Thermal conductivity of liquid UO₂ near the melting point

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Experimental determination of the thermal conductivity of liquid UO₂ is extremely difficult due to its high melting temperature (T_m =3120 K), its fast non-congruent vaporization in the vicinity of the melting point and possible chemical interaction with the crucible. In fact, the experimental values measured by different authors range from ≈ 2 to 12 W m⁻¹ K⁻¹, that indicates the presence of unknown severe perturbations in some of the experiments carried out so far.

Self crucible steady-state laser-melting [1, 2] - that can provide the cleanest melting conditions - requires sophisticated mathematical analysis along with exact knowledge of the position of the solid-liquid interface, as well as presumption of non-displacement on the liquid surface. The present state of the problem was summarized by Ronchi [3] where by detailed analysis of the thermal diffusivity dependence on temperature he suggested the value of $2.5 \pm 1 \text{ Wm}^{-1} \text{ K}^{-1}$ at the melting point.

This paper presents the results of experimental measurements by a new, sensitive method, based on controlled laser pulse heating. The sample was mounted in a small autoclave filled with 1 bar argon. A one-second long laser pulse was applied with a complex power-time shape: the power was first continuously increased to raise the surface temperature above T_m and was then terminated by a following constant power pulse established to ensure conditioning of the sample cooling-rate. The temperature measurements were realized by using both high-speed monochromatic and spectral pyrometers.

The experiment shows that the temperature vs. time evolution in the ascending part of the thermogram for $T>T_m$ is almost the same as for $T<T_m$ indicating that *no knee point is observed at melting point*. This is a first indication that thermal conductivity does not undergo discontinuities across the solid/liquid transition. Optical metallographic examination of the sample cross-section after freezing shows that the thickness of the molten zone was of the order of 150 µm, and almost constant over the area of the laser beam spot.

Finally, the heat transfer problem was simulated by a 2D numerical model. A sensitivity study, performed in order to assess the influence of the liquid thermal conductivity on the front face central-point thermogram, shows that it is possible to deduce the conductivity of the liquid. An analysis of experimental curves clearly indicates that the liquid conductivity near the melting point is equal, within ± 0.5 W m⁻¹ K⁻¹, to that in the solid just below T_m. This confirms that the conductivity of liquid UO₂ corresponds to the lowest values measured in the past.

- 1. H A Tasman, D Pel, J. Richter and H E Schmidt, *High Temp-High Press.* **15** (1983) 419-31
- 2. H A Tasman, Thermal conductivity of liquid UO₂, *Commission* of *the European Communities, Joint Research Centre, Annual Report TUAR88* (1988) Karlsruhe, Germany
- 3. C Ronchi, J. Phys.: Condens. Matter 6 (1994) L561