Specific heat measurements on transuranium systems at ITU Karlsruhe

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The Physical Properties Measurement System (Quantum Design) has been recently installed at ITU Karlsruhe. The installed options enable to measure the electrical resistivity and the heat capacity in the temperature range between 0.4 K and 380 K, in magnetic fields up to 9 T. The heat capacity is determined by a relaxation method. Here, we present an overview of possibilities for specific-heat measurements on samples containing transuranium elements.

The most straightforward way represents a direct measurement of the sample without any additional capsule, like for usual non-active material. However, this represents a strongly enhanced probability of a contamination. Therefore, we tested possibility of heat-capacity measurements of encapsulated samples, that is a possible way to how measure safely the transuranium materials.

The requirements for the capsules are clear: its mass should be as small as possible, made of a material with magnetic-field independent heat capacity, that is small at low temperatures, where most interesting physics appears in our research area, should be easy to handle and of course, should give a good protection against contamination.

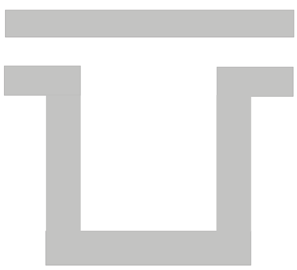

Fig. 1. Schematic drawing of the heat-capacity capsule; the inner space is cylindrical, typically 2 mm in diameter and 2 mm high.

We have tested capsules of different shapes made of two different materials: sapphire and boron nitride (BN). We found the BN capsules not suitable for our experiments. It has a relatively large specific heat at low temperatures and also its mechanical properties are not ideal. On the other hand, the sapphire - material often used in heat capacity measurements, shows excellent properties. Concerning the shape, we found as the best a cylindrical capsule shown schematically in Fig. 1. To ensure the best safety, we close the capsule by the stycast 2850 FT. This glue exhibits also very good thermal and mechanical properties down to low temperatures. We have measured the specific heat of the stycast itself in detail.

The specific heat of the sample is determined by subtracting all contributions coming from the sample platform, the capsule and the glue.

We shall note, that the measurement is done on a relatively complex system (sample + sample platform with grease + capsule + stycast). The two-τ fit used in the PPMS may be unable to fit the relaxation behaviour of such a system. Therefore, we had to verify the reliability of specific heat data obtained in this way. We have tested several materials displaying different types of specific-heat behaviour. The pure gold metal that has a very low γ-value of the electronic specific heat, URhAl with an enhanced γ-value and a second-order type magnetic phase transition, or UO_2 displaying a second-order type phase transition. We observe generally a good agreement between the data obtained without any capsule and for encapsulated samples as can be seen from Fig.2. The difference almost never exceeds 10%, and is below 5% for 90% of the data. The data obtained in external magnetic field show a similar agreement. Fig. 3 shows certain overview of the heat capacity of individual elements.

First measurements on transuranium compounds (Np- and Pu-based) have been already performed at ITU, and we get reliable data. As an example, we present the specific heat of $NpCoGa_5$ in Fig. 4.

In the case of samples containing Pu, we observe a strong self - heating effects that disable measurements down to lowest temperatures. These effects depend on the amount of Pu: the first reliable point is measured only at 3 K (2 mg of $PuGa_3$), 4.5 K (4.5 mg of $PuGa_3$) or even 6 K (10 mg of $PuGa_2$).

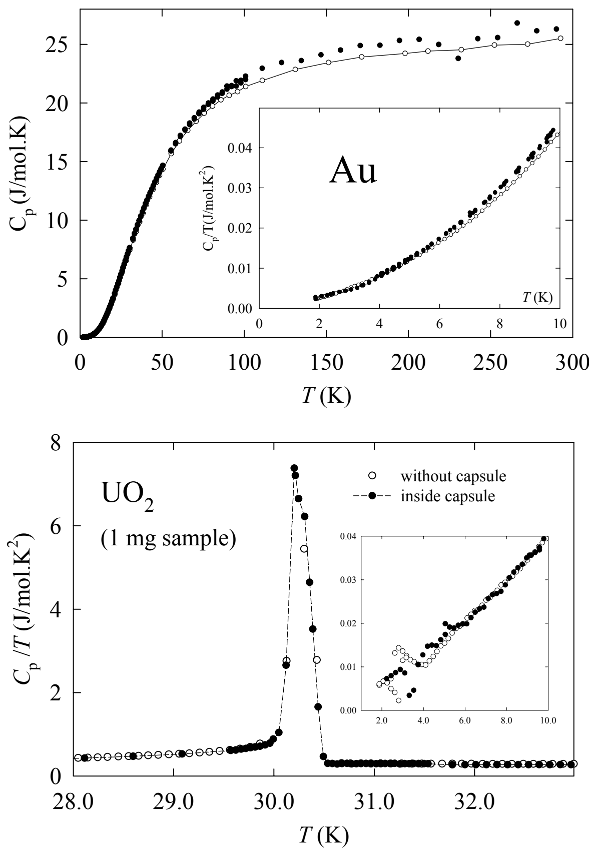

Fig. 2. Comparison of the specific heat of Au (m = 7.3 mg) and UO_2 (m = 1 mg) measured without any capsule (empty circles) and inside the capsules (filled circles).

We can conclude that the heat capacity measurements on the encapsulated transuranium samples are possible at ITU. The experimental error increases in comparison to the usual measurement, but remains acceptable in most cases. Of course, it depends strongly on the intrinsic specific heat of the given compound. The additional heat capacity of the capsule is magnetic-field independent.

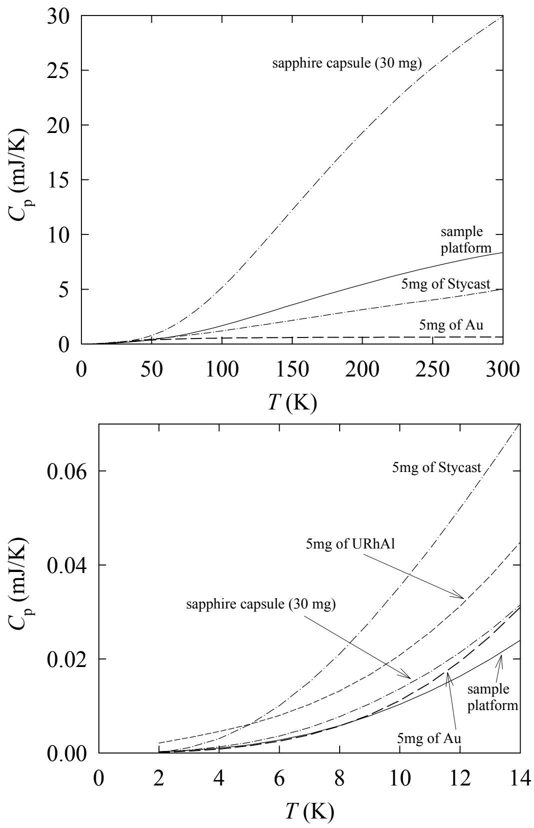

Fig. 3. Heat capacity of the capsule compared to that of selected samples and the sample platform.

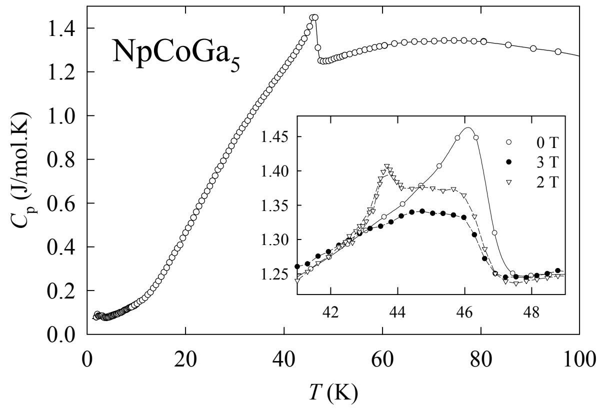

Fig. 4. Specific heat of $NpCoGa_5$.