

Experimental study of Joint-Oxide-Gaine (JOG) systems in Fast Neutron Reactors

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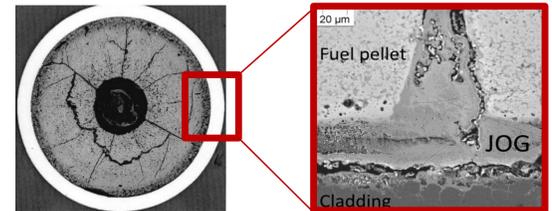
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Context of the study

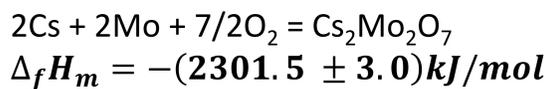
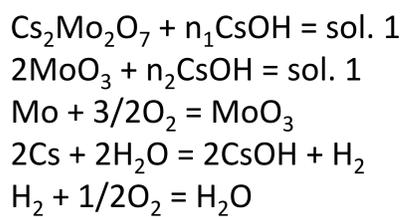
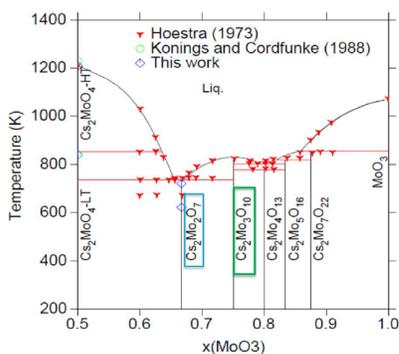
The present study was performed in the framework of the European **INSPIRE** project, dedicated to the investigation of the (U,Pu)O₂ fuel behaviour under irradiation in Generation-IV Sodium-cooled Fast Reactors (SFRs) and Lead-cooled Fast Reactors (LFRs) [1]. Due to the very high thermal gradients in SFRs and LFRs ($\sim 450 \text{ K}\cdot\text{mm}^{-1}$), the volatile fission products generated during irradiation (e.g. Cs, I, Te, Mo) migrate from the center of the fuel pin ($\sim 2300 \text{ K}$) towards the pellet edge ($\sim 973 \text{ K}$), and accumulate in the space between fuel and cladding, forming an oxide layer of a few hundred micrometers, the so-called **JOG layer (Joint Oxide Gaine)** [2].

The JOG is a very-complex multicomponent system, including Cs₂MoO₄ (major phase), Cs₂Te, CsI, Cs₂I₂, but the exact composition is still not well-known. This work aims to obtain reliable experimental data for this multi-element system and use them to develop thermodynamic models based on the CALPHAD methodology.

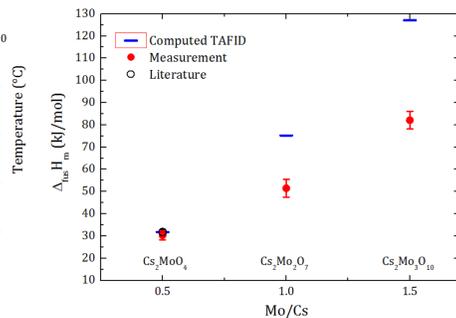
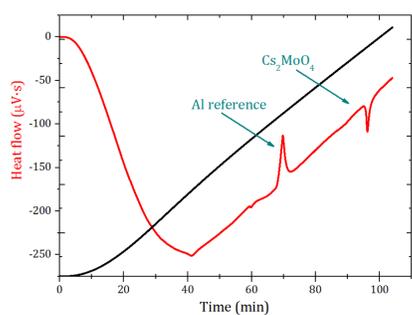


Study of the Cs-Mo-O system

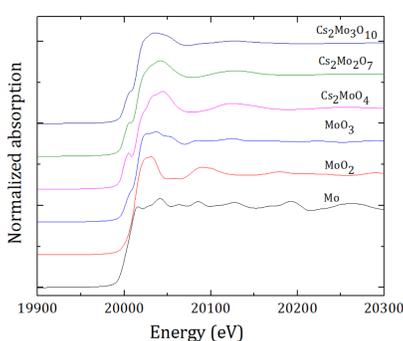
- **Enthalpies of formation** of Cs₂Mo₂O₇ and Cs₂Mo₃O₁₀ were determined using solution calorimetry.



- **Enthalpies of melting** of key compounds were measured using differential scanning calorimetry (DSC). These differed in some cases quite substantially from computed values in the available CALPHAD model [3].



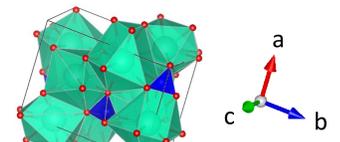
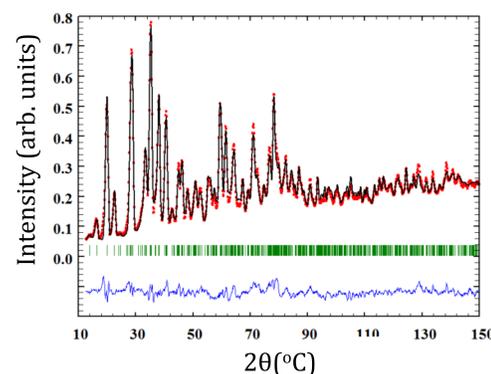
- **Valence state determination and local structure studies** were performed using XANES/EXAFS at the Mo-K edge (ESRF, France, Grenoble).



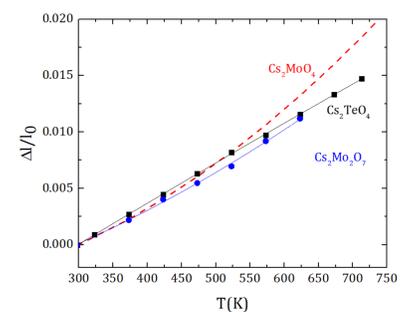
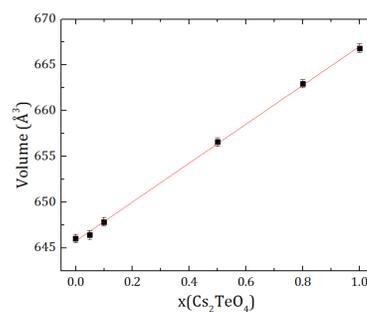
The results confirmed the Mo(VI) valence state in Cs₂MoO₄, Cs₂Mo₂O₇, Cs₂Mo₃O₁₀. A pre-edge feature [1s(Mo) → 4d(Mo)+2p(O)] was also observed, characteristic of tetrahedral MoO₄ symmetry or distorted MoO₆ octahedra.

Study of Cs₂(Te,Mo)O₄ solid solutions

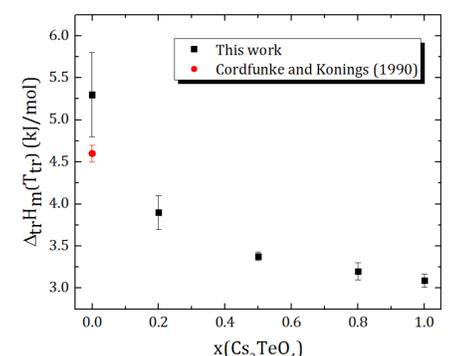
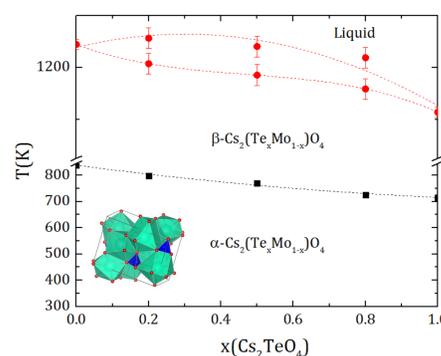
- **Structural investigations** were performed using neutron diffraction at the PEARL beamline. Cs₂TeO₄ is isostructural with Cs₂MoO₄, i.e. orthorhombic in space group *Pnma*.



- **Lattice parameters evolution with composition and temperature** were investigated using X-ray diffraction. The thermal expansion of Cs₂TeO₄ ($\alpha_V \sim 107.9 \cdot 10^{-6} \text{ K}^{-1}$) is much higher than that of UO₂ ($\alpha_V \sim 32.4 \cdot 10^{-6} \text{ K}^{-1}$) [4].



- **Phase diagram data in the Cs₂TeO₄-Cs₂MoO₄ section and enthalpy of transitions** were obtained by DSC.



Conclusions

Using these data as input for CALPHAD thermodynamic models, the composition and amount of formed phases in the JOG can be predicted as a function of oxygen potential and temperature.

[1] www.eera-jpnm.eu/inspire/

[2] M. Tourasse et al., J. Nucl. Mater. 1800 (1992) 49-57

[3] TAF-ID, www.oecd-nea.org/science/taf-id/

[4] A.C. Momin et al., J. Nucl. Mater. 185 (1991) 308-310