# CERMET FUEL BEHAVIOUR AND PROPERTIES IN ADS REACTORS

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## Abstract

Within the EUROTRANS Integrated Project, Forschungszentrum Karlsruhe (FZK) and the Institute for Transuranium Elements (ITU) are joining their efforts to study the behaviour of Mo-based CERMET non-uranium fuel for the ADS. Contributions include core safety calculations, and fuel property measurements and irradiation experiments

Safety studies for optimised EFIT core designs have concluded that, for the new low power cores of EFIT with a power class of ~ 400 MWth and a fuel power density of ~ 250 MW/m$^{3,}$ the CERMET-loaded cores behave favourably and the design limits of the fuels were not violated.

Mo-based CERMET fuel pellets and pins loaded with Pu and Am were fabricated for irradiation programmes which will start by mid-2007 in PHENIX (France) and HFR-Petten (The Netherlands). The thermal diffusivity and specific heat of the CERMET fuels (loaded with Pu and Am) were the main properties measured, and the thermal conductivity was deduced. The results were used to prepare the safety report for the irradiation experiments.

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**Keywords**

Transmutation, ADS, cermet, fuel properties

## INTRODUCTION

Within the EUROTRANS Integrated Project [1] co- financed within the 6th Framework Programme of the European commission, the sub-critical Accelerator Driven System (ADS) is now being considered as a potential means to burn long-lived transuranium nuclides. Within this programme, the domain AFTRA is responsible to develop and provide the data basis for the fuels to be used in the European Facility for Industrial Transmutation (EFIT). The preferred fuel for such a fast neutron reactor is uranium-free, highly enriched with plutonium and minor actinides. Requirements for ADS transmuter fuels are strongly linked with the core design and safety parameters, the fuel properties and the ease of fabrication and reprocessing. This study concerns the behaviour and properties of fuels with molybdenum as inert matrix.

The status of the development work was presented at the last ICENES conference [2]. Since then, the design of the European Facility for Industrial Transmutation (EFIT) was developed and the transmutation capability, the burn-up behaviour, the reactivity swing and power peaking factors, and the safety performance were determined for different cores with inert matrix fuels like MgO and Mo. For the EFIT, the CERMET with the Mo matrix is recommended as the reference fuel and CERCER with the MgO matrix as a back-up solution.

In this paper, we describe the results achieved so far, mainly for what concerns design and safety studies of the EFIT core, and the thermal properties of the Mo-CERMET fuels loaded with minor actinides.

## Design and safety aspects of Mo- related CERMET fuels

European R&D for ADS design and fuel development is driven in the 6th FP of the EU by the EUROTRANS Programme, where two ADS design routes are followed, the XT-ADS and the EFIT. The XT-ADS is designed to provide the experimental demonstration of transmutation in an Accelerator Driven System. The EFIT development, the European Facility for Industrial Transmutation, aims at a generic conceptual design of a full transmuter. A key issue of the R&D endeavour is the choice of an adequate fuel to be used in an Accelerator Driven Transmuter (ADT) like EFIT. Various fuel forms have been assessed and CERCER and CERMET fuels, specifically the matrices MgO and Mo, have finally been selected and are now under closer investigation. Pre-design studies for a 400 MWth EFIT have been performed within AFTRA to assess the generic safety performance and identify critical safety issues. In the current paper especially the safety behavior of the chosen CERMET (Mo-92) fuel is presented. The MgO matrix fuel shows advantages in the transmutation performance while the Mo-based fuel shows a superior safety behavior.

Already in the FUTURE project [3] an accelerator driven transmuter of the 800 MWth power class (ADT-800) has been developed. This ADT-800 core, used for discriminating of the various fuel forms, could be characterized by a relatively high fuel power density in the range of ~800 MW/m$^3$ for the CERMET fuel. Both the

FUTURE ADT-800 analyses and other investigations [4, 5, 6] revealed high coolant and clad temperatures for various transients in case of high power density cores. For EFIT one safety issue was therefore the investigation of the impact of a reduction of the power density, besides achieving a flat axial and radial power profile. Within AFTRA [7], single zone 2-zone and 3-zone cores with high and low power densities have been developed. The EFIT CERMET fuelled cores belong to the 400 MWth class with fuel volumetric powers ranging between 250 – 550 MW/m$^3$.

Analyses within the FUTURE project already revealed the sensitivity of the accident scenarios and outcome depending on the clad failure behaviour. To test the safety classification and safety performance, these fuels have been subjected to various transients under EFIT core conditions. Because of the unique high temperature behavior of these fuels, the investigation of design extension conditions and the identification of any cliff-edge effects are of special interest. Special emphasis has been laid on the analysis of blockage accidents. These accidents could lead to pin failures, the blow-down of fission-gas and helium, voiding and accident propagation. The SIMMER-III code [8,9] has been used as a main tool for this work and has been adapted for the conditions of innovative fuels in a heavy liquid metal cooling environment.

For a safety classification, AFTRA has provided fuel limits related to the different safety categories of the defense in depth concept [10]. A first set of failure limits for the T91 clad have also been provided by the DESIGN domain [11]. Because of the existing uncertainties in the light temperature transient behavior of these fuels it was an agreed decision not to allow any fuel ‘melting’ or disintegration up to Cat.IV. In Tab. 1 the temperature limits are displayed. Fuel melting is put in ‘parenthesis’ to reflect the processes of independent fuel/matrix melting, possible eutectic formation and disintegration and/or vaporization processes.

The values given for the CERMET fuels reflect the superior high temperature behaviour of the Mo fuel.  Safety analyses have been performed for all types of cores from single zone to three zone to identify the needs for design optimization. In the present paper the safety analysis results for a 2-zone EFIT core, developed by the AFTRA team, are displayed. The core has been designed with a nominal power of 400 MWth and achieves a minor actinide transmutation ratio of 42 kg/TWh$_{th}$, close to the theoretical limit. The subcriticality is 3000 pcm, the peak linear power is 210 W/cm. The core void worth is 6161 pcm and significantly larger than the in-built subcriticality. Reactivity swing and burn-up are still not fully satisfactory and further work has to be invested, however the core can well serve for a sound safety assessment.

For assessing the safety behavior of the different core designs several typical transients have been selected and analyzed with the SIMMER-III code [8, 9]. The current analyses include perturbations arising from the target/source (BT/I: Beam Trip/Interruption) and the core/nuclear (UTOP (Unprotected transient over power), ULOF (Unprotected loss of flow), UBA (Unprotected blockage accident)) system. Investigations of these advanced composite fuels reveal that new phenomena and scenarios have to be expected and modeled compared to ordinary MOX fast reactor fuel. The new phenomena that might be expected under such conditions are discussed e.g. in [12,13]. The significant void worth introduces a strong positive reactivity feedback mechanism from thermal coolant expansion. A stabilizing Doppler feedback is not available.

In the present paper some typical results for a ULOF and a UBA are displayed. The ULOF is of interests because of the sensitivity of the clad and coolant temperatures. Clad failure could then trigger a global gas release from the fission gas plena. Voiding and nuclear power increase could then effectively drive a pin failure propagation. The results are displayed in Fig. 1 and Fig. 2 and show that for this core design all temperatures stay below their failure limits and the core would survive a ULOF in the short term. Especially the fuel temperatures are far below the failure limits given in Tab. 1. The analyses reveal that the T91 clad used for the EFIT design is the limiting element for design optimization, because of its low creep limits of 1100 K under the EFIT boundary conditions (plenum pressure, coolant temperatures etc.). The CERMET fuel could withstand much severer transients.

In addition to the ULOF, the blockage accident (UBA) simulations are displayed in Fig. 3 and Fig. 4. A complete blocking of the subassembly inlet is assumed. The blockage represents an important scenario, as it can also lead to pin failures, the release of helium and voiding, reactivity addition and damage propagation into neighboring subassemblies. Under the heavy liquid metal conditions clad failure occurs with a blow-down of fission gases. Later the clad starts to melt and is dragged away from the failure location and refreezes in upper colder structures. Finally the fuel can become mobile and a so-called fuel sweep-out effect leads to a reactivity and power reduction (see Fig. 3 & 4). Though under external source conditions the power reduction is limited, it is helpful to stop the damage propagation and core disruption process observed in [16]. The sweep-out phenomenon has been observed experimentally in case normal fast reactor MOX fuels [14]. In case of composite fuels the issue is more complex and further investigations will be performed to take full credit of this effect. An important issue is that sufficient grace-time is provided to detect the blockage and shut-down the accelerator beam.

## Fabrication

At the Institute of Transuranium Elements, Mo based CERMET pellets have successfully been fabricated in the Minor Actinide Laboratory. The preparation of these pellets is based on the fabrication of beads containing the actinide phase by a combination of the external gelation [15], and infiltration methods [16] followed by their mixing with the molybdenum matrix. In this work the fabrication, characterization and determination of thermal properties of two cermet fuels, $PuAmO_{2-x}$-Mo (FUTURIX 5, FX5) and $PuAmZrO_{2-x}$-Mo (FUTURIX 6, FX6), is presented.  These fuels have been fabricated under the framework of the FUTURIX-FTA experiment and they will be irradiated in the Phénix reactor [17].

Highly porous beads of $PuO_2$ and $PuZrO_2$were produced by the external gelation method. After their calcination at 1073K in air the spheres were infiltrated with an Am nitrate solution to reach the required Am content. To reach sufficiently high Am concentration the infiltration step was repeated following an intermediate thermal treatment to evaporate excess water and convert the infiltrated Am nitrate phase to the oxide. The final Am content in each compound ahs been determined by calorimetric analysis of the beads. The required volume fraction of actinide phase was mixed with the required amount of Mo. Following addition of zinc stearate as lubricant, the mixtures were compacted with a biaxial press into pellets. Sintering was performed at 1873K for 6 hours in $Ar/H_2$. The final density of the both cermet pellets was higher than

95% of the theoretical value (TD). The main characteristics of the cermet fuels are shown in Table 2.

A comprehensive characterization of the fuel has been made, including density, thermal stability, open porosity, chemical composition, crystal structure and ceramograhic examinations. Pictures depicting microstructure of axial section of a FX5 and FX6 pellets are shown in Fig. 6. The macrographs show a homogenous distribution of isolated actinide-bearing beads.

This fabrication process is being further developed to increase the amount of Am incorporated in the actinide phase. Two options have been considered either to increase the number of successive infiltrations or to increase the porosity present in the original $PuO_2$ beads. A maximum of three consecutive infiltrations has been determined, which guarantee a good quality of infiltrated beads, free of agglomerates. The amount of americium infiltrated after three infiltrations is 25%weight which leads to a final composition of $Am_{0.244}Pu_{0.756}O_{2-x}$. The latter option has been also investigated and the porosity of the beads was increased by adding a pore former during the beads synthesis. The beads can incorporate up to 35% weight in only two infiltrations, however they are very soft and they do not keep their integrity during pelletisation and they become flat. Further investigations are required to optimize this promising process.

## CERMET Thermal properties

### 1. SPECIFIC HEAT

The Specific heat was measured by Differential Scanning Calorimetry (DSC) using a cover gas of Argon 6.0 (100 ml/min) and a heating rate of 25 K/min. The polynomial interpolation of the experimental Cp (in J $g^{-1}$ $K^{-1}$) are given by (with T in K):

$Cp\ (FX5) = 2.441616\ 10^{-11}\ T^3 - 7.605117\ 10^{-08}\ T^2 + 1.196540\ 10^{-04}\ T + 2.253980\ 10^{-1}$

$Cp\ (FX6) = 5.067667\ 10^{-11}\ T^3 - 1.471663\ 10^{-07}\ T^2 + 1.895171\ 10^{-04}\ T + 2.353187\ 10^{-1}$

### 2. THERMAL DIFFUSIVITY AND CONDUCTIVITY

Measurements of the thermal diffusivity were done by the laser flash technique. The samples were coated with carbon in order to avoid laser light reflexion at the surface of the samples. The effect of thermal dilatation on sample thickness and density were taken into account using a linear dilatation coefficient of 6 $10^{-6}$ $K^{-1}$ corresponding to the value of the Mo matrix. The measurement was done following ascending and descending temperature programmes, up to about 1450 K. No significant difference was observed between the results obtained during the ascending and descending temperature regimes.

The thermal conductivity was calculated from the measured thermal diffusivity, measured specific heat and density. The values correspond to the results as measured, without any correction for porosity.

The linear interpolations of the thermal conductivity $\lambda$ (in W $m^{-1}$ $K^{-1}$) are given by (with T in K):

$\lambda\ (FX5) = -0.0174x + 101.1264$

$\lambda\ (FX6) =-0.00768x + 57.21263$

The thermal conductivity of the CERMET fuel was modeled using an analytical result taking into account the high volume fraction of inclusions and the high difference in thermal conductivity between the matrix and the inclusions [18]. The microstructure of the CERMET corresponds to a Boolean distribution of spheres [19], with overlap (the centers of the spheres are randomly located and the spheres may interpenetrate, **Fig. 10**). Using $k_f$ for the conductivity of the inclusions, $k_m$ for the conductivity of the matrix, the upper bound of the equivalent conductivity is given by:

$$k_{max} = k_m \frac{1 + \left(\left(d-1\right)\left(p+\xi_1\right)-1\right)\beta_{fm} + \left(d-1\right)\left(\left(\left(d-1\right)p-q\right)\xi_1 - p\right)\beta_{fm}^2}{1-\left(1+p-\left(d-1\right)\xi_1\right)\beta_{fm} + \left(p-\left(d-1\right)\xi_1\right)\beta_{fm}^2}$$

where $\beta_{ij} = \frac{k_i - k_j}{k_i + (d-1)k_j}$ , $p=v_f$ , $q=1-v_f$

and d is the number of dimensions involved in the heat transfer (d= 3), and $\xi_1 = 0.5615\ v_f$ [20].

The volume fractions of inclusions considered in the calculations were 13.5, and 38.9 %, and the density was taken equal to 95%TD. For the thermal conductivity of the Mo matrix, the value from the review of Laurent [21] was used. The conductivity of the $Am_{0.224}Pu_{0.756}O_{2-x}$ inclusions was taken equal to the one of $PuO_2$ [22]. The conductivity of the voids is 0.026W/m.K [23]. The calculated and measured conductivities are compared on Fig. 3. The predictions are good for the lower volume fraction of inclusions over the whole temperature range. An over-prediction of about 10% is observed for the higher volume fraction at low temperatures.

Compared to oxide fuels, the use of a Mo matrix increases strongly the thermal conductivity of the target: even for a volume fraction of 50% of inclusions, the thermal conductivity at 1000K of the CERMET is about 10 times higher than the one of the oxide fuels like MOX [**2**]. Compared to the CERCER fuels, the operating temperature of CERMET fuels is expected to be low. This property allows the insertion of a larger amount of actinide in the target, without increasing significantly the temperature.

## Conclusion

This report has brought complementary results in relation to the safety calculations of the EFIT core studied in EUROTRANS. Analyses have shown that also with Mo matrix based fuels a good transmutation ratio can be reached. Mo based CERMET fuel has the advantage to behave favorably under transient and accident conditions. Even in case of a complete blockage of a subassembly under beam-on conditions fuel no radial damage propagation is observed in the calculations and fuel can be swept-out of the core resulting in a reactivity decrease. There is sufficient grace time to shut down the accelerator beam to achieve a final stable core configuration.

It has been demonstrated now that Mo-based CERMET fuels with Pu and Am could be fabricated according to reactor-grade requirements (for PHENIX and HFR), and that the main geometrical, chemical and physical specifications could be met. Moreover the good thermal performance was shown, based on the measurements of the thermal diffusivity and specific heat. These results were included as an input for the design of the CERMET irradiation programmes FUTURIX-FTA and HELIOS. These two irradiation programmes will now start soon in France and The Netherlands, for the period 2007-2009. The main goals are the demonstration of the in-pile mechanical and chemical stability of the fuel, the fission gas and helium release behavior, and its comparison with other fuel types such as CERCER oxides, nitrides and metallic fuels.

## Aknowledgments

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## Tab. 1 Categorization of Fuel Limiting Temperatures (BOL Fuel) According the Defense in Depth Concept

| | | CERCER | CERMET |
|---|---|---|---|
| "Melting" temperature | Matrix | 2150 K[*] | 2896 K |
| | Fuel | 2450 K | 2450 K |
| | | | |
| Category 1 | No melting/disintegration | 1750 K | 2300 K |
| Category 2 | No melting/disintegration | 1850 K | 2350 K |
| Category 3 | No melting/disintegration | 1950 K | 2400 K |
| Category 4 | No ‘melting’ for CERCER & CERMET fuels | 1950 K | 2400 K |
| DEC | Limited up to extended ‘melting’ | 2150 K | 2450 K |

[*] Matrix evaporation limit

**Fig. 1  ULOF power and reactivity trace for EFIT  two zone CERMET core**

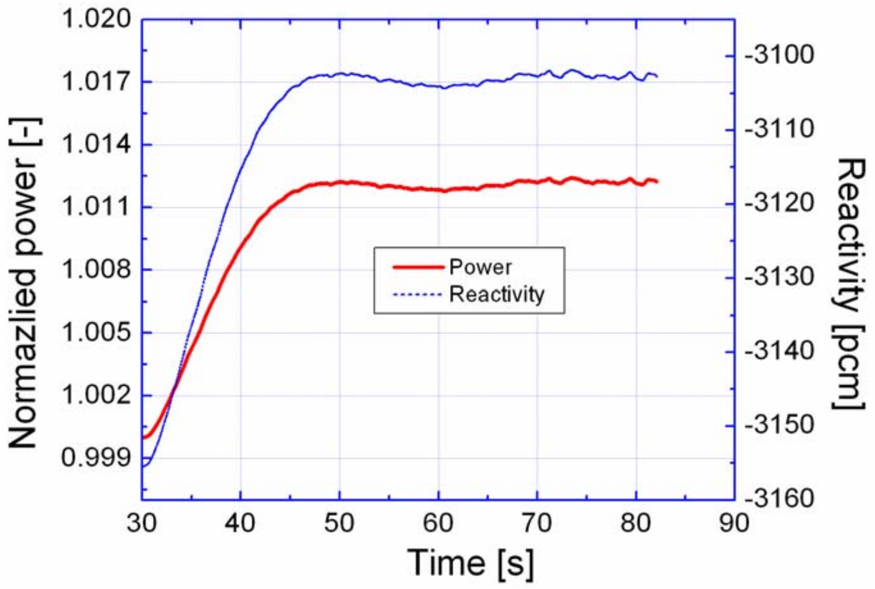


**Fig. 2 ULOF temperature development for EFIT two zone CERMET core**

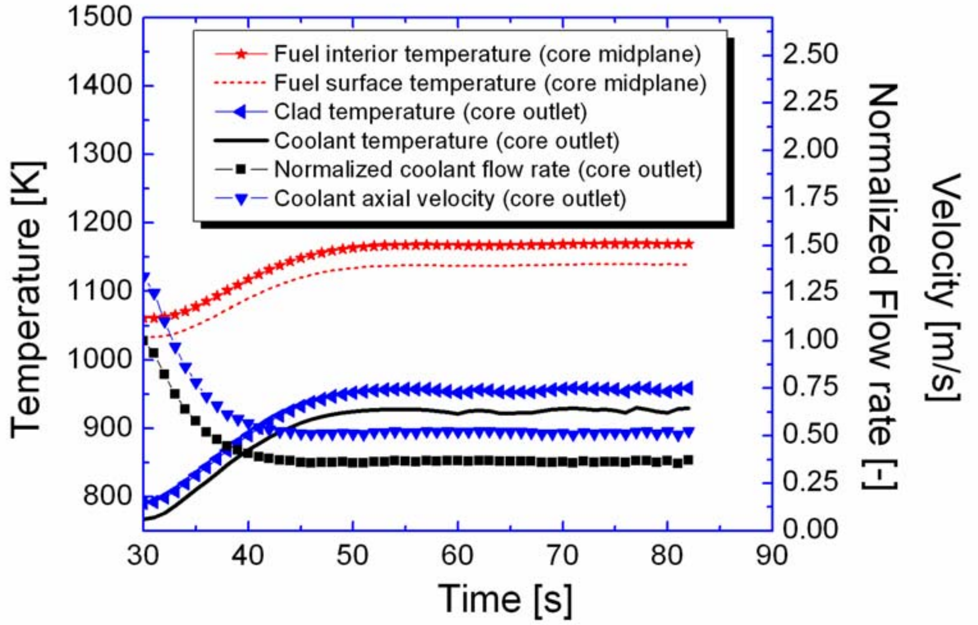


Fig. 3 Power and reactivity trace for a blockage accident in the CERMET core

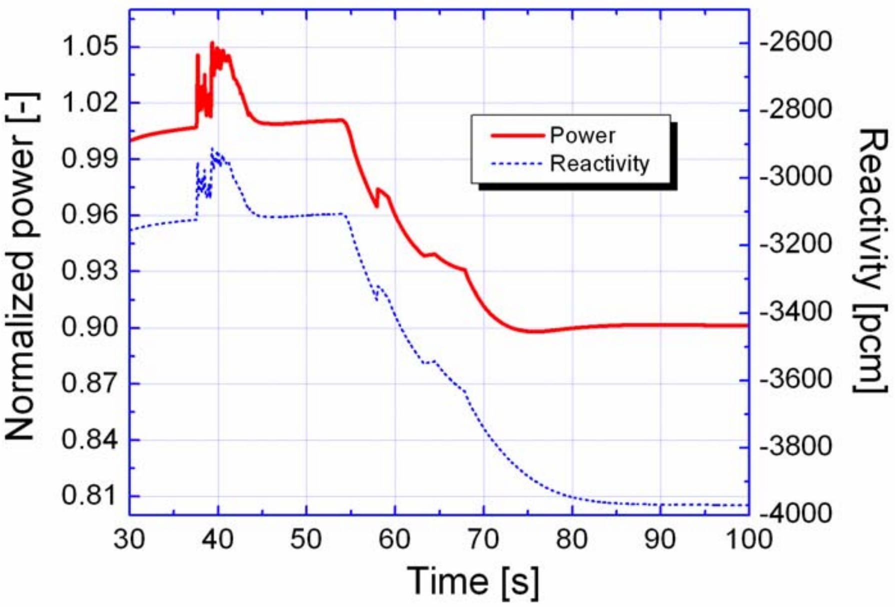


**Fig. 4 Temperature development for a blockage accident in the CERMET core**

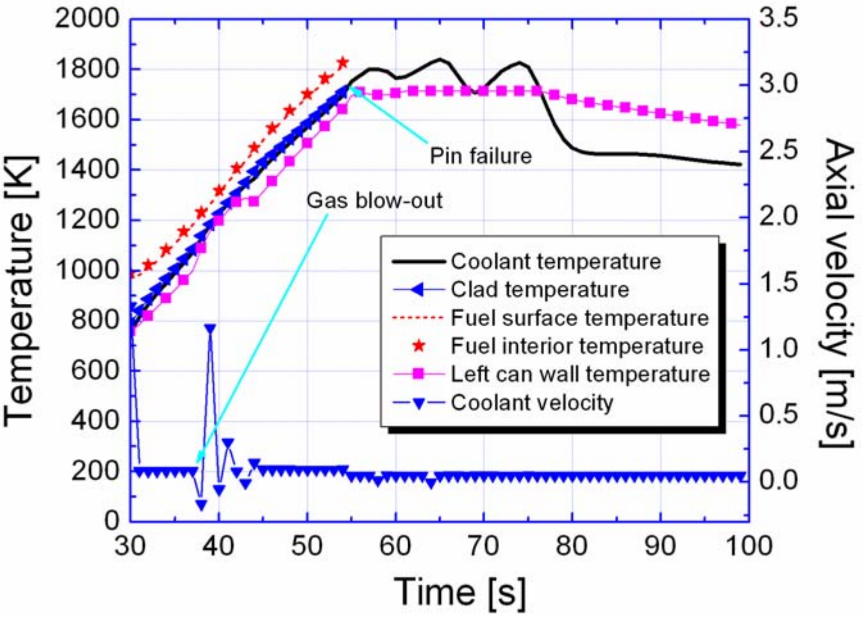
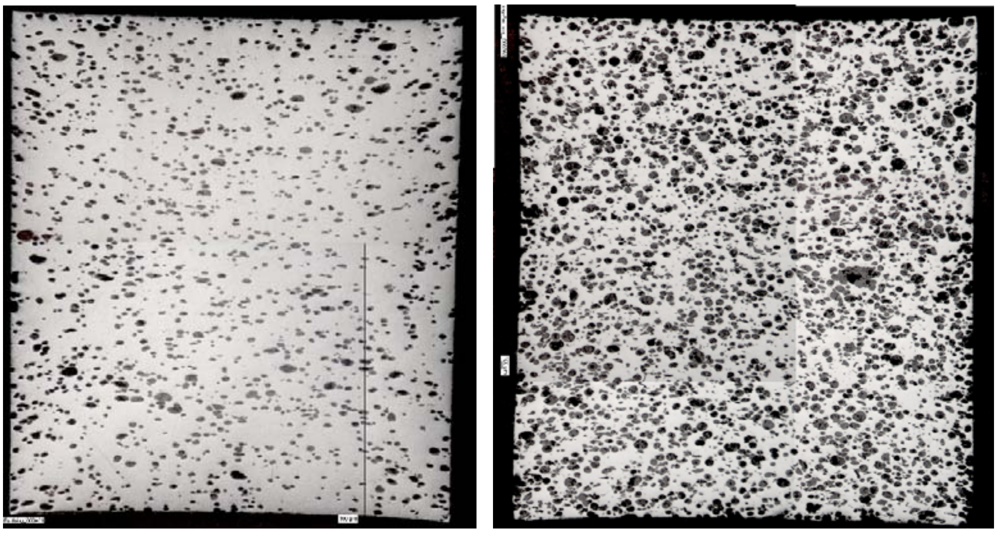


**Fig. 6 Microstructure of an $Am_{0.198}Pu_{0.797}O_{2-x}$ – Molybdenum (FX5) pellet (left) and $Am_{0.24}Pu_{0.23}Zr_{0.53}O_{2-x}$ – Molybdenum (FX6) pellet (right)**



### Table 2.- Characteristics of cermet fuels FX5 and FX6

| Actinide compound | | | | Cermet composition | |
|---|---|---|---|---|---|
| Sample | | Am infil. %weight | TD (g.cm$^3$) | Actinide Beads %vol | Density %TD |
| FX5 | $Am_{0.20}Pu_{0.80}O_{2-x}$ | 19.7 | 11.503 | 14 | 96 ± 1 |
| FX6 | $Am_{0.24}Pu_{0.23}Zr_{0.53}O_{2-x}$ | 36.8 | 8.48 | 39 | 95 ± 1 |

### Fig.7 Measured Specific heat of fuels FX5 and FX6

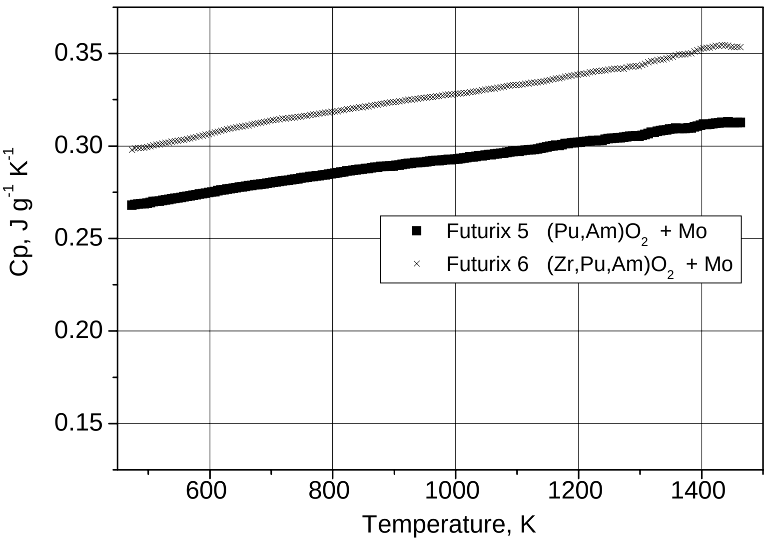


**Fig. 8: Thermal diffusivity of fuels FX5 and FX6 corrected for sample thickness change**

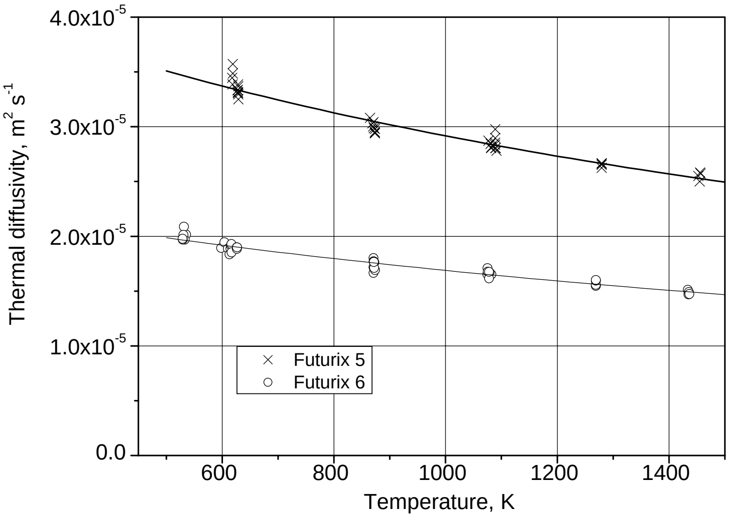


**Fig. 9: Thermal conductivity of fuels FX5 and FX6**

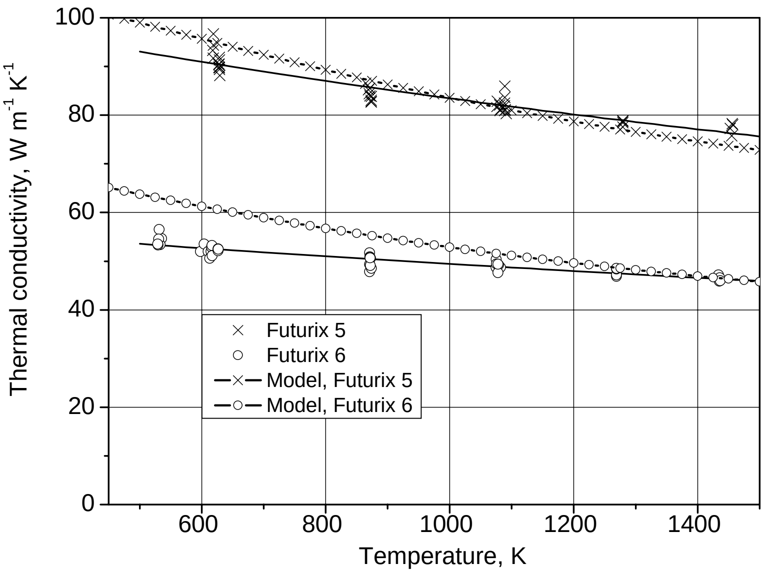


**Fig. 10 Modeled microstructure of the CERMET: Boolean distribution of spheres with overlap, the volume fraction of inclusions is 30%**