Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Electrodeposition of alloys and compounds from high-temperature molten salts

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A R T I C L E I N F O

Article history: Received 16 July 2016 Received in revised form 13 August 2016 Accepted 13 August 2016 Available online 16 August 2016

Keywords: Electrodeposition Molten salt Alloys and compounds High temperature Coatings

ABSTRACT

This paper reviews the development of the electrodeposition of a variety of types of alloys and compounds from high-temperature molten salts. The electrodeposition from high-temperature molten salts has several distinct potential advantages. Firstly, molten salts can offer a much larger operative temperature range than other electrolytes such as aqueous electrolytes, which greatly expands the material systems that can be prepared by this method. Molten-salt electrodeposition is also able to obtain reactive metals (e.g., Al and Mg) and refractory alloys and compounds that are difficult or unable to be electrodeposited from aqueous solutions. Additionally, the electrodeposition from high-temperature molten salts contributes to the formation of strongly adherent coatings because the diffusion process enables the metallurgical bonding between the coating and the substrate. Therefore, a wide variety of alloys and compounds have been prepared by the molten-salt electrodeposition. The reviewed materials in this work are categorized into light metal alloys, transition metal alloys, rare-earth containing alloys and metal-containing compounds and special attention is paid to the reactive metals and refractory alloys and compounds. The electrochemical processes and growth mechanisms of the coatings formed in molten salts are described. The relationship among the preparation parameters, structures and properties is discussed. Finally, the development trend of the molten-salt electrodeposition is summarized and future work that should be carried out is proposed.

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1. Introduction

Coatings consisting of metal-based alloys and compounds have been extensively used as protective and/or functional materials in a wide variety of industrial areas. For protection applications, coatings with desired composition and structure can be used to enhance the surface properties such as the wear resistance [1,2], the corrosion resistance [3,4] and the high-temperature oxidation resistance [5-8] of the substrates. On the other hand, for functional applications, coatings with attractive physical and chemical properties (e.g., ferromagnetic [9,10], optical [11] and catalytic [12–14] properties) have attracted great interest. To date, a large number of technologies, such as the chemical vapor deposition (CVD) [15,16], physical vapor deposition (PVD) [17,18], plasma spray (PS) [19], hot dipping [20] and electrochemical deposition [21], have been developed to prepare coatings. Among these methods, the electrochemical deposition method has received great attention due to its distinct advantages. For instance, the electrodeposition method has good flexibility for coating the objects with big size and complicated shape and the required equipment is relatively simple and economic [6,22]. In addition, various parameters of the coatings, such as the composition, structure and thickness, can be controlled through rationally selecting the electrodeposition conditions such as the composition of the electrolytes, electrodeposition current, potential and mode (e.g., constant or varied current/potential).

The electrodeposition method can be briefly divided into four categories based on the electrolytes which are aqueous solutions, organic solvents, ionic liquids (also known as "room-temperature molten salt" or "low-temperature molten salt") and hightemperature molten salts. The electrodeposition from hightemperature molten salts has drawn particular interest because of the following reasons: 1) Compared to other electrolytes, molten salt systems can provide a much wider operating temperature range from 150 to 1050 °C depending on the composition of the molten salt [23,24]. 2) It is possible to use the molten-salt electrodeposition to obtain various reactive metals such as Al, Mg and Ti as well as refractory compounds such as borides and carbides, which cannot be electrochemically deposited from aqueous solutions [25–27]. 3) Molten-salt electrodeposition enables the formation of the metallurgical bonding between the coating and the substrate due to the high-temperature diffusion process, which can lead to the formation of strongly adherent coatings [28].

Based on the above reasons, the electrodeposition from molten salts has aroused considerable interest among researchers and great progress has been made in this area. This article reviews the development of the electrodeposition of various types of metals, alloys and compounds from molten salts. Since the electrodeposition from ionic liquids has already been reviewed in our previous work [29], the present work focuses on the high-temperature molten salts with the temperature above 100 °C.

2. Electrodeposition of light alloys

2.1. Aluminium (Al)-based alloys

Al-Mn allovs have attracted considerable interests due to their quasicrystal phenomena as well as their promising chemical, physical, mechanical and corrosion resistant properties [30,31]. A large number of literature have focused on the microstructure of Al-Mn alloys electrodeposited from AlCl₃-based molten salts containing MnCl₂ with particular focus on the effects of Mn content on the structure of coatings [30–33]. It has been found that the amount of Mn in the electrodeposited Al-Mn alloys play an important role in the microstructure. Small amounts of Mn (less than 17 wt.%) resulted in the formation of a supersaturated facecentered cubic (fcc) aluminium solid solution, and amorphous phase was obtained when the Mn content was higher than 25 wt.% [30–33]. The effects of electrodeposition parameters on the formed deposits have also been studied. For example, Li et al. [34] reported that Mn content decreased while the thickness of the plating layer increased as the electrodeposition current density increased from 10 to 45 mA cm⁻² in AlCl₃–NaCl–MnCl₂ molten salts. To improve the surface quality of the deposited coating, Uchida et al. [25] employed forced fluid flow to electrodeposit the Al-Mn alloys and found that the electrodeposits with smooth surface could be obtained from AlC13-NaCl-KCl molten salt with about 25 wt.% Mn at a high current density up to 600 mA cm^{-2} . Additionally, some studies have been paid to the investigation of ferromagnetic properties of Al–Mn τ phase prepared by the molten salt electrodeposition. For instance, Stafford et al. [9] prepared ferromagnetic Al–Mn τ phase by the electrodeposition from AlCl₃–NaCl (55:45 mol%) molten salt electrolyte containing MnCl₂. It is interesting to note that the measured coercivity (1.0 T) was over twice that reported by other work (0.46 T) [35]. The authors attributed this to the reason that the deposits having (112) texture that was positioned perpendicular to the applied field [9].

Some investigations have focused on fundamentally understanding the formation mechanism of Al–Mn coatings during the electrodeposition process in molten salts. For example, Jafarian et al. [36] studied the mechanism of the Al–Mn electrodeposition by electrochemical methods including cyclic voltammetry, and potential and current transient. A three-dimensional (3D) diffusion-controlled nucleation and growth mode was found for the Mn electrodeposition on Al in AlCl₃–NaCl–KCl (66:20:14 wt.%) with the addition of 0.3 wt.% MnCl₂ [36]. Stafford Gery [37] reported different rate-controlling steps depending on the electrodeposition overpotential. It was revealed that the electrodeposition process of the alloy formation was kinetically controlled at small cathodic overpotentials while it was limited by the mass transport (first Mn^{2+} and then Al_2Cl_7) at larger cathodic overpotentials.

Besides Al–Mn alloys, Al–Ni and Al–Cr alloys have also been investigated because of their potential advantages such as the excellent corrosion resistance and high-temperature oxidation resistance [7,26,38–42]. Moffat [38,39] electrodeposited Al–Ni

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