



Effect of Al₂O₃ mole fraction and cooling method on vitrification of an artificial hazardous material. Part 1: Variation of crystalline phases and slag structures

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ABSTRACT

This study investigated how Al ions affect slag structure. During vitrification, pure Al₂O₃, CaO, and SiO₂ served as the encapsulation phases with the use of Al mol% as an operating parameter. All specimens with the same basicity (mass ratio of CaO to SiO₂) of 2/3 were vitrified at 1400 °C and cooled by air cooling or water quenching. XRD was used to measure the volume fractions of crystalline and amorphous phases. In a non-Al environment, CaSiO₃ was formed in air-cooled and water-quenched slags. With the addition of Al₂O₃, no crystalline phases were observed in water-quenched slags. With the increase of Al mol% in specimens, the Al ions in air-cooled slags initially acted as an intermediate linking one tetrahedron chain to another and reducing the amount of crystalline phase, then behaved as a network former making the slags amorphous, and finally replaced Si ions in silicate frames to generate a large amount of CaAl₂Si₂O₈. Air cooling improved the formation of crystallize structures with more leachable metal ions. A highly crystallized Al-framed structure is not suitable for encapsulating hazardous metals in vitrified slags.

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1. Introduction

Vitrification is widely used to treat hazardous materials such as fly ashes, electroplating sludge, and nuclear wastes [1–3], because it may be applied to destroy organic toxics, recover metals, and stabilize hazardous metals [4–6]. Slags obtained from vitrification are commonly reused as additives for building materials, glassy ceramics, or cements [7,8]. For such cases, the immobilization of hazardous metals is especially important from an environmental point of view. To vitrify the wastes containing toxic metals, the ternary glass matrix system of CaO–Al₂O₃–SiO₂ (CAS) plays an important role in encapsulating hazardous metals. The mass ratio of CaO to SiO₂ in an original specimen is defined as its basicity, which is usually adopted to evaluate the control of vitrification processes. In a glass matrix, the most important components for glass networks are Si and O, and these two basic elements often influence the slag structure. Ca ions also affect the slag structure because they randomly distribute through the structure to provide local charge sites, and therefore, they act as network modifiers to modify the network structure. For years, many studies have been conducted to explore how basicity governs vitrification processes [9,10].

In addition to Si and Ca cations, Al³⁺ also plays an important role in the formation of crystalline structures because it acts as both

a network modifier and an intermediate in silicate minerals [11]. However, little attention has been paid to the effect of Al₂O₃ on the structures of slags or the immobilization of hazardous metals.

In a random glass network model, the coordination number of O ions of SiO₄ tetrahedra and (AlO₄)[−] tetrahedra is 4. The Si and Al ions are both network formers. However, Al ions, with a coordination number of 6, may link tetrahedral silicate groupings through a simple ionic bond and are thus referred to as intermediates [12].

A few researchers have investigated how Al₂O₃ addition affects physical properties of slags. One study reported that increasing the amount of Al₂O₃ enhanced early hydration of cement but excessive amounts of Al₂O₃ decreased strength of a slag when the slag was blended with Portland cement [13]. The amount of Al₂O₃ greatly affects the crystallization of slags in a CAS system [14]. Al₂O₃ governs the structure of a slag and is significantly associated with strength of the slag, but little is known about how Al₂O₃ influences the immobilization of hazardous materials. When recycling slags vitrified from hazardous materials, encapsulation is more important than physical properties. Therefore, we explored the effect of Al ions and two cooling ways on the crystalline characteristics of slags and the encapsulation of metals. The results are provided by two sections: Part 1: variation of crystalline phases and slag structures, and Part 2: encapsulation of metals and resistance to acid. The former, presented in this paper, discussed the volume fractions of crystalline phases and amorphous phases of slags. The influences of Al₂O₃ mole fraction and the cooling method on the formation of slag structures were also investigated. The microstructures of slags

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Table 1
Physical properties of targeted metals.

Targeted metal	Boiling point (°C)	Specific weight
Cr	2200	7.1
Cu	2300	8.9
Mn	1900	7.2
Ni	2900	8.9
Pb	1620	11.3

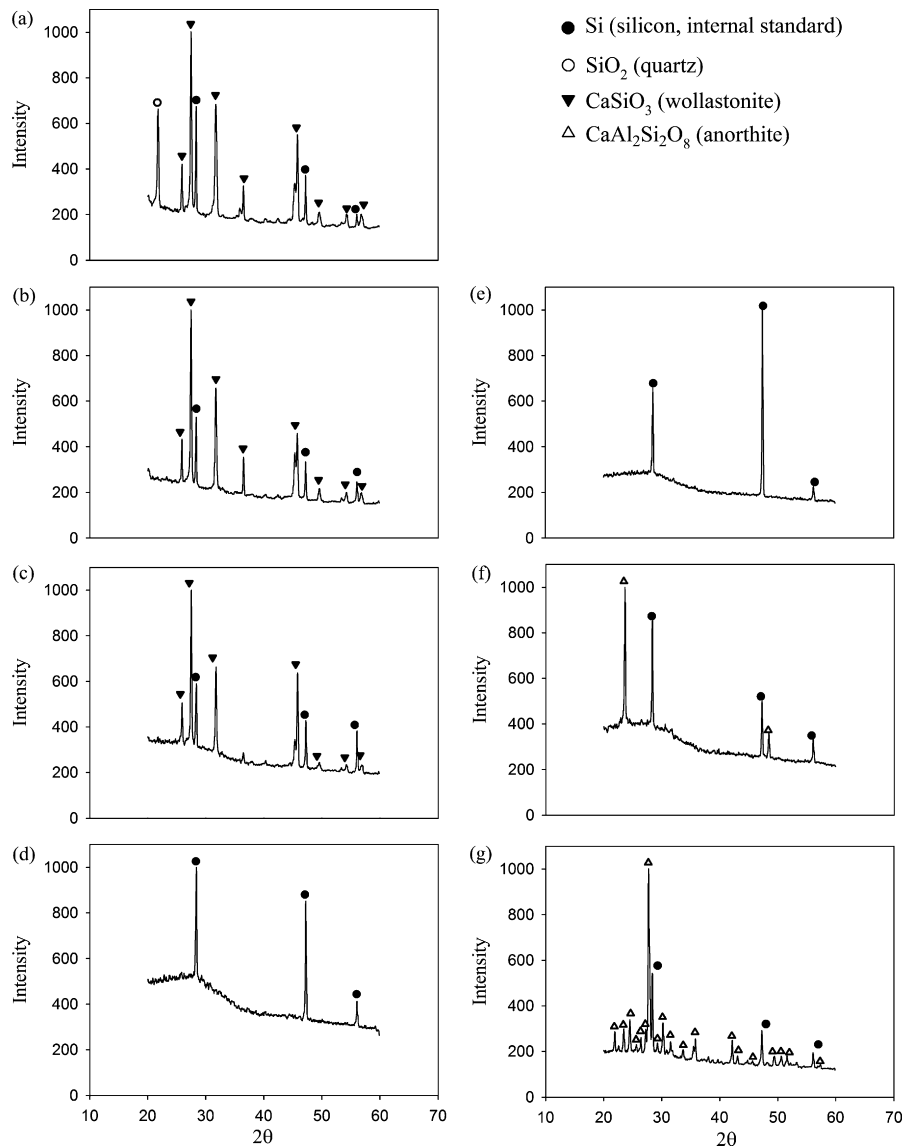
Table 2
Composition in mol% of the glass matrix of the original specimens.

Specimen (%)	Al ₂ O ₃ (as Al ions)	CaO (as Ca ions)	SiO ₂ (as Si ions)
A-0, W-0	0.0	41.7	58.3
A-1, W-1	5.7	39.3	55.0
A-2, W-2	11.3	37.0	51.8
A-3, W-3	16.8	34.7	48.5
A-4, W-4	22.2	32.4	45.4
A-5, W-5	27.6	30.2	42.2
A-6, W-6	32.9	28.0	39.1

Table 3
Compositions and percent mass retained (PMR) data of slags.

Targeted metal	A-0				W-0			
	Range (mg/kg)	Average (mg/kg)	RSD (%)	PMR	Range (mg/kg)	Average (mg/kg)	RSD (%)	PMR
Cr	1220–1260	1240	1.90	1.23	969–1070	1020	5.12	1.23
Cu	767–902	855	8.91	0.847	451–563	517	11.4	0.512
Mn	1410–1530	1450	4.50	1.44	1170–1370	1270	7.71	1.26
Ni	1020–1250	1130	9.87	1.12	726–854	809	8.88	0.801
Pb	176–181	178	3.30	0.176	193–200	197	2.80	0.199

RSD: relative standard deviation(= standard deviation/average × 100%).

**Fig. 1.** XRD patterns of air-cooled slags: (a) A-0, (b) A-1, (c) A-2, (d) A-3, (e) A-4, (f) A-5, and (g) A-6.

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