Leaching of Metals from Dental Silver Alloy Wastes

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Abstract: Environmental issues of hazardous metal wastes as well as growing demand for metals has increased focus on the forthcoming provision of metals. Therefore, the recovery and recycling processes of precious metals from secondary resources have become more prominent in the last years. Silver is one of the precious metals which can be recovered from wastes such as electronic wastes, coin and medal production losses, photographic films, and dental filling materials known as amalgam, which has the highest silver content. The present paper investigates the acid leaching of metals from a waste sample of dental silver alloy generated during the melt spray process. The alloy constitutes of 42.13% Ag, 31.03% Sn, and 26.84 Cu. The phase composition of amalgam generally consists of Ag_2Hg_3, Ag_3Sn, Sn_xHg, Cu_6Sn_5, and Cu_3Sn. The effects of the system temperature (25-80°C), nitric acid concentration as the leachate (13.75-65%), pulp density (33-200 g/l), and reaction time (0-240 min) on the dissolution recovery of silver, copper, and tin have been investigated. In the best case, we recovered 100% of silver and 98% of copper as soluble nitrates while tin was isolated as solid stannic oxide.

Keywords: Amalgam, Dental alloy, Leaching, Recycling, Silver

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1 INTRODUCTION

Dental amalgam is a solid solution alloy mainly consisting of silver, tin, and copper, combined ¬– amalgamated– with mercury, and usually is classified by its copper content. The commercial amalgam composition has changed during decades, but the composition of high-copper amalgam typically includes 40-60% silver, 27-30% tin, 13-30% copper, 1% zinc, and mercury [1]. The phase composition of amalgam generally consists of Ag_2Hg_3, Ag_3Sn, Sn_xHg, Cu_6Sn_5, and Cu_3Sn [1-2]. During the production processes of dental alloys often losses and wastages take place which are known as dental alloy wastes. Environmental legislations for heavy metals besides the economic benefits make this sort of waste a valuable secondary source of metal recovery.

Several studies have been reported around the recovery of metal values from dental alloy wastes [1-4]. Lee and Fung [2] have reported an A-to-Z procedure of metals recovery from amalgam wastes including the sublimation of mercury by heating, a two-stage leaching of the residue in nitric acid while tin remained solid as $SnO_2.xH_2O$, isolating the silver as AgCl by adding chloride to the solution, and reduction of sliver through reacting with ammonia water or sodium hydroxide. Yilmaz et al. [1] have investigated many influential factors in the recovery of silver from amalgam wastes through leaching in nitric acid and consequent cementation of silver by copper, showing an almost complete recovery of silver while tin was isolated in the leaching residue. The recovery of metal values from lead-free solder wastes (mostly containing tin, silver, and copper) is also a chemically-similar case [5-7].

The aim of the present paper is a parametric leaching study of metals from dental silver alloy.

2 MATERIALS AND METHODS

In the present study, a sample of dental alloy waste was generated during the melt spray process before the amalgamation (Faghihi Company,Karaj, Iran) to investigate the leaching parameters. The chemical and phase compositions of the alloy are given in "Table 1" and "Fig. 1", respectively.

Table 1 The chemical composition of the dental alloy.

Element	Ag	Sn	Cu
wt%	42.13	31.03	26.84

All the leaching experiments were conducted using 100 ml-acid solution batches in a 250 ml three-neck glass reactor balloon on a heating mantle equipped with magnetic stirrer, illustrated in "Fig. 2". The acid solution batches were prepared using laboratory-grade nitric acid (65%) and deionized water.

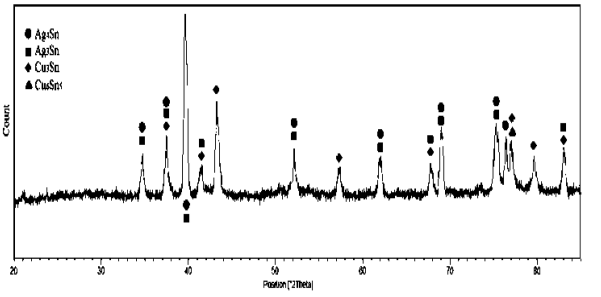

Fig. 1 The x-ray diffraction pattern of the dental alloy.

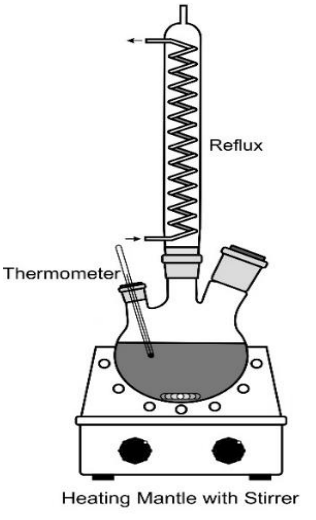

Fig. 2 Setup of the leaching experiments.

The effects of four parameters on the dissolution percent of the metals were investigated: (1) temperature: 25-80°C, (2) acid concentration: 13.75-65%, (3) pulp density: 33-200 g/l, (4) time: 0-240 min. The chemical analysis of the metals in the solutions was determined by using Vista-PRO Simultaneous ICP-OES (Varian Inc.) device.

3 RESULTS AND DISCUSSION

3.1. Effect of Temperature

The effect of temperature on the dissolution recovery of silver, copper, and tin is represented in "Fig. 3". According to the results, the recovery of silver has been increased from 88% to almost 100% by a temperature rise from 25°C to 50°C, while copper has not completely dissolved unless the experiment temperature was set to

70°C. Thermodynamically, we may expect copper to be dissolved more easily than silver because copper is a more active metal. However, the alloy does not consist of separate pure metals, but intermetallic compounds, which is a thermodynamically-different case. On the other hand, despite the significant temperature rise, tin is seemed to be intact. Practically, tin reacts with nitric acid resulting in stannic oxide and nitrogen oxides as given in "Eq. (1-2)" [8]:

$$3Sn + 4HNO_3 + H_2O = 3SnO_2 \cdot H_2O + 4NO \quad (1)$$

$$Sn + 4HNO_3 = SnO_2 \cdot H_2O + 4NO_2 + H_2O \quad (2)$$

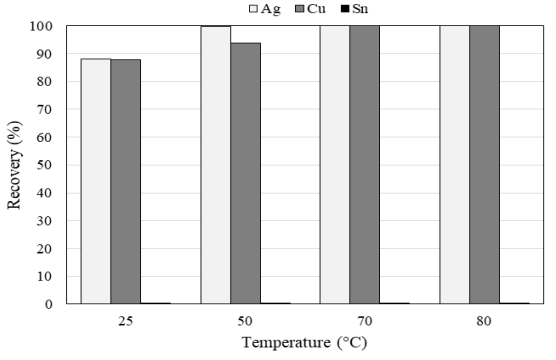

Fig. 3 Dissolution recovery of Ag, Cu, and Sn from the dental silver alloy at different temperatures (27.5% HNO¬3, 50 g/l pulp density, 180 min, 300 rpm).

3.2. Effect of Acid Concentration

As seen in "Fig. 4", a nitric acid concentration of 27.5% is adequate for almost complete recovery of silver while copper needs at least 55% to approach the complete dissolution (99%) in the experimental conditions. The maximum yield of tin dissolution is about 1% at the acid concentration of 55%.

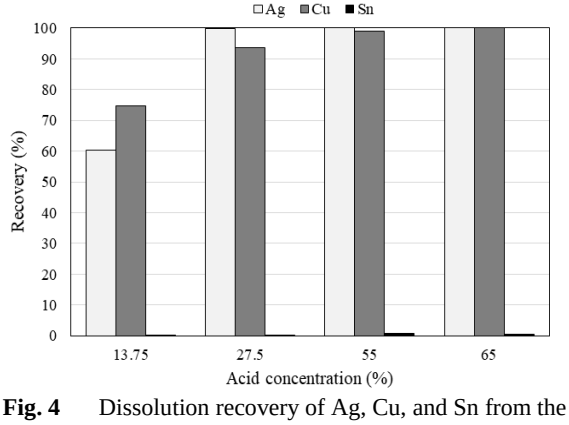

Fig. 4 Dissolution recovery of Ag, Cu, and Sn from the dental silver alloy at different nitric acid concentrations (50°C, 50 g/l pulp density, 180 min, 300 rpm).

3.3. Effect of Pulp Density

The investigation of pulp density effect is of practical importance because the results determine the highest solid to liquid ratio not affecting the leaching recovery. As concluded from "Fig. 5", pulp density, generally, has no significant effect on the leaching recovery of the metals; therefore, economically, the highest is the best. We calculated that at the tested pulp densities, there are over-stoichiometric nitric acid quantities based on "Eq. (3-6)":

$$3Ag + 4HNO_3 = 3AgNO_3 + NO + 2H_2O \quad (3)$$

$$Ag + 2HNO_3 = AgNO_3 + NO_2 + 2H_2O \quad (4)$$

$$3Cu + 8HNO_3 = 3Cu(NO_3)_2 + 2NO + 4H_2O \quad (5)$$

$$Cu + 4HNO_3 = Cu(NO_3)_2 + 2NO_2 + 2H_2O \quad (6)$$

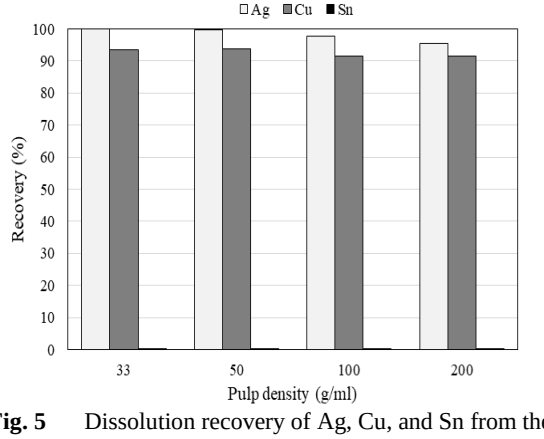

Fig. 5 Dissolution recovery of Ag, Cu, and Sn from the dental silver alloy at different pulp densities (50°C, 27.5% HNO3, 180 min, 300 rpm).

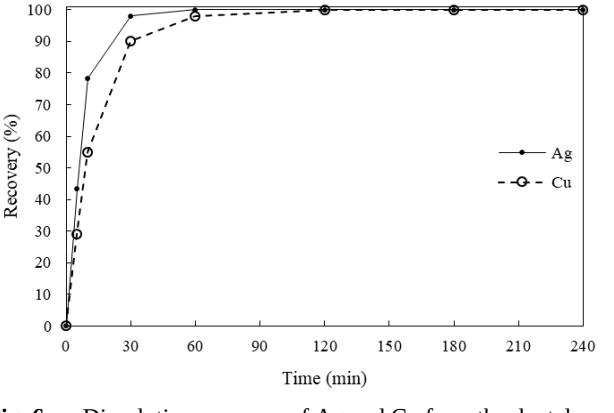

Fig. 6 Dissolution recovery of Ag and Cu from the dental silver alloy versus time (50°C, 27.5% HNO3, 200 g/l pulp density, 300 rpm).

3.4. Effect of Time

We performed a continuous leaching test for 240 minutes to determine the minimum time required for dissolution of silver and copper. The total dissolution yield of tin did not exceed 1%, and hence, we disregarded it. The result illustrated in "Fig. 6" reveals that 60 minutes suffices an almost complete recovery of silver (100%) and copper (98%) as soluble nitrates.

4 CONCLUSION

The leaching of silver, copper, and tin from a waste sample of dental silver alloy (Fighihi co.) was investigated at different conditions of temperature (25-80°C), nitric acid concentration (13.75-65%), pulp density (33-200 g/l), and time (0-240 min). The results demonstrate that the most influential parameters are acid concentration, temperature, time, and pulp density, respectively. The best-case experimental conditions according to the results are 50°C, 27.5% HNO3, 200 g/l pulp density, and 60 minutes resulting in the dissolution yield of 100% Ag, 98% Cu, and 1% Sn. While silver and copper successfully dissolved as nitrates, tin isolated in the leaching residue as solid stannic oxide.

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