

High temperature interaction between liquid Gd-containing alloys and selected oxides

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The sessile drop method was applied for investigation of high temperature interaction of liquid Gd, Ti, binary Gd-Ti and ternary Gd-Ti-Zr alloys in contact with dense polycrystalline Y_2O_3 and $ZrO_2+3\%Y_2O_3$ substrates. During high temperature tests performed under flowing Ar, the contact angles were determined from the images of drop/substrate couples recorded with CCD camera. The effect of metal chemistry on its melting, wetting behaviour and reactivity in contact with oxide substrates are analysed.

Keywords: wettability, reactivity, Gd-alloys, oxides

1. Introduction

Gadolinium (Gd) is widely used in advanced alloys to improve their functional and utility properties. However, high melting temperature, high oxygen affinity and chemical aggressiveness against most available refractory materials makes difficult melting, casting as well as high-temperature testing of Gd-containing alloys using container-assisted methods (e.g. calorimetric studies, measurements of surface tension of liquid alloys) [1,2].

The paper is focused on investigation of high temperature interaction between liquid Gd-containing alloys and selected oxides.

2. Experimental

The materials were pure Gd (99.95%), Ti (99.98%) and their binary and ternary alloys produced by arc melting (Table 1). Dense polycrystalline oxide substrates were made from Y_2O_3 and ZrO_2 powders by HT-HP synthesis and polished up to a roughness of about 120 nm.

Real-time melting, wetting, spreading and solidification behaviour of selected Gd/oxide systems were examined by the sessile drop method [3,4] using experimental facility described in [4,5].

During high temperature tests, the selected metal/substrate couples were contact-heated to the test temperature under protective atmosphere (flowing Ar) and their images were recorded with a high-speed high-resolution CCD camera. The test temperatures were about 50 to 100°C above the melting points of the metal samples.

The structure and chemistry of solidified couples were examined both on the top surfaces and on cross-sections using optical microscopy (OM) and scanning electron microscopy (SEM) coupled with energy dispersive X-ray analysis.

3. Results

Wetting phenomenon was observed between Y_2O_3 substrate and pure Gd immediately after its melting (1312°C) showing $\theta \sim 65^\circ$. Upon 2 min heating to 1360°C, a contact angle degreased to a final value of 33°. Similar wetting behavior was observed for $Gd_{60}Ti_{40}$ alloy at 1473°C.

Table 1. Contact angles of selected Gd/oxide couples

Metal	Substrate	Temp. °C	$\theta, ^\circ$
Gd	Y_2O_3	1360	33
$Gd_{60}Ti_{40}$	Y_2O_3	1473	33
$Gd_{30}Ti_{70}$	Y_2O_3	1500	157
Ti	Y_2O_3	1760	77
Gd	$ZrO_2+3\%Y_2O_3$	1362/1412	68
Ti	$ZrO_2+3\%Y_2O_3$	1720/1770	122
$Gd_{60}Ti_{40}$	$ZrO_2+3\%Y_2O_3$	1697	92
$Gd_{40}Ti_{60}$	$ZrO_2+3\%Y_2O_3$	1760	80
$Gd_{40}Ti_{30}Zr_{30}$	$ZrO_2+3\%Y_2O_3$	1650	104
$Gd_{40}Ti_{30}Zr_{30}$	$ZrO_2+3\%Y_2O_3$	1660	101
$Gd_{40}Ti_{40}Zr_{20}$	$ZrO_2+3\%Y_2O_3$	1690	95
$Gd_{40}Ti_{20}Zr_{40}$	$ZrO_2+3\%Y_2O_3$	1700	97
$Gd_{40}Ti_{20}Zr_{40}$	$ZrO_2+3\%Y_2O_3$	1717	94

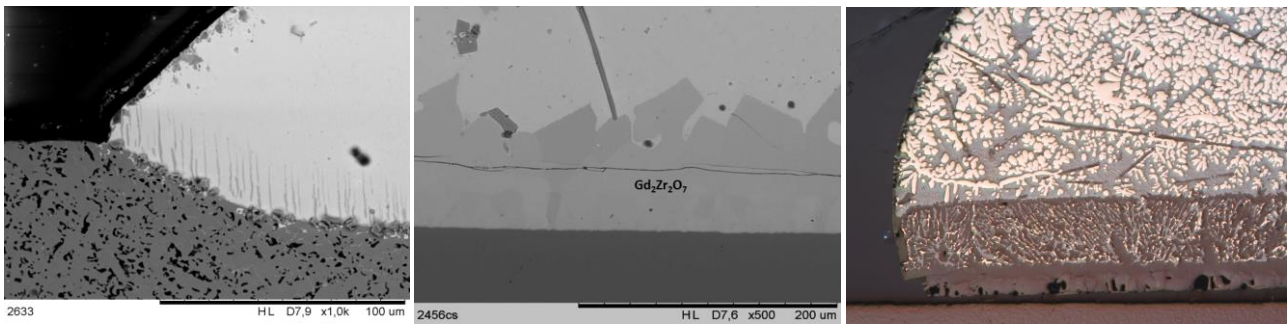


Fig. 1 Microstructures of cross-sectioned sessile drop couples:
a) Gd/Y₂O₃; b) Gd/ZrO₂+3% Y₂O₃; c) Gd₄₀Ti₄₀Zr₂₀/ZrO₂+3% Y₂O₃; a,b - SEM; c - OM

Y₂O₃ was also wetted by pure Ti, contrary to the high Ti-containing alloy Gd₃₀Ti₇₀ showing non-wetting behavior and a very high contact angle.

The ZrO₂ substrates containing 3% Y₂O₃ exhibited much worse wettability with both pure metals and their alloys. These substrates were not wettable by pure Ti at the experimental temperatures of 1720°C and 1770°C, contrary to pure Gd showing a good wetting at 1362°C and 1412°C. Alloying Gd with 40% Ti caused significant increase in the contact angle and non-wetting behavior at 1697°C. Further increase of the Ti content to 60% made ZrO₂+3% Y₂O₃ substrate wettable at 1760°C. For Gd-Ti-Zr alloys with 40% Gd, when Ti was partially replaced by Zr, no wetting was observed at selected temperatures.

Structural characterization of solidified couples revealed significant difference in the mechanisms of high temperature interaction in the studied systems. In the Gd/Y₂O₃ couples, the interaction is dominated by a strong dissolution of the substrate in the liquid metal resulting in the formation of a deep crater in the substrate, alloying Gd drop with Y and the growth of directionally solidified interfacial region during cooling (Fig. 1a). On the contrary, the interaction of both pure Gd and selected Gd-containing alloys with ZrO₂+3% Y₂O₃ substrates is controlled by the formation of continuous interfacial reaction product Gd₂Zr₂O₇ (Fig. 1b).

Additionally in the Gd-Ti-Zr/ZrO₂+3% Y₂O₃ couples, a Gd-rich layer is evidenced in the Gd-Ti-Zr drop near the reactively formed interfacial layer while the other part of the drop is almost free of Gd (Fig. 1c). This effect is related to the liquid metal de-mixing since the Gd-Ti-Zr alloys exhibit miscibility gap in the temperature range studied in this work.

4. Summary

Dissolutive wetting mechanism is responsible for a good wetting of liquid Gd and Gd-rich alloys on Y₂O₃ substrates.

Wetting phenomenon in the Gd/ZrO₂+3% Y₂O₃ couple is dominated by the formation of continuous and wettable layer of a new phase Gd₂Zr₂O₇ at the interface.

Alloying Gd with Ti or Ti+Zr reduces reactivity of liquid metal with both Y₂O₃ and ZrO₂+3% Y₂O₃ substrates thus resulting in higher contact angles.

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