A METHOD TO DETERMINE A START OF POROSITY AND DEFECT FORMATION IN THE FUEL ELEMENTS WITH U–Mo FUEL

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ABSTRACT

Fuel elements (FE) with dispersed U-Mo fuel and Al matrix were tested in the research reactor IVV–2M. At a certain value of fuel burn up gas pores formed and united with test time leading to a formation of defects as extended gas cavities and ruptures in a fuel meat and pillowing of FE claddings. The moment the pores start to form and unite with the ruptures in the fuel meat is very difficult to fix while measuring their sizes even in plate FE. Since this formation is very important for the analysis of U–Mo fuel behavior under different operation conditions and for working out the measures to extend the operation life of the FE with U-Mo fuel relevant to high values of burn up. A gas pores form changing with burn up and changes of a thickness of an $(U,Mo)Al_x$ layer near the pores were analyzed and the method to determine the moment of the gas pore formation and ruptures of the fuel meat was suggested.

1. Introduction

The Reduced Enrichment for Research and Tests Reactors program (RERTR) is being implemented in several countries. In the experiments for the RERTR program the dispersed U-Mo fuel revealed a formation of gas pores, their coalescence and a formation of gas micro-cavities and a rupture of a fuel meat that lead to swelling of fuel element claddings [1-4].

The experiments showed that the higher the fuel test parameters were (mainly, a temperature and a fuel fission rate), the lower the burn up values were for the defects to form in the dispersed U-Mo fuel. In the experiments IRIS-1 where the test parameters were not high, i.e. a thermal flux was ~140 W/cm^2, a fission rate was ~$2.4\cdot10^{14}$ fis./(cm^3·s), a maximum temperature of fuel cladding was ~75°C and pillowing was observed at ~67% burn up. In IRIS-2 the parameters were higher, a thermal flux was ~240 W/cm^2, a fission rate was ~$4.8\cdot10^{14}$ fis./(cm^3·s), a maximum temperature of fuel claddings was ~100°C and pillowing was observed at ~40 % burn up. In the experiments FUTURE pillowing was detected at ~33% burn up at a thermal flux of ~340 Wcm^{-2}, a fission rate of ~$6.5\cdot10^{14}$ fis./(cm^3·s) and a fuel cladding temperature of ~130 °C. In the UMUS tests, where a thermal flux was ~250 W/cm^2, a fission rate was ~$4.0\cdot10^{14}$ fis./(cm^3·s) and a fuel cladding temperature was ~110 °C, the fuel elements of 35% enrichment damaged at 20% burn up.

At the IVV-2M reactor two full-size fuel elements of low enrichment were incorporated into standard fuel element assemblies designated FEA # KM003 and FEA # KM004 and tested within the frames of the Russian RERTR program. For the tests the fuel elements were fabricated by extrusion and contained the dispersed fuel composition (U-9%Mo)/Al of 36 % enrichment ^{235}U with a uranium density of 5.4 g/cm^3. The size fraction of the atomized fuel particles was 63÷160 μm. The fuel claddings were made of the aluminum alloy SAV-1 and AMg-2.

The irradiation tests of the first series were performed on KM003 to attain a fission fragment quantity to be equivalent to an average fuel element burn up of ~40 % for the fuel of 19.7 % enrichment [3]. In these tests a maximum thermal flux was ~80 W/cm^2, a cladding's surface

temperature was not more than 80 °C and a maximum fission rate was $4.2 \cdot 10^{14}$ fis./($cm^3 \cdot s$). After the exposure during 107 effective days the fuel elements kept the same shape, their volume increased by ~1.6 % and a change in a "turn-key" size was not more than 0.4 %. A detected interaction of fuel particles with an aluminum matrix was insufficient, the interaction area was observed in the central parts of the fuel elements and its maximum width was ~4 μm. In the fuel meat there were no pores and gas cavities caused by irradiation.

The tests of the second series were performed on FEA # KM004 to reach an average equivalent burn up of ~80 %. The mean power of this FEA # KM004 was 30 % higher than that of FEA # KM003. In that test a maximum thermal flux was 120 W/cm^2, a cladding's surface temperature was 90°C and a maximum fission rate was $5.7 \cdot 10^{14}$ fis./($cm^3 \cdot s$). Both fuel elements in KM004 revealed gas pores in the fuel meat, an accumulation of gas pores and fuel meat ruptures as well as one bulging in a central part of each fuel element.

In the irradiation tests the integrity of the cladding of fuel elements is monitored, they are inspected visually and their size is measured. The irradiation tests are followed by destructive post-irradiation investigations and because of their complexity and high cost, they are carried out until a maximum burn up is reached. Therefore the destructive test results reflect, in many cases, a summarized state of the fuel at the end of the test and do not describe the evolution of fuel behavior under these conditions. However, such information is necessary for predicting a fuel behavior in different operation conditions and for working out the measures for increasing the burn up.

The full size fuel elements show a variation in length within certain limits of fuel irradiation parameters such as temperature, fission rate and burn up. This peculiarity permits to trace the behavior of U-Mo fuel under different irradiation conditions while investigating different areas of the fuel elements. Changes in the shape of the forming gas pores and in the $(U,Mo)Al_x$ layer thickness near these pores with burn up are analyzed and the method to determine the moment of gas pores formation and fuel meat ruptures is suggested in this paper.

2. Results and Discussions

The post-irradiation investigations of the fuel elements with U-Mo fuel as incorporated into the full size FEA # KM003 and FEA # KM 004 [3, 4] and tested in the IVV-2M reactor showed that the $(U,Mo)Al_x$ layer was formed around the U-9%Mo fuel particles; the thickness of the layer was changing along the fuel element length (Fig. 1).

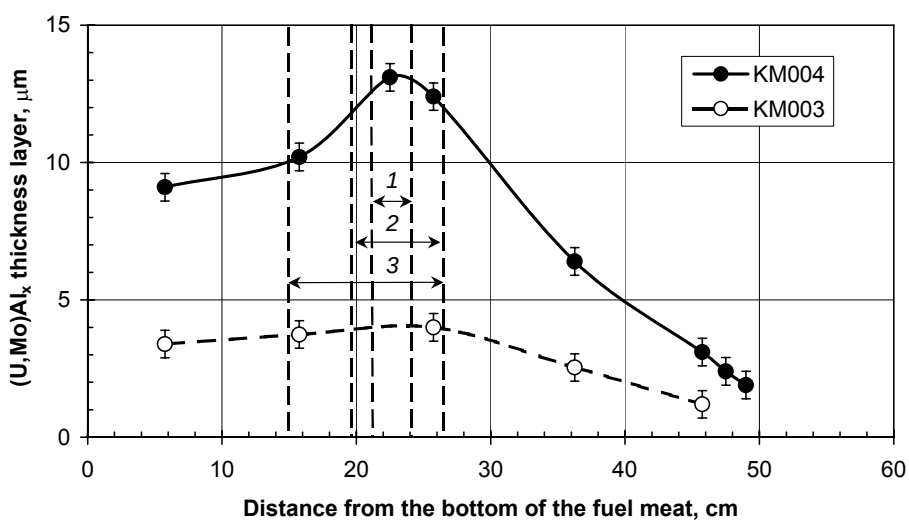


Fig. 1. Changes in the $(U,Mo)Al_x$ thickness layer along the fuel element length

The changes in the thickness depend on the main test parameters and are described by the expression:

$$\delta = 1.03\cdot\tau^{1.09}\cdot(\varphi_{BOL}/\varphi_o)^{0.96}\cdot\exp(-5665/RT_{BOL});\ R^2=0{,}99;\ n=13, \quad (1)$$

where δ is the $(U,Mo)Al_x$ thickness layer (μm); τ is the time (h); φ_{BOL} is the fission rate in the UMo particles (f/(cm^3·s)) at the beginning of life; $\varphi_O = 1\cdot10^{14}$ f/(cm^3·s); T_{BOL} is the temperature of fuel claddings (K) at beginning of life.

According to (1) the dependence of the $(U,Mo)Al_x$ thickness layer on the test time and the fuel fission rate at beginning of test is practically linear and it was stable during all the test [5].

In addition to the formation of the $(U,Mo)Al_x$ layer the gas pores (Fig. 2a) and their accumulations together with the extended gas cavities (Fig. 2b) are formed in the central areas of the fuel elements of FEA # KM004. Finally, they result in ruptures of the fuel meat (Fig. 2c) and swelling of the fuel claddings [5].

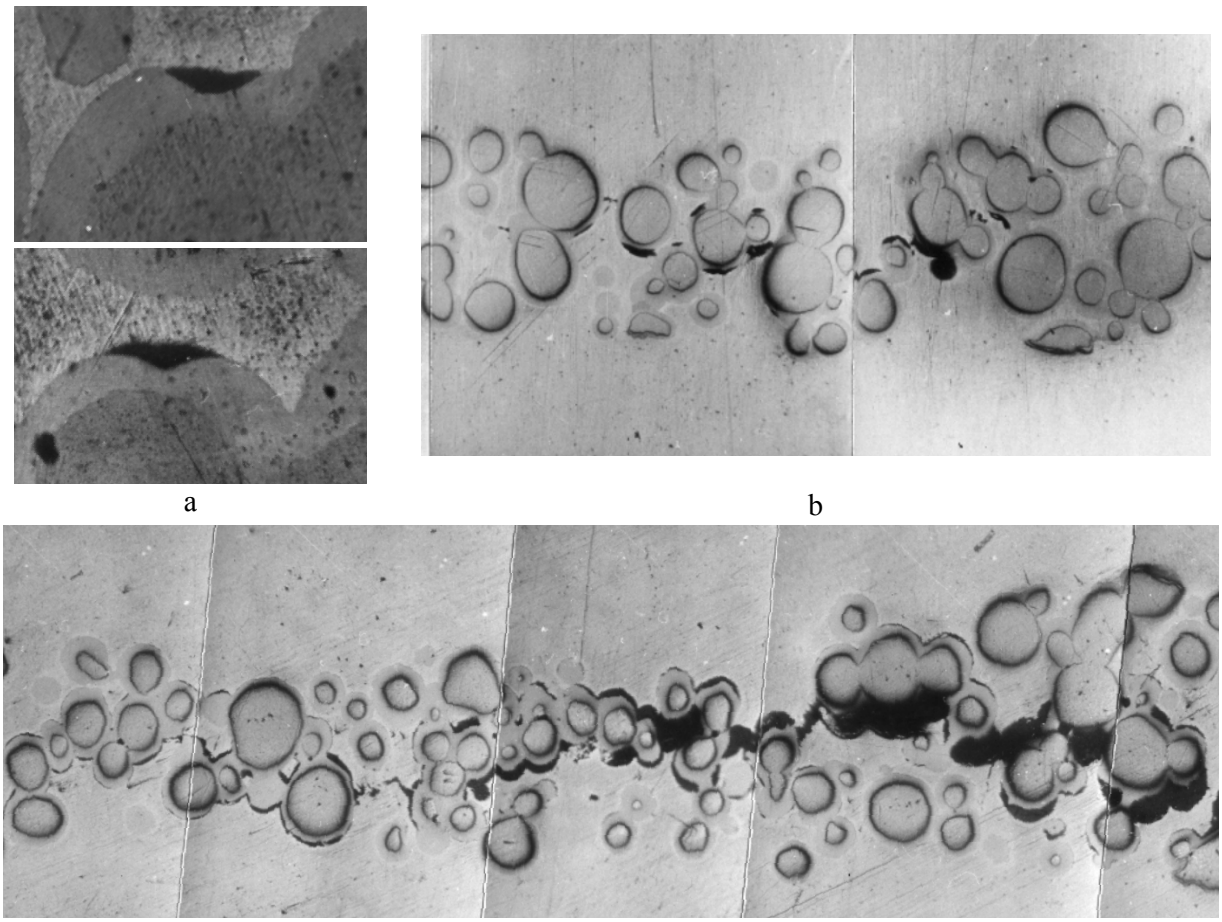


Fig. 2. Individual gas pores (2a) and extended gas cavities (2b, 2c) with a micro rupture of fuel core (2c)

Individual gas pores are formed in the area designated as *"1"* in Fig. 1; the accumulations of the gas pores with 2-mm ruptures in the fuel meat are in area *"2"* and swelling of the claddings is revealed in area *"3"*.

The individual gas pores, their accumulations and ruptures occur mainly at the phase boundary $(U,Mo)Al_x$ layer/Al matrix.

A peculiar feature of the individual gas pores is that $(U,Mo)Al_x$ layer is narrower in the location of the gas pores (Fig. 2a).

This phenomenon is of interest because it allows to estimate a moment in time corresponding to the pores initiation, i. e. when the continuity between the layer and the matrix is disturbed. This can be estimated from the ratio of $(U,Mo)Al_x$ thickness layer, δ_i, where central areas of gas pores are located, to all the interaction layer thickness that is near the pores, δ_{max}, (Fig. 3).

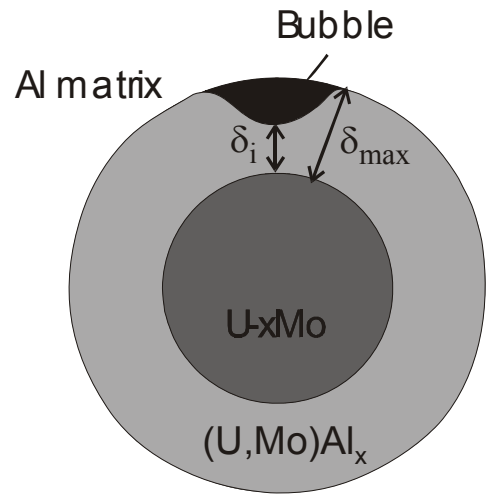


Fig. 3. A schematic of fuel particle with a gas bubble to determine δ_i/δ_{max} ratio

This is based on the following. First, it is supposed that, if there is a discontinuity between the $(U,Mo)Al_x$ layer and the matrix (where pores are initiated) a growth of the interaction product $(U,Mo)Al_x$ is practically stopped and, as a result, a diffusion path of reacting components to these areas of the layer becomes longer. Second, according to (1) the thickness δ of the $(U,Mo)Al_x$ layer depends on time to the $\mathbf{n}^{th}$ - power, that permits to write the ratio δ_i to δ_{max} as

$$\delta_i / \delta_{\max} \approx (\tau_i / \tau_{\max})^n \qquad (2)$$

where τ_i is the moment of pore initiation and τ_{max} is the total test duration.

Expression (2) is true if all the factors affecting the speed of growth of the interaction product $(U,Mo,)Al_x$, such as temperature, fuel fission rate, phase content, structure of reacting components and etc., are equal in time.

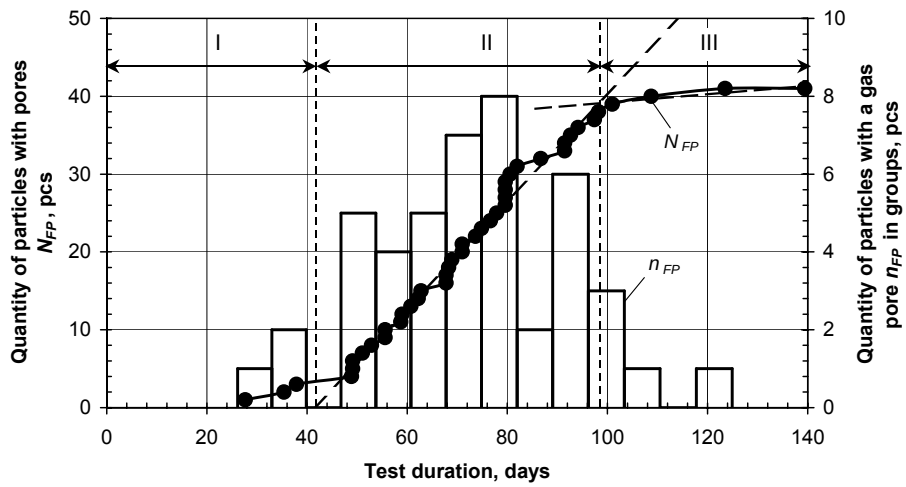
The time corresponding to the pore initiation moment can be obtained from expression (2)

$$\tau_i = \tau_{\max} \cdot (\delta_i / \delta_{\max})^{1/n} = \tau_{\max} \cdot (\delta_i / \delta_{\max})^{0.92}. \qquad (3)$$

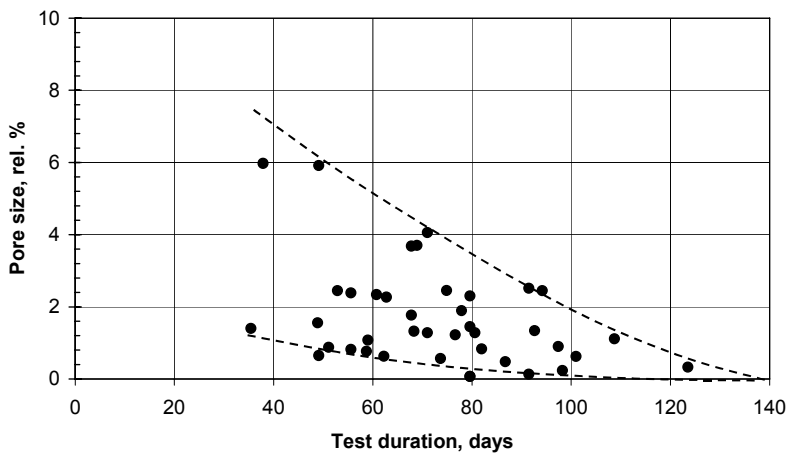
Forty one fuel particles with individual gas pores were measured and the obtained data are presented in Fig. 4.a as a plot of an increasing total amount N_{FP} of the fuel particles with the gas pores and as a diagram of the quantity of the pores n_{FP} forming within the time intervals, which are equal to 5% of the total duration of the test.

It should be noted that only the fuel particles with individual gas pores were analyzed. The particle with pores located near the extended gas cavities in the swelling area were not considered because the $(U,Mo)Al_x$ thickness layer, the shape and size of the pores themselves were changing there due to deformation processes.

According to the data in Fig. 4.a the test duration consists of three intervals, they are designated as I, II and III. The initial one, without gas pores, lasts from 0 to ~40 days; it can be an incubation period when gas fission fragments are accumulating in the aluminum matrix and conditions for pore nucleation are forming.



a



b

Fig. 4. Changes of gas pores quantity (4a) and size (4b) vs test duration

The second interval lasts from ~40 to ~100 days when new pores are forming intensively and the existing pores are growing in size. The latter is proved by the data in Fig. 4b, where the pores formed at the early phases of the second interval have, as a rule, larger sizes. In addition, a gas pressure in the pores, probably, also changes because the pores absorb the xenon and krypton atoms, both the newly formed ones (as a result of fuel fission), and those located in the matrix areas adjacent to the pores.

The third interval lasts more than ~105 days when a dramatic retardation of a new pores formation is observed through an increasing slope of the N_{FP} curve and an increasing number of the pores n_{FP} forming within the intervals equal to 5% of total duration of the test (Fig. 4.a). That is, probably, due to the occurrence of the processes related to a coalescence of the pores and a rupture in the fuel meat. These processes lead to the formation of the extended gas pores and the involvement of the fuel particles which have the highest probability of formation both nuclei and pores.

It should be noted that the transition boundary from the second phase to the third one practically coincides with the scam med rods in the reactor (Fig. 5) caused by the temporary stop of coolant circulation. An abrupt decrease of the reactor power and a short disturbance of heat removal from the fuel elements can cause thermal stresses in the fuel meat and an increase of a gas pressure in pores and provoke a formation of ruptures in fuel meat along with the extended gas cavities and swelling of fuel element claddings.

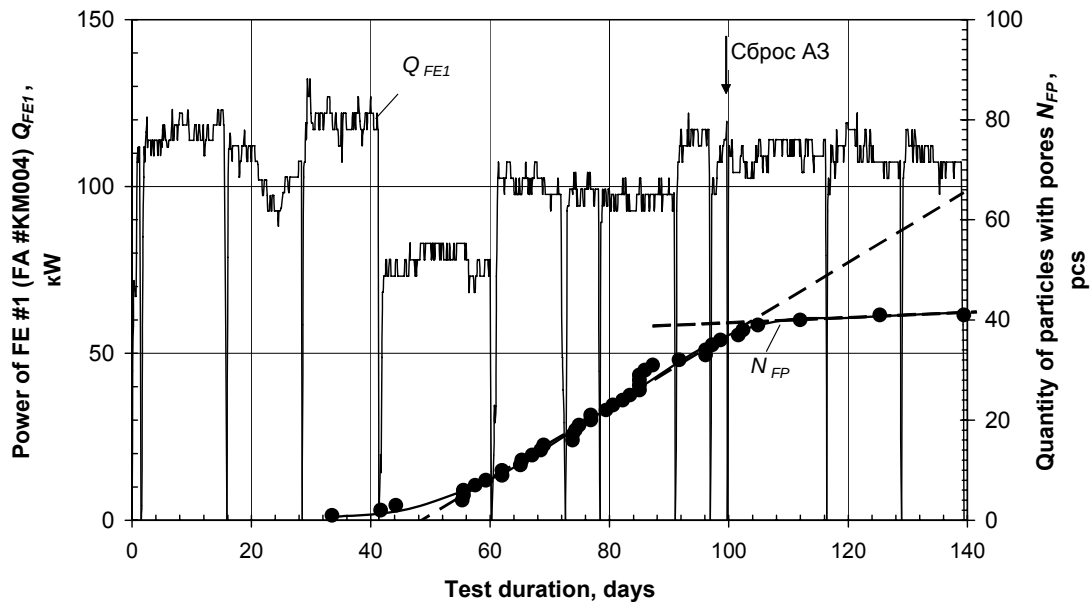


Fig. 5. Changes of gas pores quantity vs test duration imposed on the plot of fuel element power

The plot of changing gas pore quantity with fuel burn up is given in Fig.6. There is also a plot of the changing thickness of the $(U,Mo)Al_x$ layer obtained from dependence (1) for temperature values and fission rates corresponding to the fuel element areas under study.

The data show that the incubation period of ^{235}U burn up, which is necessary for an accumulation of sufficient gas fission fragments in the Al-matrix and providing conditions for a pore nucleation is equal to 30 %. In this period the $(U,Mo)Al_x$ layer of the thickness ~3.5 μm is formed.
When burn up is within ~30 to ~70 % new pores are forming and the size of the existing pores is increasing; the existing pores are accumulating a critical gas pressure sufficient for a rupture to occur in the fuel meat matrix. In this test period the thickness of the $(U,Mo)Al_x$ layer increases from ~3.5 to ~9 μm.

It is mentioned above that the extended gas pore formation along with the rupture of the fuel meat is possible in the area designated as III. The burn-up corresponding to the beginning of this area is ~70 %. The thickness of the layer within this test interval increases from ~9 to ~12.2 μm.

These data are in good agreement with the test results obtained in the program IRIS-1 [1] of plate fuel element with U-Mo fuel of 19.75% enrichment ^{235}U and a uranium density of 8.0 g/cm^3. The thickness of the plates was controlled after each cycle of irradiation tests. In IRIS-1 the tests parameters were close to those of FEA # KM004, i.e. a thermal flux was ~140 W/cm^2, a fission rate was ~$2.4 \cdot 10^{14}$ fis./(cm^3·s), a maximum temperature of fuel claddings was ~75°C and swelling occurred at ~70% burn up.

The boundary values of the $(U,Mo)Al_x$ layer thickness relevant to the fuel meat transition from one state into another, permit to make a diagram of the stability areas, and initiation of porosity an swelling in the dispersed U-Mo fuel (Fig. 6).

Thus, the method suggested for the analysis of the metallographic data on the fuel particles studied in this work permits a more precise estimation of the initial conditions of porosity and swelling formation in the dispersed U-Mo fuel.

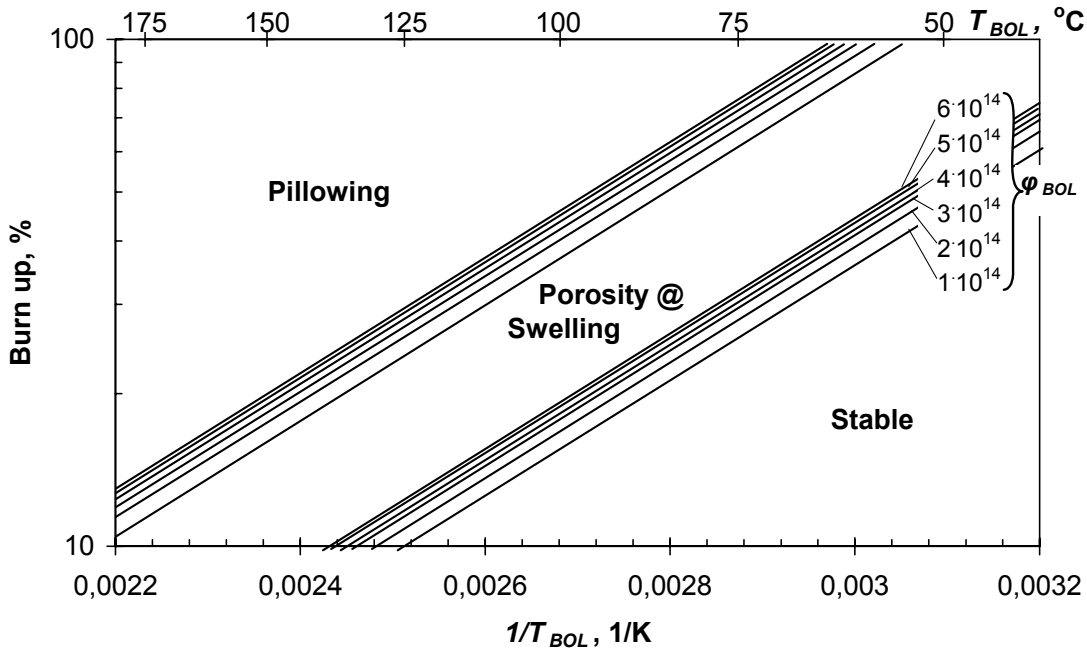


Fig. 6. Influence of operation parameters of fuel elements with dispersed U-Mo fuel on formation conditions of areas with stable and porous structure as well as on pillowing formation conditions

3. Conclusions

The formation of gas pores and the changing of their shape with burn up were analyzed. These gas pores were formed in the fuel meat of the tubular fuel elements of hexagon shape, which were tested in the IVV-2M reactor.

The fuel elements contained the dispersed uranium-molybdenum fuel (U-9% alloy) of 36.6 % enrichment ^{235}U and of a uranium density of 5.4 g/cm^3. During the tests maximum thermal flux was 120 W/cm^2, the temperature of the fuel elements claddings was not more than 90 °C, the maximum fission rate was $5.7\cdot10^{14}$ fis./(cm^3·s). The method to determine the start of gas pore formation at the phase boundary (U,Mo)Al_x/Al-matrix was suggested; the method was based on the determination of the ratio of the (U, Mo)Al_x thickness values in the center and at the edges of gas pores and allowed to build a dependence of f changing pore quantity on the test duration and fuel burn up.

Three areas were distinguished on the curve of the changing gas pores quantity vs the test duration and the burn up that corresponded to different conditions of the fuel meat. It was shown that at the initial (incubation) period which lasted till 30 % burn up ^{235}U was attained, there were processes associated with the solid solution accumulation of gas fission fragments in the Al matrix and a formation of the conditions necessary for a pore nuclei formation. The (U,Mo)Al_x thickness layer of 3.5 µm was formed at that test interval.

At the next stage when the fuel burn up was from ~30 to ~70 % ^{235}U the individual gas pores were formed and they were intensively growing with the gas accumulation inside the pores until the pressure sufficient for fuel rupture was reached. In this test interval the thickness of the (U,Mo)Al_x layer increased from ~3.5 to 9 µm.

At the final stage of the tests when the fuel burn up exceeded 70 % ^{235}U the processes related to the coalescence of individual gas pores, the formation of extended gas cavities, the ruptures of the fuel meat and swelling of the fuel elements claddings started.

4. References

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