

High temperature behaviour of liquid Ni-based alloys with oxide ceramics

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High temperature interaction between liquid Ni and its alloys (NiCo10, NiCr10, NiW10, Inconel 740, Haynes 282) and refractory oxide-based ceramics containing Al₂O₃, MgO, MgAl₂O₄, Mg₂SiO₄, ZrO₂ was examined by a sessile drop method at 1500°C in argon. A specially developed procedure for *in situ* opening of the metal/ceramic interfaces directly at the testing temperature was used. Structure and chemistry of interfaces were examined using SEM+EDS combined with scanning probe microscopy analysis. The results are discussed in terms of chemical interactions in relation to the role of alloying elements and substrate chemistry on reactivity and interface formation in selected metal/oxide systems.

Keywords: *wetting, reactivity, Ni alloys, oxides*

1. Introduction

Information on high temperature behaviour of refractory materials in contact with Ni-based alloys, particularly the effect of alloying additions on wettability and reactivity between an alloy and a refractory, is of great practical importance for metal casting applications [1-6].

The paper is focused on applicability of oxide-based ceramics for making foundry appliances needed for melting and casting of the new generation of Ni-based superalloys in order to manufacture cast boiler and turbine components for Advanced UltraSuperCritical Power Plants (A-USC).

2. Experimental procedure

Wettability and reactivity in selected metal/oxide couples were examined by a sessile drop method allowing the direct measurements of contact angle (θ) [7]. The materials used are shown in Table 1. Oxides were in the form of single crystals (sc) of a roughness $R_a < 1$ nm or dense polycrystalline (pc) substrates produced by HT-HP sintering from high-purity powder and polished up to $R_a < 150$ nm.

The tests were performed in an experimental system specially designed for high temperature studies [8]. Its details together with testing procedures are described in [3-8]. The following testing conditions were used: high-purity flowing Ar (5N), 1500°C, contact heating with a rate of 10 K min⁻¹. Real time behavior of an alloy on a substrate was recorded by a high-speed high-resolution CCD camera. In order to get the drop/substrate interfaces unaffected by cooling history, the interfaces were exposed at 1500°C directly in the UHV chamber by drop pushing from the 1st to 2nd position on a substrate [4-6]. Structural analysis of solidified sessile drop samples was performed by scanning electron microscopy (SEM), energy-dispersive X-ray spectroscopy (EDS) and scanning probe microscopy (SPM).

3. Results

The results from a comparative investigation of the effects of alloying additions to Ni and oxide type on wettability in selected metal/oxide couples are summarized in Table 1. Liquid Ni and its binary alloys do not wet ($\theta > 90^\circ$) any of the selected oxide substrates. An exception is NiCr10/MgO couple since, after melting, liquid NiCr10 wets and spreads over the substrate but after a few seconds it again de-wets it. The wetting-dewetting phenomenon takes place several times during heating up to 1500°C and it is accompanied with drop movement. Most probably, this phenomenon is related with periodic formation and disappearance of the wettable interfacial phase (MgCr₂O₄ [6]) due to its dissolution in the alloy and further transfer of Cr to flowing gas. Real time observations of melting, wetting, spreading and solidification of Cr-containing alloys (NiCr10, IN740, H282) on MgO-containing substrates (MgO, MgAl₂O₄, Mg₂SiO₄) evidenced drop movement, a larger scatter in contact angle measurements and a greenish color of the substrate surface that had been in contact with or in the vicinity of the drop.

Table 1 Contact angles of alloy/oxide couples at 1500°C
("→" corresponds to the tests using drop pushing from the 1st to 2nd position on the substrate at 1500°C)

| Substrate | Contact angle (degree) of selected alloys (chemical composition in wt%) | | | | | |
|--|---|---------|------------|-------------|--------------------|-------------------|
| | Ni (99.99%) | NiCo10 | NiCr10 | NiW10 | IN740 ^a | H282 ^b |
| Al ₂ O ₃ (sc) | 107 | 118 | 114 | 112 | 107 | 84 |
| MgO (sc) | 111→111 | 124 | 62-118* | 128 | 91±5 | 80-95*→85* |
| MgAl ₂ O ₄ (sc) | 93 | 110→110 | 148→110±10 | 135±5→130±5 | 95±5 | 85*→87* |
| Mg ₂ SiO ₄ (pc) | | | | | 154→154 | 91* |
| ZrO ₂ +MgO (pc) | | | | | | 81→86 |
| ZrO ₂ +3%Y ₂ O ₃ (pc) | 100±10* | 120→110 | 100→100 | | 94±5 | 82→81 |
| ZrO ₂ +Y ₂ O ₃ ^{mp} (pc) | 110 | | | | 147 | 134→109 |
| ZrO ₂ +ZrO ₂ ^{mp} (pc) | 120 | | | | 127 | 131→106 |

^a INCONEL®740: 20Co, 25Cr, 0.5Mo, 0.9Al, 1.8Ti, 2Nb, 0.7Fe, 0.3Mn, 0.5Si, 0.03C (Special Metals Corp., USA)
^b HAYNES®282: 10Co, 20Cr, 8.5Mo, 1.5Al, 2.1Ti, 1.5Fe, 0.3Mn, 0.15Si, 0.06C, 0.005B (Haynes International Inc., USA)
* apparent values due to drop movement or strong secondary substrate roughening

IN740 and H282 alloys do not wet polycrystalline Mg₂SiO₄, however, the formation of an Al₂O₃-rich phase was noted to occur. Especially with H282 alloy, Al₂O₃ was evidenced both at the interface and on the substrate surface near the drop, forming a continuous network along substrate grain boundaries. The worst wetting and the smallest reactivity were observed for IN740 and H282 on ZrO₂ substrates containing additions of Y₂O₃ or ZrO₂ nanoparticles (np). SEM, EDS and SPM characterization of interfaces showed that initially smooth substrate surfaces became rough due to a high temperature interaction with all alloys. Secondary roughening is particularly pronounced for MgO^{sc} after wettability tests with NiCr10, IN740 and H282 alloys showing two orders of magnitude increase in drop-side substrate surface roughness.

4. Summary

Due to high melting temperature and the presence of reactive alloying elements, the interaction between A-USC candidate alloys (IN740, H282) and oxide-based ceramics is accompanied with substrate dissolution in the alloy and the formation of interfacial reaction products. It results in secondary substrate roughening, a decrease in the contact angle values and an increase in adhesion of the liquid alloy to the oxide substrate. Unusual wetting behavior of liquid Cr-rich alloys on MgO-rich substrates, accompanied with the drop movement, is caused from continuous change in the substrate surface chemistry. The above phenomena may contribute to the formation of casting defects responsible for a significant reduction in mechanical properties of products.

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