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# USE OF MOLYBDENUM AS A STRUCTURAL MATERIAL OF FUEL ELEMENTS IN LEAD OR LEAD-BISMUTH EUTECTIC COOLED FAST REACTOR TO IMPROVE ITS SAFETY

**Abstract.** In our previous works has been described that the mean prompt neutron lifetime in fast reactors can be significantly elongated [1,2]. But if the last parameter is not comparable with the time required for the heat generated in the fuel elements to transport from a fuel to a coolant, feedback on the effect of changes in coolant parameters on reactivity during this time will not have time to manifest. Here was studied that the thermal constants of fuel elements can be shortened by using molybdenum as a structural material of fuel elements. Other aspects related to the use of molybdenum in nuclear reactors have also been studied.

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%.
4. Mo has a good corrosion resistance to Pb and Pb-Bi eutectic

A decrease in thermal constants of fuel elements, in combination with an increase in the mean prompt neutron lifetimes in the absence of the heat transfer crisis in fast reactors, can be promising way for a better reactor operation safety even under the conditions of the prompt neutron excursions.

**Keywords:** improvement of fast reactor safety, physic-mechanical characteristics of molybdenum, thermal constant of fuel elements, corrosion resistance in lead and LBE.

## 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 [3]. 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 [4]. 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.

## Physico – mechanical characteristics of molybdenum and some peculiarities of its nuclear – physical properties

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

Natural isotopic composition of molybdenum is represented by seven stable isotopes with mass numbers 92, 94–98, and 100, of which $^{98}$Mo is the most common (23.75%).

Molybdenum has a favorable complex of physico - mechanical characteristics, due to which it is one of the best structural metals (Table 1).

Table 1 - Physico-mechanical characteristics of molybdenum

| Characteristic | unit of measurement | Value |
|---|---|---|
| Crystal cell, $\alpha_0$ | nm | 0,314737 |
| Atomic radius $r_a$ | nm | 0,139 |
| Atomic volume Ω | $м^3$/mole | 9,42E-06 |
| Atomic mass A | a.t.u. | 95,941 |
| Ionization potential U | eV | 7,29 |
| Density at $20^0$C ρ | кg/$m^3$ | 10,2E+03 |
| Melting temperature $T_{melt}$ | $^0$C | 2587(2625) |
| Boiling temperature $T_{boiling}$ | $^0$C | 5227 |
| Spesific heat of fusion $L_m$ | J/кg | 0,382 |
| Spesific heat of evaporation L at $T_{boiling}$ | J/кg | 6,191 |
| Thermal conductivity at $20^0$C λ | W/(m*К) | 162 |
| Heat capacity at $20^0$C $C_c$ | J/(кg*К) | 240-250 |
| Thermal expansion at $20^0$C $\alpha_t$ | $К^{-1}$ | 5,1-5,2E-06 |
| Vapor pressure при $T_{пл}$ | Pa | 2,94(3,47)E-02 |
| Electrical resistivity at $20^0$C $\rho_e$ | Om*m | 5,0-5,7E-08 |
| Magnetic susceptibility при $20^0$C $\chi_{ms}$ | $m^3$/кg | 0,82-0,93E-09 |
| Electron work function A | eV | 4,33 |
| Emissivity on a smooth surface ρ | | 0,4 |
| Modulus of normal elasticity at $20^0$C | kgs/$mm^2$ | 32000 |
| Shear modulus at $20^0$C | кgs/$mm^2$ | 12200 |

Important advantages of Mo are its high values of melting point, normal elasticity modulus and thermal conductivity with a relatively low density and low coefficient of linear expansion [5,6,7,8,9,10,11]. Since the density of Mo (10200 kg / m3) is almost two times less than the density of W (19300 kg / m3), the Mo-based alloys have a much higher specific strength (at temperatures below 1370 ° C). Molybdenum has a rather low neutron capture cross section; the capture values of some molybdenum isotopes, in the thermal spectrum is smaller than the zirconium (zirconium alloys are used as a basic structural material in the core of thermal reactors) capture cross-section ($\sigma_{(n,\ y)thermal}$(Zr) = 0.18 barn), and in the fast spectrum is smaller than iron capture cross-section ($\sigma_{(n,\ y)1\ MeV}$(Fe) = 0.06 barn) (Table 2) [12,13]. Mo has good heat resistance and high radiation resistance [14,15,16], it is characterized by high corrosion resistance in most alkaline solutions, in liquid metals, as well as in sulfuric, hydrochloric and hydrofluoric acids at different temperatures and concentrations.

Table 2: Isotopic composition and radiative neutron capture cross-sections of molybdenum at thermal point (En = 0.025 eV) and in the fast spectrum

| Nuclide, atomic number of molybdenum isotope | Natural composition [%] | nuclei concentration ρ, 1-E24 $cm^{-3}$ | $\sigma_{(n,y)}$, barn ($E_n$=0,0253 eV) | $\sigma_{(n,y)}$, barn ($E_n$=1 MeV) |
|---|---|---|---|---|
| Monat | | 0,06403 | 2,55 | 0,035 |
| 92 | 14,8 | | 0,019 | 0,033 |
| 94 | 9,3 | | 0,015 | 0,036 |
| 95 | 15,9 | | 14 | 0,054 |
| 96 | 16,7 | | 0,5 | 0,028 |
| 97 | 9,6 | | 2,1 | 0,055 |
| 98 | 24,1 | | 0,13 | 0,028 |
| 100 | 9,6 | | 0,199 | 0,017 |

At the same time molybdenum of a natural isotopic composition is characterized by a significantly larger neutron capture cross-section in the thermal energy range

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 [12].

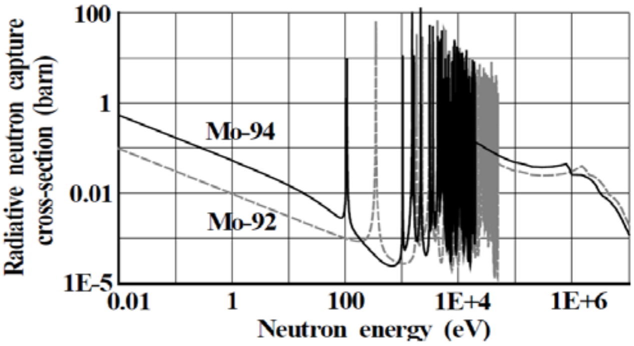

Figure 1: Radiative neutron capture cross-section of molybdenum isotopes $^{92}$Mo and $^{94}$Mo on neutron energy (nuclear data library JENDL-4.0)

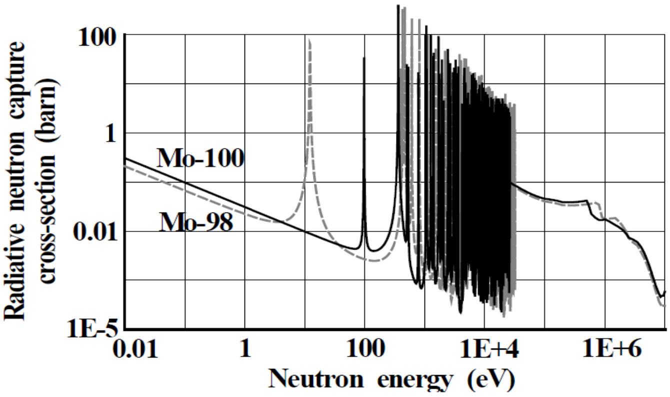

Figure 2. Radiative neutron capture cross-section of molybdenum isotopes $^{98}$Mo and $^{100}$Mo on neutron energy (nuclear data library JENDL-4.0)

**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 [17] 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 [17].

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

Table 3. Shows some thermo-physical parameters of molybdenum and zirconium that are important for heat transfer in the fuel element [4, 12, 18].

Table 3 - Nuclear-physical and thermo-physical properties of fuel and structural materials.
[a]Granules – U-9%Mo; matrix – Mo; the proportion of granules $V_f = 0.5$

| Material | Density [g/cm$^3$] (20°C) | $T_{melting}$ [°C] | Heat capacity [$10^7$J/(m$^3$·K] | Thermal conductivity [W/(m·K)] (1000°K) | 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 |

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 [3] 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 3.

### 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 3).

It can be seen that among materials presented in table 3, 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 [19].

Experimental studies [20] 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 Vf = 0.5 in dispersion fuel material, the average coefficient of thermal conductivity of the fuel rod is 71.5 W/(m·K) [18], and the time constant of the fuel element with a diameter d = 9.1 mm will be $\tau_{th} = 0.26$ sec (see table 3).

### Prospects for the use of dispersion fuel elements with molybdenum as a structural material in fast reactors and corrosion resistance of molybdenum to lead and lead-bismuth eutectic

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.

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 [5,21,22,23,24,25,26,27] in fast reactors. Here some results of different studies, that shows high corrosion resistance to Pb and Pb-Bi coolants.

In work [27] The relative resistance of 21 metals and alloys to mass transfer in liquid lead has been measured. Tests were performed in small, quartz thermal-convection loops. The test temperature was about 800°C, with a thermal gradient of 300 $^0$C existing across the loops. All the metals and alloys studied, only niobium and molybdenum exhibited a high resistance to mass transfer (Figure 3). Neither of these metals suffered noticeable mass transfer or corrosion under the test conditions. A transverse section of the hot-leg specimen from a molybdenum loop is shown in Fig. 4.

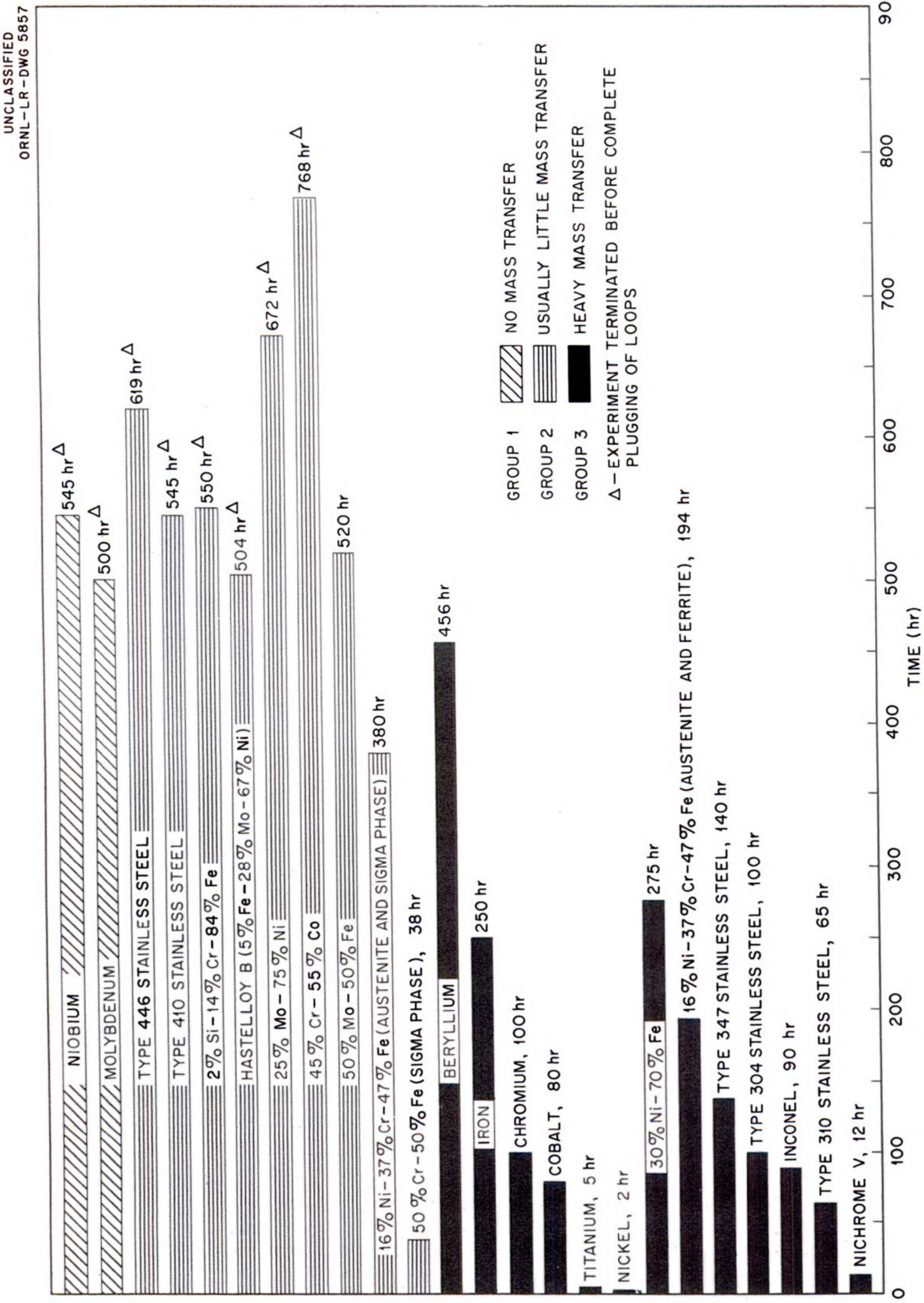

Figure 3: Mass transfer in liquid lead

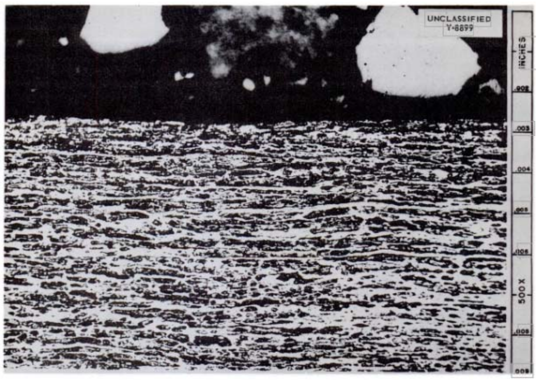

Figure 4 - Transverse Section of Hot-Leg Specimen from
a Molybdenum Loop in Which Liquid Lead Was Circulated. 500X

The specimens of molybdenum were immersed in the stirred lead-bismuth for 1000 h, that was heated up to 700 $^0$C [28]. Oxygen concentration was $5*10^{-6}$ wt.%.

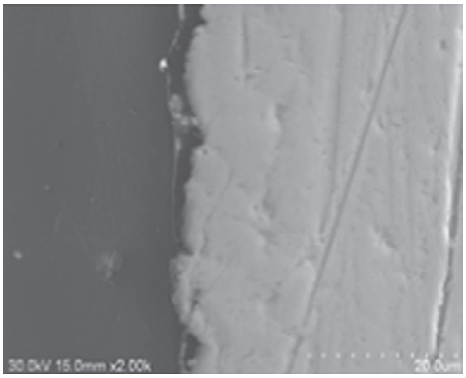

Figure 5 - SEM micrograph of Mo after immersion in 700 $^0$C LBE for 1000 h

Fig. 5 shows the SEM micrograph of the cross section of the molybdenum. There is no significant weight change of the molybdenum specimen. These results show that after the immersion in high temperature LBE for 1000 h. At lower temperature, high corrosion resistance of molybdenum in liquid LBE has been also reported in another works. Fazio et al. (2003) reported that in flowing LBE at 400 $^0$C molybdenum exhibited smooth surface with no evidence of LBE on the surface and growth of oxide layer. Hata and Takahashi (2005) reported that in the stirred LBE pool at 450 $^0$C molybdenum had good corrosion resistance.

The result of the molybdenum after immersion in Pb–Bi is shown in Fig. 6 [29]. Oxygen concentration was $10^{-6}$ wt.%. (550 $^0$C) - 2* $10^{-}5$(800 $^0$C)).

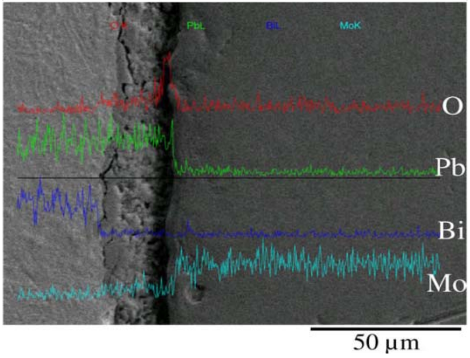

Figure: 6 SEM-EDX micrograph analysis of molybdenum after immersion in Pb–Bi at 550 $^0$C for 12 h
and then the temperature was increased up to 800 $^0$C and kept there for 12 h

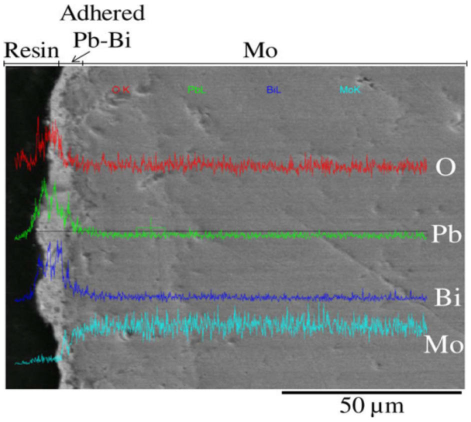

Figure 7 - SEM-EDX micrograph analysis of molybdenum after immersion in Pb–Bi at 550 $^0$C for around 500 h and then the temperature was increased up to 800 $^0$C and kept there for 15 h.

Fig. 7 shows the SEM-EDX micrograph analysis of the cross section of the molybdenum after immersion in Pb–Bi [29]. Oxygen concentration was $10^{-6}$ wt.% (550 $^0$C - 800 $^0$C).

The compatibility test was performed at 673 K and the corrosion and tensile results reported concern the first 1500-h run of the loop operation [30]. All the materials tested suffered from liquid metal attack exhibiting a weight loss. The consequent evaluation of the corrosion rate showed that, under the given test conditions, the refractory metals are more resistant than the steels. The measured weight loss of molybdenum sample was $9.1*10^{-7}$mg/mm$^2$ ($4.7*10^{-5}$ µm/h). From a metallographic point of view, as is shown in the SEM micrograph of Fig. 8 metal exhibited an almost smooth interface with the lead–bismuth alloy. No evidence of liquid metal attack on the surface of material, or of the growth of an oxide layer, could be detected.

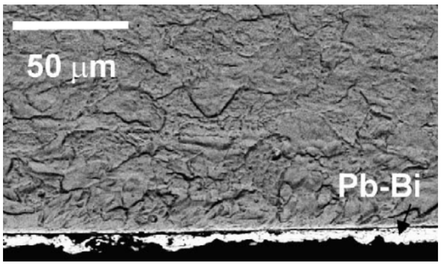

Figure 8: SEM – micrograph of the Mo cross section

The weight loss measured could be associated with both the uniform dissolution and the low solubility of Mo in Pb–Bi liquid at 673 K.

In another work [31] the specimens of molybdenum were immersed in the lead for 500 h, that was heated up to 700 $^0$C. Oxygen concentration was $4.5*10^{-7}$ wt.%.

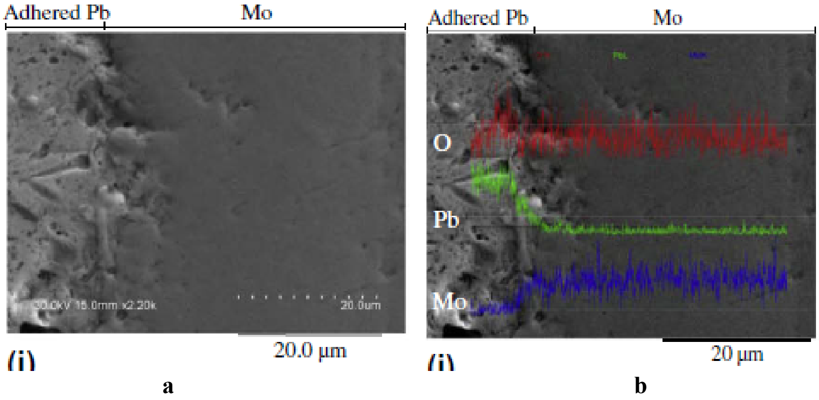

Figure 9 - SEM(a)-EDX(b) micrograph of cross section of tested molybdenum specimen after immersion in lead at 700 $^0$C for 500 h

The figures 4, 5, 6, 7, 8, 9 shows that no penetration of Pb–Bi or Pb into the molybdenum matrix and no dissolution of molybdenum atoms from specimen into Pb–Bi/Pb. Moreover, no crack on the surface of the specimens after immersion in Pb–Bi/Pb under transient temperature up to 800 $^0$C/700 $^0$C was observed.

Fig. 8a shows that neither cracks nor a friable molybdenum oxide layer on the surface of the specimen occurred.

This results showed that the molybdenum exhibited high corrosion resistance to Pb–Bi up to 800 $^0$C and to Pb up to 700 $^0$C. In all this work molybdenum specimen has a purity of at least 99.95 wt.% .

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 [19] 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) [32].

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.

**Summary conclusion**

All the results presented above allowed us to make the following conclusions:

1. 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.
2. The use of the isotopic molybdenum would be more than 50%.
3. Molybdenum has the good resistance to lead and LBE.

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# ҚАУІПСІЗДІКТІ ЖОҒАРЫЛАТУ ҮШІН, ҚОРҒАСЫН НЕМЕСЕ ҚОРҒАСЫН-ЕВТЕТИВТІ САЛҚЫНДАТЫЛҒАН ЖЫЛДАМ РЕАКТОРДА, ОТЫН ЭЛЕМЕНТТЕРІНІҢ ҚҰРЫЛЫМДЫҚ МАТЕРИАЛЫ РЕТІНДЕ МОЛИБДЕНДІ ПАЙДАЛАНУ

**Аннотация.** Зерттеудің негізгі мақсаты – ядролық реакторлардың қауіпсіздігін арттыру үшін, отын элементтерінің құрылымдық материалы ретінде, молибденді пайдалануды түсіндіру болып табылады. Қолданылған молибденнің ерекшелігі, оның изотоптық құрамы, қатерлі ісіктің медициналық диагностикасында пайдаланылатын, бөлу каскадының жұмысы кезінде қалдық ретінде алынған молибденнің құрамына сәйкес келеді.

Зерттеу барысында табиғи молибденнің нейтронды-физикалық қасиеттері (JENDL-4.0 ядролық деректер кітапханасы) және металл молибденінің жылулық қасиеттері пайдаланылды.

Келесі нәтижелер алынды:

1. Шар тәрізді микротвэлдарды пайдалану жолымен жүретін жеңіл сулы реакторлар үшін, сонымен қатар, молибденнің әлсіз жұтылатын изотоптарымен байытылған Мо-матрицасында металл U–Mo-түйіршіктерінің құймасынан тұратын, дисперсті отын пайдалану жолымен жүретін жылдам реакторлар үшін де, твэлдардың тұрақты уақытын азайту әдісі ұсынылған.
2. Молибденді әлсіз жұтылатын изотоптармен байытудың қажеттілігі көрсетілген.
3. Молибденнің изотоптық құрамының жалпы қолданылуы 50%-дан жоғары.
4. Мо-нің Pb және Pb-Bi евтетивінде коррозияға жақсы төзімділігі бар екендігін дәлелдейтін жұмыстарға шолу жасалынды.

Молибденнің әлсіз жұтылатын изотоптарымен байытылған Мо-матрицасында металл U–Mo-түйіршіктерінің құймасынан тұратын, цилиндр тәрізді отынды білікшелердегі дисперстік отынды пайдалану жолымен жүретін, жылу бөлетін элементтер твэлдарының тұрақты уақытын азайту, жеңіл сулы және жылдам ядролық реакторлардың қауіпсіздігін арттырудың лайықты тәсілі болуы мүмкін.

**Түйін сөздер:** жылдам реакторлардың қауіпсіздігін арттыру, молибденнің физика-механикалық сипаттамалары, жылу бөлетін элементтердің жылулық тұрақтысы, қорғасын мен LBE-дегі коррозияға төзімділігі.

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## ИСПОЛЬЗОВАНИЕ МОЛИБДЕНА В КАЧЕСТВЕ СТРУКТУРНОГО МАТЕРИАЛА ТОПЛИВНЫХ ЭЛЕМЕНТОВ В СВИНЦЕВОМ ИЛИ СВИНЦЕВНОМ-ЭВТЕТИЧЕСКОМ ОХЛАЖДЕННОМ БЫСТРОМ РЕАКТОРЕ ДЛЯ ПОВЫШЕНИЯ ЕГО БЕЗОПАСНОСТИ

**Аннотация.** Основной целью исследования является обоснование использования молибдена в качестве конструкционного материала топливных элементов для повышения безопасности ядерных реакторов. Особенностью используемого молибдена является то, что его изотопный состав соответствует молибдену, который получается в качестве хвоста при работе разделительного каскада для изготовления материала для медицинской диагностики рака.

При проведении исследования использовались нейтронно-физические свойства изотопов природного молибдена (библиотека ядерных данных JENDL-4.0) и теплофизические свойства металлического молибдена.

Были получены следующие результаты:

1. Предложен способ уменьшения постоянной времени твэлов как для легководных реакторов путем использования шаровых микротвэлов, так и для быстрых реакторов путем использования дисперсного топлива, содержащего, например, гранулы металлического U–Mo - сплава в Mo - матрице с обогащением слабопоглощающими изотопами молибдена.
2. Показана необходимость обогащения молибдена слабо поглощающими изотопами.
3. Общее использование изотопного состава молибдена составит более 50%.
4. Выполнен обзор по работам, где доказывается, что Mo имеет хорошую коррозионную стойкость в Pb и евтетике Pb-Bi.

Уменьшение постоянной времени твэлов тепловыделяющих элементов, путем использования дисперсионного топлива в цилиндрических топливных стержнях, содержащих, например, гранулы металлического U-Mo-сплава в Mo-матрице с обогащением слабо поглощающими изотопами молибдена, может оказаться заслуживающим внимание способом повышения безопасности легководных и быстрых ядерных реакторов.

**Ключевые слова:** повышение безопасности быстрых реакторов, физико-механические характеристики молибдена, тепловая постоянная тепловыделяющих элементов, коррозионная стойкость в свинце и LBE.

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