國立臺北科技大學
資源工程研究所

碩士學位論文

以無機聚合材料固化/穩定化焚化飛灰之研究

Study on MSWI Fly Ash Solidified / Stabilized with Geopolymer Material

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摘　要

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都市垃圾焚化飛灰含有大量之重金屬，其處理方法以水泥系固化法為主，因水泥固化體之抗壓強度低，且重金屬在長期酸性環境下仍有再溶出之虞，為改善水泥固化法之缺點，本研究將焚化飛灰參配變高嶺石及鹼性活化液共同製作無機聚合固化體，探討不同養護時間、SiO_2/Al_2O_3 比及 SiO_2/Na_2O 比對於無機聚合固化體之抗壓強度、固化重金屬及其顯微結構之影響，並進一步評析焚化飛灰無機聚合固化體之長期穩定性。

由研究結果顯示，以 SiO_2/Al_2O_3 比 3.0 及 SiO_2/Na_2O 比 0.75 製得之焚化飛灰無機聚合固化體具有較佳之抗壓強度及重金屬 Pb 固化效能，固化體於養護 28 天後，抗壓強度達 14.6 MPa，而固化體中重金屬 Pb 之 TCLP 溶出濃度低於 0.1 mg/L。由 ^{29}Si、^{27}Al NMR 及 XRD 之分析結果顯示，焚化飛灰無機聚合固化體皆具有非晶質之 CSH 鏈狀矽酸鹽結構及非晶質之三維鋁矽酸鹽架狀結構，主要結晶相為 NaCl、$Ca(OH)_2$、SiO_2、$Ca_2Al(OH)_6Cl \cdot 2H_2O$ 及 $CaCO_3$。而由 FTIR 分析結果顯示，固化體中 Si-O-T (T=Si 或 Al) 鍵之非對稱伸張振動吸收峰位於 941~957cm^{-1} 間。由 SEM/EDS 顯微結構分析結果顯示，固化體為表面呈現緻密而帶有細小粉狀顆粒，

元素組成以 Na、Cl、Ca、Si 及 Al 為主。

由 SiO_2/Al_2O_3 比 3.0、SiO_2/Na_2O 比 0.75 合成並經養護 28 天的焚化飛灰無機聚合固化體之半動態溶出試驗結果顯示，固化體於連續 60 天萃取下，其抗壓強度雖由原始的 14.6 MPa 下降至 0.8 MPa，但仍有 60%以上的重金屬 Pb 仍未溶出，顯示焚化飛灰無機聚合固化體中重金屬 Pb 具有長期穩定性。而由不同萃取時間固化體顯微結構分析結果顯示，固化體萃取至 60 天後，其表面顯微形態呈現一絲絲多孔狀，元素組成主要以 Si 元素為主。由 FTIR 分析結果顯示，隨著萃取時間之增加，固化體中 Si-O-T (T=Si 或 Al) 鍵非對稱伸張振動之吸收峰會愈往高波數移動。此外，固化體中共存有非晶質之 CSH 鏈狀矽酸鹽結構及非晶質之三維鋁矽酸鹽架狀結構，但隨著萃取時間之增加，固化體中逐漸轉變成以非晶質之三維鋁矽酸鹽架狀結構為主。

ABSTRACT

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Because of amounts of heavy metals in municipal solid waste incinerator fly ash (MSWI fly ash), MSWI fly ash solidified/stabilized with cement and chelating agents was the major treatment technology of MSWI fly ash. However, even after solidification/stabilization, heavy metals in MSWI fly ash could be leached in the acidic condition. Therefore, it was utilized fly ash, metakaolinite, sodium silicate and alkaline solution to prepare fly ash geopolymerized solidification/stabilization matrices (FAGPSSM) in this study. The effects of curing time, SiO_2/Al_2O_3 ratio and SiO_2/Na_2O ratio to compressive strength, heavy metals leachability and micro-structure of FAGPSSM would be investigated and long-term stability of heavy metals in FAGPSSM would be evaluated.

The results indicated that the compressive strength and heavy metal leachability of FAGPSSM prepared with SiO_2/Al_2O_3 and SiO_2/Na_2O ratio respectively 3.0 and 0.75

would be better. The leaching concentration of Pb in FAGPSSM cured after 28 days by TCLP method was lower than 0.1 mg/L and it showed that FAGPSSM could effectively immobilize Pb in MSWI fly ash. The spectra of ^{29}Si and ^{27}Al NMR implied that there were amorphous calcium silicate hydrate (CSH) gels and aluminosilicate gels both coexisting in FAGPSSM. The XRD patterns showed that the amount of crystallinity present was mainly caused by NaCl, $Ca(OH)_2$, SiO_2, $CaCO_3$ and $Ca_2Al(OH)_6Cl$ phases present in FAGPSSM. The spectra of FTIR indicated that the vibrational band at 941～957 cm^{-1} was attributed to the Si-O-T (T=Si or Al) asymmetric stretching mode. The microstructures of FAGPSSM showed that the grain of FAGPSSM was compact and powder present on the surface of particle.

The semi-dynamic leaching test (SDLT) was conducted to evaluate the long-term stability of Pb in FAGPSSM prepared with SiO_2/Al_2O_3 and SiO_2/Na_2O ratio respectively 3.0 and 0.75 and cured after 28 days. The results carried out by SDLT showed that after 60 days extraction, the compressive strength of FAGPSSM would decrease from 14.6 MPa to 0.8 MPa, but the cumulative leaching percentage of the total Pb in FAGPSSM only reached to 40%. Therefore, it inferred that the long-term stability of Pb in FAGPSSM was better than in the cementitious solidified/stabilized matrix. The microstructures of FAGPSSM showed that the grain of FAGPSSM would become porous after 60 days extraction. The spectra of FTIR indicated that as extraction time increased, the vibrational band of Si-O-T (T=Si or Al) asymmetric stretching mode would shift to higher wavenumbers. After 60 days extraction, the structure of FAGPSSM would turn from the coexistence of the amorphous CSH gels and aluminosilicate gels to the aluminosilicate gels.