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Self-organizing carbon nitride coatings on steel from molten lead–magnesium eutectic

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Abstract

Based on the results of calculation and experimental studies zirconium saturated eutectic Pb–Mg alloy is recommended as liquid metal coolant in fuel elements loaded with nitride fuel. Test stand was developed and manufactured for carrying out studies of deposition of nitride and/or carbide protective coatings from molten eutectic Pb–Mg within narrow gap between coaxially arranged tubes. Pilot testing has been performed.

Keywords: Lead; Magnesium; Zirconium nitride; Liquid-metal sublayer; Corrosion protection.

Introduction

Lead-cooled fast reactor loaded with nitride fuel is designed based on the principles of "inherent safety" and cost effectiveness [1]. At present fuel elements with nitride fuel and liquid metal sublayer allowing reducing temperature of nitride fuel to the levels ensuring reliable fuel operation at high burnup with suppressed swelling and release of gases are regarded as promising fuel element for BREST-OD-300 fast reactor units. Results of studies of fuel element with gap (initial gap size is equal to 250 μm) filled with He and Pb after irradiation in BOR-60 reactor are presented in [2], where calculated temperature at the center of fuel element with gas sublayer can reach 2033 K (60% Pu); that on the fuel element cladding is equal to 860 K. Gap between fuel and cladding (≈10 μm) in cold state remained only in the upper and lower sections of the fuel element loaded with 45% PuN + 55% UN fuel with burnup equal to 5.4% of heavy nuclei. The average rate of increase of diameter of fuel pellets observed for the section with maximum increase of fuel element diameter amounts to 1.1–1.2 ± 0.1 per 1% burnup for $U_{0.55}Pu_{0.45}N$ and $U_{0.4}Pu_{0.6}N$ fuel, respectively. Gas-filled pores and impurities were observed.

Contact between fuel and cladding in fuels element takes place when local burnup reaches about 5% of heavy nuclei. After the contact average values of hoop stresses in the cladding calculated according to the contribution of radiation-induced creeping in the increase of their diameters, are equal to 100 and 220 MPa, respectively.

At the same time in the presence of inside TVEL of a lead sublayer the temperature of the center of fuel does not exceed 1153 K, and on an internal surface of a cover – 908 K. There were no gas pores and allocations. However there is a problem of compatibility of a liquid metal inside sublayer with a cover of TVEL in the presence of nitride fuel. Results of research of fuel elements (Fig. 1) after radiation showed the progressing interaction of steel cover with a Pb sublayer which is characterized by its dissolution in one parts and formation of a layer of metal deposits in other parts. Owing to dissolution of a cover its thickness on separate sites decreased approximately by 40% (at burning out of 5.5% of heavy nuclei, 23 dpa) [2].

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Table 1
Element composition (in atomic %) of the surface and cross-cut microsection (for points 1–4 in Fig. 2) of 16Cr12WMoSiVNbB steel (INCA) with ZrN coating formed at 993 K during 50 h in Pb–Mg–Zr alloy under nitrogen pressure.

Element	Surface of steel		Surface of cross-cut microsection			
	Mean value averaged over 3 points	Standard deviation	1	2	3	4
Fe	54.6	6.4	78.9	77.4	79.9	85.2
Cr	9.1	1.1	10.3	10.3	9.7	85.2
V	0.4	0.1	0.2	0.4	0.3	0.2
PB	0.0	0.00	0.0	0.0	0.0	0.0
Mo	0.3	0.04	0.3	0.2	0.2	0.3
Zr	6.4	0.8	3.3	1.5	2.4	0.2
Si	1.3	0.2	1.3	1.5	1.2	1.5
Mg	0.7	0.06	0.1	0.2	0.2	0.0
O	10.8	1.3	4.4	7.5	5.3	2.0
N	15.8	2.1	0.0	0.0	0.0	0.0
Ni	0.5	0.06	0.6	0.9	0.7	0.8

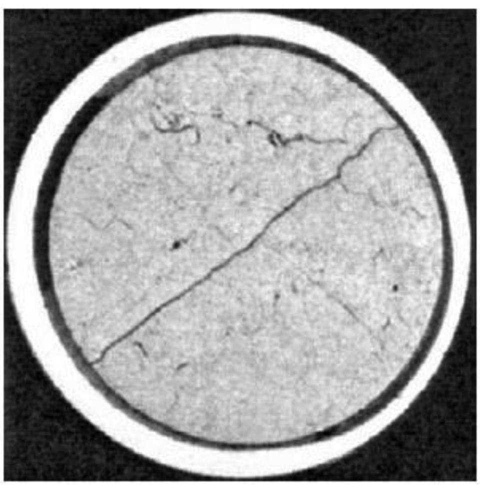

Fig. 1. General view of cross-section of fuel element with lead sublayer [2].

The solution of the problem of compatibility of a cover of TVEL with nitride fuel and a sublayer Pb–Mg is proposed in this paper.

Compatibility of lead–magnesium eutectic with steel

Ampoule and loop testing of samples, as well as testing short experimental models of fuel element with uranium nitride core were performed in order to investigate corrosion of steel placed in lead-based alloy. Pb saturated with zirconium and lead alloys with magnesium rectified from impurities and saturated with zirconium were suggested as the testing alloys. Use of the alloys resulted in the reduction of corrosive interaction with structural materials, as compared with purified lead [3].

The best solution of the problem of liquid-metal sublayer compatibility with fuel element cladding in the presence of nitride fuel is ensured by spontaneous formation on the surface of steel of protective layer of zirconium nitride from lead-based eutectic with concentration of magnesium equal to 2.25% by mass (0.164 atomic fraction) and with up to 0.2% by mass of nitrogen in the presence of nitrogen within the gas-filled cavity. This ensures self-healing of accidental damages of the coating. Corrosion damages and changes of mass of samples of 16Cr12WMoSiVNbB and Cr13Mo2Si2 steels were not detected during ampoule and loop testing of these samples conducted at temperatures equal to 813–1023 K in eutectic consisting of 2.25% Mg and 0.2% Zr with remaining being Pb, as well as after testing at 973 K during 5700 h of short testing model of fuel element with uranium nitride core with heat transferring sublayer having the above indicated composition and with cladding made of 16Cr12WMoSiVNbB steel with already formed zirconium nitride coating [4].

Ampoule testing steels in lead–magnesium eutectic at temperatures equal to 873–1023 K

Formation of protective zirconium nitride coating on the surface of steel was achieved in lead-based eutectic with magnesium concentration equal to 2.25% by mass and up to 0.2% by mass of zirconium within the temperature interval of 813–1023 K during up to 500 h. Analysis of variation of mass, X-ray diffraction phase and element analysis of steel samples were implemented to demonstrate formation of the protective coating.

Composition and structure of the coating formed on the surface of 16Cr12WMoSiVNbB steel at 973 K during 50 h in the alloy consisting of 2.25% Mg, 0.2% Zr with remaining being Pb under N_2 pressure were investigated by X-ray diffusion analysis using DRON-1 facility (structural characteristic typical for ZrN and ZrC was identified) on the electron-scan microscope (REM) with application of wave X-ray spectrum microanalysis INCA, as well as using nuclear microanalysis methods [3,4]. Results of studies performed using REM are represented in Table 1. Zirconium (6.7 ± 0.2 atomic %) and nitrogen (15.6 ± 0.2 atomic %) were found on the sample surface. In the points marked with asterisks near the surface of the cross-cut microsection in Fig. 2 zirconium was found in significantly lower quantities (0.2–3.3 atomic %), while nitrogen was not found at all [4]. According to the data of the much finer nuclear microanalysis nitrogen was found within small depth (depth of nitrogen penetration amounted to