ANALYSIS OF FACTORS INFLUENCING FORMATION OF PORES AND PILLOWS IN UMO FUEL

A.M. Savchenko, A.V. Vatulin, I.V. Dobrikova, G.V. Kulakov, S.A. Ershov, Y.V. Konovalov

A.A. Bochvar All-Russia Research Institute of Inorganic Materials (VNIINM)
123060, P.O.B. 369, Rogova St., 5A, Moscow, Russia, sav@bochvar.ru

ABSTRACT

Consideration is given to stages of irradiation induced UMo fuel-aluminium matrix interaction resulting in pores and pillows. They involve fission fragments releases out of fuel into matrix to form a damage rim, growth of interaction layer in the matrix direction with fission fragments displacement to interaction layer – matrix boundary, fission products concentration at interaction layer – matrix boundary, formation of "shears" on interaction layer, fission gas products piling up in voids, pressure increases, formation of pillows. Interaction mechanism at each stage and factors influencing interaction are discussed in detail.

The analyses of the results in pile test investigations take us to the conclusion that additional factor, influencing interaction is technologic factor, i. e. the local pileups of fuel particles. Irradiation produced pores and discontinuities are most often encountered either between UMo fuel particles or within their pileups. In those regions rims of fission fragment effected damages overlap and the value of the local burnup in those regions related not to an individual particle but to the microregion of a fuel component is much higher. Estimation of pileups influence for various types of fuel elements is made and methods reducing the non-uniformity of fuel in local regions are suggested.

The expense of thermal spikes induced by fission products leads to formation of an intricate structure of $(U,Mo)Al_7$ type with excess of Al. To our view it is a finely dispersed mixture of phases consisting of UAl_3 phase, pure Al and a small amount of UMo_2Al_{20}. It has been shown that depending on irradiation conditions such a structure might transform to a thermodynamically stable structure of $(U,Mo)Al_3$ type. Experimentally acquired data are presented, that pertain to formation of identical type structures with excess Al in binary U-Al system without Mo. Under out of pile conditions structures were obtained that simulate those of intermetallic compounds having Al microinterlayers.

1. Introduction

Currently the interaction between U-Mo fuel and Al is the basic factor that limits the serviceability of fuel elements. The interaction results in extra swelling of fuels, disappearance of a heat conducting matrix, a temperature rise in the fuel element centre and penetration porosity and so on. Intermetallic compounds formed via interaction under irradiation have not been produced so far under out-of-pile conditions, which makes studies of those processes difficult. Due to interaction UMo fuel has limitations under some conditions, which requires searching for new solutions in this direction.

It might be gathered from examining the dynamics of the UMo fuel - aluminium interaction under irradiation that after some time the stage of relatively uniform fuel swelling is followed by the commencing stage of accelerated swelling. It is attended with matrix disappearing, through pores and so-called pillows formation, which results in a fuel element fracture [1-3]. At this stage the largest part is played by the fission gas product (FGP) behaviour. The fuel elements serviceability is determined by the onset of this stage.

At each stage the interaction processes have specific features and distinctive characteristics and effect the fuel element serviceability in different ways. The duration of each stage depends on irradiation conditions. Some factors that provide for the interaction layer growth, e.g., irradiation temperature, when specifically combined with other irradiation characteristics (time, burnup, heat flux, fission rate) as well as with technologic and structural features may prolong the initial stages of interaction, elongating in this way the time of the transition to the accelerated swelling stage. Hence, studies into the interaction specific features may reveal extra ways of improving the serviceability of fuel elements.

Many factors influence on the commence stage of accelerated swelling. They are irradiation temperature and time, burnup, burn ups rate, which is determined by fission rate and heat flux.

Some technologic factor such as fractional composition, volume fraction of fuel and uniformity of its distribution can be also added. The latter due to availability of local pileups of fuel particles, strongly influences on interaction rate, since the concentration of thermal spikes in area of fuel - matrix interaction increases.
Irradiation scenario has can be also taken into account as one of the factors influence on the commence stage of accelerated swelling. Schematically factors influence on the commence stage of accelerated swelling is illustrated in fig. 1.

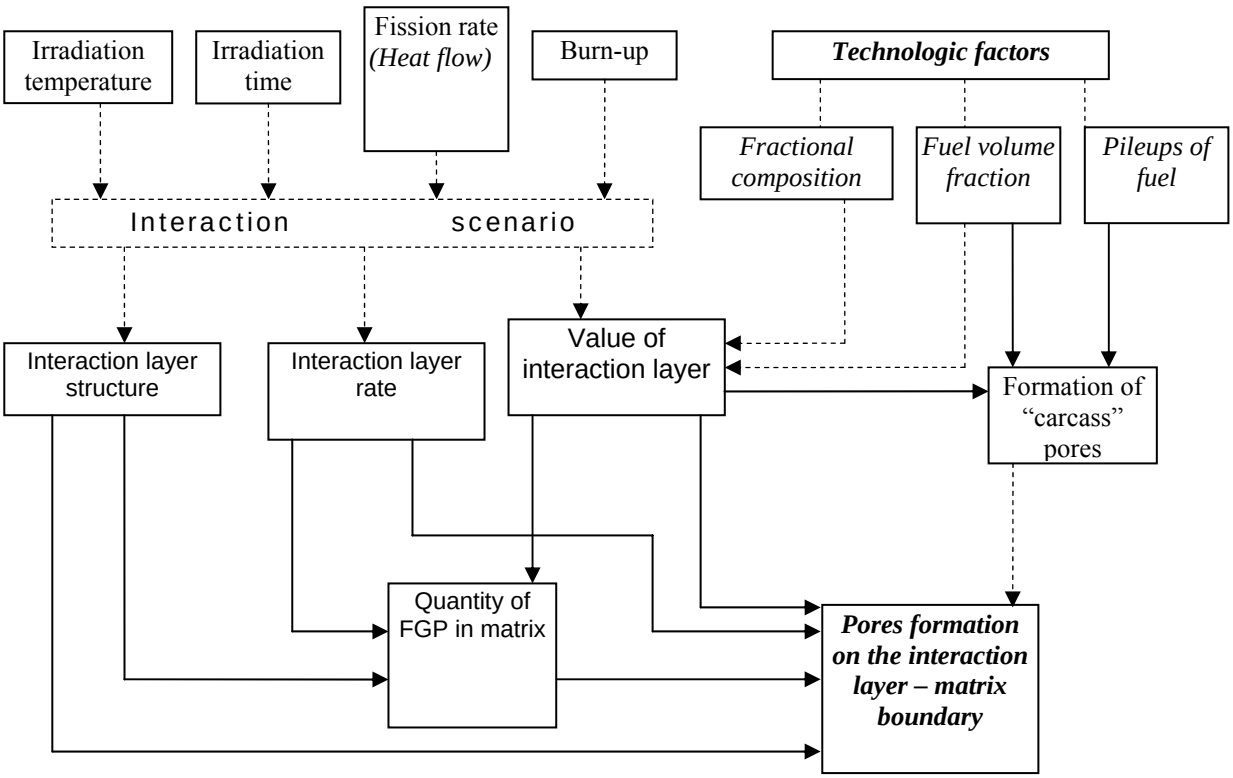


Fig. 1. Factors influence on the commence stage of accelerated swelling

Therefore determination of fuel – matrix interaction mechanisms and investigation of the interaction processes will lead to determine the optimal ways of reducing interaction.

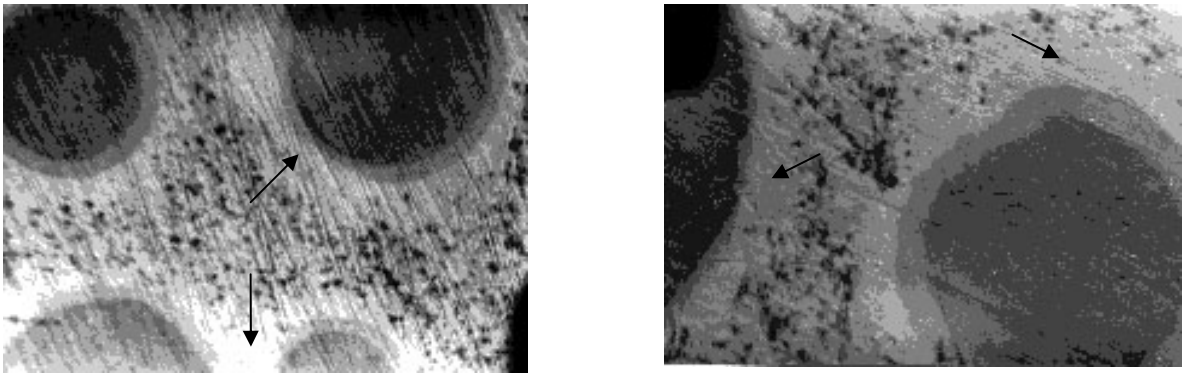
2. Stages and Mechanisms of Interaction

Now consider the interaction stages leading in the end to FGP piling up in resultant pores and pillowing.
The following interaction stages might be arbitrarily singled out:
1. Fission fragment releases out of fuel into matrix.
2. Growth of interaction layer in the matrix direction with fission fragment displacement to interaction layer – matrix boundary.
3. Fission fragment piling up on interaction phase – matrix boundary, "shears" formed at the interfaces with Al matrix as well as FGP concentrating within "carcass" pores.
4. FGP build-up in voids, pressure increases, formation of pillows.
Consider those stages in more detail.

2.1. Fission fragment releases into matrix

Due to small sizes of fuel granules (60-150 μm) significant amounts of fission fragment get into an aluminium matrix. The free path of a fission fragment in a U-Mo alloy makes up 6-10 μm, in

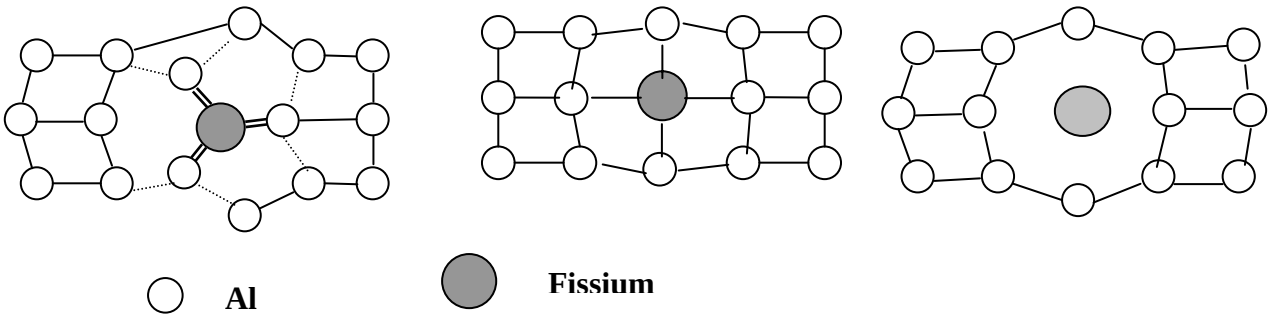
aluminium it is 10-15 μm. In aluminium matrix fission fragments form the so-called rim of damages up to 15 μm thick that is clearly visible in fig. 2. The total volume fraction of fission fragments in an aluminium matrix is the larger, the less is the fuel particle size.



a b

Fig. 2. Damage rim formed around fine (a) and coarse (b) fuel granules, 40 % burnup, IVV-2M reactor, Zarechny [4-5].

The release of fission fragments into the matrix from interaction layer is several times less and is dependent of the interaction phase composition. It is usually described by the $(U,Mo)Al_x$ formula where x is equal to 3-7. That is why, a compound of the $(U,Mo)Al_7$ type shall release lower quantities of fission fragments into a matrix in comparison to $(U,Mo)Al_3$.
Fission fragments available in the matrix might form either aluminides or substitutional solid solutions or, as is in the case of FGP, might be more or less distributed within a damage rim (fig. 3). In this case they are scattered within the damage rim and do not induce significant swelling increases or a lower serviceability of a fuel element.

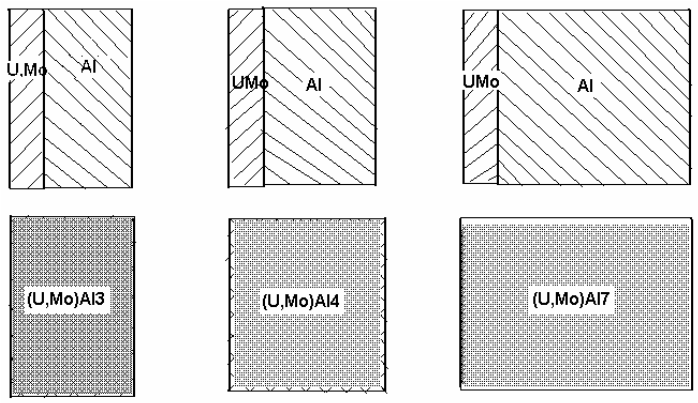


a b c

Fig. 3. Schematic fission fragment distribution within damage rim;
a – formation of aluminides, b – formation of substitutional supersaturated solid solutions,
c – isolated state (FGP)

2.2. Growth of Interaction Layer

The next stage of fuel composition structure change is an interaction layer growth in the matrix direction with fission fragment displacement from the damage rim to the interaction layer – matrix boundary.
The interaction layer is primarily growing in the direction to the aluminium, since its content of aluminium is a factor of 3-7 higher than that of uranium. In this way the damage rim around fuel particles decreases when the interaction phase is growing. Fig. 4 is a schematic presentation of the formation of interaction layers of the $(U,Mo)Al_x$ type (with account for volume changes proceeding at the time).



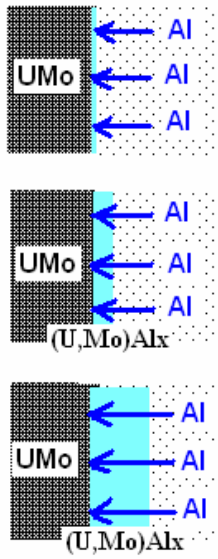


Fig. 4. Schematic presentation of (U,Mo)Al_x type structure with account for volume changes.

Fig. 5. Growth dynamics of interaction phase

Since aluminium has a higher diffusion mobility in comparison to uranium, upon interacting it is the process of the aluminium diffusion into uranium that prevails, not the reverse. As it will be shown below it is due to this fact fission fragments pile up in intergranular pores. The growth dynamics of the interaction layer is illustrated in fig. 5.

First, a thin layer of (U,Mo)Al_x is formed, then aluminium atoms diffuse through this layer and reach UMo to form the next layer putting off the previous one to the Al matrix side. Thus, the interaction layer continues its growth in the matrix direction.

Now, consider what occurs to fission products specifically FGP, available in the damage rim that is left by aluminium for UMo fuel.

Fission gas products do not interact with uranium, hence, do not diffuse through the intermetallic uranium – aluminium layer in the direction of U-Mo fuel. Therefore, as the interaction layer is growing they are gradually displaced to the interaction layer – Al matrix boundary. This process is schematically presented in fig. 6.

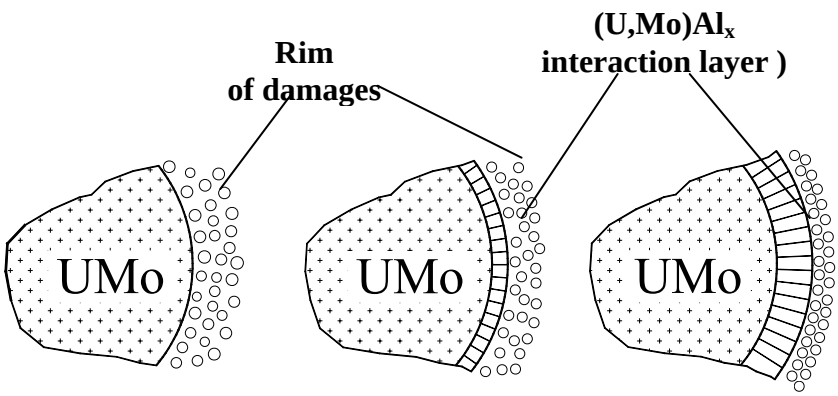


Fig. 6. Schematic pileup of fission fragments on fuel-matrix boundary.

As the interaction layer is growing fission gas products are piling up in a higher concentration on the interaction phase – aluminium boundary which is repeatedly pointed out in the CEA work [2] (fig. 7).

The boundary concentration of fission fragments also depends, in particular, on the growth dynamics of an interaction layer phase. The longer time it grows the more fission fragments manage to leave fuel granules for an aluminium matrix before it “absorbs” the damage rim. The quantity of fission fragments that are added from the interaction layer itself is much less.

Hence, the most acceptable version would be a version, when the interaction layer is quickly growing at the beginning and is decelerating at the end. This scenario is realized with the most intensive irradiation at the beginning of a cycle at the low concentration of fission fragments in a matrix.

However, the worst irradiation scenario was realized in IVV-2M reactor tests, where at the 40% burn up the main part of fission fragments released out of fuel into matrix. The irradiation layer don't impede this process since of its small value (3-4 μm). That is why it situated inside the damage rim. Then, during the last third part of irradiation at accelerating fission rate from 3.6 $x10^{14}$ to $4.6x10^{14}$ $f/cm^3/s$ interaction layer grew rapidly up to 12 μm, with fission fragments displacements (concentration) to the interaction layer – matrix boundary. Interaction layer growth dynamics via burn up is shown in fig. 8.

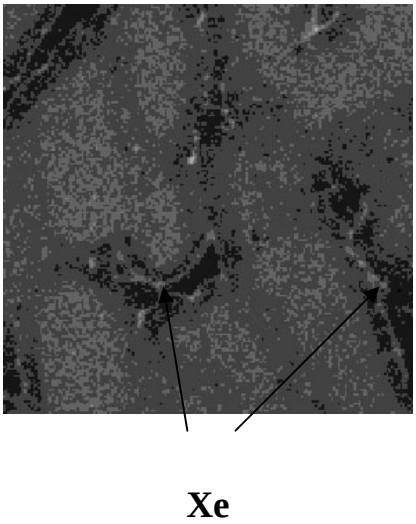


Fig. 7. Higher concentration of fission gas products on interaction layer – aluminium matrix boundary [2]

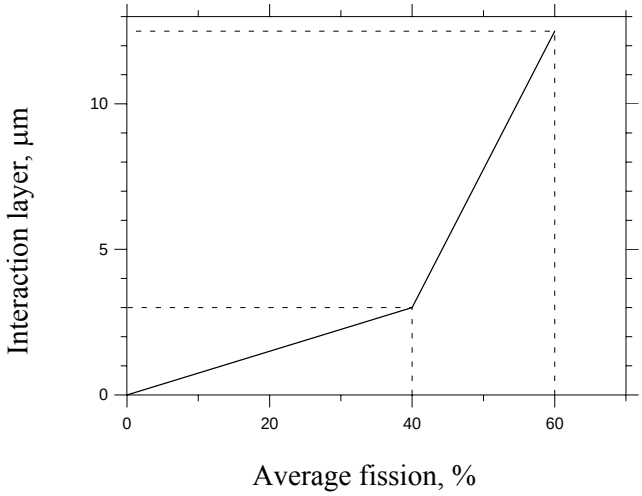


Fig. 8. Interaction layer growth dynamics via burn up

One of the most "successful" versions of design and irradiation scenario is a rod type fuel element designed in KAERI [6], where the critical interaction layer value of 10-13 μm that is a free path of a fission fragment in a matrix is attained rather quickly. This is not feasible in standard plate fuel (ANL and CEA) having the volume content of fuel more than 50 %. In this instance, the matrix completely disappears at the interaction layer thickness of 10-15 μm.

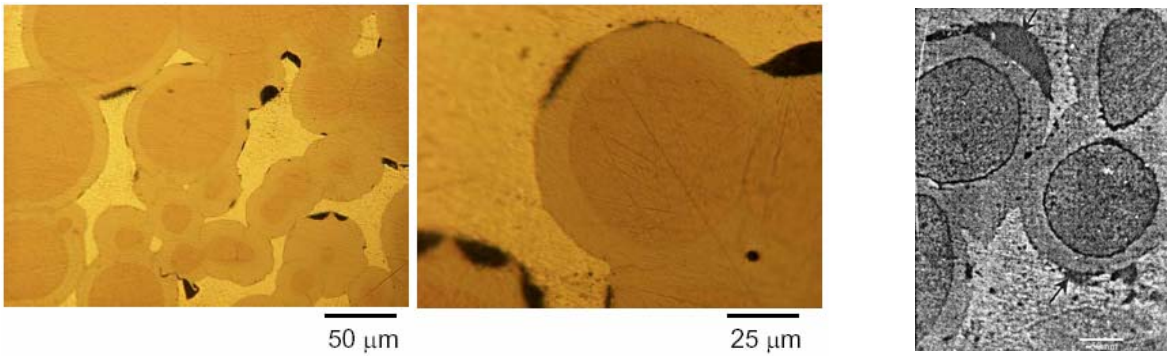
2.3. Fission fragment concentration on interaction layer – matrix boundary, formation of "shears" on interaction layer.

The next interaction stage results in concentrating fission fragments on the interaction layer – matrix boundary, the interaction phase "shearing" and fission fragments concentrating in "carcass" pores.

Shearing is clearly visible in fig. 9a [3], in CEA tests as well as in fig. 9b where the similar effect was obtained on fuel tubes in reactor tests carried out in Zarechny [4-5].

It might be assumed that the FGP piled up in some sites of the fuel – matrix boundary restricts the access of aluminium to the fuel and serves the barrier against aluminium diffusion. Hence, the local growth of the interaction phase in the particular site terminates. At the expense of diffusion aluminium atoms bypass the obstacle and a void remains in place of aluminium atoms and this leads to the formation of cavities – shears in the interaction phase.

It can be pointed out that concentration of FGP forming barrier around fuel particles is larger in cold regions. That is why voids appear preferential in the heat flow direction, i. e. in the direction to the cladding and to excess of Al matrix. This is clearly evident from fig. 9a.



a b

Fig. 9. Lenticular shape porosities at the interfaces with Al matrix as a result of piled up fission fragments, a – CEA tests [3], b – Zarechny tests [5].

Another sinking site for FGP is intergranular pores that might sometimes form by the so-called "carcass" mechanism. It consists in the fact that due to the growth of the interaction layer the fuel particles commence to repulse with the resultant formulation of intergranular pores. This mechanism is realized under out-of-pile conditions completely and partially under irradiation since irradiation induced creep leads to little changes in fuel particle shapes and filling the pores formed.

2.4. Pileup of FGP in voids, pressure increase, pillowing.

The resultant fission fragment concentration on the interaction layer – aluminium matrix boundary produced a higher gas pressure. This pressure results in the propagation of a continuous inner void along the fuel rod height. Gas filled voids of individual fuel particles merge, and brittle intermetallic links between particles break down. After the attainment of the critical pressure that depends on the crack length, strength characteristics of a fuel rod, its shape (plates or rods) etc, the internal pressure gives rise to pillowing and lowering down the serviceability of a fuel rod (fig. 10) [5].

It is to be noted that the region of a continuous crack in the fuel component covers not only the pillowing region but also the adjoining regions. That is why fission gas products available in a pillows come from both the pillowing region and the boundary ones (fig. 11) [5].

The critical pressure resulting in pillowing depends on a defect length. Small-size defects are known to be capable of withstanding high pressures of FGP without substantial plastic strains or pillows. It is natural that the occurrence of pillows on the rigid design fuels (rod ones) is unlikely [8].

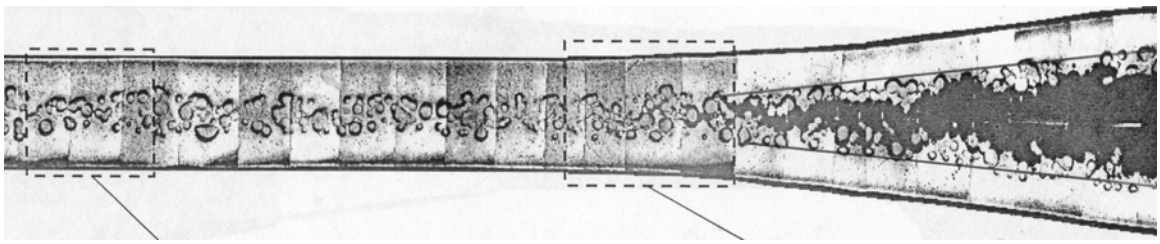


Fig. 10. Pillows formation in fuel component [5]

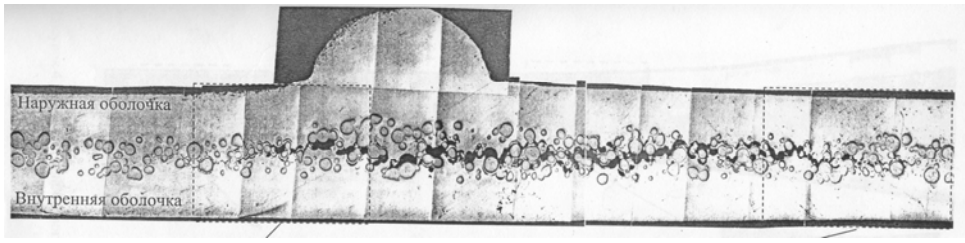


Fig. 11. Continuous porosity in continuous pillowing region of fuel component [5]

3. Technologic factors, influencing UMo fuel – Al matrix interaction

Ones more factor, that influence on character and extent of interaction and as well as on the porosity formation in the fuel component is technologic factor, i. e. fuel element fabricating technology. We bear in mind not only fractional composition and volume fraction of fuel, but practically haven't taken into account earlier the local pileups of fuel particles, which are determined by fuel element fabricating technology. A specific part of fuel composition in-pile tested in IVV-2M reactor (Zarechny) is shown in fig. 12. It can be seen that many local micro regions exist in the fuel, where fuel particles concentration is distinguished by several times. Naturally, the less is volume fraction of fuel the more is segregations of fuel particles.

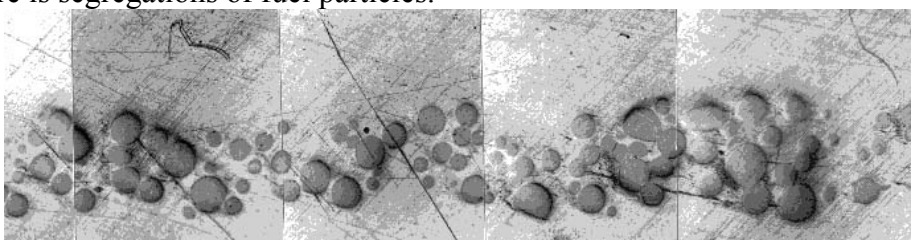


Fig. 12. Longitudinal section of fuel composition at 0,25H of fuel element [5]

As it was be observed after investigation of irradiated fuel tubes in IVV-2M reactor, that pores and discontinuities, resulting from interaction under irradiation are most often encountered either between fuel particles or within their aggregates. Just there the processes of gas swelling and arise of stresses in fuel component start up, that latter spread along the whole fuel element.

In other words serviceability of fuel is determined not only by the average characteristics of the whole fuel, but precisely by the characteristics of the local regions of fuel, where the maximum location of the fission gaps exists. For average uranium density of 5,4 g/cm^3 the deviation from uniformity estimates ~110%, that at uranium density of 8,0 g/cm^3, applicable in CEA and ANL fuel elements, the deviation from uniformity estimates ~40%.

These estimations conform by the results of metallographic analyses as well as by the measurements of fuel composition thickness along the fuel element (fig. 12, 13) [5].

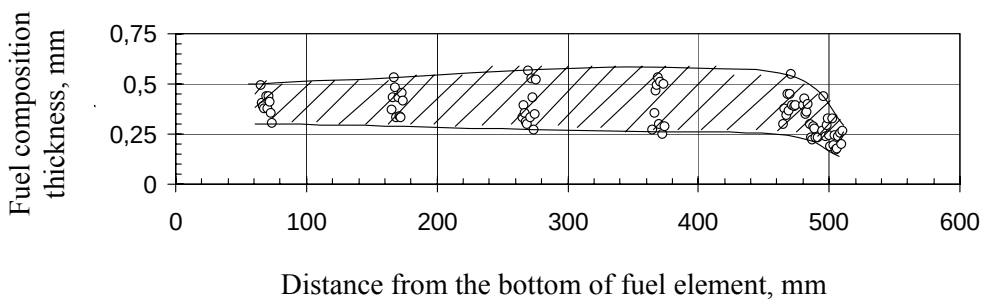


Fig. 13. Changes in thickness of fuel element, fuel cladding and fuel composion along the inner fuel tube, face N1 [5]

It can be seen from fig.13 that practically in each cross section the thickness of fuel composion ranges from 0.25 to 0.5 mm, i. e. the deviation makes up 100%.

Although the temperature, burnup in atomic percent and fission rate in those local pileups regions practically the same as in average fuel composition, however the value of local burn ups related to the micro region of fuel composion, not to the isolated particle itself, is much larger.

In those local regions at large areas, rims of damages from the neighbour particles overlap and local burnup increases. It is well known fact that UMo fuel – Al matrix interaction propagates at large extent by FGP, than by temperature. Without irradiation the interaction doesn't start even at 400^0C.
Several ways of reducing the interaction influenced by the pileups of fuel particles are under consideration now. The simplest one – to use the mono fraction of fuel granules. For instance, mono fuel size fraction of 160, 100 and 60 μm respectively can be used separately in fuel elements (for fuel elements, fabricated in Russia).
The other method – coating fuel granules with aluminium. This method eliminates the occurrence of fuel granules conglomeration.
Elimination of fuel granules contact may be reached by application of super fine powder (10 – 15 μm) (Si for instance). When fabricating fuel composion (mixing and rolling) Si powder occupies intergranular space and move fuel granules apart. Necessary quantity of powder – 2-3%.

4. Analyses of UMo – Al interaction structures

One of the impotent aspects that determinate the character of interaction is the structure and composition of UMo – Al interaction layer.
According to the U-Al phase diagram only three intermetallics, viz., UAl_2, UAl_3 and UAl_4, are available. However, in a reactor irradiated at certain conditions, intermetallic compounds had the content of Al varied from 6 [10] to 7-8 [1, 11]. This structure has not been produced under out-of-pile conditions. It has been assumed [1] that molybdenum stabilizes this structure since the Al-Mo system contains both $MoAl_6$ and $MoAl_7$. Latter investigations of the phase diagram of U-Mo-Al system [9] revealed a ternary Al rich intermetallic compound UMo_2Al_{20}, which approximately corresponds to the $(U,Mo)Al_7$ formula. However, this equilibrium phase in the U-Mo-Al system is not identical to the UMo/Al interaction layer for the following reasons:
1. In U-Mo alloy, e. g., UMo9 the U:Mo ratio equals 8:2, while in the novel equilibrium compound it is 1:2. Hence, for it to form, Mo has to concentrate in a local zone where an interaction with Al shall proceed. This is impossible in terms of thermodynamics since the Al diffusion rate is higher than that of molybdenum.
2. The U-Mo ratio in the detected interaction layer and on UMo fuel is the same as in original UMo fuel, i.e., 8:2, in other words Mo available in the interaction layer is much lower than U. Hence, the open ternary UMo_2Al_{20} compound has nothing to do with the interaction layer.
In our view, $(U,Mo)Al_x$ where x=6-8 resulting from the Al-UMo alloy interaction is actually not a stoichiometric compound of $(U,Mo)Al_7$ type but is a complex structure that consists of a fine mixture of $(U,Mo)Al_4$ or $(U,Mo)Al_3$ phases with aluminium, i. e. $(U,Mo)Al_x/Al$, where x=3-4. Schematically the formation of such structure is represented in fig 14 [12]. A small amount of UMo_2Al_{20} in this finely dispersed mixture of phases is possibly available. The formation of metastable supersaturated solid solution based on uranium aluminide compound is also available, but in this case its formation is thermodynamically unprofitable.
Such a structure is only formed under reactor irradiation at the expense of thermal spikes induced by fission products at the $(U,Mo)Al_4$-Al or $(U,Mo)Al_3$-Al boundary that lead to melting down and quick crystallization of this zone to form a finely dispersed $(U,Mo)Al_x/Al$ alloy. The driving force of this process is the formation of an alloy of the same phase composition consisting of two heterogeneous components – UAl_4 and Al or UAl_3 and Al, since when the alloy is formed through the energy of alloy formation releases, the energy of a heterogeneous system is lowered down [13]. Due to the availability of aluminium interlayers, the interaction phase has higher thermal conductivity than that of a pure intermetallic phase and under irradiation is fairly sinterable with other similar layers available on other fuel particles to form a thermal contact.
The disadvantages of such structures refer to the fact that it is neither a barrier against the diffusion of Al into fuel nor slows down the interaction. Its drawback is also the fact that for it to form a large quantity of the heat-conducting matrix is spent.

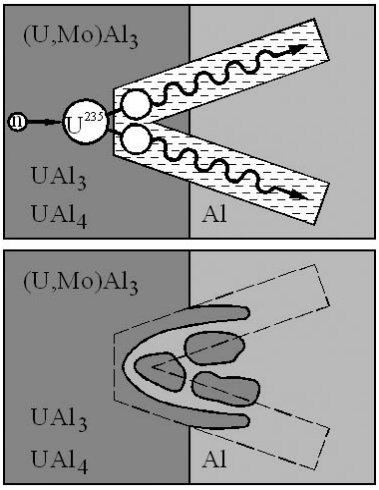
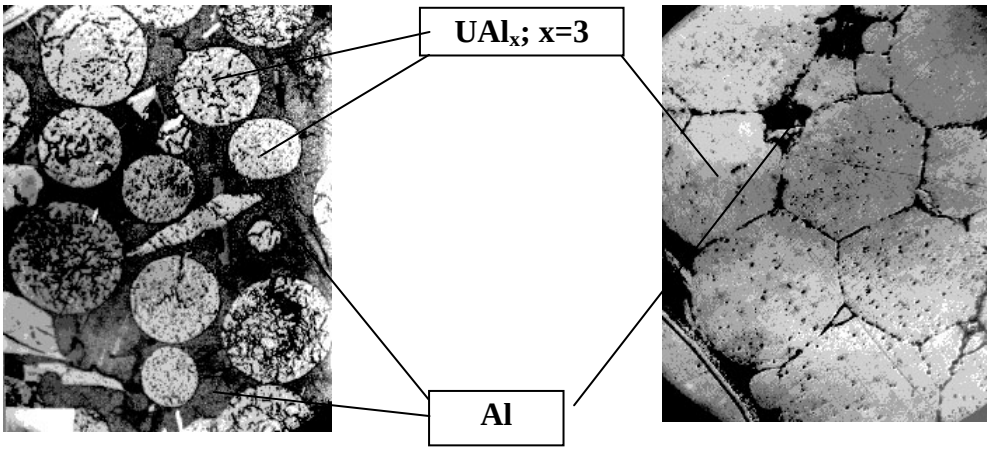


Fig 14. Formation of $(U,Mo)Al_x/Al$ type structure with excess of Al by the expense of thermal spikes from fission fragments along $(U,Mo)Al_x$-Al boundary, a) melting boundary zone by the expense of thermal spikes, b) rapid crystallization of this zone to form finely dispersed mixture of phases $(U,Mo)Al_x/Al$ [12]

The structure of a similar type although under different irradiation condition was experimentally obtained by us via the UAl_3-Al interaction; UAl_3 not containing molybdenum. Figure 15 illustrated as fabricated and after irradiation UAl_3+Al microstructure [12]. X-Ray diffraction analyses found the existence of only 2 phases UAl_3 and Al under irradiation.



a **b**

Fig. 15. Formation of UAl_3/Al type structure under irradiation, a) as fabricated; b) after irradiation [12]

The existence of such structures is determined by irradiation conditions, since two processes occur simultaneously – formation of such structures by thermal spikes and Al diffusion process into UMo fuel with formation of typical interaction phase, similar to those, that takes place in out of pile conditions. If the latter process predominates, the $(U,Mo)Al_3$/Al type structure might transform to a thermodynamically stable structure of $(U,Mo)Al_3$, containing approximately 10 volume percent of micro precipitates UMo_2Al_{20} phase. Therefore the ratio fission rate – temperature determines the structure of interaction phase.

Our experiments evidence that the structure of the UAlx type where x>4 may not be obtained under out-of-pile conditions by sintering U-Mo and Al even at high temperatures and long exposures. This structure results only under the mutual action of temperature and irradiation. Therefore, at Bochvar Institute to investigate the properties of the like structures the procedure was tried out involving simulators of similar structures, in which the crystal structure of the UAlx type contains excess aluminium and x might be varied within 4 to 6 [12-14]. This type of the structure might be obtained

not only as interaction layers on U-Mo fuel but also as compacts, which facilitates studying their properties. Structures of simulators based on UAl_3 compound are shown in fig 16.

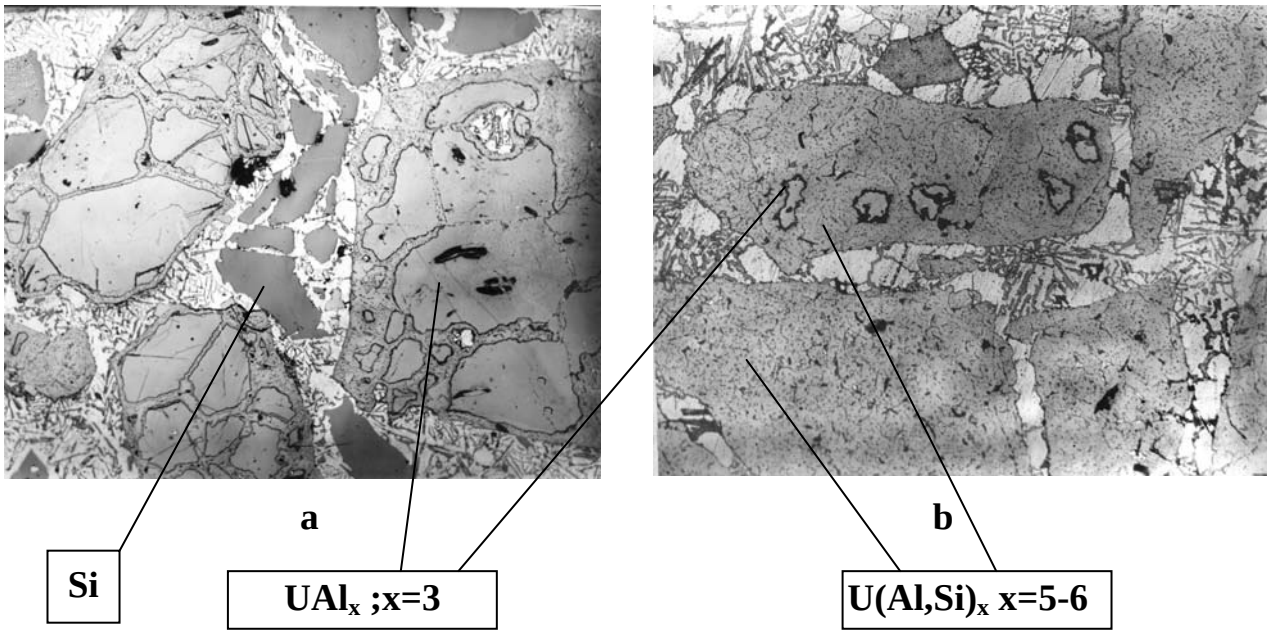


Fig. 16. Structure of simulator of interaction phase UAl_3/Al, a) as fabricated UAl_3 + Al structure, b) after heat treatment - UAl_3/Al type structure [12-14].

It can be mentioned that the initial value of measured microhardness for UAl_3 phase was 5000 – 5500 MPa, as for simulated structure formed in out pile conditions and represented in fig. 14b was the two times lower 2500 – 3000 MPa due to micro interlayers of Al in it.

5. Conclusion

1. Consideration is given to stages of irradiation induced UMo fuel-aluminium matrix interaction resulting in pores and pillows. They involve fission fragments releases out of fuel into matrix to form a damage rim, growth of interaction layer in the matrix direction with fission fragments displacement to interaction layer – matrix boundary, fission products concentration at interaction layer – matrix boundary, formation of "shears" on interaction layer, fission gas products piling up in voids, pressure increases, formation of pillows.
2. The importance of the initial stage of interaction, when the main quantity of FGP released out of fuel into matrix with formation of damage rim is shown. This stage creates pre-conditions for FGP pilling up on interaction phase – matrix boundary, which leads afterwards to fuel composition gas swelling. FGP concentration in Al matrix can be decreased either by reducing the interaction, which is the separate direction of investigations, or by accelerating interaction layer growth at the initial stages of in-pile testing up to value, equal to the fission fragment free path in matrix (which was put into practice in the KAERI tests). The latter route is not always feasible at the high volume fraction of fuel.
3. The analyses of the results in pile test investigations take us to the conclusion that additional factor, influencing interaction is technologic factor, i. e. the local pileups of fuel particles. Irradiation produced pores and discontinuities are most often encountered either between UMo fuel particles or within their pileups. In those regions rims of fission fragment effected damages overlap and the value of the local burnup in those regions related not to an individual particle but to the microregion of a fuel component is much higher. The less are the volume fraction of the fuel in the fuel composition, the more is the influence of fuel pileups in it. Estimation of pileups influence for various types of fuel elements is made and methods reducing the non-uniformity of fuel in local regions are suggested.

4. The expense of thermal spikes induced by fission products leads to formation of an intricate structure of $(U,Mo)Al_7$ type with excess of Al. To our view it is a finely dispersed mixture of phases consisting of UAl_3 phase, pure Al and a small amount of UMo_2Al_{20}. The driving force of this process is formation of an alloy since with alloy formed via release of formation enthalpy, the energy of heterogeneous system is lowered down. It has been shown that depending on irradiation conditions such a structure might transform to a thermodynamically stable structure of $(U,Mo)Al_3$ type. Experimentally acquired data are presented, that pertain to formation of identical type structures with excess Al in binary U-Al system without Mo. Under out of pile conditions structures were obtained that simulate those of intermetallic compounds having Al microinterlayers.

4. References

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