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One-step preparation of TaIr₃-based material and its ablation performance under extreme environmental conditions

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ABSTRACT

The solid-state interaction of TaC with Ir within the 1000–1600 °C temperature range leads to the formation of TaIr₃ intermetallics. The durability of the TaIr₃-based system with a silicon additive was studied under extreme environmental conditions. The TaIr₃-based material displays satisfactory ablation resistance at 2000 °C in air in arc-jet. The post-test microstructural and XRD analysis of the developed material allowed us to propose the role of different constituents of the Ta – C – Ir – Si system. The iridium-containing phases play the most significant role in favorable ablation behavior of the developed system.

1. Introduction

The trend towards the optimization of efficiency in modern gas turbines, combustors and nose tips requires the development of high-strength materials with structural stability and corrosion resistance during operation at temperatures higher than 2000 °C [1,2]. Platinum group metal-based refractory alloys and intermetallics, especially those composed of iridium and refractory metals of IV-V groups, attract permanent attention of material scientists involved in the development of ultra-high temperature materials [3–5]. Indeed, these materials have a number of very attractive properties, in particular high melting points, often exceeding 2000 °C, high strengths and incompressibility, which are retained up to elevated temperatures, good oxidation resistance due to extremely low recession rate of iridium even at temperatures close to its melting point (2443 °C) [3,6–11].

Traditionally, iridium-based intermetallics composed of iridium and refractory metals of IV-V groups are synthesized from the mixtures of transition metals and iridium using high-temperature methods, e.g. arc melting [3,5,12,13]. High-temperature reaction conditions are combined with long annealing times and frequent regrinding steps for the preparation of powder intermetallics. An alternative approach to synthesize the above-mentioned intermetallics is a solid-state reaction of carbides, borides, and nitrides of corresponding refractory metals [14–17]. The reactions of carbides with iridium had been receiving increased attention in the 60-70's [14]. Reactions between HfC, ZrC, ThC₂ and iridium or rhodium were studied in connection with the development of the protective coating for graphite by Criscione and co-

workers [14]. The proposed approach to design the protective coatings for graphite involved iridium and rhodium as the external coating separated from the substrate (graphite) by an intermediate layer of some refractory materials that would be chemically stable both with respect to carbon and the external layer. However, as was shown by Criscione and co-workers [14], the reaction between metal carbide and iridium occurs within the temperature range of 1200–2000 °C with the formation of MeIr₃ phases alone, even when metal carbides were in a great excess with respect to iridium.

Simultaneously, Raub and Falkenburg studied the stability of some carbides including tantalum carbide towards platinum-group metals at high temperatures [18,19]. Later, Strife et al. [17] and Pierre [20] carried out a thorough study of the mechanism of the solid-state reaction of HfC with iridium at temperatures higher than 1650 °C. Based on the calculations of hafnium activity in HfC, HfIr, and HfIr₃, they concluded that when stoichiometric HfC is in contact with iridium, only the formation of cubic HfIr₃ should be observed. Contrary, the study of the solid-state reaction of TiC_x with iridium showed that TiIr₃ together with TiIr are formed, and the composition of the obtained product is strongly dependent on the stoichiometry of initial titanium carbide [17].

Unfortunately, another promising intermetallic compound for high-temperature applications, namely, TaIr₃ ($T_m = 2380 \pm 25$ °C [21]), has been overlooked. Only a brief mention of the formation of cubic TaIr₃ through the reaction of TaC with iridium was presented by Holleck in his Thesis [15] and Raub and Falkenburg [19]. Meanwhile, TaIr₃ deserves more attention due to a set of very attractive properties. TaIr₃ has a high melting point, high bulk modulus (327 GPa), and hardness

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(~26 GPa) [10,22]. One can note that other intermetallic phases of the Ta – Ir system, namely, σ (Ta₃Ir) and α_1 (TaIr) phases have also melting points higher than 2000 °C, which differs them advantageously from the corresponding analogs of the Zr – Ir and Hf – Ir systems. In addition, TaIr₃ appears to be able to withstand high-temperature corrosion in air because one of the components, tantalum, is oxidized with the formation of refractory tantalum oxide ($T_m = 1877$ °C for Ta₂O₅), whereas the other component of intermetallics, iridium, forms gaseous iridium oxide with very low velocity even at 2000 °C [7,23–25]. Unfortunately, no data about the oxidation resistance of TaIr₃ at 2000 °C were presented before.

This work is dedicated to study the peculiarities of the solid-state interaction of tantalum carbide with iridium depending on the experimental conditions, and to test the ablation resistance of the TaIr₃-based system obtained by the solid-state reaction of TaC and iridium under severe conditions at 2000 °C.

2. Experimental Section

2.1. Materials and preparation of the powders

The following powder materials were used as the initial substances, tantalum carbide (TU 6-09-03-443-77, Russia) and iridium (GOST 12338-81; purity > 99.96%, Russia). According to the XRD data, the as-received tantalum carbide phase is nonstoichiometric, and its composition corresponds to TaC_{0.98}. The SEM image of initial TaC powder is shown in Fig. S1 (Supplementary Material). The powders of TaC and Ir were weighted to reach the 1:1 and 1:3 ratios and mixed. The mixtures were placed in grafoil boxes and heated up to a given temperature, T , in the vacuum of 10^{-5} torr, kept at a given temperature for different time, τ , from 1 to 4 h. The products formed in the TaC – Ir system within the temperature range 1000–1600 °C were analyzed by SEM, XRD, and Raman to study the influence of the composition, the temperature and the duration of the heat-treatment.

2.2. Characterization

X-ray diffraction (XRD) patterns of the powders were recorded with a D8 Advance (Bruker AXS, Germany) powder diffractometer using Cu-K α radiation ($\lambda_1 = 1.54056$ Å, $\lambda_2 = 1.54439$ Å in the $30^\circ < 2\theta < 130^\circ$ interval. Quantitative phase analysis and lattice parameter refinement were performed by the Rietveld method using the Topas 4.2 software (Bruker AXS, Germany) [26] and Inorganic Crystal Structure Database ICSD (FIZ Karlsruhe, Germany, 1996).

The morphology and local elemental compositions of samples were examined with high-resolution scanning electron microscopes MIRA 3 LMU, (TESCAN, Czech Republic) equipped with energy dispersive X-ray spectroscopy (EDS) detector (INCA Energy 450 XMax 80) capable of detecting low Z elements. The variation coefficient characterizing the reproducibility of a single determination was found to be ~ 1% for EDS within the compositional range of the main components. Additionally, TM-1000 (Hitachi Ltd., Japan) scanning electron microscopy coupled with EDS detector Swift-TM (Oxford Instruments Analytical Ltd., GB) was used for the analysis of the morphology.

The oxidation tests were carried out in plasma generator EDG-200 M. The working current range was 120–400 A; the voltage of the plasma generator was 120 ± 1 V. The tested system was composed of the powdered TaC and Ir mixture with silicon additive for improved sintering of the mixture, the TaC:Ir:Si = 14:16:1 M ratio being used. A powder mixture was loaded into steel die and compacted as a pellet with sizes of $\varnothing 30 \times 10$ mm. The mixture was compacted and heated up to 1600 °C under vacuum. Then the compact was exposed to the arc-jet in the perpendicular direction. The maximum temperature at the ablation center reached 2000 °C, which was measured with an optical pyrometer focused on the hot zone of the specimen.

3. Results and discussion

3.1. Phase composition and morphology of products obtained by solid-state reaction

The XRD patterns of TaC:Ir mixture with the 1:1 ratio depending on temperature are presented in Fig. 1. The first features of the appearance of new TaIr₃ (cubic AuCu₃ structure) phase become noticeable at 1100 °C (Fig. 1, insert a). No intermetallic compounds other than TaIr₃ are formed. In addition to the new phase, unreacted TaC and iridium phases were observed too. The lattice parameter of TaIr₃ intermetallics was determined to be equal to 3.886 ± 0.001 Å which coincides with the value for stoichiometric TaIr₃ [21]. With an increase in the treatment temperature to 1300 °C, the lattice parameter of TaIr₃ intermetallics slightly increases and becomes equal to 3.890 Å, which corresponds to the composition TaIr_{2.86} (74.1% at. Ir). Subsequent temperature rise to 1600 °C, as well as an increase in exposure time to 4 h at this temperature, does not result in the change of the lattice parameter (Fig. 1 insert b; Supplementary Material, Fig. S2). It is noteworthy that the content of TaIr₃ phase calculated from the XRD data attains 96% (wt.) for the product obtained at 1600 °C for 4 h (TaC:Ir = 1:3).

No carbon phases were found by the XRD analysis. However, intense peaks in the regions of $1200 - 1600$ cm⁻¹ and $2500 - 3000$ cm⁻¹ were detected in the Raman spectra of products obtained under different experimental conditions (Supplementary Material, Fig. S5). The Raman peaks centered at about 1280 (D-peak) and 1592 (G-peak) cm⁻¹ can be assigned to the carbon phase.

As was mentioned above, unreacted initial iridium and TaC phases are present in the products obtained at 1100–1200 °C. As follows from Fig. 1 d (Supplementary Material, Fig. S3a, the 1:3 mixture), the positions of the XRD peaks of iridium are shifted towards the small-angle direction. This suggests that the lattice parameter of iridium is increased ($a = 3.846$ Å at 1200 °C), which could be explained by partial substitution of iridium atoms by tantalum atoms in the crystal structure of iridium ($R_{Ta} = 2.00$ Å vs. $R_{Ir} = 1.80$ Å). No traces of iridium were detected in the XRD patterns of products obtained at 1300 °C and higher (Fig. 1d). Thus, starting from this temperature, iridium was fully consumed by TaC, whereas tantalum carbide was detected in all products including those obtained upon long-term exposure at 1600 °C. The excess of TaC phase can be explained by the fact that only TaIr₃, i.e., intermetallic compound with the highest iridium content is formed during the reaction of TaC with iridium. As a consequence, unreacted TaC is detected in the reaction products. Taking into account a shift of the K α_1 and K α_2 doublet for the (422) peak (Fig. 1c), one can consider that the composition of unreacted TaC approaches the stoichiometric one, as the reaction between TaC_x and iridium proceeds. It could be discreetly proposed that carbon eliminating in the course of the reaction of nonstoichiometric TaC_x with iridium diffuses inward the TaC_x lattice and fills the carbon vacancies. The peculiarities of the formation of TaIr₃ for the TaC:Ir = 1:3 powder mixture are the same as those deduced from the XRD results for the TaC:Ir = 1:1 mixture (Supplementary Material, Figs. S3 and S4).

Analysis of the SEM images of TaIr₃ product (1600 °C, 4 h, TaC:Ir = 1:3) shows that it is composed of grains with irregular shape (Fig. 2 a, b). The sizes of the grains fall within the range 1–10 μ m. Together with separate grains, rather large-size aggregates composed of sintered grains are also observed. According to EDS analysis data (Table 1), the composition of product determined in different areas of several samples was estimated to be TaIr_{2.89} – TaIr_{3.02}, which is in good correspondence with the composition determined by the XRD analysis, namely, TaIr_{2.86} – TaIr_{3.15} (74.1–75.9% at.). Also, in some SEM images one can observe that transparent thin sheets are present on the surface of the product grains (Fig. 2 b). These sheets could be related to the carbon phase. One can remind that the presence of carbon phase as transparent sheets was detected in the products obtained from HfC and Ir mixture [27].

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