

Inert matrix fuel with low melting point zirconium brazing alloys

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Abstract

Several versions of inert matrix fuel (IMF) using low melting point zirconium brazing alloys are suggested to improve their utilisation. The alloys are incorporated into a fuel component. Upon heating they melt down and due to capillary properties they penetrate fuel particle joints and ensure metallurgical contact between cladding and fuel elements as well as between fuel components themselves. The brazing alloys developed at A.A. Bochvar Institute have melting temperatures of 963–1133 K and are produced in the form of both granules and amorphous (metglass) strips. Zirconium brazing alloys used in IMF increase the thermal conductivity and allow an improved bonding to the fuel cladding, which will result in higher serviceability of fuels under transient conditions. Several versions of low melting point zirconium brazing alloy IMF are reviewed with respect to the feasibility of their utilisation.

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1. Introduction

Presently there are numerous concepts and versions of Inert Matrix Fuel (IMF) element designs for burning civil and weapon grade plutonium [1–4]. The most currently studied versions comprise pellet type fuel elements with an inert matrix based on zirconia or spinel.

The disadvantages inherent in those versions of IMF, i.e. low thermal conductivity and the absence of metallurgical bond between cladding

and fuel pellets reduce their serviceability during transients.

To improve the properties of IMF it is suggested to use low melting point Zr brazing alloys, which have been developed at A.A. Bochvar Institute [5–7]. They could be directly introduced into a fuel component (Fig. 1(a)), serve as a contact between fuel pellets and a Zr cladding of a fuel element (Fig. 1(b)) or be a matrix for Pu based fuel (Fig. 1(c)). When sufficiently heated the alloy melts down and at the expense of capillary forces, deeply penetrates the fuel and metallurgically bonds the fuel element and the cladding. The alloy could also be used in place of an aluminium matrix in the designs of heterogeneously distributed fuel elements [8,9].

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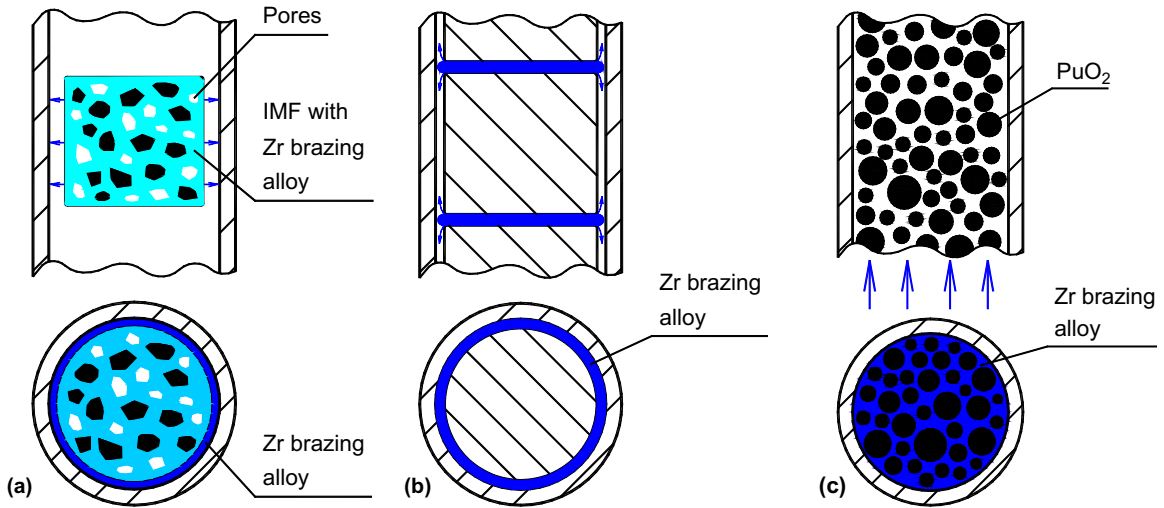


Fig. 1. Some modes of Zr brazing alloys application: (a) introduction of Zr brazing alloy into IMF pellet; (b) metallurgical bonding of cladding to IMF pellet; (c) as a matrix for IMF granules. The arrows show the motion of Zr-base molten brazing alloy in the process of a fuel element fabrication.

2. Characterization of Zr brazing alloys and specific feature of their application

There are currently several classes of novel Zr-base brazing alloys. All of them are deep ternary or quaternary eutectics having relatively low temperatures of melting, i.e. from 963 to 1133 K. The alloy compositions, the temperatures of melting and brazing are summarized in Table 1 [5–7].

The alloys can be manufactured as granules or amorphous strips. When quickly quenched the alloys easily become amorphous with the maximal thickness of amorphous strip produced by spinning of 250 μm (Fig. 2).

Due to the low melting points the alloys might also be employed as a matrix alloy in the process of impregnating fuel granules, loaded into a fuel element cladding, with molten zirconium brazing alloy [10,11]. One of the examples of UO₂ fuel composition produced in this way is illustrated in Fig. 3.

Another more efficient mode of applying Zr brazing alloys is to make use of their capillary properties. The alloys of this type were successfully used to braze end caps to fuel claddings; with a cladding – end to cap gap of several microns (Fig. 4).

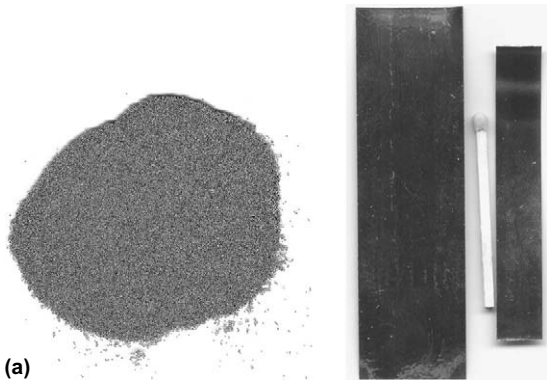
The alloys as amorphous strips were also used to braze dissimilar materials, e.g. ceramics and a metal, graphite and a metal, to impregnate porous graphite, etc. The brazing alloy always spontaneously entered micrometer size gaps.

Fig. 5 presents the results of one of the experiments to examine the capillary properties of a Zr–Fe–Cu system alloy. Two densely compressed Zr plates having slots 0.05–0.35 mm thick were immersed into a molten alloy and held at 1183 K for several minutes. Because of capillary forces the molten alloy rose to the height of up to 50 mm and entered the sites where no gap was available.

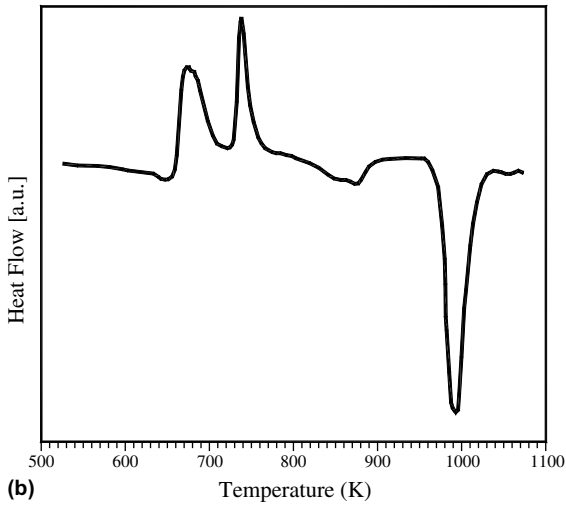
Based on the capillary properties of Zr brazing alloys the fuel element fabrication technology was developed and is called the capillary impregnation

Table 1
Alloy compositions, melting and brazing temperatures (T)

| Number of group | Alloy composition (wt%) | | | | | | Melting T (K) | Brazing T (K) |
|-----------------|-------------------------|------|------|---------|---------|-----|---------------|---------------|
| | Zr | Ti | Fe | Cu | Be | Nb | | |
| 1 | Base | 5–20 | 4–7 | 1–3 | 1.5–2.5 | – | 963–993 | 1053–1083 |
| 2 | Base | – | 4–8 | 0.5–3.0 | 2–3 | 1–3 | 1053–1083 | 1123–1143 |
| 3 | Base | 5–10 | 8–14 | 8–14 | – | – | 1083–1093 | 1153–1173 |
| 4 | Base | – | 6–12 | 6–12 | – | – | 1123–1133 | 1173–1183 |



(a)



(b)

Fig. 2. Appearances of granules and amorphous strips (a) and differential thermal analysis of one of the Zr–Ti–Fe–Cu–Be system alloys (b).

(Cap. Imr.) [9–11]. It consists of two stages and is schematically presented in Fig. 6.

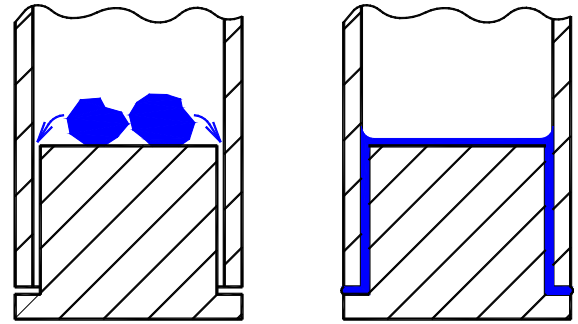


Fig. 4. Schematic presentation of brazing zirconium end cap to fuel cladding with zirconium brazing alloy.

1. Vibroloading of mixed fuel and matrix granules into cladding (Fig. 6(a)). In this case, coarse granules of fuel and fine granules of a zirconium alloy matrix (zirconium brazing alloy) are used. The volume ratios of fuel components are:

| | |
|---|--------|
| The fuel forms a skeleton | 55–60% |
| The matrix within the interstices of the skeleton | 10–22% |
| The pores | 16–30% |

The ratios between the diameters of the fuel and matrix granules range from 3:1 to 10:1.

2. Vacuum anneal above the melting temperature of the matrix alloy (capillary impregnation, Fig. 6(b)). While melting down under the action of capillary forces, the zirconium brazing alloy fills the joints between particles of fuel and the joint between fuel and cladding to form the so-called bridges, which increase the thermal conductivity of the fuel component.

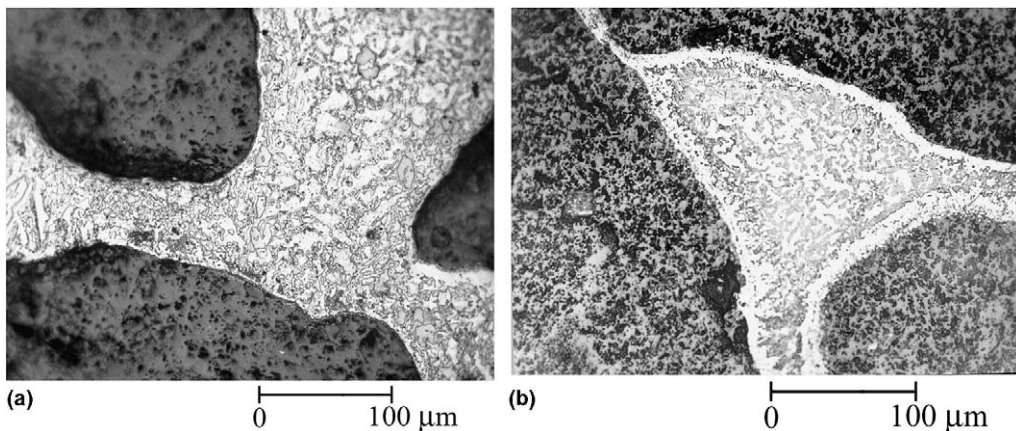


Fig. 3. Microstructure of $\text{UO}_2 + \text{Zr–Fe–Be}$ alloy fuel composition, (a) as impregnated, (b) as isothermally annealed at 1023 K for 6000 h.

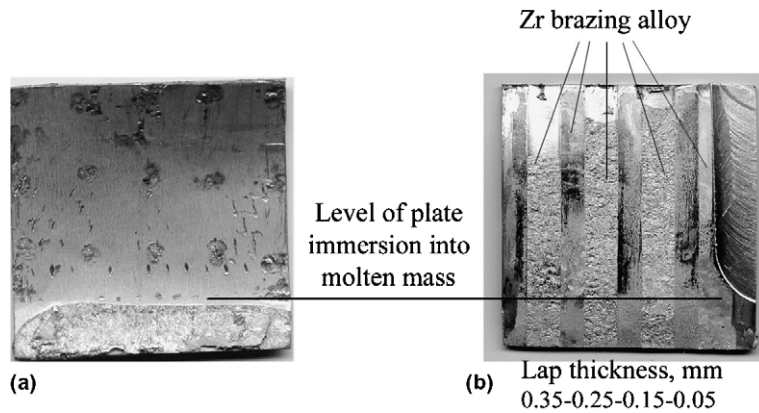


Fig. 5. Capillary properties of Zr–Fe–Cu alloy at 1183 K; (a) outer surface of plate, (b) inner surface of plate.

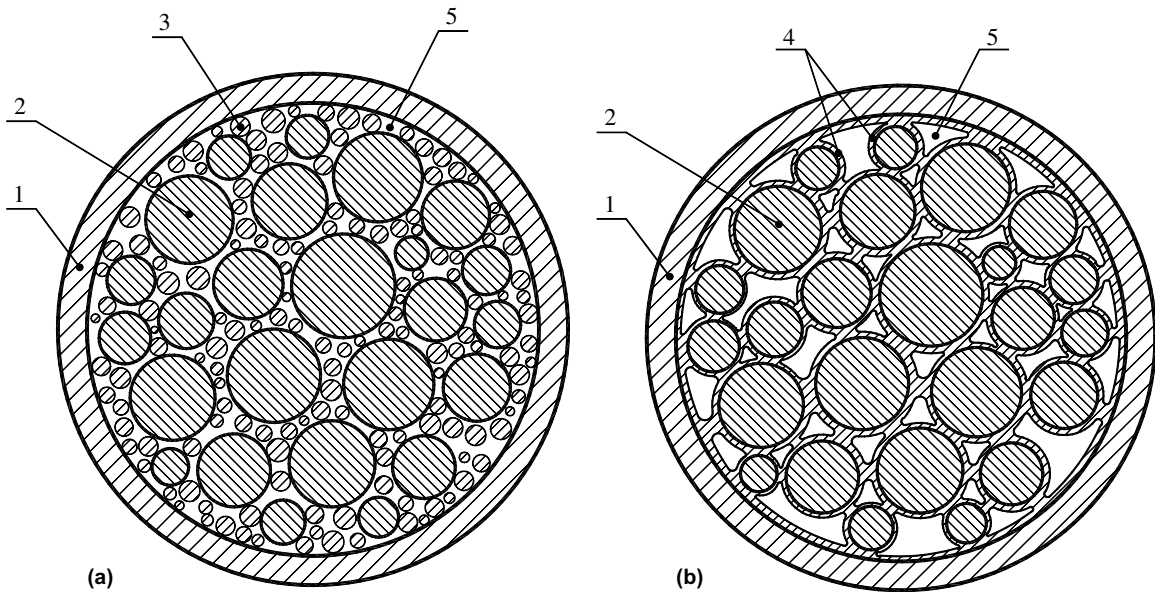


Fig. 6. Schematic cross-section representation of fuel element fabricated by capillary impregnation method [9,10]. (a) As vibroloaded. Consecutive loading fuel and matrix granules (infiltration method); (b) as capillary impregnated, (1) fuel cladding, (2) fuel granules, (3) Zr alloy matrix granules, (4) ‘bridges’ and matrix alloy coats on fuel granules after heating and cooling, (5) pores.

The application of this technology to fabricate the VVER-1000 type fuels using the high-density uranium fuel of the U-9Mo alloy type, promotes a high uranium density (up to 10 g U cm^{-3} in the fuel composition) as well as a high thermal conductivity of the fuel component ($15\text{--}20 \text{ W m}^{-1} \text{ K}^{-1}$). According to the assessing calculations, the pores available in the fuel composition accommodate fuel swelling under the cladding up to a burn-up corresponding to a fission density of 0.8 g cm^{-3} [10,11].

The appearance of the fuel composition and the macrostructure of the fuels fabricated

by the capillary impregnation are illustrated in Fig. 7.

3. Versions of applying zirconium brazing alloys in IMF elements

3.1. Metallurgical bonding between fuel element cladding and IMF pellets

To achieve the fuel pellet–cladding bonding a brazing alloy in the form of strips is placed between the pellets and upon melting fills the cladding–pellet gap (Fig. 1(b)).

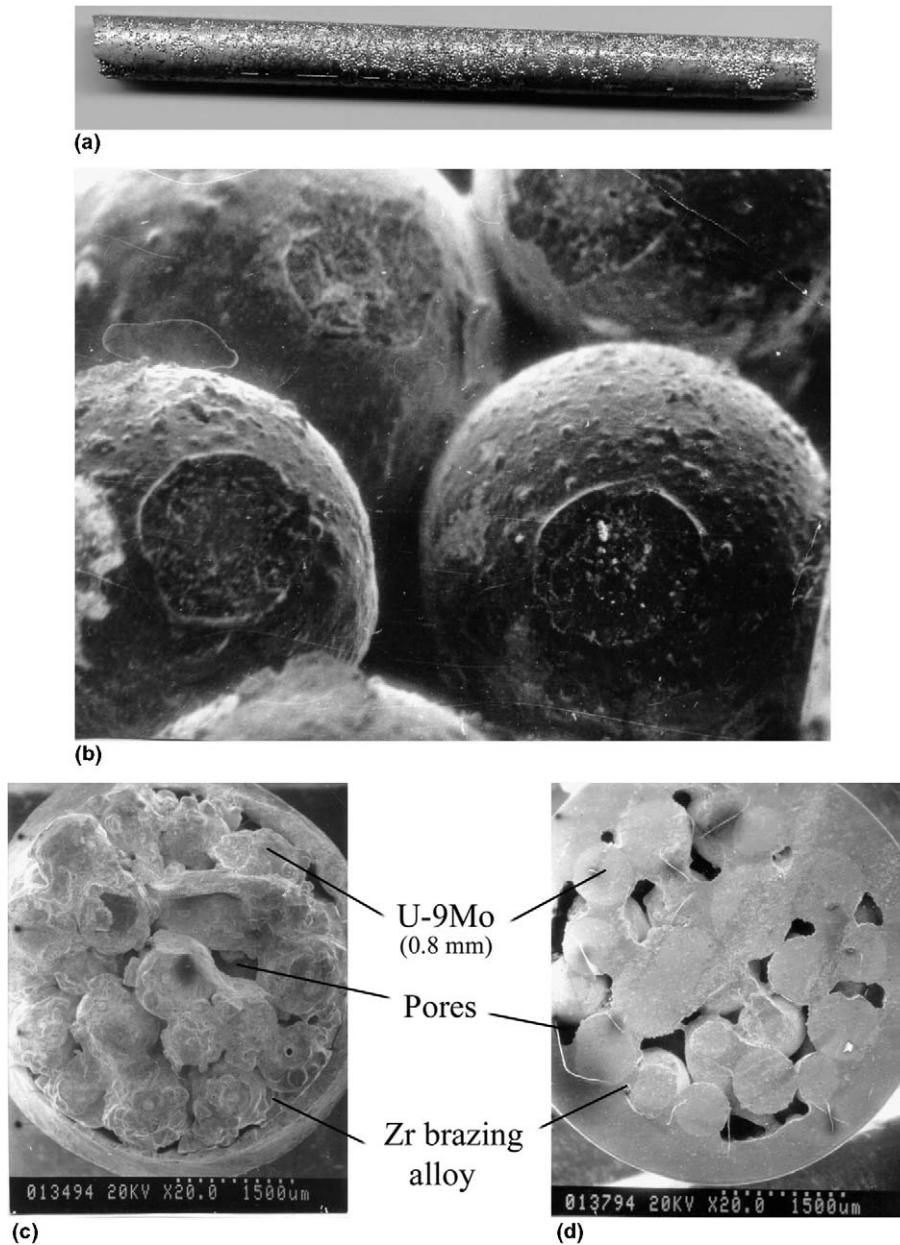


Fig. 7. Structure of (U-9Mo + Zr-Fe-Cu) fuel elements fabricated by capillary impregnation for VVER-1000 reactor type; (a) appearance of uncladded fuel component (25% porosity, 7.7 mm outer diameter); (b), (c) fractogram of fuel fractures (34% and 18% porosity, respectively), (d) fuel microstructure (22% porosity).

3.2. Brazing alloy introduction into IMF pellets

Another way of applying Zr-base brazing alloys is its direct introduction into a pellet as a component of an IMF pellet, composed for example of zirconium, plutonium dioxide and zirconium brazing alloy powders (Figs. 1(a) and 8). Pressed pellets are stacked inside a cladding and annealing (capillary impregna-

tion) is carried out while the zirconium brazing alloy is melting down it bonds the zirconium powder to PuO_2 within the pellet as well as the pellet itself to the zirconium cladding. In this case the pellet porosity is retained to accommodate swelling.

The optimized volume ratios between the pellet components are as follows:

| | |
|------------------|-----|
| PuO ₂ | 10% |
| Zr | 60% |
| Brazing alloy | 20% |
| Pores | 10% |

The as-pressed pellet might not be directly loaded into the cladding of a fuel element, instead, it might be pre-subjected to the capillary impregnation to increase its strength (Fig. 8). Then the pellet is loaded into the fuel element cladding and the capillary impregnation is repeated to promote pellet-cladding bonding using the brazing zirconium alloy available in the pellet.

3.3. Capillary impregnation of (Er, Y, Pu, Zr)O_{2-x} granules

In this option the fuel is composed of (Er, Y, Pu, Zr)O_{2-x} microspheres obtained by using the internal gelation fabrication process that was developed at PSI [1,2]. Here a Zr brazing alloy is used as a matrix applying the standard capillary impregnation [9,11] as the fabrication method. The fuel element cross-section is schematically shown in Fig. 9.

3.4. Isolated arrangement of fuel

One of the most promising versions of an IMF element is the one for which the plutonium fuel, primarily PuO₂, is arranged separately in fuel mini-elements that are arranged within a fuel element, the inner space of which is filled with a matrix material. The design and advantages of such a fuel element are described in more detail in [8,12]. However, these designs use a relatively low melting aluminium alloy as a matrix.

In addition to the high reliability of such a design that meets the ‘Rock Fuel’ requirement, and a feasi-

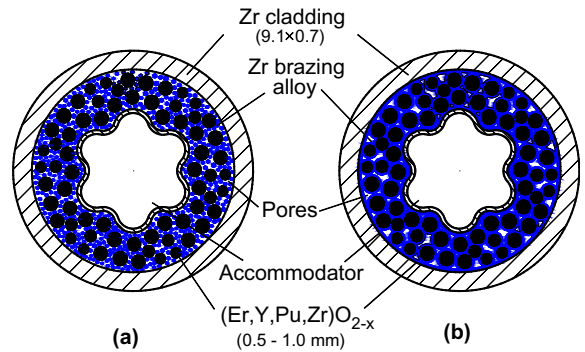


Fig. 9. Schematic presentation of cross-section of (Er, Y, Pu, Zr)O_{2-x} microspheres fuel element fabricated by capillary impregnation; (a) after vibroloading, (b) after capillary impregnation.

ble extended burn-up, its advantage is the simple and environmentally friendly process of the fuel element fabrication, i.e., the minimal quantity of the process operations related to Pu fuel processing.

The likely options of fuels using fuel mini-elements and Zr-base brazing alloys as a matrix material are illustrated in Fig. 10 [8,9,11,12].

One of the versions of a fuel element of this type contains fuel mini-elements where a porous Zr matrix fills and bonds the cladding to fuel mini-elements. The matrix is produced by the capillary impregnation method and consists of the bonded Zr and a Zr brazing alloy (Fig. 11).

The porous Zr matrix has two functions:

1. Medium for heat transfer from fuel mini-element cladding to the cladding of a fuel element. Its thermal conductivity makes up 10–15 W m⁻¹ K⁻¹. The temperature of the fuel mini-element cladding not exceeding 823 K, while that of the fuel does not exceed 1223 K.

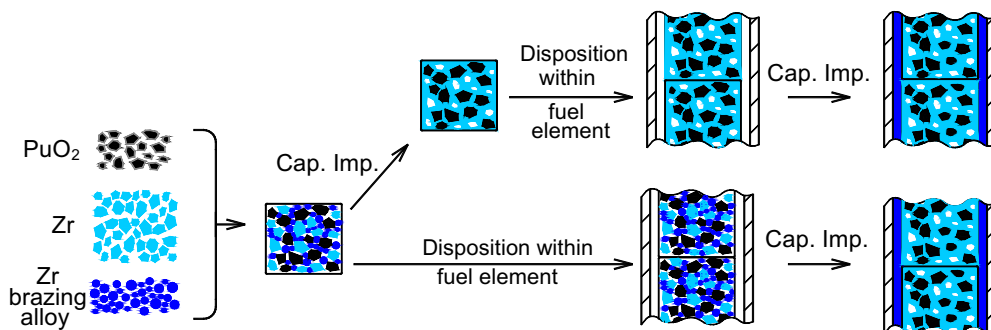


Fig. 8. Stages of IMF element fabrication with directly introduced Zr brazing alloys into IMF pellet.

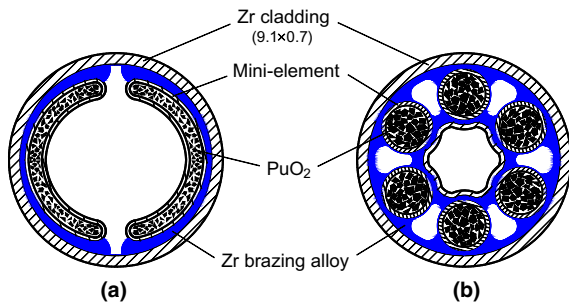


Fig. 10. Schematic presentations of various versions of fuels with fuel mini-elements and Zr-base brazing alloys; (a) flat fuel mini-elements, (b) cylindrical fuel mini-elements.

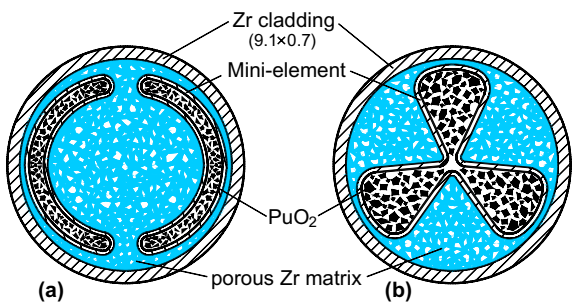


Fig. 11. Schematic presentation of different versions of fuel element with fuel mini-elements and Zr porous matrix (a, b) and microstructure of porous matrix (c).

2. Accommodation of fuel mini-element swelling. Hence, the fuel mini-element has not to be necessarily rigid, restraining swelling, it might be of any shape having a developed surface to increase the heat transfer.

4. Conclusion

The A.A. Bochvar Institute developed zirconium brazing alloys, with melting temperatures of

963–1133 K and the methods of their application to IMF fuel elements, allowing the extension of feasible designs using IMF and the improvement of their properties. These improvements primarily concern the thermal conductivity that makes the operation under transient conditions feasible.

The most promising design is the use of fuels with fuel mini-elements.

The advantages of this design are:

- possibility of extending burn-up;
- fuel element reliability, no contact of matrix with FGP, high corrosion resistance of fuel element components during operation and long-term storage that allows their categorization as 'Rock Fuel' to be directly disposed of in geologic formations,
- dust-free process of fuel element fabrication (reduced quantity of irradiation hazard cycles) and environment friendly technology.

Currently, a complex of activities are under way to design, validate and choose the optimized fuel element design. The technological studies relevant to the fabrication of fuel element simulators are in progress.

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