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High temperature oxidation behaviour of Ag–36.35 wt.% Zn and Ag–38.50 wt.% Zn–0.60 wt.% Al

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ABSTRACT

Ag–36.35 wt.% Zn and Ag–38.50 wt.% Zn–0.60 wt.% Al were oxidised in air at 450–650 °C up to 48 h. The oxidation kinetics of both alloys were parabolic with the lower rate constant for the latter alloy. After oxidation for 48 h, thermal oxide scale on Ag–36.35 wt.% Zn was ZnO at all studied temperatures. When 0.60 wt.% Al was added to Ag–38.50 wt.% Zn, ZnO and ZnAl₂O₄ were formed at all studied temperatures. Al₂O₃ was evidently observed at 550 and 650 °C. The formation of Al-containing oxides helped reduce the oxidation rate of Ag–38.50 wt.% Zn–0.60 wt.% Al.

1. Introduction

A silver alloy commonly used in jewellery industry is a sterling silver which contains Ag at least 92.5 wt.% with Cu as a main alloying element [1]. For an aesthetic purpose, Zn is alternatively added instead of Cu to make the alloy shine pink. T. Minemura et al. [2] reported that Ag–50 wt.% Zn could shine pink when AgZn in the CsCl structure was formed. However, in the silver casting and metal working processes at high temperatures, metal loss due to thermal oxide scale formation occurs and this must be avoided. It was reported that the addition of Al to Fe–Cr [3,4] and Co–Cr [5] could increase the high temperature corrosion resistance of the alloys. It was then hypothesised in the present work that the addition of Al to Ag–Zn might reduce the alloy oxidation rate.

Concerning the oxidation of silver alloys, the high temperature oxidation behaviour of the conventional sterling silver [6], Ag–Sn [7], Ag–Sn–Cu [8] and Ag–VC composite [9] were studied. The effect of Zn on the high temperature oxidation of Cu [10,11], Mg [12] and Sn [13] alloys were also examined. The recent work has synthesised Ag and Al co-doped ZnO with the emphasis to enhance its charge transport properties [14]. However, the behaviour of thermal oxide scale on Ag–Zn and Ag–Zn–Al which could be the oxides containing Zn and/or Al has never been investigated. Thus the objective of this paper is to study the high temperature oxidation behaviour of the Ag–Zn and Ag–Zn–Al alloys.

2. Experimental procedures

The studied alloys were silver alloys with the compositions shown in Table 1. For the Ag–36.35 wt.% Zn, it consisted of 63.62 wt.% of Ag, 36.35 wt.% of Zn with a small amount of Cu with the content of 0.03 wt.%. For the Ag–38.50 wt.% Zn–0.60 wt.% Al, it consisted of 60.84 wt.% of Ag, 38.50 wt.% of Zn, 0.60 wt.% of Al, also with a small amount of Cu with the content of 0.06 wt.%.

For the high temperature oxidation test, the sample was cut as a disk with the diameter of 16 mm and the thickness of 2 mm as exemplified in Fig. 1. The sample was ground on SiC papers up to 1200 grit, cleaned by alcohol in an ultrasonic machine and dried in forced air. The sample was further placed in the horizontal furnace. It was heated with the heating rate of 42 °C min^{−1} in synthetic air (79%N₂–21%O₂) to the studied temperatures (450, 550 and 650 °C), and at these studied temperatures the oxidation proceeded with different periods up to 48 h. After these periods, the sample was cooled down in furnace under laboratory air to room temperature.

For the physico-chemical characterisation, an X-ray diffractometer (D8 ADVANCE BRUKER) using Cu Kα (λ = 1.5406 Å) radiation was used for phase identification. A scanning electron microscope (SEM, JSM-6610LV) equipped with an energy dispersive X-ray analyser was used for imaging and elemental analyses.

3. Results

Fig. 2 shows mass gains of Ag–36.35 wt.% Zn and Ag–38.50 wt.% Zn–0.60 wt.% Al oxidised at 450, 550 and 650 °C. At each temperature,

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Table 1
Chemical compositions (wt.%) of the studied alloys.

Alloy	Composition			
	Ag	Zn	Al	Cu
Ag-36.35 wt.% Zn	63.62	36.35	-	0.03
Ag-38.50 wt.%Zn-0.60 wt.% Al	60.84	38.50	0.60	0.06

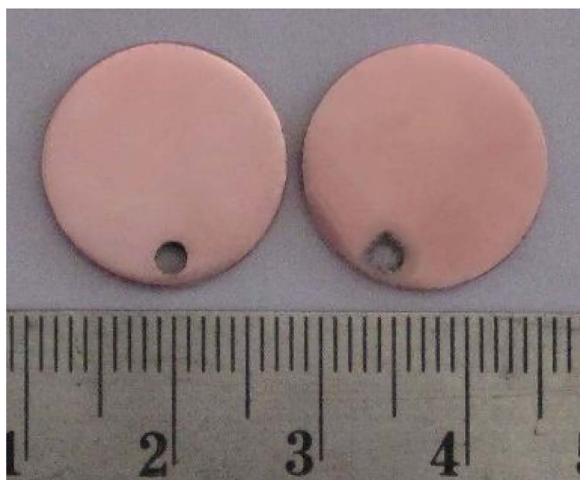


Fig. 1. The Ag-38.50 wt.% Zn-0.60 wt.% Al sample.

mass gain of Ag-38.50 wt.% Zn-0.60 wt.% Al was lower than that of Ag-36.35 wt.% Zn. For example, at 550 °C and 48 h of oxidation, the average mass gain of Ag-38.50 wt.% Zn-0.60 wt.% Al was 0.05 mg cm⁻² while it was 0.47 mg cm⁻² for Ag-36.35 wt.% Zn. It was also found for each alloy that when the oxidising temperature was raised, mass gain at a given oxidation period was increased. For example, the average mass gains of the Ag-36.35 wt.% Zn were 0.10 and 0.47 mg cm⁻² when it was oxidised for 48 h at 450 and 550 °C respectively. It is noted that the oxide scales on the studied alloys oxidised at 450 and 550 °C for the period up to 48 h, as well as at 650 °C for the period up to 30 h, were adherent to the metal substrates as exemplified in Figs. 3 and 4. Fig. 3 depicts the SEM micrograph and energy dispersive X-ray spectroscopy (EDS) mapping results of the cross section of the Ag-36.35 wt.% Zn after oxidation at 550 °C for 48 h. The scale was adherent to the alloy substrate with the thickness averaged from two images of 4.96 ± 0.06 μm. The EDS mapping results showed the distribution of Zn and O in the oxide scale. Fig. 4 shows the SEM micrograph and EDS mapping results of the cross section of the Ag-38.50 wt.% Zn-0.60 wt.% Al after oxidation at 550 °C for 48 h. The average thickness of the oxide scale on this alloy was 0.96 ± 0.05 μm. The EDS mapping showed the distribution of Zn, O and Al in the oxide scale. However, when the two alloys were oxidised at the highest studied temperature of 650 °C for 48 h the oxide scale spallation was observed. Mass gains of the alloys in that condition were not then plotted in Fig. 2.

Fig. 5 depicts the X-ray diffraction (XRD) patterns of Ag-36.35 wt.% Zn oxidised at 450, 550 and 650 °C for 48 h. At all studied temperatures, we observed peaks of ZnO which corresponded to ICDD 01-1136 reference peaks and AgZn which corresponded to ICDD 29-1155 and ICDD 29-1156. Ag₅Zn₈ corresponding to ICDD 26-0956 was also observed when the sample was oxidised at 450 and 550 °C but it was not detected when the sample was oxidised at 650 °C.

Fig. 6 shows the XRD patterns of Ag-38.50 wt.% Zn-0.60 wt.% Al oxidised at 450, 550 and 650 °C for 48 h. Peaks of ZnO, AgZn, Ag₅Zn₈ were detected in the same manner as in the case of Ag-36.35 wt.% Zn. This is to say that ZnO and AgZn were observed at all studied

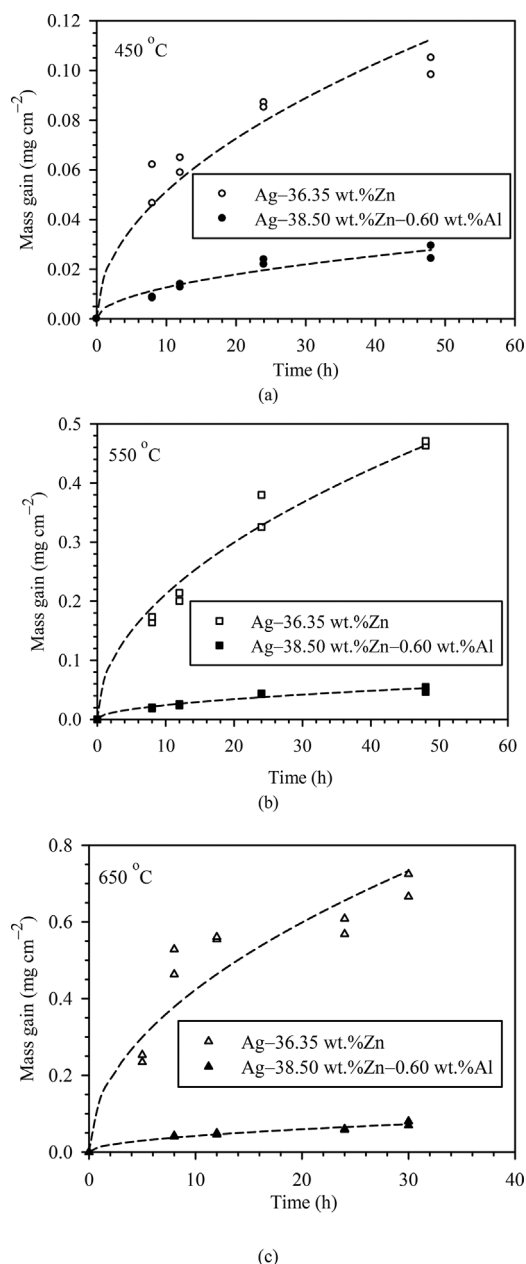


Fig. 2. Mass gain of Ag-36.35 wt.% Zn and Ag-38.50 wt.% Zn-0.60 wt.% Al oxidised at (a) 450 °C, (b) 550 °C and (c) 650 °C.

temperatures while Ag₅Zn₈ was detected when the sample was oxidised at 450 and 550 °C but was not found when the sample was oxidised at 650 °C. In addition, Al-containing oxides were detected in the oxide scale, i.e. ZnAl₂O₄ peaks corresponding to ICDD 74-1138 were found at all studied temperatures while Al₂O₃ peaks corresponding to ICDD 50-0741 were detected when the sample was oxidised at 550 and 650 °C.

4. Discussion

4.1. Oxidation kinetics

Mass gain in Fig. 2 was plotted as a function of the square root of time as shown in Fig. 7. Fig. 7(a) and (b) present the results for Ag-36.35 wt.% Zn and Ag-38.50 wt.% Zn-0.60 wt.% Al respectively. It is noted that these plots excluded data points in the condition where the excessive scale spallation was observed, i.e. 650 °C for 48 h. From Fig. 7, the linear relationship between mass gain and square root of

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