ESRF	**Experiment title:** Extension of the melting curves of Ta and Pb beyond 1 Mbar	**Experiment number**: HS-3829
Beamline: ID27	**Date of experiment**: from: 2/10/2009 to: 7/10/2009	**Date of report**: 10/12/2009
Shifts: 15	**Local contact(s)**: M. Mezouar	*Received at ESRF:*

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Report:

This work is the continuation of HS-3159 and HS-3532 (15 shifts allocated).
Our main goal was to confirm, or infirm, the discrepancy between the melting curves of tantalum measured in laser-heated diamond anvil cell, LHDAC and in shock-compression experiments (see [1], [2], **Figure 1**). The reasons for this discrepancy have been largely debated in the recent literature [3]; in particular, ab initio calculations confirmed shock-compression melting point [2].
During this experiment, various sample assemblies have been used in the conditions summarized in Table 1. The sample was a tantalum foil of ~5 microns thickness or sample powder grains. New pressure transmitting media, MgO and Al_2O_3, have been tested because the ones used in our previous experiment (HS-3532: Ne, Ar, NaCl) reacted with Ta, or did not prevent the reaction of Ta with the diamond anvil (to form TaC). In some runs, diamond anvils coated with a few microns of alumina have been used.
The pressure was first increased to the desired range. Then it was gradually heated by the two lasers; X-ray and pyrometry signal recorded every ~4s during each heating series. Several heating series were performed at each pressure step. Alignment of X-ray, laser beams and pyrometry were checked for each heating series.

Run	Anvils culet size (μm)	Al_2O_3 coating	P range (GPa)	T range (K)	Pressure medium	Gasket	Importance of Ta-C reaction	Ta-pressure medium reaction
5	200 x 300	N	55-60	300-~5000	Al_2O_3	Re	XXX	XX
6	100 x 300	N	70-120	300-~5000	MgO	Re	XXX	
7	70 x 300	N+pits	~80	300-~4000	NaCl	Re	X	XX
8	200 x 300	N	60-70	300-~4500	MgO	Re	XX	
9	200 x 300	Y	60-70	300-5000	Ne	Re	X	
10	150 x 300	Y	40	300	Ne	Re+ Cu	No heating – gasket instability	No heating
11	100 x 300	Y	50-65	300-3000	MgO	Re	XXX	

Table 1: conditions of each experimental run.

The current experiments, together with the previous HS-3532 run, show that the Ta-C reaction cannot be completely prevented, but only minimized by a careful sample preparation (the sample should not touch the anvils) and at best, with the the use of rare-gas solids as pressure media. Al_2O_3 coating of diamond anvil are not efficient to protect them, because Al_2O_3 also strongly reacts with Ta.
The melting criterion was the observation of diffuse x-ray scattered signal; the P/T conditions of melting were determined using pyrometry and the equation of state of tantalum. The pyrometry temperature was checked

using Ta high P-high T EoS. Temperature measurement was considered reliable when pyrometry and EoS temperatures were similar and forming a plateau.

During run 5, only one melting point could be measured before the complete reaction of Ta with both Al_2O_3 and C. During runs 6, 8, 11, several temperature plateaus on the T *vs.* laser power curve have been observed, but no X-ray evidence of melting was measured; we thus believe that these plateaux corresponded to the Ta+C->TaC reaction rather than the melting of tantalum. The best results in this experiment have been obtained with Ne pressure medium, but it has the drawback of not fluorescing under X-ray irradiations, which makes the alignment procedure of X-ray and laser beam less precise. In addition, the melting of neon around Ta results in large changes of the Ta sample geometry, which is not desirable. Overall we found that the best pressure media are neon and argon (used above ~60 GPa: below, it reacts with Ta). Microprobe analysis performed on samples 5 and 6 showed that no alloying between tantalum and rhenium gasket occurred (amount of rhenium in the tantalum sample smaller than the detection limit, ~1at%), despite the closeness of the sample and the gasket. Reaction with the gasket thus does not seem to be an issue in our experiments.

Despite these numerous experimental difficulties we have been able to measure a few melting points in the 50-120 GPa pressure range. Several heating series showed that no melting occurred for Ta largely above the previously published melting curve [1]. Our melting points are in reasonable agreeement with theoretical predictions [2]. The P-T conditions of our runs, with the melting observation, are summarized Figure 1. We suspect that previous results [1] could have been biased by undetected chemical reaction between Ta and/or diamond anvil. In fact, these results were obtained by visual observation of melting in LHDAC, and the recording of a temperature plateau. The current work emphasizes the necessity of an *in situ* x-ray diagnostic to undertand the changes undergone by laser-heated samples in diamond anvil cells, in particular for the study of melting phenomena. We believe that we have found the best experimental conditions for the tantalum melting problem and we wish to pursue this work to obtain more reliable melting points.

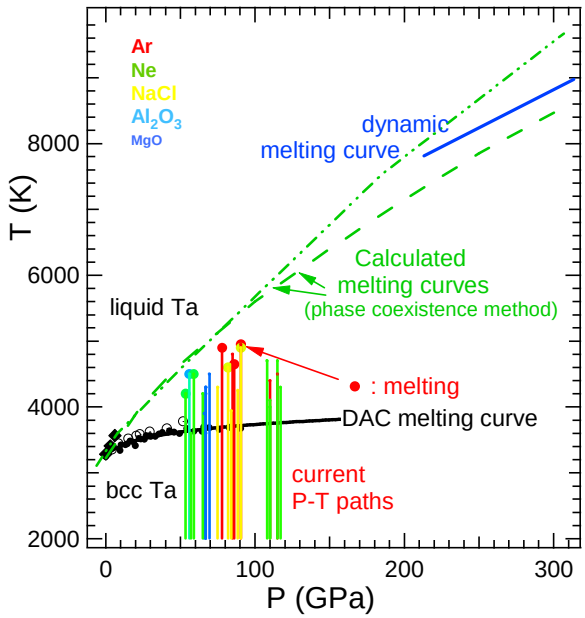

Figure 1: current P-T paths for various pressure media (Ar, Ne, NaCl, Al_2O_3, MgO) and melting points compared to previously measured/calculated melting curves [1,2]. The paths which do not finish with a circle correspond to heating series in which no melting has been observed. The temperature uncertainty is approximately 500 K.

References :

[1] D. Errandonea *et al.*, Phys. Rev. B **63**, 132104, 2001
[2] S. Taioli *et al.*, Phys. Rev. B 75, 214103, 2007 ; Z.-L. Liu *et al.*, Phys. Rev. B 77, 024103, 2008
[3] C.J. Wu *et al.*, Nat. Mat. 8, 223, 2009