



Preparation of niobium carbide powder by electrochemical reduction in molten salt



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ABSTRACT

The niobium carbide powder was prepared via electrochemical reduction of the mixture of Nb₂O₅ and carbon in molten CaCl₂–NaCl. The reaction pathway from the sintered precursor to the final product has been investigated. The effect of the working temperature on the reduction of the Nb₂O₅/C composite precursor was considered. The role of carbon during the electrochemical reduction of the composite pellet was discussed. The samples were analysed by XRD and SEM. The results indicated that the NbC powder was approximately 200 nm after the reduction. Nb₂O₅ was gradually reduced to Nb, and NbC was subsequently obtained by the reaction of carbon with Nb metal. In addition, Nb₂O₅ could spontaneously react with CaO in the melt to form a series of calcium niobates. The participation of carbon was available for the efficiency of electro-reduction of Nb₂O₅.

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1. Introduction

Niobium carbide is an attractive material that exhibits unique properties, such as high melting point (about 3610 °C), high microhardness (more than 235 GPa) and Young's modulus (about 3.38×10^5 N mm⁻¹), good wear resistance and chemical inertness. Due to these excellent properties, niobium carbide is a promising material for a wide range of applications including cutting tools, protective coating for instruments, microelectronic devices, heating element of electric furnace, and a catalyst for decomposition of ammonia [1–4]. In particular, niobium carbide powder is used as an additive in other metals or alloys as a hard phase to improve the material properties [1,5].

Some methods have been adopted to prepare niobium carbide including carbothermic reduction of niobium oxides [6,7], self-propagating combustion of niobium and carbon [8], gas phase reaction of NbCl₅ and carbon-containing reductants [9,10], and mechanochemical synthesis from niobium and carbon [11,12]. However, the relatively high temperature or hazardous substances,

which are not environmentally friendly, are typically employed in most of these methods. As a novel method to obtain metals and alloys, electro-deoxidation of metal oxides in molten alkali halides has generated much interest, and this method can produce metals and alloys in a simple, low temperature and environmentally friendly way. Previously, researchers investigated the preparation of metal carbides by electro-deoxidation of metal oxides/carbon precursors in molten salts. Abdelkader et al. [13] investigated the electrochemical synthesis of hafnium carbide powder in molten chloride, and fabricated fully dense monolithic HfC ceramics from the electro-deoxidized product. Yan et al. [14] prepared TiC powders and coatings by electro-deoxidation of solid TiO₂ in molten salt. Lang et al. [15] obtained Cr₃C₂ by electrochemical reduction of the self-sintered cathode in molten CaCl₂. Zou et al. [16] reported the direct electrosynthesis of Ti₅Si₃/TiC composites in a CaCl₂ melt. However, there have been limited studies on the electrochemical preparation of niobium carbide.

In this study, niobium carbide was obtained by electro-reduction of Nb₂O₅ in the presence of carbon, and the reaction mechanism of the electrochemical reduction of Nb₂O₅ as well as the formation of NbC were investigated. The objective was to prepare refractory metal carbides through a more facile approach.

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2. Experimental

Nb_2O_5 and carbon powder were well mixed at a molar ratio of Nb: C = 1:1.5, and about 10 wt. % of polyvinyl alcohol was added to the mixture. The mixed powder, which weighed 1.0 g, was pressed into a cylindrical pellet under 6 MPa of pressure (15 mm in diameter and 1 mm in thickness) followed by sintering at 1000 °C for 6 h in an argon atmosphere. A eutectic mixture of CaCl_2 and NaCl (about 400 g) was used as the electrolyte, which was dried at 350 °C for 24 h. All of the chemical reagents employed in this study were of analytical grade.

The electrochemical experiments were carried out in a molten salt system. An alumina crucible (90 mm in diameter, 100 mm in height) was filled with the mixed salts and placed at the bottom of a sealed tubular stainless steel reactor. Then, the reactor was heated to the target temperature with argon gas circulation. The $\text{Nb}_2\text{O}_5/\text{C}$ composite pellet was used as the cathode. A high-density graphite rod (12 mm in diameter, 70 mm in length) was employed as the anode. Both of the electrodes were connected to 304 stainless steel wires (2 mm in diameter), which were slowly lowered into the melt. A constant voltage of 3.1 V was applied between the anode and the cathode, and the current was recorded. After the reduction, the reactor was cooled to room temperature. The sample was vigorously rinsed with tap water to remove the solidified salt and dried for examination. The phase composition of each sample was determined using a D/Max-2500PC X-ray diffractometer (XRD) with Cu-K α radiation. The morphology of the samples was investigated using a JSM-6360LV scanning electron microscope (SEM) equipped with energy dispersive X-ray analysis (EDX).

3. Results and discussion

Fig. 1a–c shows the SEM images of Nb_2O_5 , the carbon powder

and the composite pellet sintered at 1000 °C for 6 h. The sintered Nb_2O_5 pellet contains crystallites that are approximately 2 μm , and void space exists between the individual particles. The carbon powder employed for the carbonization of niobium is composed of nano-sized particles that are approximately 100 nm in size. After the sintering of the mixture, the Nb_2O_5 and carbon particles were uniformly distributed across the pellet. Most of the voids were filled with nano-sized carbon particles in comparison with the microstructure of the Nb_2O_5 pellet shown Fig. 1a. This phenomenon is probably less favourable for the penetration of molten salt and diffusion of the ionized O^{2-} ions during the electro-deoxidation of Nb_2O_5 . Fig. 1d shows the XRD patterns of the Nb_2O_5 pellet, carbon additive, and the sintered composite. The XRD pattern of the carbon additive contains two bumps at approximately $2\theta \approx 26$ and 44° , which implies that the additive is predominantly composed of amorphous carbon. Due to its non-crystalline structure, the amorphous carbon is available for the formation of NbC. A new phase was not detected after the addition of the carbon powder to the pellet based on the comparison with the XRD pattern of Nb_2O_5 . Moreover, the bumps corresponding to amorphous carbon were barely noticeable.

Electrochemical reduction of the $\text{Nb}_2\text{O}_5/\text{C}$ composite pellets was carried out at 740, 800 and 900 °C under 3.1 V for 13.5 h. Fig. 2a shows the XRD patterns of the as-prepared products obtained at different temperature. The pattern for the Nb product reduced from the direct electro-deoxidation of Nb_2O_5 is also presented. The sample gained at 740 °C was determined to primarily consist of CaNb_2O_6 , and the diffraction peaks for NbC were also observed. This result suggests that niobium carbide has been formed. The calcium niobates, such as CaNb_2O_6 , also appeared due to the incorporation of CaO into the Nb_2O_5 structure. However, the relatively low temperature is inadequate for the complete electro-deoxidation of the calcium niobates. As the working temperature increased to 800 °C,

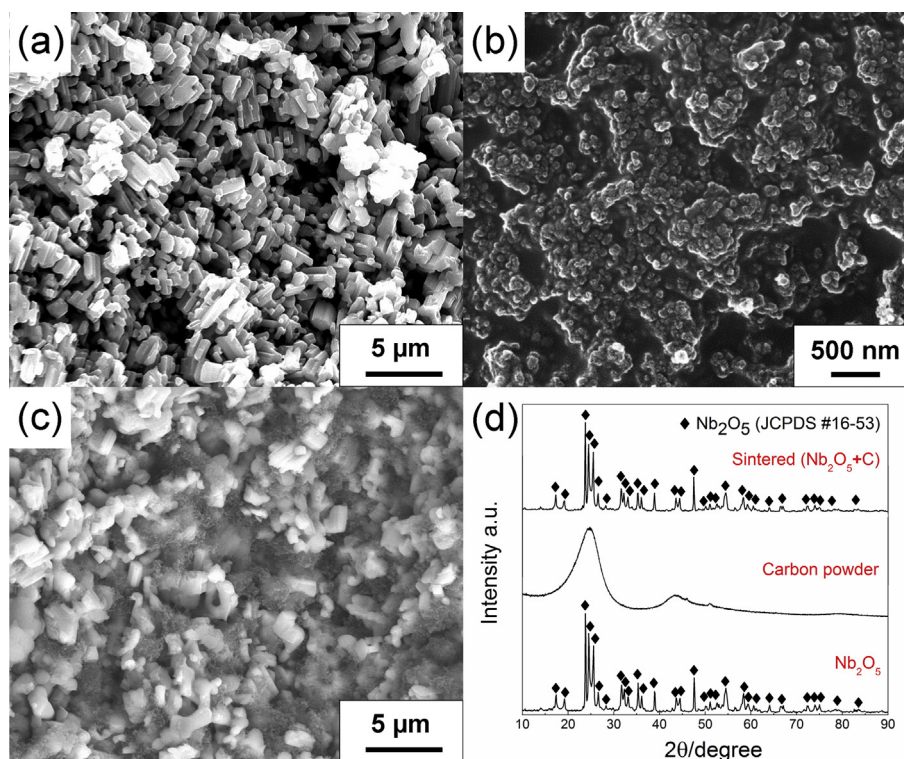


Fig. 1. SEM images of the (a) Nb_2O_5 ; (b) carbon powder; and (c) the composite pellet sintered at 1000 °C for 6 h; (d) XRD patterns of the Nb_2O_5 , carbon powder and composite pellet.

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