

行政院國家科學委員會專題研究計畫 成果報告

都市垃圾焚化底渣燒製多孔性輕質材料受熱膨脹機制之研究 研究成果報告(精簡版)

計畫類別：個別型
計畫編號：NSC 96-2221-E-032-008-
執行期間：96年08月01日至97年11月30日
執行單位：淡江大學水資源及環境工程學系

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報告附件：出席國際會議研究心得報告及發表論文

處理方式：本計畫可公開查詢

中華民國 97 年 12 月 29 日

行政院國家科學委員會補助專題研究計畫 成果報告
 期中進度報告

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成果報告類型(依經費核定清單規定繳交)： 精簡報告 完整報告

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漢技術學院環境資源管理系

中 華 民 國 9 6 年 1 1 月 2 8 日

一、前言

過去十多年來台灣經濟高度的成長國民生活水準不斷提高，都市廢棄物的產生在人口都市化的今日，是相當嚴重的問題。目前台灣將面臨都市垃圾處理的重大考驗，由於台灣地小人稠，而環保意識的高漲，更造成廢棄物處理設施興建上的困難，因此垃圾掩埋場和焚化爐設立便困難重重。

目前關於焚化灰渣處理方式已逐漸發展成資源再利用，根據行政院環保署於民國九十二年 12 月公告「一般廢棄物一垃圾焚化廠焚化底渣再利用管理方式」一案，已然確立灰渣處理方向，唯國人對灰渣中重金屬及戴奧辛之危害存有疑慮，因此對灰渣再生性產品望之退怯，故未來除了政府既定政策需落實執行，更應加強研究低污染、無害化和對人體無傷害的乾淨產品，且應修法保障再生性材料之出路，逐步達成源頭減廢、全回收及零廢棄之目標。

二、文獻回顧

輕質骨材為輕質化工程材料的一支，混凝土添加輕質骨材後即成所謂的輕質混凝土(形成輕質混凝土方式有多種，如輸入氣泡，添加高分子聚合顆粒等)，可增強建築物之耐震能力(郭氏，2000)，還具有質輕、可隔熱、隔音之優點(王氏，2005)，作為隔間材亦有相當優良之工程表現。

輕質骨材歐美發展時間較台灣來的早，以美國為例，Hayde 於 1917 年以旋窯燒結膨脹黏土及膨脹頁岩取得專利，隨後於 1919 年設廠開始生產，於十年後設立移動式爐架燒結系統，至此為輕質骨材商業化之濫觴。1950 年代所產製之輕質混凝土其抗壓強度可達 45 MPa，比早期輕質混凝土的抗壓強度 14 MPa 要高出許多，二次大戰結束後，美國輕質化材料快速發展，所研發出低自重、耐久性佳之輕質磚和輕質骨材，因此於工程上大量使用(林氏，2006)。

亞洲對於輕質骨材研究以日本起步較早，中國輕質粒料發展自 1956 年山東地區首次採用水泥回轉窯成功試燒黏土陶粒，於今日也有 50 年光陰，目前中國當局有心推廣及政府強力導向，目前中國輕質材料的使用已經相當盛行。至於台灣輕質骨材研究起源則是於 1970 年代成功大學土木系材料實驗室和台灣大學土木系材料實驗室進行開發，成大王櫻茂教授以旋轉窯試燒膨脹性頁岩，並進行造粒型和非造粒型及破碎型輕質骨材製作；1980 年代台大高健章教授採用破碎頁岩，試燒非造粒型輕質骨材；1990 年代台灣工業技術學院營建系(台灣科技大學前身)黃兆龍教授發展出冷結型輕質骨材，並取得專利(黃氏，2003)。

在輕質骨材之製程中，由 Riley(1951)等人陸續提出在溫度升高時有兩個不可或缺的發泡機制與條件，方能使原料膨脹製成輕質骨材(Bhatty, 1989)。目前已被各界接受原料之膨脹或發泡機制主要條件為：

- (1)原料經加熱至高溫時，表面須先生成黏性之玻璃相特性，並有足夠之時間來封閉自骨材內部放出之氣體。
- (2)在高溫生成黏性玻璃相後，原料中必須含有釋放出氣體之物質，並認定為材料之膨脹或發泡現象是輕質骨材製造上重要機制。

由一些以天然材料燒製成輕質骨材的文獻中發現原料以黏土、頁岩及板岩等，其中礦物相以伊萊石(Illite)受眾多文章討論，伊萊石化學式大概是 $K_x(Al,Fe,Mg)_2(Si,Al)_xO_{10}(OH)_2$ ，而陶瓷技術中也確定添加高嶺石和細粒雲母碎片可有效達成膨脹之目的(程氏，1984)，而根據窯業專業著作中指出黏土在不同溫度之下，不同的化合物會受熱分解(Norton F.H. 原著，1977)，由王氏之研究中針對黏土礦物添加膨脹劑如

赤鐵礦、黃鐵礦、白雲石和方解石來增加其膨脹性，在加熱時觀察顏色，當膨脹溫度會造成顏色轉棕色，由此可知三價鐵離子產生變化所形成的（王、顏，1992）。

三、研究目的

為了能進一步確定焚化底渣燒製輕質的機制，本實驗將利用都市垃圾焚化底渣，添加氧化鐵和氧化矽，利用不同的燒結技巧，觀察骨材受熱膨脹的情況，進一步設計輕質骨材膨脹氣體收集裝置，針對不同加熱溫度所產生出的氣體進行分析，則將氣體成分和原料成份於輕質骨材膨脹三相圖內進行比較，藉此了解成份和膨脹氣體的關係。本研究之研究目標有以下三點。

- 底渣之基本物理和化學組成之分析：底渣之外觀、灼燒減量、粒徑分析、pH、和金屬元素組成。
- 將骨材膨脹之發泡氣體進行收集，進行逸氣分析：逸氣成份分析和骨材原料之關係進一步分析，將逸氣成份和氧化鐵添加量進行比較，確立氧化鐵於輕質粒料燒製中所扮演之角色。
- 建立輕質骨材燒製之參考指標。由於傳統輕質骨材燒製時只有單純的研究原料—膨脹頁岩、黏土及浮石等，如今轉變為成份複雜的都市垃圾焚化灰渣，操作上之困難度不可同日而語，也增添了許多變數。因此建立更多的燒結條件作為燒製骨材的操作依據有其必要性，如此也可加速商業化之腳步。

四、研究方法

本實驗將使用前處理後之底渣利用 SiO_2 及 Fe_2O_3 進行調質，改變其燒製條件，目標為製作出具有工程性之輕質骨材，實驗分成「前處理」和「骨材膨脹機制探討」兩部份。

1. 都市垃圾焚化底渣前處理：定北區某大型都市垃圾焚化底渣資源再生廠作為研究材料，原料經由兩次水萃，每次 5 分鐘，液固比為 2 (wt%)，水萃後以 #200 進行過篩，篩上物顎碎後球磨變成粗底渣，篩下物則為細底渣。
2. 前處理完底渣進行調質和各種相關機制的探討：本實驗將粗底渣添加添加氧化矽和氧化鐵，利用不同燒結條件觀察粒料受熱膨脹之情況，進一步設計膨脹氣體收集裝置，將燒製過程所產生之氣體進行逸氣分析。添加的方式及比例如下：

(a) **實驗一**：固定底渣含量，燒製過程中控制粒料不熔坍變形，研究氧化鐵添加對膨脹氣體產生之影響。調質比例：將 10 克的粗底渣添加 1 克之 SiO_2 之後，再行添加 0.1、0.2、0.3、0.4、0.5 克之 Fe_2O_3 ，樣品名稱訂為 NO.1~NO.5。燒製條件：燒製溫度訂為 1080°C ，由室溫為初始溫度，加熱速率為 $15^\circ\text{C}/\text{min}$ ，達到 1080°C 時進行持溫，持溫時間隨 Fe_2O_3 的添加量增加而逐次減少，爐體氣氛為 N_2 ，爐內壓力為 $1.5\text{Kg}/\text{cm}^2$ 。

(b) **實驗二**：將粗底渣與 Fe_2O_3 依比例添加，每件樣品之燒製條件控制不變，研究未添加 SiO_2 下之影響。調質比例：將粗粒徑底渣添加 Fe_2O_3 (未添加 SiO_2)，比例為 1%、2%、3%、4%、5% (重量百分比)，粒料總重為 10 克，樣品名稱訂為 NO.1-F~NO.5-F。燒製條件：燒製溫度訂為 1070°C ，由室溫為初始溫度，加熱速率為 $15^\circ\text{C}/\text{min}$ ，達到 1070°C 時進行持溫，持溫時間為 6 分鐘，爐體氣氛為 N_2 ，爐內壓力為 $1.5\text{Kg}/\text{cm}^2$ 。

(c) **實驗三**：將粗底渣混入 10%wt.之細底渣，再添加 Fe_2O_3 調質，每件樣品之燒製條件不變，研究粒料加入細底渣之變化。調質比例：將粗底渣添加 10%之細底渣，添加完成後再依比例加入 Fe_2O_3 進行調質，比例為 1%、2%、3%、4%、5%、10%、20% (重量百分比)，粒料總重為 10 克，樣品名稱訂為 NO.1A-F~NO.5A-F、NO.10A-F、NO.20A-F。燒製條件：燒製溫度訂為 1060°C ，由室溫為初始溫度，加熱速率為 $15^\circ\text{C}/\text{min}$ ，達到 1060°C 時進行持

溫，持溫時間為 7 分鐘，爐體氣氛為 N_2 ，爐內壓力為 1.5 Kg/cm^2 。

(d) **實驗四**：將粗底渣和混入 10% 細底渣之粗底渣，進行不同燒結氣氛之研究。調質比例：取粗底渣添加 10% 細底渣之混合灰進行燒製，粒料總重為 10 克，樣品名稱訂為 BA-10FI (Air)、BA-10FI(N_2)，純粗底渣樣品名稱訂為 BA (Air)、BA(N_2)。燒製條件：燒製條件分為兩者：(A) 純粗底渣：燒製溫度訂為 1070°C ，由室溫為初始溫度，加熱速率為 $15^\circ\text{C}/\text{min}$ ，達到 1070°C 時進行持溫，持溫時間為 6 分鐘，爐體氧化氣氛為 Air (壓縮空氣鋼瓶氣)，還原氣氛為 N_2 ，爐內壓力為 1.5 Kg/cm^2 。(B) 粗底渣添加 10% 細底渣：燒製溫度訂為 1060°C ，由室溫為初始溫度，加熱速率為 $15^\circ\text{C}/\text{min}$ ，達到 1060°C 時進行持溫，持溫時間為 7 分鐘，爐體氧化氣氛為 Air (空氣壓縮鋼瓶氣)，還原氣氛為 N_2 ，爐內壓力為 1.5 Kg/cm^2 。

逸氣分析裝置設立

逸氣分析系統主要分為兩個主要設備，首先是氣相層析儀設備，其次為電加熱式管狀高溫爐和兩設備之間的連接系統，逸氣分析系統圖見圖 1。分析過程分為兩階段，第一階段為採樣控制，採樣時間為 60 sec，採樣器溫度為 40°C 恆溫，待分析氣體填充於 1 mL 之 Loop，開始進行第二階段為注入，將樣品注入氣相層析儀之 Porapak Q 填充管柱進行分離，最後利用熱導偵測器 (TCD) 偵測二氧化碳之含量。

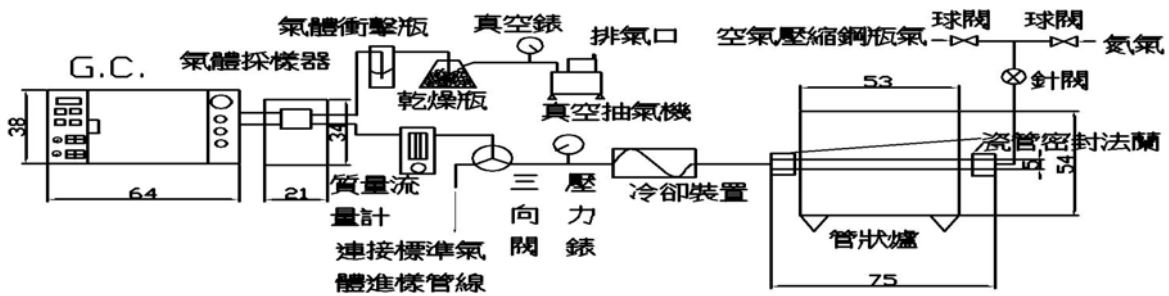


圖 1 逸氣分析系統圖

烘箱溫度 50°C 持溫，偵測器溫度 100°C ，偵測器流速 $26.5 \text{ mL}/\text{min}$ ，載氣為 He，載氣壓力為 5 Kg/cm^2 ，出口端流速為 $143 \text{ mL}/\text{min}$ 。

燒結體之工程性質分析

檢測燒結物之視比重、體密度、吸水率及視孔隙率等各項物理性質：參考耐火磚視孔隙度、吸水率及比重試驗 (CNS 619 R3013)。毒性特性溶出程序試驗依 NIEA R201.11C 進行。

五、結果討論

1. 逸氣分析結果

• 實驗一之逸氣分析成果

CO_2 圖譜數據如表 1 所示，可觀察到當底渣量固定時，不同添加量之 Fe_2O_3 確實在這組實驗中產氣有所變化，由於利用超純氦作為層析儀之載氣，提供了 TCD 較好的感度 (responds)，因此 Peak Height 值良好，且載氣流量和壓力控制適中，沖提 (elution) 時間對於分離氮氣和二氧化碳是相當理想，層析管柱在合適的溫控下，解析力 (resolution) R 值皆大於 1 (例如：NO.1 之 $R=2.714$)，證實了優良的離析效能。

表 1 實驗一之層析圖譜數據

Sample Name	Retention Time(min)	Area	Peak Height	Furnace Temp.
NO.1	1.033	462.74	38.797	1080
NO.2	1.050	845.097	100.563	1036
NO.3	1.050	479.763	46.466	1080
NO.4	0.733	1088.902	128.784	1080
NO.5	0.966	985.642	108.414	1080

註：CO₂最高濃度之情況

本實驗也針對粒料於受熱過程中，二氧化碳產生情況利用連續式線上分析來檢測，由圖 2 和圖 3 兩組可看出利用底渣燒製輕質骨材過程中，產氣情況皆於高溫區發生。

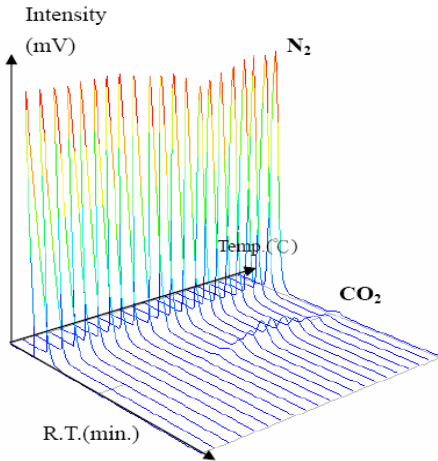


圖 2 NO.1 於 430°C~1080°C 層析圖譜 (最後一組為 1061°C)

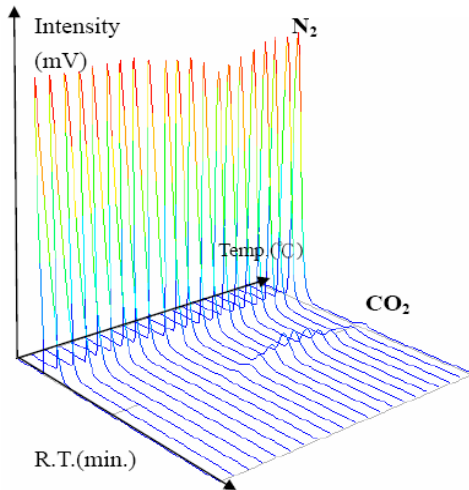


圖 3 NO.5 於 430°C~1080°C 層析圖譜 (最後一組為 925°C)

圖中兩 peak 之間溫度差約為 40°C，於加熱停止後再進一次樣，觀測燒結後產氣情況。二氧化碳之產生情況約在 800°C 後才有峰譜出現，於最高持溫之時，常有較大量二氧化碳之產生，因此可充分證明在高溫時膨脹氣體是呈現液相抑制其逸出的動態平衡之情況。

• 實驗二之逸氣分析成果

本段實驗停止添加氧化矽，Fe₂O₃ 和底渣依百分比進行添加，一方面觀察 SiO₂ 對於膨脹氣體之影響，一方面了解 Fe₂O₃ 於粒料燒結過程中扮演之角色。可由 peak area (表 2) 發現氣體濃度於實驗一之結果相比則濃度低了許多，而 Fe₂O₃ 的添加造成氣體量之改變未若實驗一如此明顯，因而推斷 Fe₂O₃ 對於二氧化碳變化之影響遠較底渣來的小。

表 2 實驗二之層析圖譜數據

Sample Name	Retention Time(min)	Area	Peak Height	Furnace Temp.
NO.1-F	0.966	713.563	86.625	1026
NO.2-F	1.016	459.647	59.993	1026
NO.3-F	0.983	599.8505	74.942	1069
NO.4-F	1.016	546.055	70.526	1026
NO.5-F	0.983	558.406	71.364	1069

註：CO₂最高濃度之情況

由加熱過程中之連續測量的結果中膨脹氣體大量出現位置也是大約在 800°C，但和前者不同之處在於停止加熱後，仍有不少的二氧化碳再冷卻過程中被偵測到，這對於粒料之膨脹是有正向幫助的，這就表示原料之活性是足夠的。

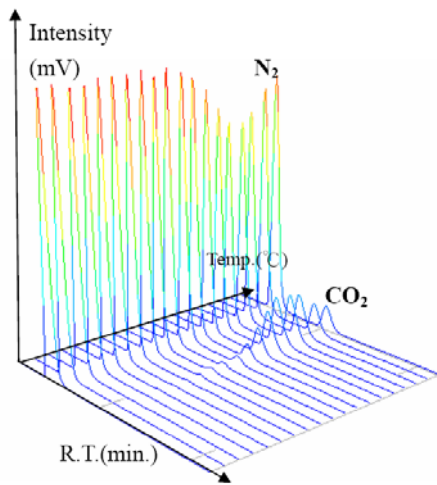


圖 4 NO.1-F 於 430°C~1070°C 層析圖譜 (最後一組為 983°C)

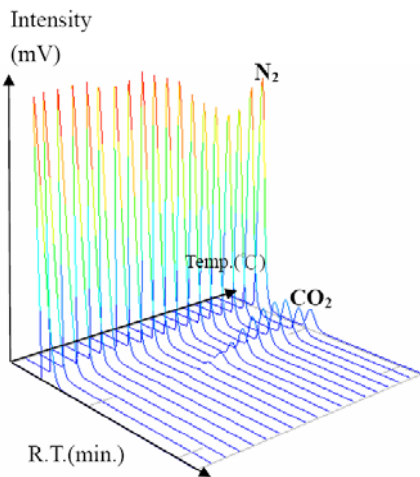


圖 5 NO.5-F 於 430°C~1070°C 層析圖譜 (最後一組為 979°C)

• 實驗三之逸氣分析成果

本階段實驗可了解細底渣對於粒料燒結乃至於膨脹氣體生成之影響，更將 Fe₂O₃ 添加量增加至 20%，如此可更有利於偵測 Fe₂O₃ 釋放二氧化碳之情況。將添加 10% 細底渣之原料，加入 Fe₂O₃ 後，二氧化碳產生量隨 Fe₂O₃ 添加量提升，然而添加 10% 和 20% 之 Fe₂O₃ 卻非二氧化碳最高濃度 (見表 3)，這種情況可能和 Fe₂O₃ (助熔劑) 添加量過多，造成粒料於加熱過程中提早熔塌，因此高量添加 Fe₂O₃ 所製成的粒料輕質化效果不及添加 1%~5%。

表 3 實驗三之層析圖譜數據

Sample Name	Retention Time(min)	Area	Peak Height	Furnace Temp.
NO.1A-F	0.983	789.841	95.327	1060
NO.2A-F	0.983	599.8805	76.26	1018
NO.3A-F	1.016	664.629	82.325	1018
NO.4A-F	0.983	778.424	92.835	1060
NO.5A-F	1.066	1161.458	107.894	1060
NO.10A-F	1.033	850.2785	99.223	1060
NO.20A-F	1.05	622.5255	77.458	976

註：CO₂最高濃度之情況

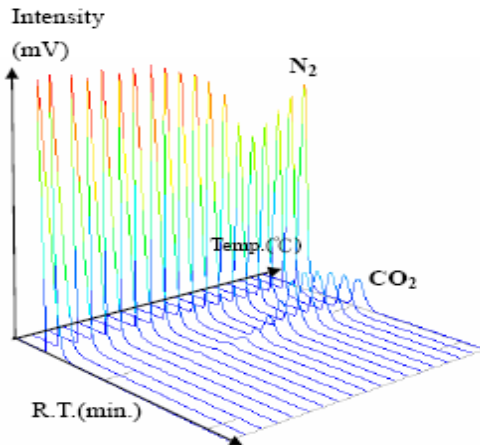


圖 6 NO.1A-F 於 430°C~1060°C 層析圖譜（最後一組為 878°C）

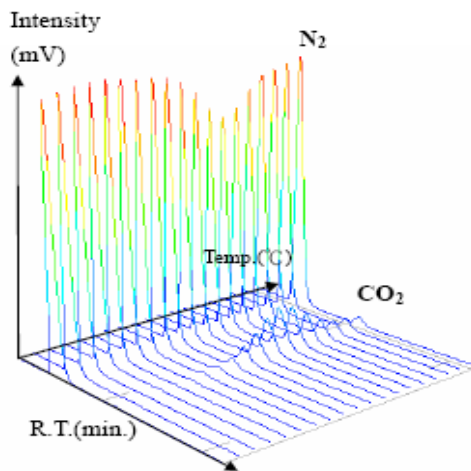


圖 7 NO.20A-F 於 430°C~1060°C 層析圖譜（最後一組為 921°C）

• 實驗四之逸氣分析成果

主要為測試不同燒結氣氛對於底渣和添加細底渣後，粒料燒製過成之變化，由表 4 中清楚得知粗底渣確實能造成較大量之二氧化碳生成，但導入空氣是否對二氧化碳產生改變仍無法由此得知，這是由於無法得知提供之空氣是否已經達到理論空氣量，因此由單一溫度下之氣體分析依然無法解決此問題。

表 4 實驗四之層析圖譜數據

Sample Name	Retention Time(min)	Area	Peak Height	Furnace Temp.
BA(Air)	1	500.182	67.524	941
BA(N ₂)	1.016	473.086	64.721	1069
BA-10FI(Air)	1	666.828	83.082	933
BA-10FI(N ₂)	0.95	1128.419	123.919	1069

註：CO₂最高濃度之情況

而從圖 8 和圖 9 及圖 10 和圖 11 兩相比較，發現通入空氣之樣品，其氣體產生最大量於九百多至一千度已經產生，而氮氣氣氛之樣品於加熱停止後仍舊可偵測到二氧化碳，由此可推測氮氣氣氛可得到較優良之輕質骨材（質輕、外觀完整、吸水率低）。這是因為在空氣氣氛下，二氧化碳出現時間過早，粒料在一千度之前，表面並無法達到黏性玻璃相，因此即使有氣體產生，粒料也未能含有足以包覆氣體之融相黏度，導致粒料全無膨脹情況。而氮氣氣氛燒結則是在高溫持溫時，氣體依然穩定生成，如此則可在高溫黏性玻璃相形成足夠剪力時，將氣體包覆住，達成抑制氣體逸出的動態平衡，因此可得到優良之輕質骨材。

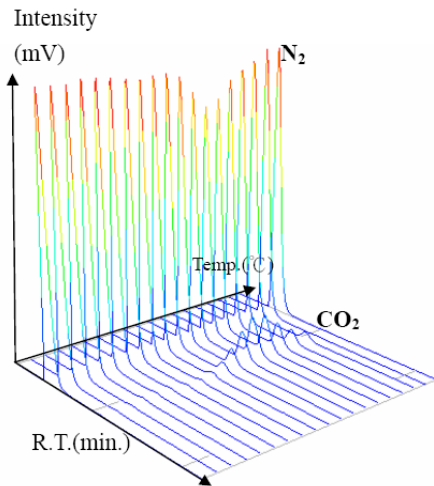


圖 8 BA(Air)於 430°C~1080°C 層析圖譜（最後一組為 1061°C）

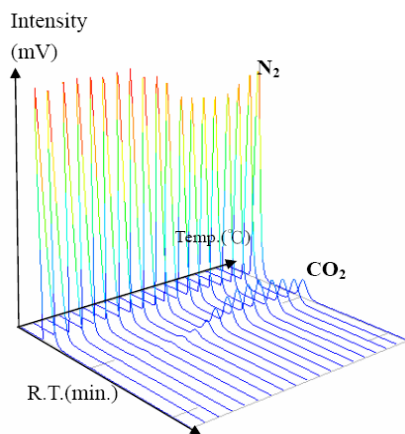


圖 9 BA(N₂)於 430°C~1080°C 層析圖譜（最後一組為 893°C）

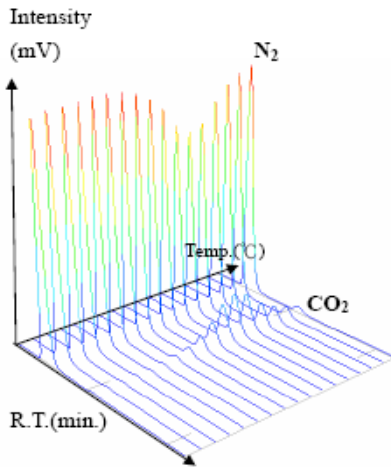


圖 10 BA-10FI(Air)於 430°C~1080°C 層析圖譜 (最後一組為 837°C)

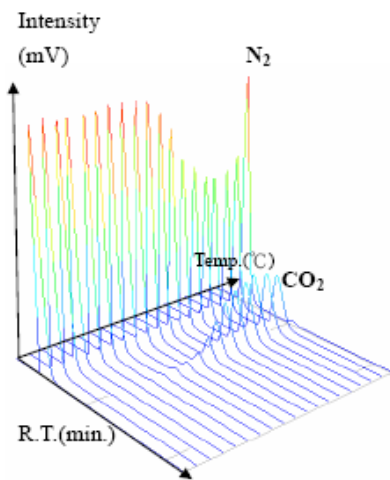


圖 11 BA-10FI(N₂)於 430°C~1080°C 層析圖譜 (最後一組為 875°C)

2. 燒結體工程性質分析

● 燒結體之視比重

實驗一中粒料皆達成輕質化之目標，甚至有比重小於 1，回顧 NO.5 的各項數據，不但孔隙多且均勻，氣體於高溫持溫段氣體仍不斷產出，因此確實達成輕質化之目標。

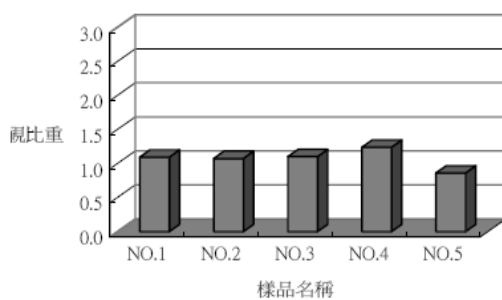


圖 12 實驗一之燒結體視比重

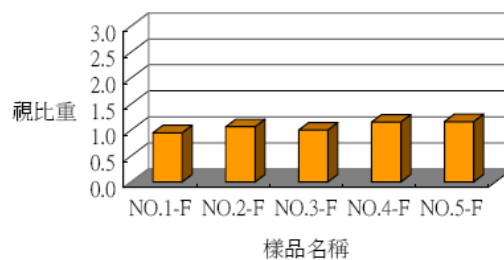


圖 13 實驗二之燒結體視比重

實驗二中粒料也順利達成輕質化目標，因此可推測添加 SiO₂ 於粗底渣內所造成的膨脹效應並未呈現，而一般添加矽化合物主要會造成網狀結構物，除了可形成完整之晶相外，於高溫之下還可形成黏度。實驗三中粒料之比重依然低於 1.8，輕質化特性明顯，唯獨高量添加 Fe₂O₃ 由於發生熔坍情況，因此損失不少孔洞，比重略為升高。

實驗四中兩種氣氛對於粒料之燒製可謂大相逕庭，綜合先前幾節之討論中，不難預料通入 Air 之氣氛，粒料無輕質化發生，比重接近於 2.5，和通入氮氣後比重降至為 1.0 左右，

氮氣氣氛的效果可見一般。

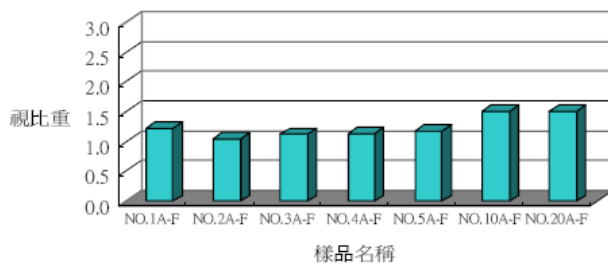


圖 14 實驗三之燒結體視比重

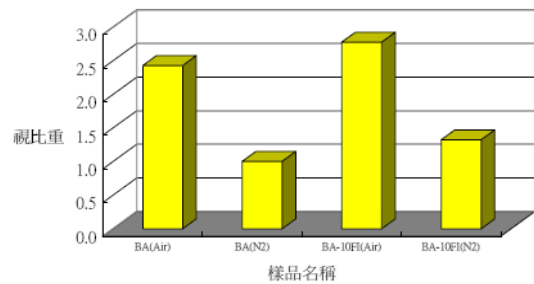


圖 15 實驗四之燒結體視比重

● 燒結體之 TCLP 及重金屬含量變化

燒結體之 TCLP 施作則選擇本實驗中吸水率和視孔隙度較高之燒結體，由於較高之吸水率可能會增加燒結體內部和液體接觸之機會，燒結體選定為 NO.3A-F，其視孔隙度為 7.57%，吸水率高達 7.29%，為相同燒結條件下之最高值。TCLP 及重金屬含量變化實驗結果見表 5。

表 5 燒結體之 TCLP 試驗

element	NO.3A-F(mg/L)	法規限制
Pb	0.214±0.00	5
Zn	0.934±0.03	-
Cu	0.433±0.01	15
Cd	N.D.	1
Cr	0.098±0.00	5

註：ave±SD，樣品數：3，燒結體：NO.3A-F，終點 pH：4.60
 N.D.：低於偵測極限；Zn 目前無管制，法規限制皆為總金屬量
 Detection limit：Pb = 5 ppb、Zn = 0.3 ppb、Cu = 0.6 ppb、Cd = 0.35 ppb、Cr = 0.5 ppb.

燒結體之 TCLP 量相當低，不到法規限制的十分一，因此安全性相當可靠，而燒結體之重金屬含量變化中，仍然以鋅為最大量，約有 19%，其他變化量則相當低，可了結燒製過程重金屬移動性低，因此造成二次污染的情況也較小，是相當穩定的再生材料。

表 6 燒結體之燒前、後重金屬含量變化

Elements	燒前含量(μg)	燒後含量(μg)
Zn	43±0.5	35±1.6
Cr	5±0.0	5±0.0
Pb	10±0.5	11±0.0
Cd	N.D.	N.D.
Cu	17±0.5	23±1.1

註：ave±SD，樣品數：3，燒結體：NO.3A-F
 N.D.：低於偵測極限；燒前離粒重：9.70 g；燒後燒結體重：8.90 g
 Detection limit：Pb = 5 ppb、Zn = 0.3 ppb、Cu = 0.6 ppb、Cd = 0.35 ppb、Cr = 0.5 ppb.

六、結論

(1) #200 以上之粗粒徑底渣經研磨後，均質的粗底渣確實可燒製出輕質骨材，而粗骨材也較細骨材更適合燒製再生產物。

(2) 利用氣相層析儀所建置的逸氣分析系統經過實際操作後是可行，未來可透過更換層析管柱和偵測器對其他可能產生的氣體成份進行觀測。

(3) 在氮氣氣氛下，圖譜上出峰之溫度約為 750~800°C，而於高溫持溫段氣體產出量最大，停止加熱後則 CO₂ 濃度逐漸降低。

(4) 若於停止加熱後仍可量測到 CO₂ 時，則可預估氣體於高溫時和黏性玻璃相達成動態平衡，因此骨材應該含大量孔隙，有助於輕質化。

(5) 空氣氣氛下燒結時，由於 CO₂ 氣體提早出現，因此高溫黏性玻璃相並無法產生足夠剪力來對氣體進行包覆，膨脹效果差。

(6) 若能加以分析其他氣體之生成，則較能了解釐清其膨脹和其成份之關係。而底渣本身之原料特性可能會造成產氣的差異，此方面應可加以研究，而燒失量和產氣情況所呈現之關係仍需研究。

(7) 灰渣本身差異性頗大，可對不同採樣點之灰渣進行分析，針對氣體膨脹情況和成份進行比較，以循求出更佳的發泡條件和技術。

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註：與本篇計畫相關著作

陳政綱，「都市垃圾焚化底渣燒製輕質骨材膨脹機制之研究」，碩士論文，淡江大學水資源及環境工程學系，2007。

陳政綱、何志軒、孫常榮、高思懷，「都市垃圾焚化底渣燒製輕質骨材之膨脹機制研究」，第三屆全國輕質骨材混凝土研討會，2007，台中。

陳政綱、何志軒、孫常榮、高思懷，「即時分析都市垃圾焚化底渣燒製輕質骨材生成之 CO₂」*中華民國環境工程學會第十九屆廢棄物處理技術研討會*，2007，高雄。

八、計畫成果自評

本研究主要探討都市垃圾焚化底渣燒製輕質骨材過程中，對於骨材的膨脹機制進行深

入的研究，研究初期包括原料前處理之研究，內容含蓋了底渣的兩段式水萃以及過篩和研磨的步驟，前處理之研究主要為整合本研究團隊對於原料前處理上之諸多研究，研究的主軸在於骨材原料的調質乃至於骨材燒製過程中膨脹氣體之逸氣分析，同時針對骨材表面的晶相和燒結條件之間的關係有更全面性的瞭解，因此在未來的研究中可依循本研究的成果，添加其他廢棄物進行調質，增加資源化之價值。而本計畫在學術研究之貢獻

- (1) 灰渣燒製輕質骨材的可行性之掌握。
- (2) 了解灰渣原料中的產氣因素。
- (3) 建立廢棄材料燒成多孔發泡材的操作參數。
- (4) 研究在不同燒成溫度操作條件下，試體受熱膨脹現象之影響。

在國家發展及其他應用方面預期之貢獻

- (1) 提升灰渣再利用之價值。
- (2) 協助解決國內大量底渣之處置問題，降低環境風險。
- (3) 協助國內實場發展輕質骨材最佳操作條件與生產操作技術。
- (4) 發展自有環保技術，強化資源再生途徑。
- (5) 建立發泡劑添加之成因與掌握產品品質。

出席國際學術會議心得報告

計畫編號	NSC 96-2221-E-032 -008 -
計畫名稱	都市垃圾焚化底渣燒製多孔性輕質材料受熱膨脹機制之研究
出國人員姓名	陳政綱
服務機關及職稱	淡江大學水資源及環境工程博士生兼任助理
會議時間地點	清華大學，北京
會議名稱	The 3 rd International Conference on Waste Management and Technology
發表論文題目	Considering the Sludge Stabilization during the Wastewater Treatment Process Selection from Water Extraction of MSWI Fly Ash

一、參加會議經過

- 11/05：大會開幕與致詞、各項議題報告、晚宴
- 11/06：中國環境科學學會固體廢棄物分會學術會議、中國固體廢棄物處理利用產業發展論壇、中荷固體廢棄物環境管理高級別論壇
- 11/07：各項專題報告
- 11/08：工程參觀：北京金隅紅樹林環保技術有限責任公司

二、與會心得

此次由設立於北京清華大學環境科學與工程學系之「聯合國環境規劃署巴塞爾公約亞太地區協調中心」所舉辦的「第三屆固體廢棄物管理與技術國際會議」，舉辦的規模較往年要盛大，約有來自 22 個國家的專家出席，5 日開幕式後緊接著是各項專業報告，緊接 6 日的議程是以有害廢棄物及電子廢棄物的處置與管理為主，7 日則是各項論文報告，8 日早晨參訪北京最大的有害廢棄物處理廠—北京金隅紅樹林環保技術有限公司。

固體廢棄物越境處理問題在中國越來越被重視，而這是由於近年來有許多非法越境運送的問題不斷在中國發生，特別是有害物質及電子廢棄物的運送量為大宗，有鑑於此，中國地區近年來也加快對電子廢棄物及有害廢棄物的立法及相關組織的設立，而設立於清華大學環境科學與工程學系內的巴塞爾公約亞太地區協調中心則亞太地區廢棄物越境問題之管理與研究，而且是亞太地區唯一的協調中心。

在 5 日及 6 日的專業報告中可發現，中國近年也開始注重中國境內電子廢棄物及有害廢棄物之管理，於本次大會中也始見「四機一腦」的回收口號，及「生產者付費」、「從搖籃到搖籃」的管理原則之倡導，足見中國已經開始引進歐美國家的廢棄物管理概念，此外，與會專家也發表許多回收再利用的生產技術，目的是希望建立一套適合中國自己的再利用流程。

對於電子廢棄物的管理，於大會中也有題出許多問題，例如外國進口家電的回收清運量要如何管理，要如何向廠商收取回收金，及回收金要如何運作等問題，都引起在場學者相當大的關切。由於近年中國工商業發達，中國的消費型態已經改變，且民眾對於電子商品的需求量大增，因此電子廢棄物的生產量早已超過官方估計的數目，此外，中方對於廢棄物的統計數據仍不甚正確，這可能是普查及管理制度的不完善亦或是地方行政效率不彰所造成，因此也有中國當地的專家題出相關配套方式的建議。

有害廢棄物管理方面，除了工業所生產之廢棄物外，中國這幾年都市垃圾焚化灰渣的處理壓力也日趨緊迫，中國目前已經興建了六十幾座的都市垃圾焚化廠，未來仍會持續興建，因此有許多論文發表之題目是和都市垃圾焚化灰渣有關，會後有部份學者及中國當地的研究人員前來討論及交換意見。

由此次會議可知，中國當地急需可以有效管理有害廢棄物及電子廢棄物的方法和相關法令，其中如廢棄物的運送及運送機具的管理、廢棄物的申報及流向管控、廢棄物的貯存和清除許可的管理、廢棄物的再利用等各項問題，而台灣方面的經驗正好可以提供借鏡，如能摒除雙方政治的歧見，兩岸的交流從環保開始，不敏感又很務實，相信兩岸都可得到寶貴之經驗。

Considering the Sludge Stabilization during the Wastewater Treatment Process Selection from Water Extraction of MSWI Fly Ash

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ABSTRACT: Washing is a general pre-treatment process for municipal solid waste incinerator (MSWI) fly ash, which is aimed to remove the harmful soluble salts, to improve the regeneration process. The wastewater separated contains heavy metals and high concentration of chloride, is needed to be handled properly. In this article, Electro-aggregation method was applied to compare with traditional coagulation, and assessed the stabilization ability of heavy metals in the sludge. The parameters of the operation include the voltage, hydraulic retention time, pH, liquid-to-solid ratio of extraction, different electrode connection mode, and the stabilization reagent dosage. The removal efficiency of SS, Cu, Pb, and Zn during the wastewater treatment was around 85%, 97%, 97%, 98-100%, and 100% respectively.

In order to make the electro-aggregation sludge will be safer and lower leachability, the addition of phosphate before entering the electrical reaction unit. When added 200 mg/L of NaH_2PO_4 , 100% Pb could be removed under the conditions of 100 V, 40 sec HRT, and heavy metals in the sludge could be stabilized to satisfy the TCLP limitations.

KEYWORDS: MSWI fly-ash, water extraction, electro-aggregation, heavy metals stabilization, fly-ash regeneration.

Introduction

In the past several decades, high growth economy and dense population in Taiwan have resulted in a great quantity of municipal solid waste (MSW) production. Sanitary landfill is the simplest way of waste treatment, but in Taiwan, it is almost impossible to find suitable place to setup the landfill site since 1990.

Incineration of MSW offers a reduction in the volume and weight effectively, the residues discharged from the end of the furnace are defined as bottom ash, and residues removed from the air pollution control (APC) system are mainly fine particle, are named as fly ash. Base on chemical and physical characteristics, municipal solid waste incinerator (MSWI) fly ash can be categorized as boiler ash and reaction product (reaction ash). Ash removed from the particles adhered on heat exchanger are generally called boiler ash; reaction product is produced from the dry scrubber or semi-dry scrubber of acid gas neutralization, and separated by bag house.

MSWI fly ash contains not only large amounts of chlorides, but also a lot of oxides, such as silicates, sulfates, carbonate etc. Heavy metals existed as chloride compound has the characteristic of high mobility, which induced higher evaporation rate in the thermal treatment[1], and soluble in the washing operation. Three types of acids (acetic acid, sulfuric acid, hydrochloric acid) were tested for the leaching experiments and hydrochloric acid was found to be the most effective for lead leaching. Observably, concentration of lead leaching resulted easily from the form of chloride compound [2]. Base on the chloride content has a

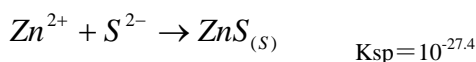
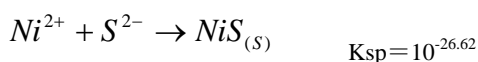
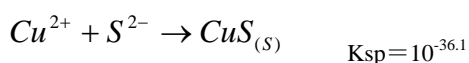
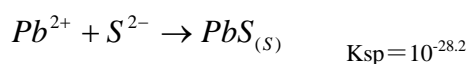
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great affect on metals leaching, water extraction process should be applied, in order to stabilize the fly ash and reduced the risks of heavy metals leaching during fly ash recycling.

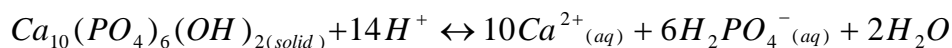
Removal of the soluble salts by extraction with water is quite simple, which has been proven to be feasible by many studies [3-6]. In order to evaluate the extracted quantity of soluble material, the washing conditions were adopted as liquid to solid ratio (L/S) of 25 with 15 min mixing, water extraction process could decrease the amount of chlorides and sulfates effectively [7]. Abbas et al. investigated the performance of water extraction conditions in liquid to solid ratio, extraction time, and multiple extraction steps, they found the triple extraction steps at L/S of 2, and extraction for 5 min had the best result for extracting of MSWI fly ash[8].

However, water extraction process had some problems in the extracted waste water treatments. Because of high concentration of heavy metals in the wastewater, the sludge produced from coagulation and flocculation process could not pass the limitation of Toxic Characteristic Leaching Procedure (TCLP) usually, which will be classified as hazardous waste. The principal mechanisms of heavy metals stabilization treatment is to form less soluble mineral phase and more stable in natural environment for long period [9, 10]. Research has shown that stabilization of lead and cadmium in fly ash by using 5% and 1.8% of sodium sulfide by weight could be satisfied the TCLP standard [9]. The stabilization chemical formulas of heavy metals are shown as follows:

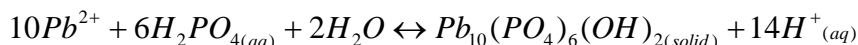


The precipitation rate of the solid particles reacted by phosphate with fly ash was very fast. After phosphoric acid was added in 10 min, 50% of phosphoric acid was consumed for forming metal phosphate [11]. Lead ion react with phosphate will produce the most stable precipitate in the environment within short reaction time. Laperche et al. addressed the chemical equation formulas of metal phosphate formation as follows[12]:

Dissolution process



Precipitation process



Considering about the stability of wastewater treatment sludge, this study investigated the reaction production resulting from chemical stabilization of sludge during the wastewater treatment (chemical coagulation) by water extraction of MSWI fly ash with Na₂S and NaH₂PO₄. Furthermore, this study was used electro-aggregation for treating waste extractive water and added Na₂S and NaH₂PO₄ in the feedstock of electro-aggregation reactor in order to stable electro-aggregation sludge at the same time. Two types of sludge were analyzed heavy metals leaching after TCLP test, and the amount of heavy metals removal from waste extractive water.

Material and Method

In this study, MSWI fly ash was gathered from a large scale of MSW incinerator plant in northern Taiwan. Reaction ash and boiler ash were mixed and the mixing proportion was according to the actual production quantity of the incinerator (reaction ash : boiler ash = 2.4 : 1.4 by weight). The extracting conditions were liquid to solid ratio of 5 (L/S = 5) with twice. After water extraction process, the aqueous suspension was filtered and the filtrate (waste extractive water) was collected. After extraction the waste extractive water was treated by chemical coagulation and electro-aggregation.

Electro-aggregation system was depicted in Fig. 1. An peristaltic pump was used to feed the wastewater into the reactor. The wastewater entered the reactor from the bottom of the reactor, flowed upwards in a zigzag channel formed of the space between the iron electrodes and baffle plates. There were 38 pieces of iron plates in the electro-aggregation reactor, spacing between the plates were 10 mm, and the electrodes were connected to the D.C. power. The total effective area of the reactor was 3,300 cm², and effective capacity was 1,000 mL. The mechanism of electro-aggregation was based on three phenomenon: (1) particles were attracted each others due to the polarization, which produced by the electric field; (2) the zigzag channel offers the opportunity for the polarized particles to collide each rapidly, to become much more larger particles; (3) sacrificial electrode was undergo oxidation reaction and released the iron ions as the coagulant, and generated fine gas bubbles to entrap the flocs to float after they leave the reactor. The electro-aggregation has the advantages of very high reaction rate, better pollutant removal than conventional coagulation, suitable for treating of wide range of various pollutant concentration, major compartments were easily maintaining and occupied only a small area.

In the first stage of the experiment, the chemical coagulation process was executed with the addition of 50 and 100 mg/L as S by Na₂S, 115 and 220 mg/L as P by NaH₂PO₄. FeCl₃ has been selected for the coagulant of 200, 300, and 400 mg/L. The experiment was proceed by jar test as follows: coagulated 3 min, flocculated 30 min, adjusted pH at 9, and settled in 30 min to collect supernatant and settling sludge. Stabilization sludge leaching properties were exam by TCLP. In the second stage of the experiment, the electro-aggregation treatment were executed under the operational conditions of 50, 76 100, 125 V, hydraulic retention time in 40 sec. Reactor outlet flow was adjusted pH at 9, settled in 30 min to separate sludge. An aim for stabilizing sludge, added varied dosage of 50 and 100 mg/L as S by Na₂S, 115 and 220 mg/L as P by NaH₂PO₄ in waste extractive water and formed sludge by electro-aggregation process and then evaluated by TCLP. The elements content in samples were determined using ICP-AES (Jobin Yvon Horiba, Ultima 2000). TCLP was according to the method NIEA R201.12C in Taiwan, and it regulated maximum allowable leaching concentration 5 mg/L of Pb, 1 mg/L of Cd, 15 mg/L of Cu, and 5 mg/L of Cr.

Results and Discussion

1. Chemical coagulation process

In the coagulation treatment experiment, the coagulant added in varied dosages of 200, 300, and 400 mg/L. After the coagulation reaction, pH were be adjusted to 9, settled by 30 min, and then collected the supernatant, measured the concentration of elements and suspended solid (SS). In the results shown in Table 1, the coagulation process could reduce a large amount of heavy metal elements, and the removal of lead was 99.9%, but SS were not good. The results of heavy metals of TCLP test for the sludge was high, especially for lead (152.1 mg/L), shown in Table 2. The heavy metals in the coagulation sludge would re-leach easily, and lead would be the dominative problem during the final disposal or recovering.

Addition of 100 mg/L Na₂S as S during the coagulation operation, the lead leaching of TCLP decreased to 22.36 mg/L, but it still exceeded the TCLP standard (5 mg/L for Pb). When dosing NaH₂PO₄ of 220 mg/L as P during the coagulation operation, concentration of lead leaching could be decreased below 1 mg/L significantly, the results showed that the NaH₂PO₄ is more efficient than Na₂S for the stabilization of coagulation sludge.

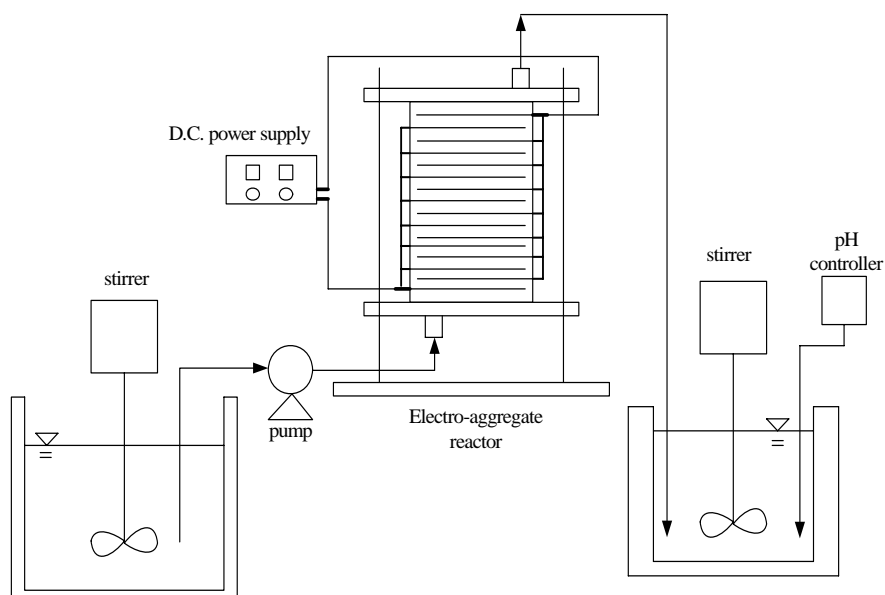


Fig 1. Schematic diagram of Electro-aggregation process.

Table 1. Concentration of the elements in the supernatant of coagulation treatment of extracted wastewater with different FeCl₃ dosage.

Elements	Extracted wastewater	FeCl ₃ 200 mg/L	FeCl ₃ 300 mg/L	FeCl ₃ 400 mg/L
Al	60	52	50	48
Ca	15,230	13,550	13,790	13,510
Fe	0.6	40	36	48
K	6,963	5,412	5,231	5,968
Mg	27	11	11	15
Na	6,675	5,692	5,541	5,478
Cd	0.63	N.D	N.D	N.D
Cr	1.58	0.02	0.02	0.02
Cu	5.32	0.04	1.06	0.14
Mn	0.12	N.D	N.D	N.D
Pb	206	1.62	0.30	1.50
Zn	42.9	0.17	0.06	1.00
Cl	51,240	31,560	32,510	32,140
SS.	580	202	202	224

Unit: mg/L; N.D.: Cd<0.2 ppb, Mn<0.1 ppb.

Table 2. The TC LP results of heavy metals concentration

for coagulation sludge stabilization by Na₂S and NaH₂PO₄ dosage.

Cons.	Coagulation only	Na ₂ S 50 mg/L as S	Na ₂ S 100 mg/L as S	NaH ₂ PO ₄ 115 mg/L as P	NaH ₂ PO ₄ 220 mg/L as P
Pb	152.1	36.45	22.36	1.23	0.95
Cd	0.54	0.42	0.21	0.08	0.05
Cr	1.87	1.02	0.54	0.32	0.12
Cu	N.D.	N.D.	N.D.	N.D.	N.D.

Unit: mg/L; N.D.: Cu<0.3 ppb.

2. Electro-aggregation process

In the electro-aggregation experiment, the reactor effluent was adjusted the pH to 9, and settled 30 min. The heavy metals remained in the supernatant at 100 V were almost non-detectable (Table 3), and the higher voltage the quality will more impression. But when it exceeds 100 V, the removal efficiency became gently and a large amount of sludge produced. Base on this phenomenon, this study would control the voltages under 100 V.

The major component of the sludge was comes from the iron floc of the sacrificial electrode. Heavy metals in the sludge leached easily in acid and alkali conditions, and caused second pollution. As shown in Table 4, after adding Na₂S in the influent of the electrical reactor, the TCLP test of lead in the sludge was still as high as 14.83 mg/L, which doesn't meet the limitation of the regulation, but Cd, Cr and Cu leaching were quit low.

Table 3. Concentration of the elements in the effluent of electro-aggregation treatment with different voltage.

Elements	Extracted wastewater	50 V	75 V	100 V	125 V
Al	60	38	42	48	60
Ca	15,230	12,420	13,010	11,140	13,010
Fe	0.6	8	6	7	15
K	6,963	5,324	5,356	4,923	5,213
Mg	27	5	6	4	6
Na	6,675	5,012	5,114	5,009	5,123
Cd	0.63	N.D	N.D	N.D	N.D
Cr	1.58	N.D	N.D	N.D	N.D
Cu	5.32	N.D	N.D	N.D	N.D
Mn	0.12	N.D	N.D	N.D	N.D
Pb	206	0.15	0.08	N.D	0.04
Zn	42.9	N.D	N.D	N.D	N.D
Cl	51,240	35,120	31,560	32,510	32,140
S.S.	580	125	102	95	70

Unit: mg/L; N.D.: Cd<0.2 ppb, Cr<0.3 ppb, Cu<0.3 ppb, Mn<0.1 ppb, Pb<3 ppb, Zn<0.3 ppb.

Table 4. The TCLP results of heavy metals concentration for electro-aggregation sludge stabilization by Na₂S dosage.

Cons.	Electro-aggregation only	Na ₂ S as S 50 mg/L, 75 V	Na ₂ S as S 100 mg/L, 50 V
Pb	135.3	18.32	14.83
Cd	0.66	0.52	0.11
Cr	1.32	0.93	0.43
Cu	N.D.	N.D.	N.D.

Unit: mg/L

The influences of sludge stabilization on adding of NaH₂PO₄ in the influent of the reactor were in relation to parameters of voltage in electro-aggregation treatment. Lead leaching decreasing with the operational voltage increasing for the TCLP test of the sludge. In the stabilization treatment of sludge, dosing of NaH₂PO₄ could decrease the lead leaching below 1/10 of TCLP standard, as shown in Table 5, it was much

better than the effect of dosing Na_2S .

Table 5. The TCLP results of heavy metals concentration for electro-aggregation sludge stabilization by NaH_2PO_4 dosage.

Cons.	Electro-aggregate only	NaH_2PO_4 as P 115 mg/L		NaH_2PO_4 as P 220 mg/L	
		75 V	100 V	75 V	100 V
Pb	135.3	0.55	0.65	0.72	0.45
Cd	0.66	N.D.	N.D.	0.01	N.D.
Cr	0.14	N.D.	N.D.	N.D.	N.D.
Cu	0.94	N.D.	0.04	0.02	0.01

Unit: mg/L; N.D.: Cd<0.2 ppb, Cr<0.3 ppb, Cu<0.3 ppb.

3. Compare the two types of treatment processes

High concentration of heavy metals, especially for lead, dissolved into the wastewater during the water extraction operation of MSWI fly ash in this study. By using of the electro-aggregation process, the pollutant removal efficiency was better than chemical coagulation process, as shown in Table 6. The electro-aggregation process could remove from waste extractive water about 96% of heavy metal elements, and had well effect on SS removal.

As shown in Table 7, electro-aggregation treatment accompanied with the addition of Na_2S or NaH_2PO_4 had good efficiency for the heavy metals stabilization of the sludge. The stabilization effect was better than the effect of coagulation, it is due to the induce of electrical energy to enhance the stabilization reaction, let the heavy metals reacted with the stabilization reagents faster and formed much more stable mineral compounds. It is helpful to prevent the secondary pollution during the wastewater treatment, and to reduce the environmental hazard risk in the disposal or recovery of the waste sludge,

Table 6. Comparison of heavy metals removal between electro- aggregation and chemical coagulation for the wastewater.

Elements	Electro-aggregation HRT 40 sec. 50~125 V, pH 9	Chemical coagulation FeCl_3 200~400 mg/L, pH 9
SS (%)	75~85	55~65
Cu (%)	96~97	70~80
Pb (%)	100	95~100
Zn (%)	100	95~100
Cd (%)	100	100

Table 7. Comparison the effect of S/P stabilization between electro-aggregation and chemical coagulation sludge by TCLP.

Elements	Coagulation with FeCl_3 300 mg/L			Electro-aggregation		
		Na_2S as S 50 mg/L	NaH_2PO_4 as P 220 mg/L	(100 V)	Na_2S as S 100 mg/L (50 V)	NaH_2PO_4 as P 220 mg/L (100 V)
Pb	152.1	22.36	0.95	135.3	14.83	0.45
Cd	0.54	0.21	0.05	0.66	0.11	N.D.
Cr	1.87	0.54	0.12	0.14	0.43	N.D.
Cu	N.D.	N.D.	N.D.	0.94	N.D.	0.01

Unit: mg/L; N.D.: Cd<0.2 ppb, Cr<0.3 ppb, Cu<0.3 ppb.

Conclusions

In the recovering of MSWI fly ash, it is important to be pre-treated by water extraction, and then waste extracted water should be treated properly. The chemical coagulation and flocculation process could accomplish over 97 % removal efficacy of heavy metals (Pb, Zn, Cu, Cr, Cd) under the condition of 300 mg FeCl₃/L dosage at pH 9. While in the electro-aggregation system, under the operational voltage of 100 V and 40 sec of hydraulic retention time at pH 9, almost 100 % removal efficiency of heavy metals could be achieved.

In this study, both types of sludge presented a large quantity of heavy metals leaching by TCLP test. However, the leachate concentration of heavy metals decreased after adding Na₂S or NaH₂PO₄. Comparing electro-aggregation process with chemical coagulation process, the electro-aggregation had better effect upon stabilizing of heavy metals. The effect of the stabilization ability of heavy metals in the waste sludge by addition of NaH₂PO₄ was proved better than Na₂S. There was significant efficiency of removing heavy metals in waste water and stabilizing sludge by means of the electro-aggregation process accompany with addition of NaH₂PO₄. This process could not only prevent the water pollution but also decrease the environmental risk of sludge during the operation of disposal or recovering.

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