

Steel foundry electric arc furnace dust management: Stabilization by using lime and Portland cement

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Abstract

The purpose of this study was to determine an appropriate treatment for steel foundry electric arc furnace dust (EAFD) prior to permanent disposal. Lime and Portland cement (PC)-based stabilization was applied to treat the EAFD that contains lead and zinc above the landfilling limits, and is listed by USEPA as hazardous waste designation K061 and by EU as 10 02 07. Three types of paste samples were prepared with EAFD content varying between 0 and 90%. The first type contained the EAFD and Portland cement, the second contained the EAFD, Portland cement, and lime, and the third contained the EAFD and lime. All the samples were subjected to toxicity characteristics leaching procedure (TCLP) after an air-curing period of 28 days. pH changes were monitored and acid neutralization capacity of the samples were examined. Treatment effectiveness was evaluated in terms of reducing the heavy metal leachability to the levels below the USEPA landfilling criteria. An optimum composition for the EAFD stabilization was formulated as 30% EAFD +35% lime +35% Portland cement to achieve the landfilling criteria. The pH interval, where the solubility of the heavy metals in the EAFD was minimized, was found to be between 8.2 and 9.4.

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1. Introduction

Steel industry plays an important role in the industrialization and development of a country, as it has the input within all manufacturing sectors. Turkish iron and steel sector, whose base was established in the 1930s, plays an important role in the industrialization and development of Turkish economy. Turkey is the largest importer of scrap in the world, importing over 13 million tonnes of scrap and producing 21 million tonnes of steel in 2005 [1]. Sixty-nine percent of the total steel production is realized by electric arc furnaces.

One of the most important problems encountered in steel foundries throughout the world is the management of the dusts produced from the electric arc furnaces. Extremely fine dust is formed in the electric arc furnace by metal vaporization, which is collected in the baghouse. In a typical electric arc furnace operation, approximately 2% of the charge is converted to dust

[2]. Since metals such as zinc (Zn) and lead (Pb) are highly volatile at the temperature of molten steel, they are concentrated in the furnace dust.

Electric arc furnace dust (EAFD) generated during steel production is regarded as a hazardous waste because of the presence of significant amounts of leachable compounds of heavy metals such as Zn and Pb. EAFD is listed as a hazardous waste by the regulations of most of the countries.

The seriousness of the management problem arises from the fact that EAFD is generated in considerable amounts and its annual output is constantly increasing. 268,300 tonnes of electric arc furnace dust was generated in Turkey, in 2005. It is highly questionable if the EAFD generated is managed properly. The common practice in Turkey is collecting the EAFD in the open sites near the plants without taking sufficient precautions. EAFD is disposed of at some plants after wetting or pelletizing with water to facilitate its handling and to prevent wind dispersal. However, collecting the EAFD is just a partial solution to the management problem; the hazardous characteristics of the dust necessitate treatment before disposal of at landfills.

EAFD is a well-known waste. Several researchers [3–5] studied on the characterization of EAFD, in detail. The chemical

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Table 1

Chemical composition of the electric arc furnace dusts (EAFD) and Portland cement (PC) and comparison with values from the previous literature, (%w/w, dry basis)

	Studied EAFD	Studied PC	EAFD					
			Ref. [8]	Ref. [10]	Ref. [12]	Ref. [3]	Ref. [4]	Ref. [5]
Ca	6.65	46.44	9–0.7	7.88–1.87	2.48	4.95–3.61	4.68	3.28
Si	1.62	9.60	10.9–0.4	3.34–0.67	2.49	2.24–1.79	2.71	–
Al	0.22	1.48	0.2–2.2	0.33–0.06	0.42	0.28–0.20	0.19	–
Fe	11.58	1.17	66.2–9.4	22.37–18.9	8.22	48.58–41.1	13.45	48.96
Mg	4.09	0.46	15.8–0.8	1.41–0.47	1.08	2.83–2.00	2.55	1.65
P	0.03	0.03	0.2–0.1	–	1.7	–	0.00	–
S	0.88	0.97	1.6–0.2	–	0.64	1.20–0.52	–	–
Na	0.20	0.16	4.7–0.9	–	36.00	0.66–0.36	0.37	–
K	0.48	0.18	1.6–0	–	0.90	1.26–0.48	0.20	–
Cl ⁻	0.5	0.01	2.7–0.2	–	1.8	–	–	–
Zn	19.05	0.00	49.8–1.2	0.69–5.58	39.90	8.10–3.75	5.2	9.24
Pb	2.48	0.00	4.9–0	0.00–0.00	5.34	2.07–0.94	1.4	–
Cr	0.22	0.00	0.5–0.0	0.1–0.02	1.8	0.33–0.19	10.9	2.90
Ni	0.02	0.00	–	0.01–0.00	0.1	0.04–0.02	4.1	–
Cd	0.04	0.00	–	0.00	0.3	0.03–0.01	–	–
Cu	0.15	0.00	0.3–0.0	0.00	3.1	0.30–0.22	–	–

composition of the dusts varies according to the type of steel produced, and these variations can be significant. Stegemann et al. [6] investigated the leachability of six Canadian EAFD. They found that chromium and nickel contaminants in the EAFD are largely unleachable and the leachability of Zn, Pb, and Cd can be significant. They concluded that the leaching of these metals is largely controlled by pH.

Stabilization/solidification (S/S) has been applied by several researchers [7–13] as a treatment option before final landfill disposal of EAFD. S/S is a process that involves the mixing of a waste with a cementitious material to reduce the containment leachability and to convert the hazardous waste into an environmentally acceptable waste form, which goes to landfill or is used as construction units [14]. The term stabilization refers to a treatment with a stabilizer that has a buffering capacity and forces the system pH towards values in which the solubility of some heavy metals is minimized [15].

Al-Zaid et al. [7] investigated the effect of EAFD on physical properties of concrete. Hamilton and Sammes [8] stabilized the baghouse dusts of a steel foundry in New Zealand. They obtained good compressive strength results and low leachate levels for Zn and Pb when they used cement at 90% level. Pereira et al. [9] used coal fly ash as the main binder to stabilize the EAFD of a Spanish steel foundry. They found that when the pH of the TCLP leachate is within 8.0–11.3 interval, minimum solubility of metals such as Pb, Zn, Cr, and Cd is achieved. Skvara et al. [10] examined the properties of mixtures of Portland cement and steel foundry dusts from the steel foundries of Czech Republic. Pereira et al. [11] obtained promising results when they stabilized EAFD with partially zeolitised fly ash although they concluded with the necessity of some pH adjustments and other improvements for the zeolite fly ash. Fernandez et al. [12] treated the EAFD of a Spanish steel foundry with low grade MgO. They applied the DIN 38414-S4 leaching test that consisted of batch water leaching at a liquid-to-solid ratio of 10 to assess the toxicity for landfill option. Olmo et al. [16] studied on the modeling of Zn solubility in stabilized EAFD. Pereira et al. [17] investigated the

behaviour of an aged S/S product, EAFD stabilized by using fly ash, cement, and lime.

The leachability of a treated waste, an S/S product, is affected from two sets of factors: (1) those originate with the material itself, and (2) those that are a function of the leaching test [15]. The S/S technology may be more effective for some certain wastes than the others. Previously, the authors [18] investigated the stabilization of EAFD of a steel foundry in Turkey by using Portland cement as the main binder and reported higher leaching levels than the USEPA landfilling limits. The results differed from the findings of Skvara et al. [10], who studied on a dust with lower Zn content. The regulatory test of the EU, EN 12457, to verify the acceptability of treated waste for disposal of at non-hazardous landfills is based on the leaching of water-soluble substances, while the regulatory test of the USEPA, method 1311-TCLP, is based on the leaching of acid-soluble substances simulating the acidic conditions of a municipal landfill. Therefore, the leaching test choice may produce different decisions for the same waste. It is clear that tailor-made solutions are needed for each waste.

The purpose of this study was to determine an appropriate treatment of the EAFD of a steel foundry in Turkey. TCLP test was used to assess the toxicity of the treated waste in this study. Lime–Portland cement-based stabilization was applied and the results were compared with those of lime-based and Portland cement-based processes.

2. Materials and methods

2.1. Materials

The EAFD was obtained from a steel foundry in Bursa, Turkey. The foundry produces 235,000 tonnes/year of steel via the electric arc furnace method, and obtains the scrap from Russia, Ukraine, and Kazakhstan. Approximately, 4700 tonnes/year EAFD is generated by the foundry. A composite grab sample of EAFD was obtained at one time from the dust collecting system,

in the summer of 2005. The EAFD was received in a dry state, and mixed thoroughly to improve the waste uniformity before use. Type I Portland cement (PC) and commercial hydrated lime, which were used as the main binders, were obtained from the cement plant of Bursa.

The results of the chemical analysis of the EAFD and PC are presented in Table 1. Table 1 also gives a comparison of the chemical composition of the studied EAFD with the values from the literature. The chemical composition of the EAFD studied falls within the range typically observed. The pH, specific surface area, and specific gravity of the EAFD were 10.95, 1800 cm²/g, and 4.30 g/cm³, respectively. The X-ray fluorescence (XRF) characterization of the EAFD allows identifying Zn, Fe, Ca and Mg oxides as the components leading to an alkaline pH in water. Initial Zn and Pb concentrations of the EAFD after the TCLP extraction were measured as 851 and 41 mg/l, respectively. According to the USEPA landfilling limits, the leachable Zn and Pb concentrations should be reduced to below 4.30 mg/l and 0.75 mg/l, respectively. The Portland cement used can be classified as low-alkali ASTM type with Na₂O content of 0.42%, as the average alkali contents of normal ASTM type cements are reported to be between 0.91 and 0.94% [14].

2.2. Experimental work

2.2.1. Characterization

Thermo electron X-ray fluorescence spectroscopy with an ARL 8660 X-ray spectrometer (Switzerland) was used for the determination of chemical composition of the EAFD and Portland cement. The specific surface areas of the EAFD and the PC were measured by the fineness test-air permeability method (ASTM 204-84) by the Blaine apparatus Tonic Technic 72071. Initial metal concentrations in the EAFD were analyzed using a Shimadzu atomic absorption flame emission spectrophotometer (AA-6701F). European Union's compliance leaching test of EN12457-2 and United States EPA Method 1311 toxicity characteristics leaching procedure (TCLP) were applied before the determination of initial heavy metals concentrations. Deionised water was used as leaching fluid for EN12457-2 test and a liquid-to-solid ratio of 10 l/kg was applied. An extraction fluid with pH 2.88 was used for the TCLP test.

2.2.2. Sample preparation

Three types of paste samples, where the EAFD content varied from 0 to 90% of the total weight, were prepared. A water-to-solid ratio of 0.4 was applied for cement-based samples and 0.6 was applied for lime-based samples. Each sample was prepared in triplicate. The first type of the samples contained the EAFD, Portland cement, and water, and the second contained the EAFD, Portland cement, lime, and water. Lime and Portland cement were mixed with two mix design ratios of 1:1 and 1:9, for the second type of the samples. The third type of the samples contained the EAFD, lime, and water. The ingredients were mixed 5 min before water was added and mixing continued for 10 min. A kitchen mixer was used. The pastes were poured in plastic moulds for curing at room temperature for 28 days.

2.2.3. Analysis

All the samples were subjected to the TCLP extraction. Zn and Pb levels in the leachate after the TCLP were measured using a Shimadzu atomic absorption flame emission spectrophotometer (AA-6701F). The pH changes were monitored before and after the TCLP extraction. The samples were also subjected to the acid neutralization capacity (ANC) test. For the ANC, the samples were divided into seven sub-samples and placed in test tubes with an increased amount of nitric acid at a liquid-to-solid ratio of 10:1. The tubes were then rotated end over end for 48 h and centrifuged before the measurement of the extract pH using a Sartorius pH meter.

2.2.4. Evaluation

The results were evaluated according to the EU and USEPA landfilling criteria, in order to decide whether the stabilized EAFD was appropriate to be disposed of at a municipal landfill.

3. Results and discussion

Initial metal concentrations after the leaching and extraction tests of EN12457-2 and TCLP were measured in order to decide the landfill class where the EAFD could be accepted. The results are shown in Table 2. The regulatory limits in the table represent the upper limits to classify the waste to be disposed of at landfills for non-hazardous waste. Table 2 shows that Zn and Pb values obtained in the leachate of EAFD caused the waste not to

Table 2
Initial metal concentrations in the EAFD leachate obtained by applying EN12457-2 and TCLP tests

Parameter	EAFD (mg/l) (after EN12457-2)	EAFD (mg/l) (after TCLP)	Regulatory limits (for non-hazardous waste landfills)	
			EU (for EN12457-2)	USEPA (for TCLP)
Pb	1.56	41.0	1.00	0.75
Zn	57.0	851.0	5.00	4.30
Cu	0.00	3.23	5.00	–
Cr	0.00	0.23	1.00	0.60
Cd	0.02	0.09	0.10	0.11
Ni	0.03	0.98	1.00	11.00
Al	0.08	0.12	–	–
Si	1.82	1.86	–	–
Ca	470.00	680.00	–	–
Fe	0.22	650.00	–	–

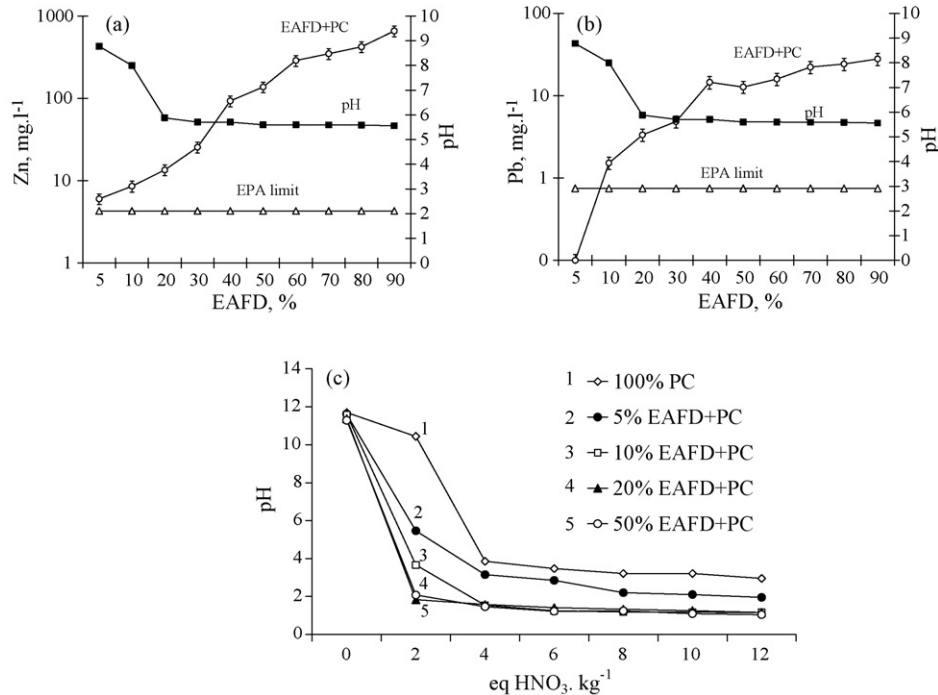


Fig. 1. EAFD stabilization with Portland cement: (a) Zn leaching and pH changes, (b) Pb leaching and pH changes and (c) acid neutralization capacity.

be disposed of at non-hazardous waste landfill. Therefore, this study aimed to stabilize the Zn and Pb in the EAFD.

3.1. Stabilization by using Portland cement

First type of the samples was treated with PC, and the TCLP test was performed to assess the stabilization performance. Fig. 1 shows the results of the PC-based treatment of the EAFD. As the EAFD content in the samples increased, Zn and Pb leaching increased (Fig. 1a and b). Zn and Pb leached from the samples after the TCLP extraction were found to be higher than the limit values. It can be inferred from the figure that the pH range of 8.78–5.56 measured at the TCLP leachate after the PC stabilization of the samples containing EAFD in varying amounts from 5 to 90% was found to be insufficient to stabilize both Zn and Pb. According to Fig. 1c, the acid neutralization capacity of the samples decreased as the EAFD content increased. ANC test was used to determine the buffering capacity of the S/S products. The higher buffering capacity of the product would increase the possibility of maintaining alkaline conditions and minimizing the leaching. The decrease in the acid neutralization capacity contributed the sudden increase in the lead leaching from 0.1 to 1.53 mg/l as the EAFD content in the samples increased from 5 to 10%. However, low amount of EAFD in the samples such as 5% might have been physically encapsulated in the cement matrix, hindering the Pb leaching.

3.2. Stabilization by using lime

Stabilization of the EAFD was carried out with lime in this part of the study. Fig. 2 shows the results of the lime-based treatment of the EAFD. It can be seen from Fig. 2a that lime,

when used as the only binder, can stabilize Zn even at high EAFD content of 70% in the paste. The leaching results after the TCLP were found to be below the landfilling criteria. However, when the EAFD content in the samples increased above 70%, the Zn leaching showed a sharp increase above the landfilling limit. This sudden increase can be correlated with the sudden decrease of the pH at the same point, as can be seen from Fig. 2a. When the EAFD content in the pastes increased from 70 to 80%, the Zn solubility increased from 0.35 to 328.8 mg/l, and the pH decreased from 11.38 to 5.72. When the EAFD content in the samples increased from 5 to 10% the Pb leaching showed a sharp increase similar to the results of the PC-based stabilization as mentioned in Section 3.1 of this study, although the pH remained constant. This suggests that although pH and acid neutralization capacity are the main controlling parameters in stabilization, other mechanisms such as physical encapsulation might have played a role in the physical entrapment of the low amount of EAFD and hindered the leaching. The samples containing more than 5% EAFD did not conform to the USEPA Pb limit. In this high pH environment of 11.4–11.9, Pb was resolubilized because of its amphoteric behaviour. Pb leaching varied between 3.79–8.76 mg/l in this high pH range. Pb leaching also increased similar to that of Zn as the EAFD content increased from 70 to 80% and as the pH decreased from 11.38 to 5.72. The acid neutralization capacity of the pastes decreased as the EAFD in the pastes increased, as can be seen from Fig. 2c.

As can be inferred from Fig. 2, the pH environment provided by lime was convenient for Zn stabilization, but high for Pb stabilization. Pb exhibited high solubility because of its amphoteric behaviour in this high pH range. Normally high pH is desirable, because metal hydroxides have minimum solubility in the range of pH 7.5 to 11 [15]. Unfortunately, all metals do not reach min-

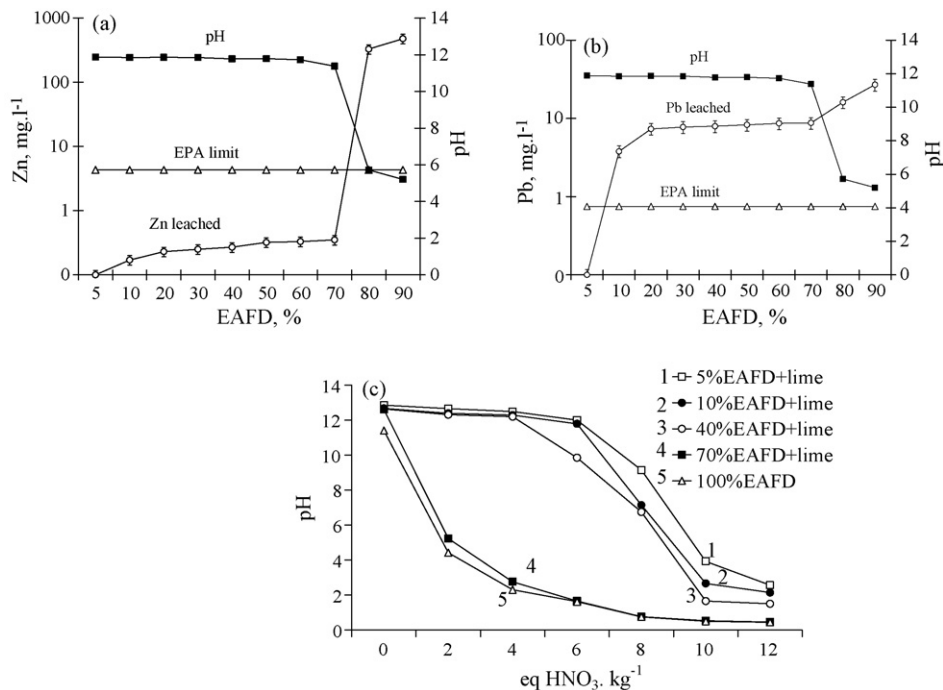


Fig. 2. EAFD stabilization with lime: (a) Zn leaching and pH changes, (b) Pb leaching and pH changes and (c) acid neutralization capacity.

imum solubility at the same pH, as was the case for Zn and Pb.

3.3. Stabilization by using lime and Portland cement

Considering that (a) the optimum pH of the system must be a compromise, (b) stabilization with only lime provides a pH above 11.4, which was high for Pb insolubility, and (c) stabilization with only Portland cement provides a pH below 5.0, which was low for both Zn and Pb insolubility, a new formulation was tried. Lime and PC were mixed together with a ratio of 1:1 to prepare a cementitious binder to stabilize the EAFD.

Fig. 3 shows the results of lime and PC-based treatment of the EAFD. Use of lime and PC mix (1:1) as the binder provided promising results in terms of Zn and Pb stabilization (Fig. 3a and b). The leaching values for both Zn and Pb were lower than those obtained by using only lime or only PC as the main binder. An increase in Pb leaching was observed when the EAFD content in the pastes increased from 30 to 40%. This increase was attributed to the decrease in the pH at the same point. As the EAFD increased from 30 to 40%, the pH decreased from 8.16 to 6.16, and the Pb leaching increased from 0.11 to 1.52, which is above the landfilling limit (Fig. 3c). The acid neutralization capacity of the system also decreased as the EAFD content increased from 30 to 40%, as can be seen from Fig. 3d.

It can be inferred from Fig. 3 that it is possible to stabilize the EAFD by using a mix of lime and PC. The mix provides a convenient pH environment for both Zn and Pb insolubility. Pb is more sensitive to pH changes and has a narrow pH range of insolubility when compared to Zn [14,15]. Therefore, Pb is the limiting heavy metal in the EAFD stabilization and the system pH should be first tailored according to Pb insolubility. This

observation confirmed some prior work from the literature [19].

As a result, an optimum composition for EAFD stabilization was formulated as: 30% EAFD + 35% lime + 35% Portland cement.

Metals may not precipitate as their single metal hydroxides, but may form other phases. This might have been the case for Zn stabilization in the present study. The hydroxy-complexes $Zn(OH)_4^{2-}$ and $Zn(OH)_3^-$ are present at pH environments above 8 [20]. Although the anionic properties of these complexes preclude their adsorption to the negative surface of the C–S–H formed during cement hydration, they may form the calcium zinc complex hydrated compound $CaZn_2(OH)_6 \cdot 2H_2O$ [20,21]. Unlike Zn, Pb does not form a new phase during cement hydration [22]. The Pb surface species are likely to be negative species in solution at high pH [20]. The dissolved species of Pb in high pH environment (12.5–13.8) are Pb^{2+} , $Pb(OH)_2$, $Pb(OH)_3^-$ [21,23]. The reason for the lower leachability of Zn than that of Pb observed in this study might have been the fact that Zn forms a new crystalline phase at high pHs.

To investigate the effect of low amount of lime on the EAFD stabilization, a different combination of lime and cement was prepared. Lime and PC were mixed together with a ratio of 1:9. Fig. 3 also shows the compared results of stabilization with two mix designs of lime and PC. Mix design of 1:9 resulted in higher Zn and Pb leaching than the mix design 1:1 (Fig. 3a and b). In this case, low amount of lime has just increased the amount of portlandite, $Ca(OH)_2$, and therefore the vulnerability of the cement matrix to the acidic leachant. Cracks can propagate through the portlandite [24,25] and the presence of cracks might have increased the sensitivity of the pastes to the acidic TCLP leachant. As a result the mix design 1:9 yielded higher Zn and Pb leaching than the mix design 1:1.

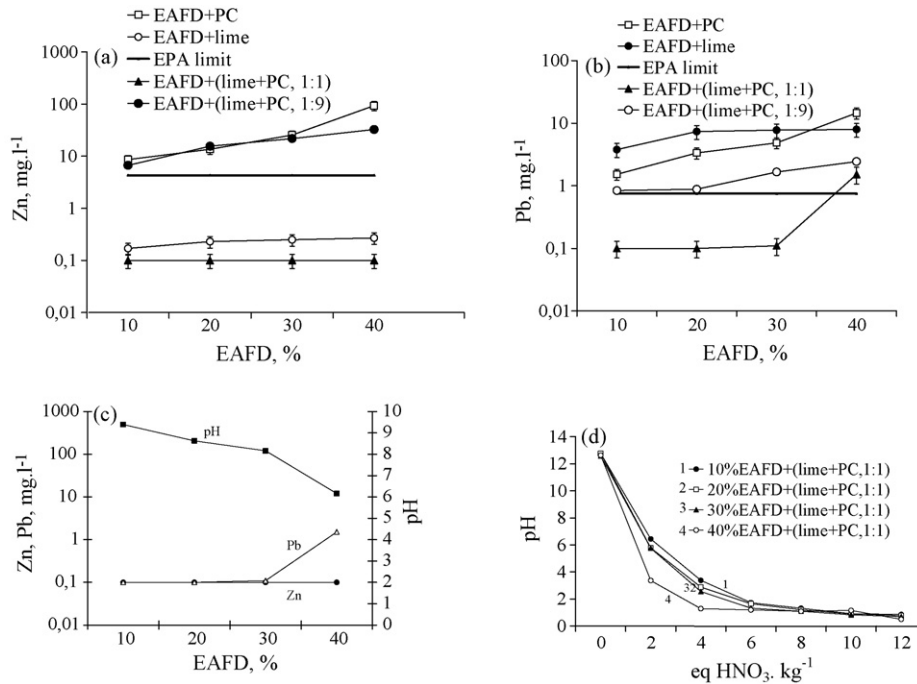


Fig. 3. EAFD stabilization with the binder containing 50% lime and 50% PC: (a) Zn leaching, (b) Pb leaching, (c) pH changes and (d) acid neutralization capacity.

3.4. Leachability as a function of pH

TCLP procedure necessitates initial adjustment of the extraction fluid to pH 2.88. However, the pH of the extraction fluid changes as it passes through the stabilization product. The buffering capacity of the stabilization product determines the leaching ratio of the metals. The pH values obtained at the TCLP leachate of the samples containing EAFD were plotted versus metal leaching results and given in Fig. 4. Zn and Pb exhibited varying leaching results that seemed to be related to the final leachate pH. According to Fig. 4, when the final leachate pH is between 6.2 and 11.9, Zn leaching below the regulatory limit obtained. Pb leaching below the regulatory limit was obtained at pH range between 8.2 and 9.4. Therefore, it can be concluded that when the pH of the stabilization system is between 8.2 and 9.4, EAFD could be successfully stabilized for landfilling purposes. This pH environment was achieved by using lime and PC with the mix design of 1:1. Up to 30% EAFD by weight

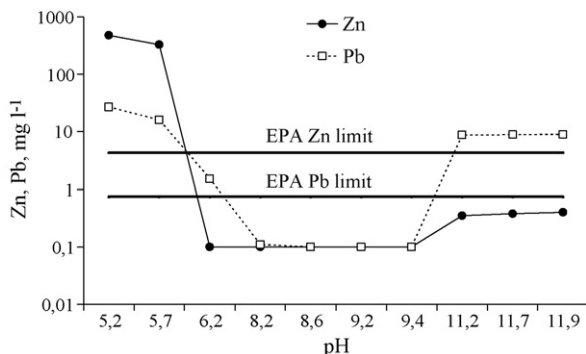


Fig. 4. Relationship between Zn and Pb leaching and pH of the leachate after TCLP.

was stabilized successfully with these binders to conform to the regulatory landfilling criteria.

3.5. General evaluation of the stabilization process for EAFD treatment

Stabilization of the EAFD yielded lower metal leaching results than the non-hazardous waste landfilling limits of USA and EU. However, the optimum formulation necessitated addition of both PC and lime to the waste, and caused a considerable increase in the waste amount. As a result, a non-hazardous but higher amount of waste was formed at the end of the process.

Table 3

Financial comparison of the two treatment options: disposal of at hazardous waste landfill or disposal of at non-hazardous waste landfill after stabilization

Item	Cost ^a (€/ton waste)
Hazardous waste landfilling	
Transportation	35
Landfilling	125
Total	160
Non-hazardous waste landfilling after stabilization	
Water	1
Lime	40
Cement	60
Electricity for mixing	1
Transportation	5
Landfilling	20
Total	127

^a The costs were calculated considering the real practical values applied in Turkey.

Hazardous waste is either incinerated or disposed of at a hazardous waste disposal site in Turkey. Since incineration is not a feasible treatment option for the EAFD because of its low loss on ignition value of 1.56%, hazardous waste landfilling is the only possible treatment option. Stabilization process brings some economical advantages over the hazardous-waste landfilling as can be seen in Table 3. Hazardous waste landfilling of the EAFD amounts to 160 €/ton, while non-hazardous waste landfilling after stabilization costs 127 €/ton according to Turkish values. The process can be considered as an alternative to the hazardous-waste landfilling, especially at places where hazardous waste landfills do not exist or are located at long distances. Steel foundries may also prefer constructing their own non-hazardous waste landfill sites rather than sending their waste to the existing landfills.

4. Conclusion

The following conclusions may be drawn from this work.

Lime, when used as the only binder, stabilized the Zn but could not stabilize the Pb in the EAFD to conform to the regulatory landfilling criteria. The pH environment provided by the lime led Pb to exhibit its amphoteric character and resolubilize.

Use of lime and PC mix (with a ratio of 1:1) as the binder provided promising results in terms of Zn and Pb stabilization. The leaching values for both Zn and Pb were lower than those obtained by using only lime or only PC as the main binder. Up to 30% EAFD could be stabilized with this mix design. An optimum composition for the EAFD stabilization can be formulated as: 30% EAFD + 35% lime + 35% Portland cement, with the findings of this study.

Final leachate pH and acid neutralization capacity was found as the most important factors governing the stabilization process of the EAFD, confirming some prior work [6,9]. According to the results of the present study, when the final leachate pH of the stabilization system is between 8.2 and 9.4, Zn and Pb in the EAFD could be stabilized successfully for landfilling purposes.

Stabilization of the EAFD is a possible treatment option prior to its landfill disposal and brings some economical advantages over hazardous waste landfilling. However, the increase in the amount of the waste as a natural result of the stabilization process should also be considered before making the final decision.

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