



## Effects of Mo additions on the corrosion behavior of WC–TiC–Ni hardmetals in acidic solutions

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### ABSTRACT

The corrosion behavior of WC–TiC–Ni cemented carbides with the additions of molybdenum was investigated in 1 M H<sub>2</sub>SO<sub>4</sub> and 1 M HCl solutions utilizing potentiodynamic polarization, long-term immersion tests and surface analytical techniques. The results show that the Mo additions can enhance the corrosion resistance of WC–TiC–Ni hardmetals owing to the formation of new (Ti, W, Mo)C phase based on TiC. Pseudopassivity was observed in the polarization curves of WC–Ni and WC–TiC–Ni hardmetals in HCl. However, the additions of molybdenum can make WC–TiC–Ni hardmetals passivated visibly in curves at 600 mV–1000 mV and enhance the pitting corrosion resistance due to the formation of MoO<sub>3</sub> film. In addition, all nickel based alloys show the typical passivation behavior in H<sub>2</sub>SO<sub>4</sub> caused by the formation of NiSO<sub>4</sub>, NiWO<sub>4</sub> and continuous metal oxide films.

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### 1. Introduction

Tungsten carbide based hardmetals, which possess high hardness and exceptional wear resistance, are widely used in industrial fields such as mining, cutting tools and wear resistant materials [1]. The importance of the corrosion resistance of hardmetals is emphasized by the tools applied in the sphere of aggressively corrosive environment including wood-cutting applications, mining in seawater [2–4]. It has been demonstrated that the preferential dissolution of the binder phase in neutral and acidic solution, caused by the galvanic coupling between carbides and binder phase [5], can inevitably bring about the sharp corrosion rate and the seriously shortened lifetime of the tools with the exposure of a fragile skeleton of carbides, which can be easily grinded off in service [6]. Compared to the hardmetals with nickel or iron binder, WC–Co based hardmetals show better mechanical and wearing properties due to the more superior wettability to the carbides and thermo-mechanical properties of cobalt binder [7–9]. However, owing to the poor corrosion, oxidation resistance [6,9] and the expensive price of cobalt, an alternative or a modified binder phase has been researched for years. As demonstrated by Aw et al. [9] and Human et al. [10], WC–Ni based hardmetals show better corrosion and oxidation resistance, thus possess more potential in terms of corrosive ambience when the mechanical properties are ameliorated.

The additions of metallic carbides such as Cr<sub>3</sub>C<sub>2</sub> [3], TiC [11–13] or Mo<sub>2</sub>C [3,13,14] have been investigated to enhance the mechanical properties of WC–Ni hardmetals. Cubic titanium carbides perform excellently due to the formation of (W, Ti)C solid solution which is more outstanding than TiC in improving the hardness, toughness, wear and oxidation resistance of the hardmetals [7,15–18]. Mori et al. [18] demonstrated that the additions of TiC can improve the corrosion resistance of WC–Co hardmetals, while little effect of TiC additions on the polarization behavior of WC-based hardmetals was found by Tomlinson et al. [22]. The addition of Mo<sub>2</sub>C or Mo can refine the microstructure and optimize the wettability between carbides and binder owing to the formation of a Mo-rich phase [17,19,20], which may influence the corrosion behavior of WC based hardmetals positively. In corrosive environment, it has been shown that the molybdenum additions may suppress the pitting corrosion and enhance the passivation behavior of both stainless steel and amorphous alloy [23,24]. In the field of hardmetals, it has been shown that 1 wt.% Mo additions can improve the properties of WC–TiC–Ni hardmetals significantly, especially the abrasion resistance, which is even better than that of commonly used WC–Co alloys [13]. Nevertheless, the effects of Mo additions on the corrosion resistance of WC–TiC–Ni hardmetals have not been investigated systematically.

In this paper, the influence of Mo additions on the corrosion behavior of WC–TiC–Ni alloys in both 1 M HCl and 1 M H<sub>2</sub>SO<sub>4</sub> was studied, as well as the role of TiC on the corrosion resistance of WC–Ni hardmetals. The knowledge about the corrosion behavior of WC–Ni hardmetals will be enriched. The techniques including potentiodynamic polarization, long-term immersion tests, scanning electron microscope (SEM), field emission electron probe microanalyzer (FE-EPMA) and X-ray photoelectron spectroscopy (XPS) were applied.

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## 2. Experiment

### 2.1. Materials

The specimens with various compositions as shown in Table 1 were manufactured by conventional powder metallurgical technology. The raw (W, Ti)C powders mainly consist of W (55.90 wt.%), Ti (31.98 wt.%) and C (11.02 wt.%). The raw powders were milled in a planetary ball mill under argon atmosphere in ethanol with a ball-to-powder weight ratio of 5:1 and a mill rotation rate of 250 rpm. After milled for 20 h, the as-prepared mixtures blended with 1 wt.% paraffin were granulated, dried and compacted into a rectangular plate under an uniaxial pressure of 200 MPa. Then the green compacts were sintered in industrial vacuum condition at 1450 °C for 60 min. Finally, the alloys were grinded and polished with diamond sand papers and diamond paste for further electrochemical measurements and immersion tests.

### 2.2. Electrochemical measurements

A three electrode cell setup with a potentiostat unit (CHI604D) was applied for the electrochemical test at room temperature. The samples were linked with copper wires and partially coated by RTV rubber with an exposing area of 0.25 cm<sup>2</sup> to the electrolyte. 1 M HCl and 1 M H<sub>2</sub>SO<sub>4</sub> were used as electrolytes open to the air, since there was no essential difference in the electrochemical corrosion behavior of target alloys in aerated or deaerated acidic solutions [6]. All the electrolyte solutions used were prepared from reagent grade chemicals and deionized water. The platinum plate served as the counter electrode (CE) and saturated calomel electrode (SCE) did as the reference electrode. All the potentials in this work were reported with respect to the SCE. After the immersion of samples in the electrolyte for 1 h, the open circuit potential ( $E_{oc}$ ) tests were conducted for 2 h immediately, which stabilized the  $E_{oc}$  values with a range of  $\pm 0.01$  v, and followed by the potentiodynamic polarization measurements. All partially coated specimens were polarized from  $-700$  mV in a positive direction to 1200 mV at a scan rate of 5 mV/s according to other reported investigations [5], while the scan rate used makes only negligible contribution to the transversal comparison [6]. Then, the chronoamperometry measurements were launched on the polarized surface to determine the properties of passivation layers. All the measurements were tested for two times with good repeatability. All corresponding parameters were calculated and simulated directly by the accompanying software of the unit and the parameters of the certain sample fluctuate within an acceptable tolerance.

### 2.3. Immersion test

For comparison with the polarization results, 1 M HCl and 1 M H<sub>2</sub>SO<sub>4</sub> were used as the static corrosion media for the immersion test at room temperature. Polished samples Ni10, Mo0 and Mo20 were immersed into the solution for 1500 h and weighed for the determination of specific mass loss after 50, 250, 600 and 1500 h. Both

**Table 1**  
The nominal composition and properties of the investigated specimens.

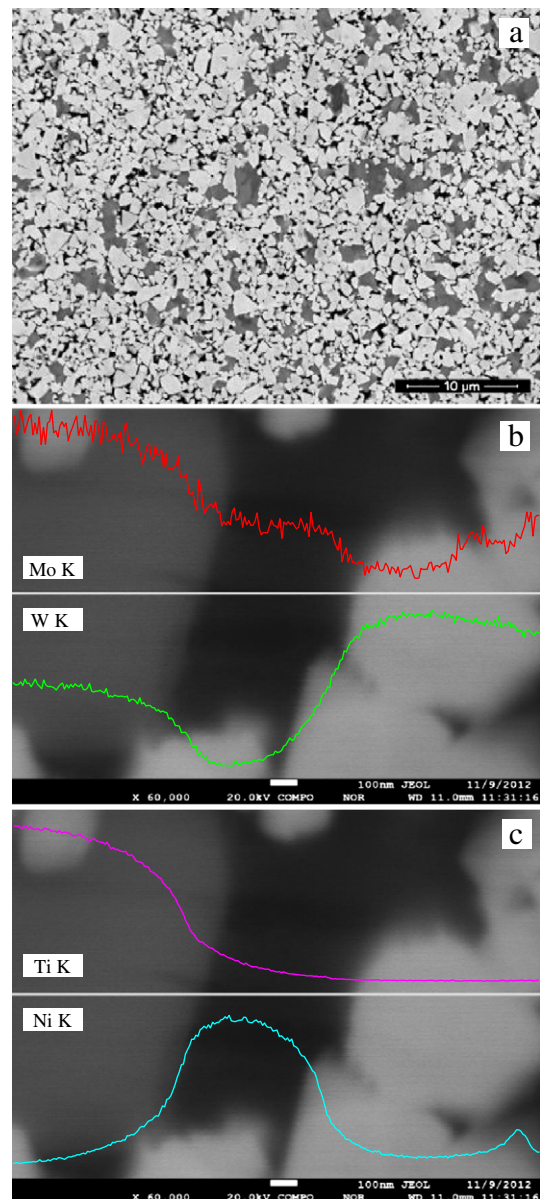
Sample identity	Composition (wt.%)					Harness (HRA)	Average WC grain size ( $\mu\text{m}$ )
	(W, Ti)C	Ni	Mo	C	WC		
Co10	–	10.0Co	–	0.3	Balance	89.6 $\pm$ 0.1	953.91
Ni10	–	10.0	–	0.3	Balance	88.8	960.17
Mo0	6.0	10.0	–	0.3	Balance	90.0	807.15
Mo5	6.0	10.0	0.5	0.3	Balance	89.7	775.12
Mo10	6.0	10.0	1.0	0.3	Balance	89.8 $\pm$ 0.1	696.56
Mo15	6.0	10.0	1.5	0.3	Balance	90.1	695.45
Mo20	6.0	10.0	2.0	0.3	Balance	90.0 $\pm$ 0.1	680.85

the as-immersed and as-polarized samples were cleaned with alcohol in ultrasonic water bath for 20 s and dried at low temperature for further characterization.

### 2.4. Microstructures and chemical characterization

The hardness of polished alloys was measured by Rockwell indentation using a 60 kg load. The morphological observations and qualitative analyses of the alloys before and after both electrochemical measurements and immersion tests were implemented using an FEI Nano230 scanning electron microscope (SEM) equipped with the Oxford X-ray energy dispersive spectroscopy (EDS). The distribution of specific elements in as-polished alloy Mo20 was characterized by line scan analyses using JEOL JXA-8530F field emission electron probe microanalyzer (FE-EPMA). The average WC grain size of alloys was measured by quantitative microscopy techniques according to the 20 k-magnification SEM micrographs.

Before and after electrochemical measurements, the X-ray diffraction (XRD: D/MAX-255) with Cu K $\alpha$  radiation (wavelength  $k = 1.5406 \text{ \AA}$ )



**Fig. 1.** (a) The SEM micrograph and (b, c) corresponding line scan analyses of the as-polished alloy Mo20 by EPMA.

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