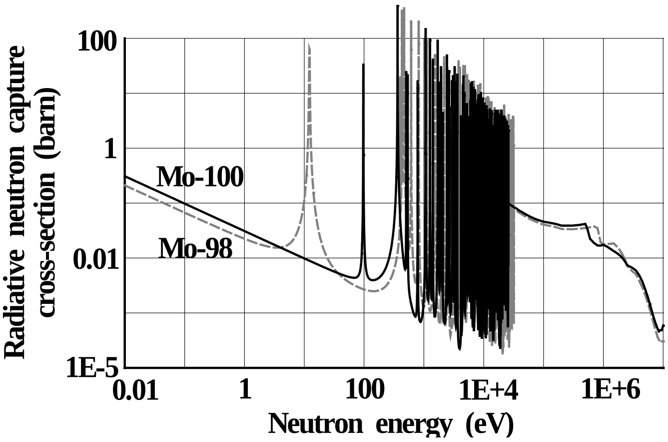


**Figure 2.** Radiative neutron capture cross-section of molybdenum isotopes $^{98}$Mo and $^{100}$Mo on neutron energy (nuclear data library JENDL-4.0).

## 3. Isotopic enrichment of molybdenum for medical purposes and for reducing its radiative neutron capture

As is known, the isotopic enrichment is a high-level technology requiring the use of multi-step separation cascades, and therefore is very expensive. However, for the case of production of enriched molybdenum there is one important factor that can significantly facilitate the solution of this issue.

There is already a commercial enrichment of molybdenum [4] with the production of the desired isotopes $^{98}$Mo and $^{100}$Mo for medical diagnosis of early formation of cancerous tumors. Currently, the diagnosis is the most advanced in the world in the field of medicine and, therefore, the need for these heavy isotopes of molybdenum is high and continues to rise.

Keeping this fact in mind, it can be assumed that the process of obtaining molybdenum enriched by the light isotopes $^{92+94}$Mo can be successfully combined with the process of producing heavy isotopes $^{98}$Mo and $^{100}$Mo. Heavy isotopes are the product at one end of enrichment cascade, while light isotopes are the product at the other end. Of course, the structure of the cascade requires to be changed to get rid of the strong neutron absorber - isotope $^{95}$Mo, atomic weight of which is intermediate in a series of atomic weights. Since this isotope is in the middle of the spectrum of the atomic weights of molybdenum natural isotopes (see table 1), a cascade of isotope separation with additional selection in the middle of the cascade should be used. This problem is considered in [4].

## 4. Thermo-physical parameters of metallic molybdenum, important for use as a structural material of the fuel element

Table 2 shows some thermo-physical parameters of molybdenum and zirconium that are important for heat transfer in the fuel element [2, 3, 5].

**Table 2.** Nuclear-physical and thermo-physical properties of fuel and structural materials.

| Material | Density [g/cm$^3$] (20ºC) | $T_{melting}$ [ºC] | Heat capacity [$10^7$ J/(m$^3$·K)] | Thermal conductivity [W/(m·K)] (1000ºK) | Time constant of cylindrical fuel element (rod) $\tau_{th}$ [s] (d = 9.1 mm) |
|---|---|---|---|---|---|
| Zr | 6.5 | 1855 | 0.23 | 21.5 | 0.36 |
| Mo | 10.22 | 2623 | 0.30 | 112 | 0.20 |
| U-9%Mo | 17.6 | 1300 | 0.35 | 40 | 0.36 |
| Dispersion fuel[a] | – | – | 0.32 | 71.5 | 0.26 |

[a] Granules – U-9%Mo; matrix – Mo; the proportion of granules $V_f$ = 0.5.

Apart from the fact that molybdenum has a melting point substantially greater, it also has a more than 5 times greater coefficient of thermal conductivity. Model fuel element of metallic molybdenum is characterized by a time constant of transferring heat to the environment [1] which is almost two times smaller than that for zirconium. These attractive properties of molybdenum can be used to create a new concept of a fuel element for thermal and fast reactors. This structural material is compatible with the well-known metallic uranium-molybdenum fuel good thermal properties of which are shown in table 2.

## 5. Rod fuel element with a small time constant $\tau_{th}$ for fast reactor and thermal reactor

As is known, fuel material of fuel elements of the world's first nuclear power plant built in the USSR (Obninsk) in 1954, was an alloy U-9%Mo. Molybdenum was chosen not only because it is able to stabilize γ-phase of uranium, but also because its alloy with uranium is characterized by a high thermal conductivity and high nuclear density of uranium (see table 2).

It can be seen that among materials presented in table 2, for creating a fuel element it is preferable to use dispersion fuel containing granules of a metal alloy U-9%Mo, dispersed into the molybdenum matrix. To improve the thermal contact of fuel with cladding, the latter also should be produced, for example, from Mo-based alloy [6]. It is known that molybdenum and its alloys are compatible with both aqueous coolant (in thermal reactors), and liquid metals Na, Pb and Pb-Bi [7, 8] in fast reactors.

Experimental studies [9] performed with a model nuclear fuel have confirmed the possibility of creating such a dispersion fuel (U-Mo - fuel granules, Mo - matrix) both in terms of compatibility over a wide temperature range of fuel granules, matrix and cladding, and at high fuel burn-up.

Since the thermal conductivity of fuel granules (U-9%Mo) is 2.8 times smaller than that of a molybdenum matrix, the average thermal conductivity of dispersion fuel material depends to a significant extent on the proportion of fuel granules, and on their forms. Assuming that fuel granules have a spherical shape, when their share $V_f = 0.5$ in dispersion fuel material, the average coefficient of thermal conductivity of the fuel rod is 71.5 W/(m·K) [5], and the time constant of the fuel element with a diameter d = 9.1 mm will be $\tau_{th} = 0.26$ sec (see table 2).

## 6. Prospects for the use of dispersion fuel elements with molybdenum as a structural material in fast reactors

As for the apparent attractiveness of the possibility of using molybdenum and its alloys as a structural material in fast reactors (fuel - uranium-molybdenum alloy, the matrix material - molybdenum in dispersion fuel elements, cladding material - possibly also refractory material), the need for use of molybdenum enriched by light isotopes still requires to be considered. However, it is important to note that in a core the combination of a high melting point liquid metal coolant (e.g., Pb, Pb-Bi et al.) and the fuel element with a cladding based on a refractory (molybdenum-based) material [6] and with a molybdenum matrix can significantly increase the stability of the core with respect to the possibility of a crisis in the heat transfer at a jump of reactivity. Using uranium-molybdenum fuel fits well into the concept of protected fuel cycle based on a mixture of ($^{233}U+^{238}U$) [10].

If two heavy isotopes $^{98+100}Mo$ would be used for medicine purposes (these isotopes account for about 1/3 of the total of molybdenum), and two light isotopes $^{92+94}Mo$ would be used for reactor purposes (these isotopes account for about 1/4 of the total of molybdenum), then the total use of the isotopic molybdenum would be over 50%. The rest of the molybdenum without damage may be used in the national economy.

## 7. Summary

A method for reducing the time constant of the fuel elements allowing us to increase the safety of light water reactors and fast reactors by using dispersion fuel in cylindrical fuel elements containing, for example, granules of metallic U-Mo-alloy into Mo-matrix with enrichment by weakly absorbing molybdenum isotopes was proposed. At the same time the use of the isotopic molybdenum would be more than 50%.

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## Acknowledgment

The authors thank the staff of the Department №9 "Physical Problems of Materials Science" of the National Research Nuclear University MEPhI Dr. A.V. Tenishev and Dr. D.P. Shornikov for useful consultations.