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# Liquid gadolinium corrosion study of TaC coating on tantalum

Nagaraj Alangi*, Jaya Mukherjee

*Laser & Plasma Technology Division, Bhabha Atomic Research Centre, Mumbai 400085, India*

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ABSTRACT

Spectroscopic information of gadolinium isotopes is necessary to exploit the full potential of various isotopes in nuclear and medical applications. Container materials to handle liquid gadolinium during spectroscopic studies is an important aspect. Tantalum carbide (TaC) has excellent stability and also exhibits superior resistance to attack by various reactive actinide metals at high temperature for long durations. Tantalum crucibles with TaC coating were prepared and tested for compatibility against liquid gadolinium at 1673 K upto 14 h in vacuum. Optical microscopy and SEM/EDS investigations were done to evaluate the micro structural features of the coating and the liquid gadolinium attack. Experimental results show that TaC coating exhibits excellent corrosion resistance against liquid gadolinium at 1673 K.

## 1. Introduction

An increased interest in the 152Gd isotope over the last decade has been associated with its potential use as burnable poison owing to its special nuclear properties. Other isotopes of gadolinium also find applications in neutron radiography, to target tumors in neutron therapy and gadolinium compounds as intravenous MRI contrast agent. Gadolinium also serves as surrogate of actinides to study selectivity aspects by spectroscopy. The key resolve of a spectroscopy activity is to obtain and enhance the essential knowledge of the basic parameters distinguishing isotopes. Gadolinium is also widely used as a surrogate in actinide spectroscopy for studying the vapor characteristics and plasma behavior.

All the engineering components required for handling liquid gadolinium in spectroscopic and vapor generator studies demand a protective barrier on metallic components as molten gadolinium is highly corrosive. Typically, these components are exposed to liquid gadolinium at 1673 K for duration up to 12–14 h. Components used for such applications should possess excellent resistance to liquid gadolinium and at the same time should not introduce any undesirable impurity into the gadolinium which might interfere in spectroscopic studies. The phase diagrams of various refractory metals with gadolinium, indicate finite solubility of refractory of metal in gadolinium at 1673 K, which makes them unfit to use in direct contact with liquid gadolinium [1]. Liquid gadolinium is highly reactive and most of the refractory metals are susceptible to inter-granular attack that can cause catastrophic failure of the component. Tantalum and tungsten both have good high-temperature strength and low solubility in molten gadolinium at process temperatures of 1673 K. Tantalum has been used a cathode material in the electrolysis of gadolinium [2]. However, tantalum is not inert against liquid gadolinium and requires a protective coating.

Thermodynamic calculation, as shown in Fig. 1, indicates that most of the common ceramic oxides such as $Al_2O_3$, $ZrO_2$, MgO, BeO and $HfO_2$ react with gadolinium metal to form $Gd_2O_3$, $Y_2O_3$ being an exception. Yttrium oxide was also reported to be used against liquid uranium for its thermodynamic stability [3]. The data on the compatibility of various refractory metals and ceramic materials with liquid gadolinium is summarized in Table 1. Although yttrium oxide is superior in high temperature corrosion resistance, engineering components for spectroscopic studies are complex in construction and monolith shapes of ceramic materials are very difficult to fabricate. Instead a protective ceramic coating on metallic components of desired shapes, is a more feasible option.

Tantalum carbide has been proven to provide corrosion resistance against actinide metal at elevated temperature [4]. As shown in Fig. 2, free energy change calculations indicate that both TaC and $Ta_2C$ are non-reactive to uranium. Hence, TaC with a melting point of 4153 K is a prospective choice for handling liquid gadolinium. The objective of this work is to prepare a tantalum substrate with TaC coating and evaluate this as a container material for liquid gadolinium handling, which in turn will be used as an atom vapor source in spectroscopy.

## 2. Materials and experimental setup

The schematic of the experiment assembly is shown in Fig. 3. The corrosion experiments were conducted in a one end closed alumina

* Corresponding author.
*E-mail address:* nagaraj@barc.gov.in (N. Alangi).

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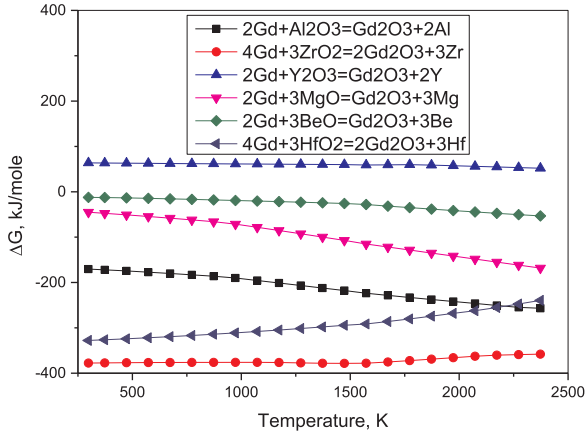



**Fig. 1.** Free energy change for reactions with liquid gadolinium and few oxides.

**Table 1**
Materials and their compatibility with liquid gadolinium.

| S.no | Material | Melting point (K) | Compatibility with uranium |
|---|---|---|---|
| 1. | Tantalum | 3290 | Solubility of ~0.1 wt% at 1673 K |
| 2. | Tungsten | 3695 | Solubility of ~0.025 wt% at 1673 K |
| 3. | Molybdenum | 2896 | Solid solution formation at 1673 K |
| 4. | $Al_2O_3$ | 2323 | Reduction reaction by gadolinium up to 2300 K |
| 5. | ZrO2 | 2900 | Reduction reaction by gadolinium up to 2300 K |
| 6. | $Y_2O_3$ | 2683 | Stable against gadolinium up to 2300 K |
| 7. | MgO | 3125 | Reduction reaction by gadolinium up to 2300 K |
| 8. | BeO | 2780 | Reduction reaction by gadolinium up to 2300 K |
| 9. | $HfO_2$ | 3031 | Reduction reaction by gadolinium up to 2300 K |

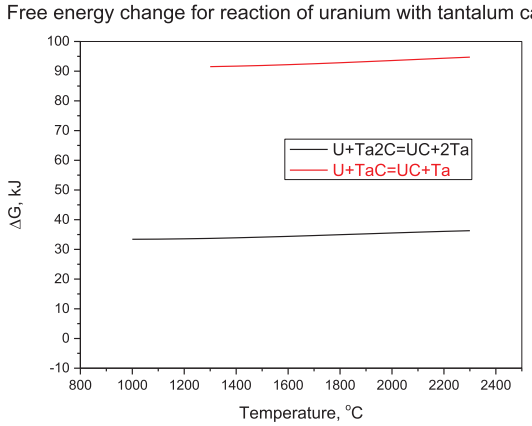
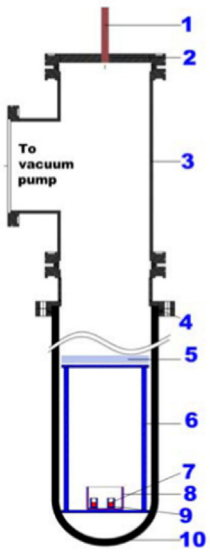



**Fig. 2.** Free energy change for reaction of with liquid uranium with carbides of tantalum.

tube with the open end connected to the vacuum system with the help of a tee. The closed end of the tube was placed inside a resistively heated furnace. Vacuum of 1E-6 mbar was maintained by an oil diffusion pump backed by rotary pump. A pack of radiation shields ensure a uniform temperature over 150 mm length in the hot zone. A tantalum cage was used to support the tantalum crucible containing gadolinium. Tantalum rods as per ASTM B708 was used to prepared crucibles for the experiments. Gadolinium of purity > 99.9% was used to study the compatibility. Crucibles were of 5 mm ID, 8 mm OD and 11 mm height with uniform thickness of 1.5 mm on all sides. TaC coating (10–20 μm thick) was formed inside this tantalum crucible by pack carburizing and




1. Stainless steel rod
2. Sealed top flange
3. Tee
4. O ring seal of alumina tube
5. Radiation shields
6. Tantalum cage
7. Gadolinium metal
8. Yttria coated tantalum crucible
9. TaC coated tantalum crucible
10. Alumina tube

**Fig. 3.** Schematic of experimental assembly.

contained about 0.7 g of gadolinium. Two crucibles with gadolinium were used in each experiment. Pre-experimental qualification for defect free crucibles was done by optical microscopy, and thermal cycling tests of a few crucibles of the batch. Detailed description of the experimental setup is discussed elsewhere [3].

## 3. Experimental procedure

The heating, cooling and holding cycles adopted in the experiments consisted of heating the setup to 873 K at 200 K/h and holding for 2 h for complete outgassing of the system. Heating was continued at 200 K/h to 1673 K to melt gadolinium and held there for 4 h and 14 h. Post experiment analysis includes observation of full crucible area under an optical microscope for defects. Defect free crucibles were cold mounted and cut with a diamond wheel of 300 μm thickness to observe the cross-section. TaC/Gd liquid interface analysis was conducted at the bottom of the crucible where gadolinium was in contact with the TaC. A Zeiss EVO 40 SEM was used along with Oxford EDX analyzer for micro-structural and chemical analysis. Photographs of inner cavity TaC coated tantalum crucibles are shown in Fig. 4.

Carburization experiments of tantalum on small test coupons were


**Fig. 4.** Tantalum crucibles with TaC coating formed inside, by pack cementation.