Journal of Hazardous Materials 166 (2009) 836–841

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Research article

Development and application of an auto-normalization program for optimal treatment of numerous metal-containing hazardous sludges

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ARTICLE INFO

Article history:
Received 10 December 2007
Received in revised form 6 October 2008
Accepted 26 November 2008
Available online 7 December 2008

Keywords:
Normalization
Program
Optimal
Metal
Hazardous sludge

ABSTRACT

Precise control of all pollutant concentrations during co-treating numerous hazardous materials is difficult because of the variety of pollutants and concentrations. An Auto-Nor program for normalizing various concentrations of numerous hazardous materials had been developed, and the normalization theory, processes, and a practical case of 20 metal-containing sludges was presented. Metal concentrations (C_{ij}) of each sludge, including Cd, Cr, Cu, Pb and Zn, were divided by toxicity characteristic leaching procedure (TCLP) regulatory limit to be relative concentrations (R_{ij}). Computations at a systematic threshold of normalization (U) by the Auto-Nor program would automatically sort out optimal co-treatment groups. In the 20-sludge case, three optimal co-treatment groups, comprising 14, 3 and 3 sludges, were sorted out and the application results show that all random samples passed the TCLP test. The Auto-Nor program can be further applied to the treatment or remediation of hazardous fly ashes and contaminated soils, improving and benefiting their concentration control, dosage management and cost reduction.

1. Introduction

Hazardous wastes, being harmful to human health and the environment, must be carefully handled during storage, transportation and treatment processes. Although different countries have different definitions of hazardous waste, most governments classify hazardous wastes into two categories: listed hazardous wastes and characteristic wastes. For instance, the United States defines a RCRA hazardous waste as one that appears on one of four hazardous wastes lists (F-list, K-list, P-list, or U-list), or exhibits at least one of four characteristics: ignitability, corrosivity, reactivity or toxicity [1]. In Taiwan, hazardous wastes are identified widely as follows—(1) listed wastes: industrial wastes generated from listed manufacturing processes, mixed metal scraps and biological/medical wastes; (2) characteristic wastes: waste revealing or containing one of the following characteristic or component: toxicity, leaching toxicity, dioxins, PCBs, corrosivity, ignitability, reactivity or asbestos [2].

According to the statistics of industrial waste in Taiwan from 2001 to 2006, hazardous waste was about 6–11% of total industrial wastes. Metal containing ash and sludge generated from air pollution control devices (APCDs), manufacturing processes, and wastewater treatment facilities are the majority of hazardous wastes, indicating that proper treatment of metal containing waste is necessary [3].

To reutilize or treat sewage sludge or industrial sludge, the basic requirements are pre-checking sludge constituents and making hazard assessment [4–7]. Some methods such as chemical extraction, electrokinetics and electrodialysis were applied to metal-containing sludges for metal removal [6,8,9]. Furthermore, hydrometallurgical or pyrometallurgical method involving a series of extraction or a combination of thermal treatment is another method for treating metal-containing sludges [10,11]. Although those methods are available, complete removing metals from sludges is not easily obtained. Thus, solidification/stabilization, a simple and practicable method using the addition of stabilization reagents (e.g., Portland cement, gypsum, polyethylene) to immobilize hazardous constituents, is the most convenient treatment for metal-containing sludges [12–15].

Hazardous sludges generated from different factories are usually transported to a co-treatment plant for treatment expediency and cost-effectiveness [16,17]. In the co-treatment plant, mixing process is indispensable and it plays an important role in regulating numerous co-treated sludges for chemical dosage in the subsequent main treatment such as solidification, stabilization and chemical extraction. However, numerous sludges containing different hazardous components and concentrations will create uncertainty in mixture concentrations. Regardless of hazardous components and concentrations, the co-treatment products often result in negative consequences; for instance, failing the toxicity characteristic

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0304-3894/$ – see front matter
doi:10.1016/j.jhazmat.2008.11.122

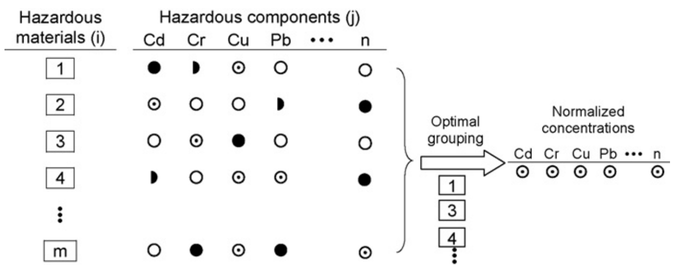

Fig. 1. Conceptual scheme of normalization. Different concentrations of hazardous components are complemented in optimal grouping materials, and lower levels of normalized mixture's concentrations are obtained (symbols of ●, ◗, ⊙ and ○ indicate high, medium, low and not detected level of concentrations, respectively).

leaching procedure (TCLP) test and leaching hazardous constituents at the disposal site. Therefore, controlling the concentrations of all hazardous sludges by pre-selecting optimal grouping sludges for normalizing their various components and concentrations is an imperative pre-work in the co-treatment plant.

An auto-normalization (abbreviated as "Auto-Nor") program had been developed, and its theoretical concept, program procedure, and an application in the treatment of 20 hazardous sludges is presented in this paper. Since TCLP is a regulatory test in most countries for identifying the leaching toxicity of a waste, all concentrations demonstrated in the application case were expressed as TCLP concentration (mg/L).

2. Materials and methods

2.1. Hazardous sludges sampling and TCLP tests

A co-treatment plant located in Miao-Li, Taiwan, was built in 2006 for treating inorganic hazardous sludges generated from wastewater treatment processes of several industries, particularly electroplating and metal surface treatment industries. In this co-treatment plant, numerous inorganic hazardous sludges containing different metals and concentrations were mixed without any pre-sorting and then blended with clay. The subsequent procedure of producing sludge-clay bricks is similar to that of making normal bricks, which involves molding, setting and drying, firing in a tunnel kiln, and cooling processes. According to governmental regulations, this sludge–clay brick treatment is classified as an intermediate treatment–stabilization/solidification. Therefore, random sampling of sludge-clay bricks and passing the TCLP test are the basic requirements for obtaining governmental permission of final disposal. Unfortunately, some of the sampled sludge-clay bricks failed the TCLP test at the first trial operation because of free selecting sludges for mixing before the major process of producing sludge-clay bricks. To improve the mixing process and control mixture concentrations within an expected range, 20 sludges were randomly selected and triple samples for each sludge were taken. Note that all sludges transported to this co-treatment plant were dewatered for saving treatment fee. TCLP tests in accordance with Method 1311, SW-846 methods of US EPA [18] were conducted for all samples. Heavy metal concentrations of Cd, Cr, Cu, Pb and Zn in TCLP leachates were detected by a flame atomic absorption spectrophotometer (Hitachi model Z-5000). The pH values of all samples were determined using a glass electrode at a sludge:water ratio of 1:2.

2.2. Concept and application of Auto-Nor program

The concept of normalization was derived from selecting optimal grouping materials to complement both of their hazardous components and concentrations. The conceptual scheme of normalization is shown in Fig. 1. Moreover, some constraints of Auto-Nor program in its application are described as follows.

1. *Applicable objects*. Objects to which the Auto-Nor program can be applied must be materials of easy mixing, such as wastewater, sludge, ash, soil, slurry and soil, so it is possible to complement different concentrations of pollutants via mixing. Furthermore, a combined mixing of different materials, such as sludge mixing with ash or soil mixing with slurry, is not allowed to avoid occurring complicated reactions in mixture.
2. *Influencing factors*. Some factors may influence the accuracy of detection results, depending on which detection method is used. For example, pH may moderately affect the detection result of metal concentration in TCLP method (Method 1311, US EPA), but pH does not significantly affect total metal content in the acid digestion method (Method 3050B, US EPA). Thus, materials with similar pH values are recommended for using the Auto-Nor program in the detection of TCLP method, ensuring the estimation accuracy of normalized concentrations.
3. *Chemical reactions*. Chemical reactions that may dramatically affect normalized concentrations in the Auto-Nor program are neglected for simplification. Therefore, similar characteristic materials and same form of pollutants species are strongly recommended in the application of Auto-Nor program for avoiding unnecessary reactions occurring in mixture.

2.3. Development and procedure of Auto-Nor program

The flowchart of auto-normalization procedure of Auto-Nor program is shown in Fig. 2. The development and procedure of Auto-Nor program are described in detail as follows.

1. Establish an $m \times n$ matrix of pollutant concentration (C_{ij}) in an Excel format for all hazardous materials, where C is the pollutant concentration (e.g., mg/L); i is the number of materials from 1 to m; and j is the pollutant such as Cd, Cr or Cu, numbered from 1 to n.
2. Set the regulatory limit of each pollutant (L_j) according to the regulatory criteria. For instance, Cd = 1.0 mg/L, Cr = 5.0 mg/L and Pb = 5.0 mg/L are the TCLP regulatory limits codified at 40 CFR §261.24 [1].
3. Divide each pollutant concentration (C_{ij}) by its regulatory limit (L_j) and transform into relative concentration (R_{ij}). That is to set all concentrations on the same basis of criteria for the convenience of comparing with regulatory limits and sorting out optimal groups

$$R_{ij} = \frac{C_{ij}}{L_j}, \tag{1}$$

where C_{ij} is the pollutant concentration and L_j is the regulatory limit of each pollutant.
4. Set threshold of normalization (U) starting from 1. U is an integer and set as the threshold, compared with the regulatory limit, for systematic comparison in the program.
5. After the setting of U, select the number of mixing materials (K) starting from m (i.e., m numbers of materials are first selected for mixing) and run program. The average of relative concentration of each pollutant ($\bar{R}_j$) and the maximum of $\bar{R}_j$ (i.e., $\bar{R}_{j,\max}$) are then determined

$$\bar{R}_j = \text{Avg.}\{R_j\}, j = 1, 2, \ldots, n;$$
$$\bar{R}_{j,\max} = \text{Max}\{\bar{R}_j\}, j = 1, 2, \ldots, n. \tag{2}$$

6. If $\bar{R}_{j,\max}$ is less than U, indicating that each $\bar{R}_j$ is less than its regulatory limit (L_j), the optimal group containing all the selected mixing materials is obtained. If $\bar{R}_{j,\max}$ is greater than or equal

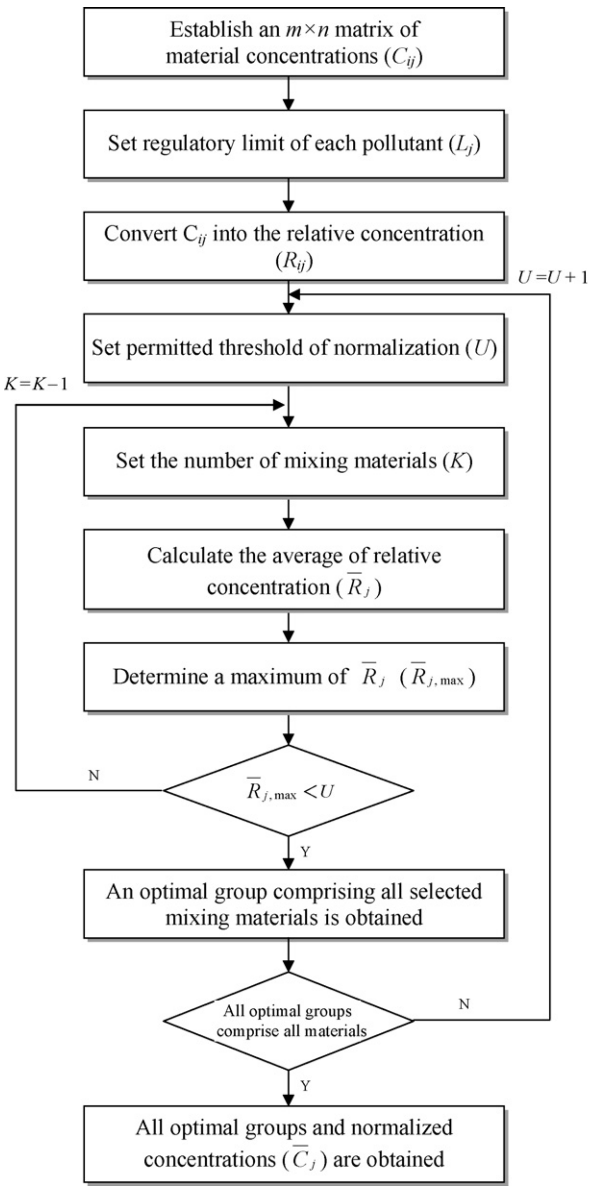

Fig. 2. Flowchart of auto-normalization procedure of Auto-Nor program.

to U, decrease the number of mixing materials by one (i.e., new $K = K - 1$) and calculate $\bar{R}_j$ again. Note that the smallest number of K is two (i.e., the smallest mixing group is made up of two materials). Do the following check and repeat this procedure until the first optimal group is obtained:

- If $\bar{R}_{j,\max} < U$, the optimal group has been found and it comprises all the selected mixing materials.
- If $\bar{R}_{j,\max} \geq U$, no optimal group is obtained. Reduce the number of mixing materials by one (i.e., new $K = K - 1$) and re-calculate $\bar{R}_j$.

7. If the first optimal group cannot be acquired at $U = 1$ or the fist optimal group does not comprise all selected materials, add one for the new U (i.e., new $U = U + 1$, indicating that the threshold is increased) and repeat steps 5 and 6 until all optimal groups are sorted out. Note that materials that had been selected in one optimal group should be deleted before the next optimal sorting.
8. In each optimal group, the normalized concentration of each pollutant ($\bar{C}_j$) is calculated by a re-conversion of Eq. (3) and all pollutant concentrations in all optimal groups are pre-estimated

$$\bar{C}_j = \bar{R}_j \times L_j, \quad j = 1, 2, \ldots, n. \tag{3}$$

where $\bar{R}_j$ is the average of relative concentration and L_j is the regulatory limit.

3. Results and discussion

3.1. Sample detections and data file creation

The pH values of 20 sludges ranging from 7.2 to 10.5 were measured, indicating that most of the metal species in these sludges were in the form of metal hydroxide precipitates. Thus, pH will not significantly affect the quality of mixtures and the accuracy of normalized concentrations can be assured. Detection results of metal concentrations of 20 metal-containing sludges and their relative concentrations are listed in Table 1. According to the TCLP criteria, all of 20 sludges were identified as hazardous wastes since each sludge exceeded at least one of TCLP regulatory limits. All metal concentration data were arranged as an $m \times n$ matrix in Excel format (C_{ij}), where m is the number of sludges and n is the number of hazardous components (i.e., metals). In the Auto-Nor program, C_{ij} was further converted into relative concentration (R_{ij}) via dividing each C_{ij} by its TCLP regulatory limit (L_j).

3.2. Program modification

The auto-normalization was executed by a series of random selecting mixing hazardous materials, consequential computing numerous concentrations of selected materials, and comparing maximum of average concentrations with thresholds. Thus, large computer memory space and time-consuming computations during auto-normalization process are necessary. In the case of 20 hazardous sludges, five metals (Cd, Cr, Cu, Pb and Zn) included in each sludge, the total number of computations during auto-normalization is 5,242,775 if optimal group is not found. During such auto-normalization process, for instance, the numbers of computation are estimated to be 77,520 and 923,780 (i.e., $C_{15}^{20} \times 5$ and $C_{10}^{20} \times 5$) at $K = 15$ and 10, respectively. Nevertheless, the Auto-Nor program was modified several times for boosting computation capability and shortening program-running time until an ideal performance was achieved.

3.3. Processes of pre-sorting optimal groups

In the pre-sorting process of 20 hazardous sludges, U was first set as 1 and the number of mixing materials (K) was initially set at 20 (i.e., the total number of sludges). The Auto-Nor program calculated the averages of relative concentration of each metal ($\bar{R}_j$), picked out $\bar{R}_{j,\max}$ from $\bar{R}_j$, and automatically compared $\bar{R}_{j,\max}$ with U; unfortunately, $\bar{R}_{j,\max}$ was greater than U, indicating that all 20 hazardous sludges were not suitable to be an optimal group for co-treatment. Therefore, K was decreased by one (i.e., new $K = K - 1$) and iterative computations and comparisons were carried out until an optimal group was obtained at $K = 14$. That means that 14 sludges, including sludge 1, 2, 3, 6, 7, 8, 9, 11, 12, 13, 14, 18, 19, and 20 as shown in Fig. 3, were the first optimal group suitable for mixing.

According to the results of the first optimal group, the averages of the relative concentration ($\bar{R}_j$) of Cd, Cr, Cu, Pb and Zn were 0.97, 0.96, 0.71, 0.96 and 0.65, respectively (Table 2). Multiplying $\bar{R}_j$ by its regulatory limit (L_j) yielded normalized concentrations ($\bar{C}_j$) of 0.97, 4.79, 10.65, 4.81 and 16.14 mg/L for Cd, Cr, Cu, Pb and Zn, respectively (Table 2). All of $\bar{R}_j$ were less than 1, indicating that all normalized concentrations of metals in the sludge mixture were below TCLP regulatory limits. Therefore, 14 sludges, 70% of total sludges, can

Table 1
Metal concentrations and relative concentrations of 20 metal-containing hazardous sludges.

No. of sludges (i)	Metal concentration (C_{ij}, mg/L) and relative concentration (R_{ij})									
	Cd		Cr		Cu		Pb		Zn	
	C_{i1}	R_{i1}	C_{i2}	R_{i2}	C_{i3}	R_{i3}	C_{i4}	R_{i4}	C_{i5}	R_{i5}
1	0.8	0.80	7.6	1.52	7.8	0.52	12.2	2.44	45.3	1.81
2	0.0	0.00	13.0	2.60	4.5	0.30	1.1	0.22	40.0	1.60
3	0.7	0.70	3.2	0.64	31.3	2.09	0.9	0.18	5.9	0.24
4	0.0	0.00	17.0	3.40	6.3	0.42	5.6	1.12	28.6	1.14
5	3.2	3.20	4.6	0.92	7.9	0.53	3.4	0.68	7.9	0.32
6	0.3	0.30	6.8	1.36	29.0	1.93	2.8	0.56	4.7	0.19
7	0.1	0.10	9.0	1.80	0.6	0.04	7.9	1.58	11.2	0.45
8	1.9	1.90	1.7	0.34	16.8	1.12	3.1	0.62	11.0	0.44
9	1.8	1.80	1.5	0.30	2.5	0.17	6.9	1.38	8.0	0.32
10	1.9	1.90	144.8	28.96	6.3	0.42	0.2	0.04	2.7	0.11
11	2.2	2.20	4.0	0.80	22.3	1.49	0.1	0.02	4.2	0.17
12	0.0	0.00	5.1	1.02	3.2	0.21	1.8	0.36	16.9	0.68
13	0.4	0.40	1.3	0.26	2.2	0.15	5.9	1.18	10.0	0.40
14	1.6	1.60	9.0	1.80	12.0	0.80	2.2	0.44	27.4	1.10
15	11.0	11.00	2.2	0.44	6.2	0.41	3.2	0.64	12.0	0.48
16	1.8	1.80	13.3	2.66	0.9	0.06	3.7	0.74	14.7	0.59
17	13.8	13.80	5.6	1.12	36.2	2.41	8.9	1.78	1.8	0.07
18	0.3	0.30	0.1	0.02	1.8	0.12	5.8	1.16	3.8	0.15
19	2.4	2.40	3.7	0.74	5.9	0.39	1.7	0.34	22.0	0.88
20	1.1	1.10	1.1	0.22	9.2	0.61	15.0	3.00	15.5	0.62
Regulatory limit (mg/L)	1.0[a]		5.0[a]		15.0[b]		5.0[a]		25.0[c]	

Metal concentrations are expressed as mean value of triplicates.
[a] TCLP criteria of US and ROC.
[b] TCLP criteria of ROC.
[c] Past TCLP criteria of ROC.

be mixed directly without any chemical dosage because metal concentrations will complement each other. From the treatment point of view, mixing optimal grouping sludges without any chemical dosage not only presents a scientifically supported method but also provides a cost-effective means of treatment for treating numerous hazardous sludges.

Before the next pre-sorting process, the concentration data of 14 sludges of the first optimal group must be deleted from data bank (C_{ij}) since they had already been sorted out. In the next pre-sorting procedure, U was increased to 2 and the number of mixing materials (K) was initially set at 6 (i.e., the total number of ungrouped sludges). Since no optimal group was obtained from $K = 6$ to 2, U was then increased to 3 and K was still started at 6 for repeating pre-sorting process. Fortunately, the second optimal group was obtained at $K = 3$, consisting of sludge 4, 5 and 16. The averages of the relative concentration ($\bar{R}_j$) of Cd, Cr, Cu, Pb and Zn in the sec-

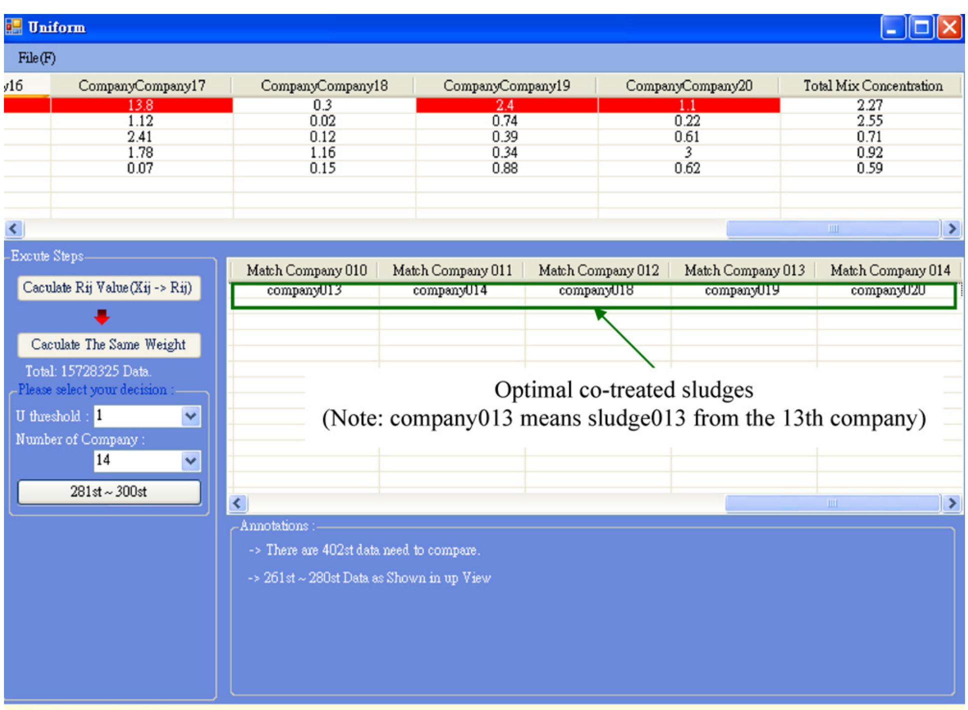

Fig. 3. Under permitted threshold $U = 1$, the first optimal group was obtained at $K = 14$, which suggested that 14 sludges (No. 1, 2, 3, 6, 7, 8, 9, 11, 12, 13, 14, 18, 19 and 20) were optimally co-treated. This figure taken from the first normalization results shows some of optimal co-treated sludges.

Table 2
Normalized concentrations in the first optimal group ($U = 1$) determined by the Auto-Nor program.

First optimal group	Sludges 1, 2, 3, 6, 7, 8, 9, 11, 12, 13, 14, 18, 19 and 20	
Metal	Average of relative concentration ($\bar{R}_j$)	Normalized concentration ($\bar{C}_j$, mg/L)
Cd	0.97	0.97
Cr	0.96	4.79
Cu	0.71	10.65
Pb	0.96	4.81
Zn	0.65	16.14

Table 3
Normalized concentrations in the second optimal group ($U = 3$) determined by the Auto-Nor program.

Second optimal group	Sludges 4, 5 and 16	
Metal	Average of relative concentration ($\bar{R}_j$)	Normalized concentration ($\bar{C}_j$, mg/L)
Cd	1.67	1.67
Cr	2.33	11.63
Cu	0.34	5.03
Pb	0.85	4.23
Zn	0.68	17.07

Table 4
Normalized concentrations in the third optimal group ($U = 11$) determined by the Auto-Nor program.

Third optimal group	Sludges 10, 15 and 17	
Metal	Average of relative concentration ($\bar{R}_j$)	Normalized concentration ($\bar{C}_j$, mg/L)
Cd	8.90	8.90
Cr	10.17	50.87
Cu	1.08	16.23
Pb	0.82	4.10
Zn	0.22	5.50

ond optimal group were 1.67, 2.33, 0.34, 0.85 and 0.68, respectively (Table 3). Among these data, the maximum of $\bar{R}_j$ was 2.33 of Cr, indicating that the highest normalized concentration was 2.33 times the regulatory Cr limit and careful control during mixing process was necessary. Normalized concentrations ($\bar{C}_j$) of Cd, Cr, Cu, Pb and Zn, obtained by the converting of $\bar{R}_j$, were 1.67, 11.63, 5.03, 4.23 and 17.07 mg/L, respectively (Table 3).

Only three sludges were left after sorting out two optimal groups. In the subsequent optimal grouping process, U was increased to 4 and K was started at 3 (i.e., the total number of ungrouped sludges). Unfortunately, no optimal group was obtained from $U = 4$–10. Finally, when U was increased to 11 and K was 3, the third optimal group (i.e., the last optimal group) comprising sludges 10, 15 and 17 was obtained. The averages of the relative concentration ($\bar{R}_j$) of Cd, Cr, Cu, Pb and Zn in the third optimal group were 8.90, 10.17, 1.08, 0.82 and 0.22, respectively (Table 4). Moreover, the normalized concentrations ($\bar{C}_j$) of Cd, Cr, Cu, Pb and Zn were converted to be 8.90, 50.87, 16.23, 4.10 and 5.50 mg/L, respectively (Table 4). Note that the highest normalized concentration of element was Cr and 10.17 times the regulatory limit implied that the third optimal grouping sludges should be cautiously controlled and treated. Several alternatives of treating these three sludges were seriously proposed as follows: (1) enhanced treatment by an addition of stabilization agent for sludge mixture; (2) temporary storage of these three sludges and future regrouping with other new coming sludges; (3) finding out the highest Cr source of sludge and reducing its amount in mixing process.

The averages of the relative concentration of each metal in the three optimal groups are shown in Fig. 4. In the first optimal group, it is very clear that chemical dosage is not required during mixing process since all $\bar{R}_j$ values are less than the TCLP regulatory limits as seen in Fig. 4. In the second and third optimal groups, $\bar{R}_j$ of Cd and Cr are higher than those of other metals, implying that these two sludge mixtures need to be properly treated; moreover, checking the concentrations of Cd and Cr after co-treatment is necessary.

3.4. Practical verification

After the development of Auto-Nor program and simulation of 20 hazardous sludges, the results were applied for practical verification at the sludge co-treatment plant, Miao-Li, Taiwan. To test the auto-normalization performance and treat all of 20 hazardous sludges soon, the general manger of co-treatment plant decided to select the first alternative, i.e., enhanced treatment by an addition of stabilization agent for optimal groups 2 and 3. Thus, 20 metal-containing sludges were classified into three optimal groups according to the simulation results, and sludges in each optimal group were completely mixed to ensure attaining the normalized concentrations. In the first optimal group, the well-mixed sludge mixture was directly blended with clay without any adjustment of chemical agent. Sludge-clay bricks were further produced as the same procedure of normal bricks. As expected, all randomly sampled sludge-clay bricks met the TCLP criteria. In the second optimal group, a proper amount of stabilization agent was added to the sludge mixture, and all sampled sludge-clay bricks passed the TCLP test. In the third sludge mixture, a fivefold amount of stabilization agent than that used in the second optimal group was added for reducing leaching risk. Subsequently, as expected, all randomly sampled sludge-clay bricks also passed the TCLP test.

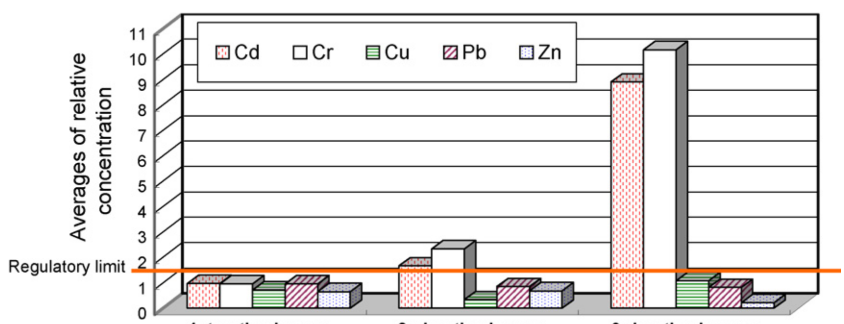

Fig. 4. Twenty metal-containing hazardous sludges were categorized into three optimal groups by the Auto-Nor program, and the averages of relative concentration ($\bar{R}_j$) of Cd, Cr, Cu, Pb and Zn in each group were pre-estimated.

4. Conclusions

To ensure treatment quality, effective controlling of the concentrations of mixture components is very important in the co-treatment of hazardous materials. In this paper, the Auto-Nor program pre-sorting optimal groups from numerous hazardous materials was developed and applied to a co-treatment of 20 metal-containing sludges. Three optimal groups, each consisting of 14, 3 and 3 sludges respectively, were sorted out and all normalized concentrations in each mixture were also pre-estimated. According to the estimated concentrations, stabilization agent for optimal groups 2 and 3 was taken into account in actual co-treatment process. In the first optimal group, co-treatment of 14 sludges (70% of total sludges) without any chemical dosage not only provided a cost-effective treatment but also ensured the success of passing TCLP test. In the second and third optimal groups, the maxima of normalized concentrations, 2.33 and 10.17 times the regulatory Cr limit, provided useful information in the addition of stabilization agent for sludges mixing. As expected, all sampled sludge-clay bricks of these three optimal groups met the TCLP regulatory criteria. In conclusion, the Auto-Nor program presenting excellent functions of optimal grouping and concentration normalization has been successful examined in the co-treatment of hazardous sludges, and it can be further applied to the co-treatment or remediation of other hazardous solid materials such as fly ash, slurry and soil.

Acknowledgements

The financial support by National Science Council, Executive Yuan, Republic of China (grant number of NSC 95-2221-E-239-020) is gratefully acknowledged. The authors also thank C.W. Tsai, general manager of co-treatment plant, for his assistance and cooperation in the optimal grouping treatment of hazardous sludges.

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