**HEAT-CONDUCTING LIQUID METAL**

**SUBLAYER IN THE FUEL ELEMENT**

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**Abstract**

In the reactor plants, which have inherent safety properties, high-heat conducting nitride uranium-plutonium fuel and lead coolant are used. Nitride fuel is less ductile than oxide one, and the resource of a nitride fuel element with a helium gap is limited primarily by the touch and deformation of the cladding by fuel (and not by its radiation damage). Use of the liquid-metal sublayer in the fuel element due to its thermal conductivity allows to increase the gap thickness and extend the fuel element resource multiple of the gap thickness. At the same time, the temperature in the center of the fuel decreases significantly (by hundreds of degrees), due to a decrease in temperature, fuel swelling decreases, the level of safety during accidents UTOP (uncontrolled increase in power), ULOF (unprotected loss of flow) increases.

However, experimental studies on the BOR-60 showed significant corrosion damage to the ferritic-martensitic cladding of the fuel element due to the transfer of the steel components dissolved in lead from the hot zone to the cold one.

A lead-based sublayer doped with magnesium and zirconium provides spontaneous formation of a protective nitride coating on the steel surface and self-healing of accidental damage of the coating, thus solving thermophysical, corrosive and conjunctural problems of the fuel element.

## INTRODUCTION

It is proposed to use dense heat-conducting nitride fuel and high-boiling, chemically inert and radiation-safe lead coolant in innovative reactor units with inherent safety properties [1–3].

At the initial stage of operation of the BREST-OD-300 experimental demonstration power unit, it is currently planned to use fuel elements with gas filling of the "fuel-cladding" gap to form the fuel element loading. At the same time, it is feasible to use a liquid metal sublayer (LMS) in the fuel elements of fast reactors with inherent safety properties in the future. To do this, a number of technological challenges must be overcome. In particular, it is necessary to develop a technology for the automated manufacturing of fuel elements with an LMS in remote production conditions, as well as improve methods for monitoring the continuity of the sublayer, etc.

The use of an LMS inside the fuel element provides certain advantages over using helium to fill the gap between the fuel and the cladding. First of all, when a heat-conducting LMS is used in the fuel element, the fuel temperature decreases noticeably, which is important during UTOP and ULOF accidents.

In addition, nitride fuel is less ductile than oxide fuel, and the service life of a nitride fuel element with a helium gap is mainly limited by the fuel's mechanical interaction with the cladding (and not by the cladding’s radiation damage). For example, in [4], it has been demonstrated that fuel elements with nitride fuel and a helium gap are serviceable until burnup, which is determined by the onset of the interaction of the fuel with the cladding. However, the subsequent course of events is difficult to predict and is determined by many factors, including the level of stresses in the cladding associated with the ratio of rates of swelling and creep of the fuel and cladding, etc.

The heat-conducting LMS makes it possible to significantly increase the thickness of the gap inside the fuel element, and thereby extend the service life of the fuel element considerably.

For example, according to estimates [5], in a fuel element with a 0,25 mm lead sublayer, when irradiated on a BOR-60 with a maximum linear thermal power of 435 W/cm, the mechanical interaction of the fuel with the cladding occurs when about 9% of heavy atoms are burned out. It is expected that higher burnup values can be achieved in the fuel elements with LMS in the future. In the conceptual work of Adamov E.O. et al., it is indicated that in a fuel element with a gaseous sublayer with burnout of more than 9-10%. thermomechanical interaction of the nitride fuel with the ferrite-martensitic (FM) cladding limiting further use of the fuel element is possible, while the LMS allows to significantly expand the gap between them without increasing the fuel temperature and, accordingly, increase burnout up to 15% or more [6]. Design of fuel element with LMS for tests in BOR-60 with length of 625 mm and height of fuel column of 390 mm with outer cladding diameter of 9.4 mm, wall thickness of 0.5 mm, fuel tablet diameter of 7.8-7.9 mm and justification of its validity are also presented in [6].

Under the guidance of E.A. Orlova, a research technique was developed and doped lead weights were made for the fuel assembly (FA) of the OU-4 presented in [6], including the composition by weight of Mg - 1.15%, Fe - 0.001%, Ni - 0.0015%, Zr - 0.015% [7]

Tests in the BOR-60 of fuel elements with unalloyed lead at burnout of 5.5% h.a. maximum temperature in fuel reached 1148 K [8], 1080 K [6], and fuel temperature in fuel element with helium sublayer is significantly hundreds of degrees higher [6].

Another problem that complicates the use of fuel elements filled with liquid metal in the cores of fast reactors is the need to ensure the corrosion compatibility of the sublayer with the cladding and the fuel.

In the course of the experiments [9], at a burnup of 5.5% in the fuel elements corrosive damage was revealed in the lead of the fuel-element cladding made of ferritic-martensitic steel, which is characterized by its dissolution in the hot zone and the formation of a layer of metal deposits in the cold zone. As a result of the cladding's dissolution, its thickness decreased by more than а third in some areas of the hot zone, according to [9].

The above-mentioned corrosion problem is resolved by alloying lead with magnesium and zirconium. The compatibility of this sublayer with a fuel element cladding made of FM steel in the temperature range of 810–1,130 K for up to 5,650 hours was justified in the works [10–15]. The concentration of magnesium $(C\_{Mg})$ in doped lead was provided in the range from 0.08 to 0.25 atomic fractions, the concentration of zirconium $ (C\_{Zr})$ in atomic fractions was maintained in the range determined from the ratio:

$C\_{Zr}=0.002+0.044C\_{Mg}^{0.7}∙exp⁡(-\frac{2895}{T})$, (1)

where T is the temperature of doped lead, K [11].

Preservation of protective coating of ZrN pre-formed on cladding of ferrite-martensitic grade in shortened model of fuel element with fuel from uranium nitride at temperature 970 K for 5,650 h confirmed in [15].

## CONCERNING THE ADVANTAGES OF USING A LIQUID METAL SUBLAYER IN A FUEL ELEMENT

## As already noted, using an LMS in a fuel leads to, in particular, the significant decrease in fuel temperature, which is important in case of accidents.

In this work a comparison of the temperature distribution over the height of the reactor core with a lead coolant in the fuel element of the central FA with a thermal power of 4,2·104 W/m, with a gap of 300 μm between the fuel and the cladding, filled with gas or lead alloyed with Mg and Zr. The comparison was made according to the method described in [16–17], with parameters described in [18–19].

The thermal conductivity of the doped lead sublayer is close to that of the unalloyed lead. [18–19]. At 810 K the thermal conductivity (λ, W/m·K) of nitride fuel is 13.9, λPb-Mg-Zr alloy = 16.9, λPb =18,1. For metal-like nitrides λZrN=13.8 and λZr =18.4 at 293 K [20].

It has been demonstrated that the temperature at the center of the fuel rod in a fuel element with the above parameters and a helium gap between the fuel and the cladding reaches 1,915 K, while when using the Pb-Mg-Zr alloy as a heat-conducting LMS, the temperature at the center of the fuel rod is equal to 1,280 K.

In recent years, new possibilities for using the liquid metal sublayer in fuel elements have also been considered. Thus, in [21–22], it is proposed to use the advantages of a liquid metal filler inside the fuel-element gap in order to ensure an acceptable reactivity run-out for the interval between refueling (comparable to the effective fraction of delayed neutrons) throughout the operation period of a fast reactor running in a closed fuel cycle in the case of its start-up using fuel with a highly non-equilibrium isotopic composition (for example, enriched uranium, as well as weapon-grade or weakly matured plutonium). For this purpose, it is proposed to vary the thickness of the sublayer inside the fuel element during refueling to control the fuel's reactivity weight during the life cycle of the reactor in the transition mode to fuel of equilibrium composition. In the case of a gas-filled gap inside the fuel element, this method would be inapplicable due to the high dependence of the fuel temperature on the thickness of the low heat-conducting gas sublayer.

## PRINCIPLES OF ensuring THE COMPATIBILITY OF THE LIQUID-METAL SUBLAYER WITH the CLADDING AND FUEL

As was mentioned in the introduction, one of the key limiting factors for the possibility of using LMS in the cores of fast reactors is the issue of corrosion damage cladding of the fuel element in the lead, which mainly occurs due to the dissolution of steel components in the lead in the hot zone and the precipitation of the same components in the cold zone.

The problem of compatibility between the sublayer and the cladding can be resolved by creating a protective coating on the surface of the steel, which serves as a diffusion barrier for the steel components.

To analyze the possibility of creating a protective coating based on nitride ceramics on the surface of steels that come into contact with alloyed lead in a fuel element with uranium nitride fuel, the thermodynamic calculations of the stability of nitrogen compounds were carried out (see Table 1). The standard thermodynamic characteristics of nitrogen compounds were taken from [23–25], and the correlation dependencies of the nitrogen pressure over uranium nitride were taken from [26–28].

It was shown that the ZrN compound is more stable than the UN one since it does not dissociate in the temperature range 600–1,000 K up to the nitrogen pressure in the system: 7·10-54 – 8·10-28 MPa. In this case, explosive lead azide Pb(N3)2 is not formed in either the solid or in the gaseous phase if the nitrogen pressure is less than 3.5·10+09 MPa, and such pressure is orders of magnitude higher than the technically feasible one.

TABLE 1. NITROGEN PRESSURE OF DISSOCIATION OF NITRIDES IN THE 600–1,000 K TEMPERATURE RANGE

|  |  |
| --- | --- |
|  | *p* (N2), MPa  |
| T, K | 2/3U+1/2N2=1/3U2N3 [23] | 3/2Mg+ 1/2N2= 1/2Mg3N2 | U+1/2N2=UN, [22] | U+1/2N2=UN, [24] | U+1/2N2=UN, [25] | Zr+1/2N2=ZrN | 1/6Pb+1/2N2=1/6Pb(N3)2 | 1/6Pb+1/2N2=1/6Pb(N3)2 vapor |
| 600 | 4.4·10-14 | 1·10-34 | 2·10-44 | 3.6·10-45 | 1·10-41 | 7·10-54 | 2.8·10+22 | 3.2·10+14 |
| 700 | 2.8·10-11 | 4·10-29 | 2·10-37 | 1.5·10-37 | 7·10-36 | 1·10-44 | 7.8·10+19 | 5.4·10+12 |
| 800 | 3.4·10-09 | 6·10-25 | 3·10-32 | 7.5·10-32 | 2·10-31 | 1·10-37 | 9.5·10+17 | 2.5·10+11 |
| 900 | 1.5·10-07 | 8·10-22 | 4·10-28 | 2.1·10-27 | 5·10-28 | 4·10-32 | 3.1·10+16 | 2.3·10+10 |
| 1,000 | 3·10-06 | 3·10-19 | 6·10-25 | 7.5·10-24 | 3·10-25 | 8·10-28 | 2.0·10+15 | 3.5·10+09 |

When steels come into contact with lead alloyed with magnesium and zirconium along with alloy saturation with zirconium in the hot zone, a protective ZrN coating is spontaneously created on the surface of steels in a nitrogen atmosphere (which contains a certain amount of ZrC as carbon moves from the steel base), with a low diffusion permeability of the steel components and the liquid metal sublayer through it. This process manifests itself most clearly at elevated temperatures; therefore, for the purpose of building a model of a protective coating creating, experimental data obtained at a maximum research temperature of 1,130 K was selected. This model (see Figure 1) was built on the basis of the results of a study of ferritic-martensitic grade steel after being in contact with lead alloyed with magnesium and zirconium for a time of 3,000 h using a Laser Atomic Emission Spectrometer (LAES), published in [10].

A vacancy curve (5) was plotted according to the difference in the iron content in the steel base and in the iron-depleted layer as a result of the release of iron into the alloyed lead (curve 6). As shown in the figure, the nature of the distribution of zirconium together with lead in the steel (curves 2 and 3) coincides with the nature of the distribution of vacancies. It can be concluded that zirconium and lead penetrate through vacancies into the steel, thus eliminating the defect and contributing to the preservation of the iron component in the steel. This is facilitated to an even greater extent by the penetration of nitrogen into the steel and the creation of lead-zirconium nitride ceramics. According to data from roentgen microanalysis, data obtained by using a scanning electron microscope and nuclear physics/LAES research methods, the coating thickness in the temperature range of 810–1,130 K does not exceed 25 μm for zirconium and 4 μm for a compound of zirconium with nitrogen. This indicates a low diffusion mobility and the permeability of the coating components in steel and, accordingly, the absence of the need to control and adjust the concentration of the coating components in the alloy.

Cr (curve 4) segregates from the steel base to its surface, which is typical for a liquid metal containing a large amount of a component that forms a stable bond with chromium.

The change in the content of elements in ferritic-martensitic grade steel along the depth of the sample after testing in the expansion tank of experimental bench test is best described by the logarithmic curve below:

Fe *I* = 0.277 ln *l* – 0.614; *R*2 = 0.850

Cr *I* = 0,519 ln *l* + 1.683; *R*2= 0.832

Zr *I* = – 6.176 ln *l* + 20.33; *R*2= 0.921

Pb *I* = – 2.197 ln *l* + 7.022; *R*2= 0.861­­

N *I* = – 3.517 ln *l* + 5.814; *R*2= 0.413

Vacancies *I* = – 0.277 ln *l* + 0.614; *R*2= 0.850,

where *I* is the difference in intensity of the spectral lines of elements near the surface of the steel and in the base of the steel, divided by the intensity in the base (for Zr, Pb, and N, the background level is taken as the base), expressed in relative units, *l* is the element detection depth in steel, *R*2 is the value of the approximation accuracy.

The logarithmic dependence of the intensity of spectral lines of elements on the depth of their penetration into steel demonstrates the inversely proportional dependence of the derivative of the intensity d*I*/d*l* on the depth of penetration of the element into steel.



*FIG. 1. Distribution of elements along the depth of ferrite-martensitic steel 16Cr12WMoSiVNbB after tests in the expansion tank of the experimental section simulating the gap in the fuel element at temperatures up to 1,130 K for a duration of 3,000 h. The analysis was carried out using LAES in the 15 J laser pumping mode with beam defocusing.*

Zirconium is introduced into steel under the influence of a chemical potential caused by a concentration gradient, nitrogen creates with zirconium the most stable compound in the system under consideration a chemical reaction with the formation of ZrN. In pre-reactor studies, nitrogen was present as a gas cushion, in the fuel element

it is formed as a result of neutron irradiation of the fuel.

An overview of publications on the study of corrosion in alloys based on Pb is given in [29], including an analysis of [30]. In this report, the following mechanism of Zr penetration into steel is recommended for study, which largely coincides with the one proposed by the authors of this report. According to [30], activated Zr was introduced into the Bi — 0.1% Zr — 0.1 % Mg alloy, and the interaction between the zirconium and the steel wall was monitored using a radioactive tracer. The following elementary stages in the chain of reactions between the Bi — 0.1% Zr — 0.1% Mg alloy and low-alloy steels are highlighted:

1 – adsorption of Zr on the sample surface;

2 – chemical interaction of the absorbed Zr and basic metal, formation of the surface layer in the form of FexZry membrane or the FexZry(C, N)z intermetallic compound stabilized with carbon and nitrogen;

3 – formation of a protective layer of ZrC carbides or ZrN nitrides;

4 – exfoliation of ZrC and ZrN layers and their subsequent regeneration.

In the experiments conducted by the authors of this report, corrosion was virtually not detected, as determined by the decrease in the mass of steel samples, which indicates the replacement of iron with zirconium and lead. Moreover, no delamination of the zirconium-rich layer was observed. On the contrary, this layer adhered firmly to the base and did not have scratches on it, even under mechanical stress. In this case, nitrogen was created from the gas phase above the alloyed lead melt.

The authors of this report used X-ray diffraction analysis and did not find intermetallic compounds of iron with zirconium. However, reflections from crystallographic planes (intensity peaks) characteristic of ZrN (ZrC) compounds were detected repeatedly, which allows us to recognize as justified the vacancy method for introducing Zr into steel as proposed by the authors of this report. This method introduces Zr under the action of the chemical potential due to the concentration gradient of Zr, i. e. saturating the liquid metal sublayer alloy with zirconium during testing and the absence of zirconium in the steel base.

The area of iron depletion of ferritic-martensitic steel at temperatures up to 1130 K for 3,000 h was about 15 μm. When recalculated in the parabolic approximation for a fuel element service life of 35,000 hours, the depth of the region of iron depletion of ferritic-martensitic steel at the same temperature is equal to 50 μm.

Considering that the surface temperature of steel in reactors with a lead coolant does not exceed 920 K and the processes of diffusion of steel components and their solubility in a lead-based alloy are described by the exponential Arrhenius dependence, a conclusion can be made about the corrosion resistance of ferritic-martensitic steel upon contact with lead alloyed with zirconium and magnesium.

In 2014-year, FA OU-4 with 4 versions of LMS (in the state of supply, purified from oxygen, purified from oxygen and doped with steel components, purified from oxygen and doped with magnesium and zirconium) was installed in BOR-60 [6]. Comparative analysis of test results is the subject of the following article.

## FINDINGS

This report analyzes the advantages of using lead liquid metal sublayer in the fuel rods in fast reactor cores and substantiates a method for ensuring the compatibility of the sublayer with the cladding and fuel by alloying the lead to fill the fuel-rod gap with magnesium and zirconium. In this case, a stable zirconium-nitride-based coating in conjunction with lead is created on the surface of the steel, which promotes the self-healing of point defects arising in the steel due to the dissolution of iron.

The authors have developed an approach to describing the process of mutual mass transfer between the steel and the liquid metal components during the creation of a protective nitride coating on the cladding surface, consistent with the results of the experiments. It can be concluded that, in the presence of magnesium - and zirconium -alloying components in the lead filling the fuel-rod gap, the problem of compatibility between the sublayer, the cladding and the fuel is generally resolved, and there is no corrosion of the cladding steel.

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