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## Stabilisation/solidification of APC residues from MSW incineration with hydraulic binders and chemical additives

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### Abstract-

This study focuses on the stabilisation/solidification (S/S) treatment of air pollution control (APC) residues from municipal solid waste (MSW) incineration. Six formulations (T1-T6) were tested based on different cements as binders, for the immobilisation of pollutants and to prevent their entering into the environment at unacceptable rates. Soluble phosphates and silicates were considered in some cases to fix heavy metals. The performance of T1-T6 products was measured in terms of initial and final setting times, mechanical strength, total availability and leaching from S/S products. Two monolithic leaching tests were used to estimate emissions of pollutants over 48 h and 64 days.

The results showed that the setting time was reduced when soluble phosphates were used. Moreover, although all the treatments have met the threshold of 1 MPa for unconfined compressive strength, this parameter was significantly reduced due to matrix dissolution during immersion. After three cycles of leaching, the limit of 10% for solubilisation was exceeded for all treatments with the exception of T5 (with phosphates).

This study demonstrated that the S/S treatment used at the industrial level can be improved with respect to toxic heavy metals, by using soluble silicates or phosphates, but not regarding soluble salts.

**Key words:** Stabilisation/solidification; APC residues; Incineration; Phosphates; Silicates, Leaching

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## Introduction

In integrated municipal solid waste (MSW) management systems, the significance of incineration differs from country to country [1]. In Portugal, more than 1 Mt of MSW per annum (about 20% of the total quantity produced) is currently incinerated. Air pollution control (APC) residues are hazardous by-products from MSW incineration according to regulations in force in developed countries, and thus they must be treated before disposed of in landfills [2]. For example, Portuguese incinerators generate more than  $30 \times 10^3$  t/year of APC residues, but in countries such as the UK this waste represented  $128 \times 10^3$  t/year in 2008 [3] and the trend is upwards. According to the European waste acceptance criteria (WAC) at landfills, APC residues must be treated prior to disposal. Several treatments have been tested for such wastes: separation processes, stabilisation/solidification (S/S) and thermal methods [2-4]. The S/S has been the most adopted technology to handle APC residues [5-13], even it is at the lower tiers of the waste management hierarchy, and it can be used successfully for many other hazardous wastes [14-23].

The current status of S/S may be found in valuable reviews [2, 4, 21, 23-29], and the main goal is to obtain a monolithic product with lower environmental impact than the initial waste. Indeed, S/S leads to a durable solid, with high strength and low porosity, where stabilisation aims to convert contaminants to less toxic forms and with low mobility [26]. In general, different binders and/or additives have been used to immobilise pollutants through chemical stabilisation and physical retention [21]. Several alkaline materials (e.g. lime, Portland cement, pozzolanic cement, etc.) have been employed in waste conditioning [29], but Portland cement (PC) is the most applied. Its composition comprises CaO (61-76%), SiO<sub>2</sub> (17-24), Al<sub>2</sub>O<sub>3</sub> (3-8), Fe<sub>2</sub>O<sub>3</sub> (1-6), MgO (0.1-4%), Na<sub>2</sub>O+K<sub>2</sub>O (0.5-1.5%) and SO<sub>3</sub> (1-3%) [24], and its main advantages are the low permeability of products, no specialized labour is required, inexpensive, tolerance to chemical stress (e.g. low pH, strong oxidizers) and the setting time can be controlled [23]. The main drawbacks are related to the large amount of cement required that increases the volume to be landfilled and the ineffectiveness to some inorganics. Also pulverized fuel ashes (PFA) from coal have been considered as pozzolanic additive [24] to treat APC residues from incineration and other wastes [14, 30, 31].

According to the literature, heavy metals such as Zn, Pb and Cd are well immobilised within pastes solidified, whereas chlorides are only partially retained [6]. In fact, the performance of S/S depends on the extent to which its hydration reactions are affected. Most of the binders (e.g. PC) induce high pH that promote adsorption, precipitation, absorption/encapsulation into nano-porous calcium silicate hydrate (C-S-H) gel, and

incorporation into crystalline components of the cement matrix [23]. In addition, it is important to note that stabilisation of contaminants may depend on their speciation [32-33], and others (e.g. sulphate and chloride) may have significant influence on strength [34.]

Some studies have investigated chemical additives to improve the performance of binders in S/S products. Those substances include surfactants, accelerators and sorbents, such as kiln dust, lime, fly ash, pulverized fly ash from coal (PFA), soluble silicates, soluble phosphates and other chemicals [22, 25, 27]. In this scope, it is important to note that whenever PC binder is used, the chemistry of S/S products is probably dominated by the dissolution of portlandite and calcium silicate hydrates [35]. Even so, soluble silicates have been added to PC, which react with polyvalent metal ions and form low solubility metal silicates [16, 23, 36]. Soluble phosphates have also been tested for immobilizing heavy metals [9, 37].

To assess the effectiveness of S/S treatment physical, chemical and mineralogical/micro-structural analyses can be conducted [27]. A number of studies have considered the leaching characteristics of S/S products [5-7, 10-13, 28, 38-44]. The long-term behaviour of S/S products is also an important point, since pollutants may be released to the environment over long periods [23, 40, 45]. The main degradation mechanisms of the solid matrix are carbonation, sulphate attack, freeze/thaw, wetting/drying and loading [23].

Despite all drawbacks, the APC residues treated by S/S are disposed of in landfills. In this scope, the present study complements our previous findings [2, 46-50] and aims to investigate alternative formulations based on the process that is currently used at the industrial scale. Thus, six formulations were evaluated with respect to setting time, compressive strength, heavy metals leaching (Pb, Cr, Cd) and soluble salts.

## **2. Materials and methods**

### **2.1- Materials and formulations tested**

The sample of APC residues was collected from a MSW incinerator plant in Portugal and comprises fly ashes from economizer, semi-dry scrubbers (including lime and activated carbon) and bag filters. The sample was extensively characterised in our previous works [46-50], where it was referred to as A4. The tested cements are classified in the standard NP EN 197-1 as CEM I 32.5R, CEM II 32.5R and CEM IV/A 32.5R. At industrial level CEM IV, known as Pozzolan cement, is usually chosen for stabilisation, since the heat given off by the hydration reaction is developed at slower rate.

The PFA were provided by a power plant located in Spain (Compostilla), whose physical, chemical and mineralogical properties can be found in [51]. In view of that, the composition is 51.2 %  $\text{SiO}_2$ ; 25.5 %  $\text{Al}_2\text{O}_3$ ; 7.5 %  $\text{Fe}_2\text{O}_3$ ; 2.8 %  $\text{CaO}$ ; 2.0% of  $\text{MgO}$ ; 0.8% of  $\text{Na}_2\text{O}$ ; 3.9% of  $\text{K}_2\text{O}$ ; 0.4 %  $\text{P}_2\text{O}_5$ ; 0.9%  $\text{Ti}_2\text{O}$ ; 0.1  $\text{MnO}$ ; 0.6%  $\text{SO}_3$ ; 0.1 % moisture; 4.3% of LOI. According to ASTM standards, the PFA used in our work is of class F ( $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3 > 70\%$ ) and has pozzolanic properties. The particle size of PFA was determined in our study by laser diffraction, Malvern- Mastersizer 2000. The size distribution is characterised by 5.2  $\mu\text{m}$  ( $d_{10}$ ), 17.8  $\mu\text{m}$  ( $d_{50}$ ) and 80.4  $\mu\text{m}$  ( $d_{90}$ ), which are in agreement with the ones reported in the literature [51].

Table 1 summarises the S/S formulations T1-T6 tested, which ones were based on industrial treatment and comprises mixing the components in two phases. Firstly, the APC residues were mixed with 79% of the total amount of water for 2 min. In a second phase, the binder (e.g. CEM IV), 21% of water and liquid reagent (LR) were blended for 2 min. The LR was prepared according to the industrial recipe, corresponding to an aqueous solution of 6 L of water, 1 kg of  $\text{Ca}(\text{NO}_3)_2$  and 0.25 kg of citric acid. The  $\text{Ca}(\text{NO}_3)_2$  was used as oxidiser to avoid the release of  $\text{H}_2$  after landfilling. The citric acid was added as retarder. The treatment T1 is equivalent to the one used in the industrial plant (reference case). In T2 a reduction of 20% of cement CEM IV was tested; T3 intends to assess the possibility of using CEM I and PFA; T4 uses CEM II as binder; T5 and T6 are tested soluble phosphate ( $\text{H}_3\text{PO}_4$ ) and soluble silicates ( $\text{Na}_2\text{O} \cdot \text{SiO}_2$ ), respectively, as described in a previous work [48]. For all blends, a Kenwood mixer with rotational-orbital movements was used. The fresh mixtures were formed into cylindrical moulds with 4 cm diameter and 11 cm height, avoiding as much as possible the formation of air voids. The specimens were formed into two steps: the half-filled moulds were compacted with 60 vertical hand beatings; after completely filled, additional 60 beatings were done.

## 2.2- Physical properties

The initial and final setting time were measured based on NP EN 196-3:2005, by using the Vicat apparatus, but not immersing the samples under water. The unconfined compressive strengths (UCS) of the monoliths at 28 days were determined according to the standard NF X31-212. Thus, specimens were cut in order to obtain the cylinders prescribed in the standard (4 cm diameter, 8 cm height). Some specimens were tested after cured into closed moulds at room temperature; others were de-moulded after 24 h and then immersed in distilled water. The UCS at 28 days was determined using a vertical strain rate of 0.63 kN/s

until specimens fail, and expressed in MPa ( $UCS = 10 F_c / (\pi D^2 / 4)$ ), where  $F_c$  is the compression force at failure (kN) and  $D$  is the cylinder diameter (cm)).

### 2.3- Chemical and leaching characterisation

The chemical characterisation of the APC residues involved the total content (TC) as described in [46], total availability (TA) according to NEN 7341, and the leaching characteristics from monoliths using the tank tests NF X31-211 and NEN 7345.

The TA test required disintegration of monolith into particles smaller than 125  $\mu\text{m}$ , and involved extracting the samples twice consecutively at liquid-to-solid (L/S) ratio 50 L/kg, at pH 7 and 4, respectively.

The test NF X31-211 is a French standard developed for leaching monolithic materials, consisting of three leaching cycles of 16 h with 8 h in between, where leaching does not occur. The dimensions of monoliths were adjusted to obtain cylinders with 4 cm diameter and 8 cm height. The leachate solution (deionised water) was under mechanical agitation in hermetically closed bottle of 2.5 L, for 16 h at L/S 10 L/kg. Before leaching, the monoliths were cured for 28 days at room temperature. The European Council Decision 2003/33/EC stated that Member States shall set criteria for monolithic wastes. However, in the recent Portuguese legislation (*Decreto-Lei* n° 183/2009) specific limits to solid massive wastes were not set, and thus French legal limit established in the *Arrêté du 30 décembre 2002* were used.

The diffusion test NEN 7345 was used for moulded materials, and allow leachant to flow around monolithic cylindrical samples placed in batch tanks for 64 days, in order to determine the cumulative emissions in  $\text{mg}/\text{m}^2$ . In this case, cylindrical specimens were cut (4 cm diameter, 4 cm height) and were leached with fresh leachant over 0.25, 1, 2.25, 4, 9, 16, 36 and 64 days, in seal buckets. The standard requires that the volume of leachant ( $V$ ) must be 2 to 5 times the volume of the monolith ( $V_p$ ). In our study,  $V_p$  of 50  $\text{cm}^3$  and  $V$  of 250 mL were used, and thus the L/S was roughly 4 L/kg (monoliths with about 60 g).

For both leaching tests, NF X31-211 and NEN 7345, two replicates were considered for treatments T1 to T6.

The total dissolved solids (TDS) were determined according to NF T90-029, by evaporating a certain amount of leachate in a crucible and measuring weight loss of the solids. The leachates were filtered through 0.45  $\mu\text{m}$  membrane filters, and the concentration of inorganic elements (Pb, Cd, Zn, Cr, Ni, Cu, K, Na, Ca, Fe, Al, Si) determined by flame

atomic absorption spectroscopy (FAAS), Perkin Elmer – 3000. The concentration of Hg in solid samples was determined by AMA-254 LECO. Graphite furnace atomic absorption spectroscopy (GFAAS), GBC - Avanta ultra Z, was used for the case of diffusion leaching solutions due to the low concentration of Cd, Cr and Pb. These analyses were performed by using matrix modifiers,  $\text{Mg}(\text{NO}_3)_2$  for Cr and  $\text{NH}_4\text{H}_2\text{PO}_4$  plus  $\text{Mg}(\text{NO}_3)_2$  for Cd and Pb. The concentration of  $\text{Cl}^-$  was measured by titration with silver nitrate.

The statistical analyses (ANOVA) were performed using the software STATISTICA (Version 6) at a significance level (*p-value*) of 0.05.

### 3. Results and Discussion

#### 3.1- Chemical composition of raw materials and the products S/S

The main raw materials used in the S/S treatments (T1-T6) were APC residues (sample A4), CEM I, CEM II, CEM IV and PFA. Table 2 summarises data about TC and TA, of minor or trace pollutants (Pb, Cd, Zn, Cr, Ni, Cu and Hg) and major species (K, Na, Ca, Fe, Al, Si and  $\text{Cl}^-$ ). The TC of Pb, Cd, Zn, Cu, Hg and  $\text{Cl}^-$  in the APC residues is significantly higher than in cements and PFA. The quantities of Ni and Cr are similar for APC residues and PFA, but lower in cements. For major elements (K, Na, Ca, Fe, Al and Si) different situations may occur. The data indicated for CEM I, CEM IV and PFA were determined in our study, while the values for CEM II were obtained from the literature [41]. In general, the amount leached from APC residues was significant for toxic metals and chloride, and thus a treatment is strictly necessary to stabilise these contaminants.

The acid neutralisation capacity (ANC) of a material may be defined as the amount of acid required during titration to reach a reference pH. Thus, NEN 7341 may be used for estimating this property, which is important because the solubility of contaminants is often pH-dependent [47]. Fig.1(a) shows the ANC for PFA, APC residues, CEM IV and CEM I, and taking pH 7 as reference, their values were 0.21, 6.5, 13.7 and 14.9 meq/g, respectively. Hence, the PFA has low ANC, the APC residues have moderate buffer capacity, whereas cements CEM I and IV reveal significant buffer capacity. In the case of S/S treatments with hydraulic binders, Fig.1(b) shows that (considering pH 7 as reference) the lowest ANC of 6.7 meq/g is observed for T3 (with PFA), followed by T5 (with phosphates) with 7.2 meq/g. For the treatments T1, T4 and T6, the buffering capacity was similar, with 9.7, 9.1 and 8.6 meq/g, respectively. These results were in agreement with the fact that when cement is partially replaced with pozzolanic materials (e.g. PFA) the ANC decreases [23]. In the case of T5, a certain amount of acid  $\text{H}_3\text{PO}_4$  was added, and thus the buffering capacity was partially

reduced in the preparation phase. On the contrary, when soluble silicates were added, the ANC was not greatly affected, since this reagent has strong buffering capacity to strong acids [23]. Thus, it is expected that the sensitivity of solids T1, T4 and T6 to external pH variation is lower than for T3 and T5, which is important in this framework in order to stabilise contaminants over a large time scale.

Leachates produced in the availability test (NEN 7341) for T1 to T6 products were analysed in order to assess the concentration of Pb, Cd and Cr (minor pollutants) and K, Na and Cl<sup>-</sup> (major elements), and the data obtained are reported in Table 3. The ratios TA/TC were determined taking into account TC indicated in Table 2 and the formulations showed in Table 1. The reference treatment T1 shows that regarding the immobilisation of Pb, only 4 % is available. It seems that S/S T3 is the worst treatment, followed by T4 and T6. The lowest availability of Pb is observed in T5 (with phosphates), since only 0.3% is likely to be leached. Cd release in adverse conditions such pH 4, is virtually complete for T1, T3 and T4, and slightly lower when phosphates and silicates (T5 and T6) were used. About 19-23% of Cr was released for T3, T5 and T6, while 32% was observed for T1. CEM II (T4) is inefficient for Cr. The behaviour of the soluble ions (K, Na and Cl<sup>-</sup>) shows that all treatments are ineffective to avoid their release. Some ratios TA/TC for K and Na are higher than 100% due to the theoretical estimation of TC regarding the cements.

### **3.2- Assessment of S/S treatment**

#### **3.2.1- Physical tests**

The main physical parameters evaluated in our study were setting times and UCS. The initial setting time (ST<sub>i</sub>) is the elapsed time between the gauging and the partial loss of plasticity, and the final setting time (ST<sub>f</sub>) is the elapsed time to acquire some compressive resistance [55]. Both parameters are important for assessing S/S treatments, not only because the blends have to be transported over considerable distances for landfilling, but also for correcting the industrial blending process based on laboratory data. The results obtained for T1 to T6 are indicated in Fig. 2. The reference treatment (T1) is characterised by ST<sub>i</sub> of 300 min and ST<sub>f</sub> of 525 min. A reduction of 20% of CEM IV (T2) has led to an increase in ST<sub>i</sub> for 345 min, but ST<sub>f</sub> has remained similar (530 min), and this retarding effect, was also observed in other studies [5, 31]. Treatments T3 and T4 showed high ST<sub>i</sub> (630 and 600 min) as well as ST<sub>f</sub> (780 and 750 min). By adding soluble phosphates (T5), a significant reduction was detected in ST<sub>i</sub> (180 min) and ST<sub>f</sub> (300 min), and thus the quality criteria mentioned in [27] may be compromised (recommended setting times are in the range 5 to 72 h). In the case of



soluble silicates (T6),  $ST_i$  increased to 360 min, but  $ST_f$  decreased to 435 min. It is important to note that these times are dependent on the amount of water, composition, temperature, and also heavy metals may be considered inhibitors of cement hydration [12, 29].

The UCS has been used as an indicator of long-term durability, and its value depends on the chemical reactions which are also dictated by the composition of the waste. Indeed, UCS is often employed as a measure of the capacity of the monolithic S/S material to resist to mechanical stresses [5, 8, 12, 13, 31]. This parameter is of great meaning since it is related to the progress of hydration reactions in the monolith, which determines its durability. In our study, UCS was determined according to NF X31-212 and the results obtained on monoliths cured for 28 days are presented in Fig. 3. According to French legislation (Arrêté du 30 décembre 2002) UCS should be greater than or equal to 1 MPa, and thus the acceptance limit was largely met by all the treatments. In practice, UCS should be as high as possible due to stresses during handling, placement and covering operations. However, to achieve higher UCS a larger amount of binder will be required [23], but the treatment becomes expensive and the landfills have a shorter lifetime. It is important to note that other possibility of increasing UCS is by reducing the water/binder ratio, but the water demand of APC residues makes this strategy impossible. Fig. 3 also shows that UCS for monoliths tested after water immersion are lower in all cases, and similar results can be found in the literature [12]. However, in the same investigation [12], it was found that in case of more than 50% of CEM I was employed, UCS did not decrease after immersion. In our study, with the exception of T2, for which UCS decreased 48%, the reduction was not superior to 20%. Indeed, the UCS decreases 20, 7, 13, 18 and 15% for T1, T3, T4, T5 and T6, respectively. Measuring the UCS after immersion is fundamental to evaluate leaching of contaminants into water and its interference with hydration reactions. In our case, disruption reactions have occurred in all treatments, mainly due to soluble salts dissolution. In addition, the results indicate that the industrial objective of decreasing the amount of CEM IV compromises the UCS criteria of 1 MPa.

### 3.2.2 – Assessment of leaching according to NF X31-211

The leaching behaviour of the S/S monoliths was analysed according to NF X31-211, after 28 days of curing, since this was the method used at the industrial plant to determine the efficiency of the treatment. Results using this test have been little reported [56-59].

Fig. 4(a)-(b) show the pH and the electrical conductivity (EC) measured in each leaching solutions. The high pH eluates for treatments T1 to T6, range from 12.3 to 12.5 and

from 11.8 to 12.2 in the first and third extractions, respectively. Despite the small differences in pH, they can induce variations in the amount of elements leached, such as Pb [47]. The treatment T5 has led to lower pH in all eluates, reflecting the acidic nature of  $H_3PO_4$  used in this treatment. Fig. 4 (b) shows that EC decreases in all cases, with a sharp decline from the first to the second extraction. This behaviour is due to the fact that soluble salts are mostly released in the first extraction from the surface, whereas in the following extractions salts can be retained by diffusion limitations. Since NF X31-211 requires EC measurement, finding a correlation with TDS is important, given that the former parameter is easily assessed. Indeed, Fig. 5(a) shows a linear relationship between TDS and EC measured in the leaching solutions, which can be useful at industrial level. Furthermore, one of the minimum criteria for S/S products is the soluble fraction that must be less than 10 % according to French legislation (Arrêté du 30 décembre 2002). This parameter corresponds to TDS expressed in percentage [ $TDS(\%) = TDS(g/L) \times L / S(L/kg_{dw}) \times 100$ ]. Fig. 5(b) shows the cumulative TDS in the leaching solutions of treatments T1 to T6, and thus it can be concluded that with the exception of T5 (with phosphates) all other treatments failed to meet the acceptance criterion. Comparing the reference case (T1) with the T2 (reducing cement content in 20%) an increase in TDS was observed. Therefore, the industrial target of reducing the consumption of cement, cannot be easily achieved. Neither PFA used as partial substitute for CEM I (T3) nor CEM II (T4) met the legal limit with respect to TDS. In addition, silicates (T6) slightly reduce the soluble fraction. One-way ANOVA used for comparing TDS from T1 to T6 showed that  $F_0$  (observed value of F-distribution) is 92.56 and  $F_{critical}$  5.19, thus there is significant difference between treatments ( $p = 7.1 \times 10^{-5} < 0.05$ ), and T5 has significant lower TDS.

The effectiveness of all treatments was also compared based on the amount of Pb, Cd, Cr, K, Na, Ca and Cl leached. Fig. 6(a) and 6(c) show, as examples, the cumulative leaching of Pb and Cr over time, in particular at 16, 32 and 48 h. Fig. 6(b) and 6(d) represent the total amount released with TA (Table 3). Fig. 6(a) displays that the release rates of Pb are similar in all cases, but according to ANOVA the amount released from T5 products is statistically lower than in the other treatments ( $p=0.015 < 0.05$ ). The cumulative mean values were 1131, 1599, 1086, 1091, 655 and 1656  $\mu g/kg$  for T1 to T6, respectively. Lower values were reported in the literature for Pb, after the S/S treatment of incineration fly ashes [56]. Fig. 6(b) highlights the difference between the quantities released and the ones available for leaching (TA) assessed on crushed material, and in general they differed by more than one order of magnitude. This figure shows that all treatments meet the threshold of 50 mg/kg. The released rates and cumulative amount of Cr were similar for all treatments with the exception of T4

( $p=0.003$ ), for which release was higher, probably due to higher Cr content in CEM II. Fig. 6(d) illustrates that there is near two orders of magnitude of difference between the observed and potentially leachable (TA) amount, and all treatments comply with the legal limit of 70 mg/kg, once the cumulative release is in the range of 0.1 to 0.28 mg/kg. The total amount of Cd released (results not shown) are statistically similar for all treatments ( $p=0.311$ ), TA varied between 44 and 50 mg/kg and the cumulative leaching values range from 4.9 to 6.6  $\mu\text{g/kg}$ . Thus, it is unlikely that Cd becomes an environmental problem.

The results obtained for K, Na, Ca and Cl<sup>-</sup> from T1-T6 are shown in Fig. 7(a)-(d). The release rates of soluble salts seem similar for all treatments, but they are indeed statistically different. In fact, the release of major elements was lower for T5, which is consistent with the behaviour observed previously for soluble fraction (Fig. 5(b)). Filippov et al. [58] reported that ortho-phosphates addition to cement pastes can form a continuous and weakly soluble network, which can decrease the release of salts. Other researchers stressed that chlorides are not effectively immobilised if binder additions were lower than 50%, and even monolithic tests exceed the relevant WAC limits [12]. Nonetheless, a washing pre-treatment is feasible and may increase the incorporation of MSW residues in cementitious materials [5].

### 3.2.3 – Assessment of leaching according to NEN 7345

In contrast to NF X31-211, the standard NEN 7345 has been used to evaluate the release of inorganic pollutants from monolithic materials, and it is often referred to as tank test [3, 6, 10, 42, 50, 59, 60]. This standard is the former diffusion test developed to simulate the leaching of moulded materials over a period of 64 days, but in 2004 this norm was revised and a similar test (NEN 7375:2004) was proposed [44].

The pH and EC measured in eluates is shown in Fig. 8(a)-(b), where results obtained using NF X31-211 are also represented. The pH remained close to 12.5 over the 8 extractions (64 days), which gives an indication of the stability of the materials. Moreover, no significant differences are observed between treatments. Comparing these results with those obtained by NF X31-211 for T1, some differences can be detected, which are related to the experimental conditions used in each test, namely agitation conditions, L/S ratio and replenishment times. The evolution of conductivity over time is similar for T1-T4 and T6, but lower values were observed for T5, especially in the early times. Fig. 8(b) shows that EC is lower in the case of X 31-211, due to the differences in L/S ratio (10 L/kg in the former case and 4 L/kg in NEN 7345).

Fig. 9(a) displays TDS determined over 64 days, where it can be observed that all the treatments follow a linear correlation with EC. The rate of released TDS(%) over time is presented in Fig. 9(b), and although differences between T1-T6 were low, the lowest values were observed for T5 and the highest for T2.

Table 4 summarises the average cumulative leached quantities over 64 days, for Pb, Cd and Cr (in  $\mu\text{g}/\text{m}^2$ ), for K, Na, Ca,  $\text{Cl}^-$  (in  $\text{g}/\text{m}^2$ ) and TDS (%). The concentrations of trace elements were obtained by GFAAS. One-way ANOVA showed that T1 to T6 cumulative data are similar for Pb ( $p=0.092$ ) and Cd ( $p=0.65$ ) but different for Cr, K, Na, Ca,  $\text{Cl}^-$  and TDS ( $p<0.05$ ).

Comparison of T1 to T6 in terms of Cd, Cr and Pb release, Fig. 10(a)-(c), indicates two regions with different rates of leaching, occurring later depletion (after the first 4 days of leaching). In the literature there are limited data regarding releasing profiles along time, possibly as result of low concentrations observed. Nevertheless, the profiles over time are similar to those indicated in [10, 13, 42, 60]. Cd was released at low concentrations (about  $1 \mu\text{g}/\text{L}$ ) in the first four eluates, and then about  $0.1 \mu\text{g}/\text{L}$  were detected. Cumulative emissions of Cd were about  $90 \mu\text{g}/\text{m}^2$  for all treatments, thus in accordance with the literature [44]. Cr concentrations ranged from 10 to  $30 \mu\text{g}/\text{L}$ . Pb release (main metal of concern in APC residues) ranged between 10 and  $100 \mu\text{g}/\text{L}$ , and the cumulative leaching ranged from 4 to  $19 \text{ mg}/\text{m}^2$ . Comparing the emissions of Pb with the WAC limit for hazardous waste landfill of  $20 \text{ mg}/\text{m}^2$  [13], it can be concluded that all treatments met that threshold. Nevertheless, the best results were achieved by the treatment with soluble phosphates and silicates, and one of these formulations was recommended to be further implemented in industry. Lampris et al. [13] reported similar releases for Pb, and depending on the S/S formulation the cumulative results were within the range 3 to  $72 \text{ mg}/\text{m}^2$ . In contrast with the Pb results obtained according to NF X31-211 for T5 (with phosphates), the cumulative leaching from NEN 7345 do not indicate the same. Indeed, it seems that T6 exhibited the lowest Pb release. However, some eluates of T6 have revealed to be particularly challenging in terms of Pb analytical quantification through GFAAS, and thus there is a possibility of the cumulative release was underestimated.

Fig. 11(a)-(b) show as example of soluble species, the concentrations and cumulative leaching of Na over 64 days, and Fig. 11(c)-(d) for  $\text{Cl}^-$ , for treatments T1 to T6 and also for monoliths T1 de-moulded after 464 days (T1\*) instead of 28 days. Results show that although the behaviour is similar for T1-T6, as mentioned above they are statistically different, and T5 revealed lower release. Monoliths T1\* exhibit lower leaching in the first 4 days, and the cumulative release is substantially smaller than T1. These results show that the cure time can

have a significant influence on the leaching processes as well as the UCS [24, 27]. Indeed, blending agents can react slowly and the physicochemical properties of the solid matrix are time dependent [24].

The leaching results according to NEN 7345 were previously analysed based on the Dutch Building Materials Decree (BMD) [50], which is a valuable regulatory tool to avoid polluting soil, ground and surface waters. From this study, it was clear that the amount of soluble salts (namely chlorides) present in the APC residues leads to the conclusion that this material must be prohibited as building material in any of the scenarios investigated. Indeed, the maximum permissible load for chlorides is largely exceeded for both unmoulded and moulded materials applications. The calculations showed that the concentration of chlorides in APC residues should be reduced at least four times of its initial concentration, and even in this case isolation measures should be taken into consideration.

#### 4. Conclusions

The possibility of improving S/S treatment used at industrial level was investigated, in order to immobilise contaminants contained in APC residues from MSW incineration over long time-scales. Six formulations (T1-T6) were tested and the effectiveness of S/S products was assessed. The formulations involved different types of cement, CEM I, CEM II, CEM IV, and PFA (pulverized fuel ash) as well as chemical additives (soluble phosphates and silicates). Results showed that the setting time was reduced when phosphates were used, and thus also the time available to dispose of S/S products. All treatments meet the limit of 1 MPa for UCS, but a strength loss after immersion was observed, indicating partial matrix dissolution. The leaching behaviour showed that the industrial formulation (T1) with CEM IV has good performance, but the soluble fraction may be a critical parameter. Lower binder additions (formulation T2) did not seem feasible due to the negative consequences observed in all parameters. The use of CEM I plus PFA (T3) or CEM II (T4) instead of CEM IV (T1) did not appear attractive since UCS was significantly reduced, soluble fraction exceeded the limit of 10% and the leaching was not improved when compared to the reference case (T1). By using chemical additives (soluble silicates and soluble phosphates), a positive effect was observed for most parameters, but soluble salts are not effectively immobilised.

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# FIGURES

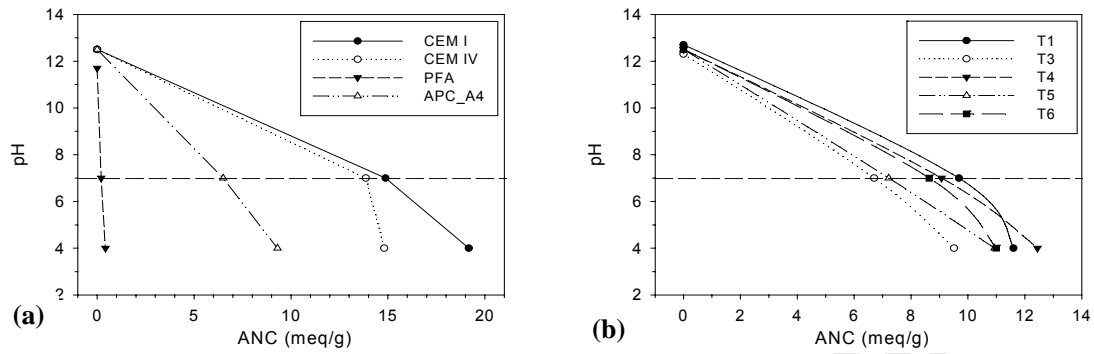


Fig. 1- Acid neutralisation capacity estimated based on NEN 7341 for a)- CEM I, CEM IV, PFA, APC residues (A4); b) S/S products obtained after treatment T1, T2, T3, T4, T5 and T6.

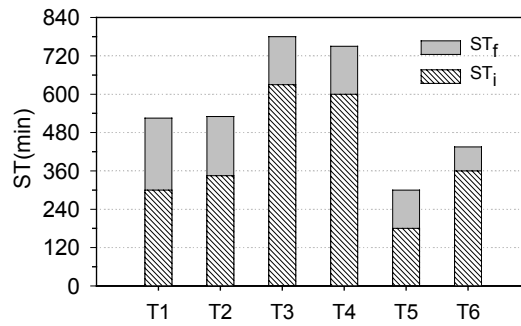


Fig. 2- Initial,  $ST_i$ , and final,  $ST_f$ , setting times for the treatments T1-T6.

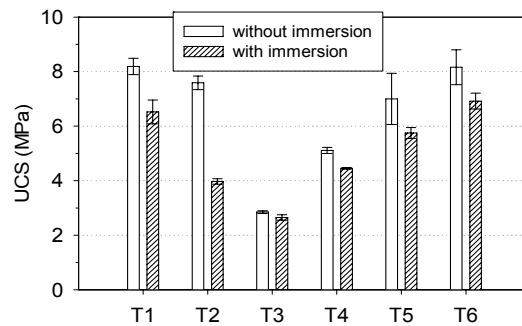


Fig. 3- Unconfined compressive strength (UCS) for treatments T1-T6, after 28 days of cure with and without water immersion ( $\pm$ sd).

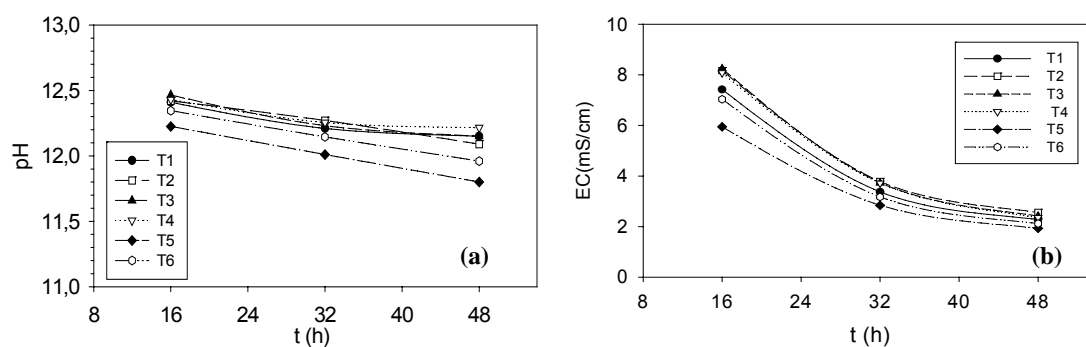


Fig. 4- a)- pH; b)- Electrical conductivity in each of the three leaching solutions from treatments T1 to T6, according X31-211.

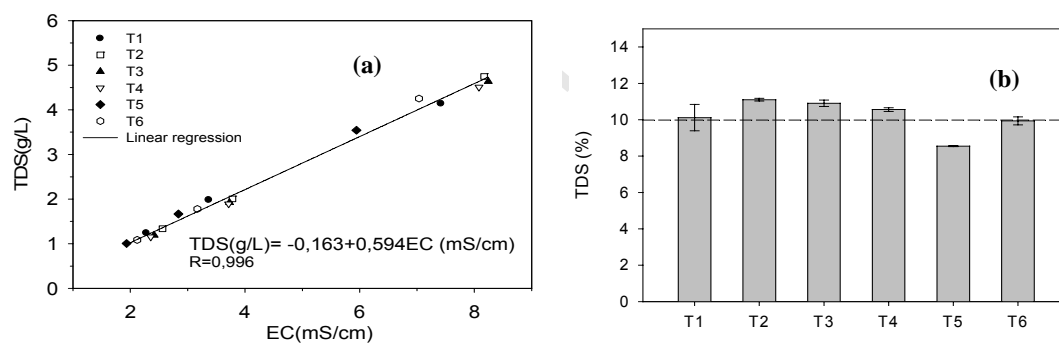


Fig. 5- a)- Relationship between TDS and EC; b)- Cumulative TDS in leaching solutions for T1 to T6.

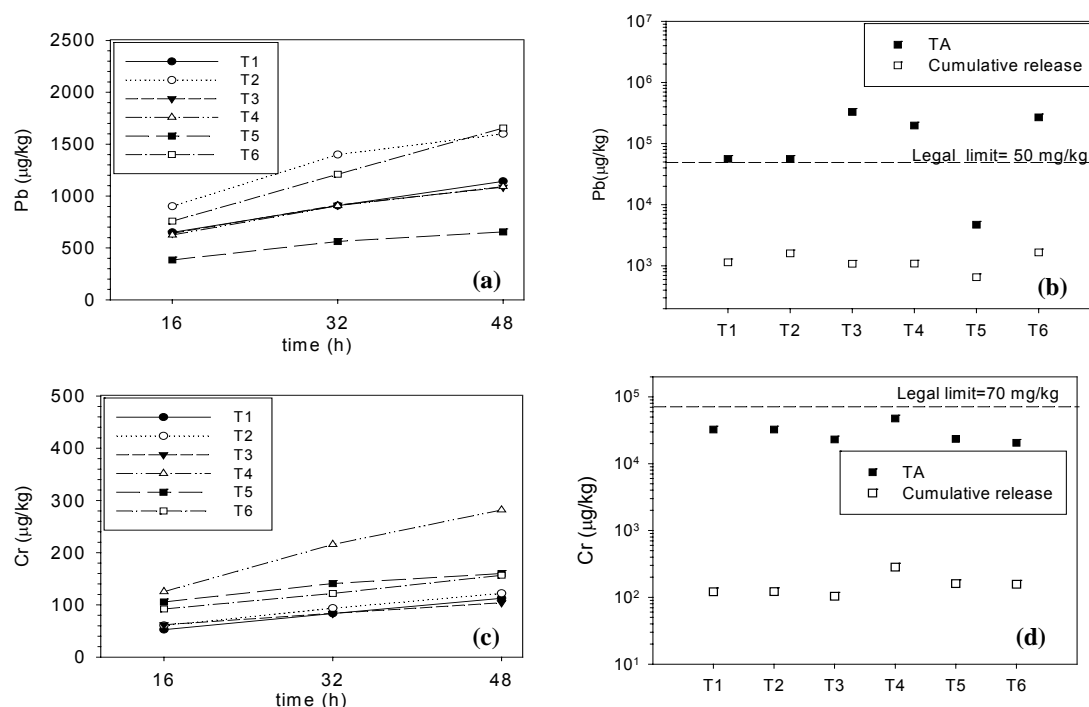


Fig. 6- Cumulative leaching for T1 to T6 products of a)- Pb according NF X31-211; b)- Pb at the end of the test and total availability; c)- Cr according NF X31-211; d)- Cr at the end of the test and total availability.

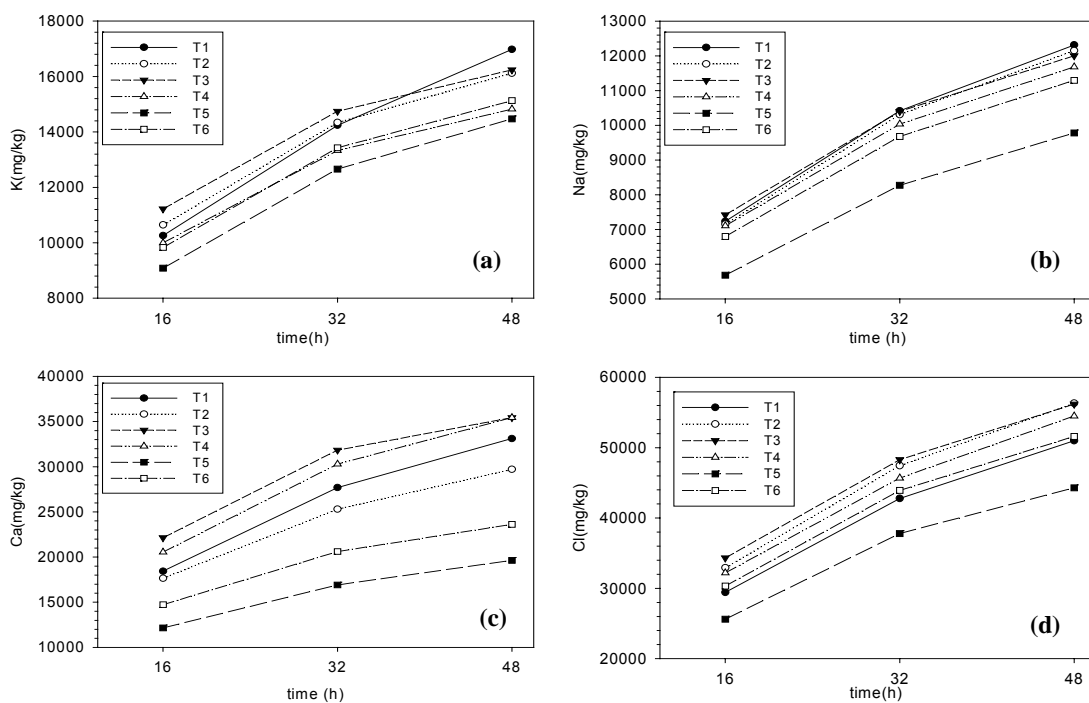


Fig. 7- Cumulative leaching according NF X31-211 for T1 to T6 products of a)- K; b)- Na; c)- Ca and d)- Cl.

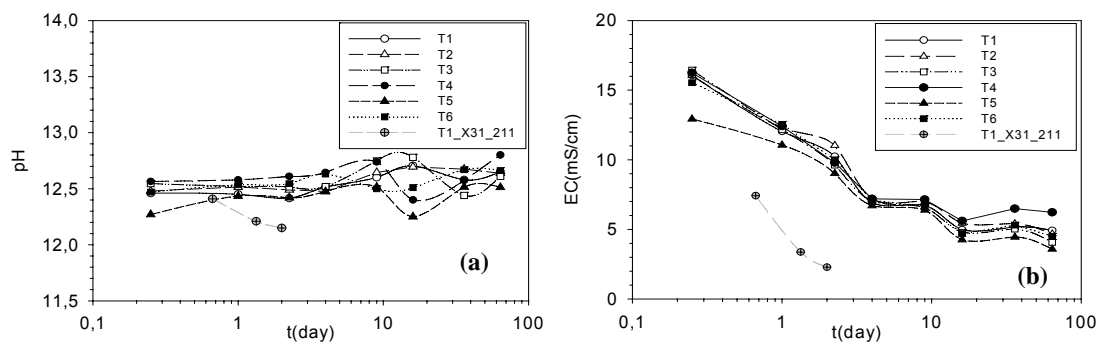


Fig. 8- a)- pH; b)- Electrical conductivity in each leaching solutions from treatments T1 to T6 according NEN 7345.

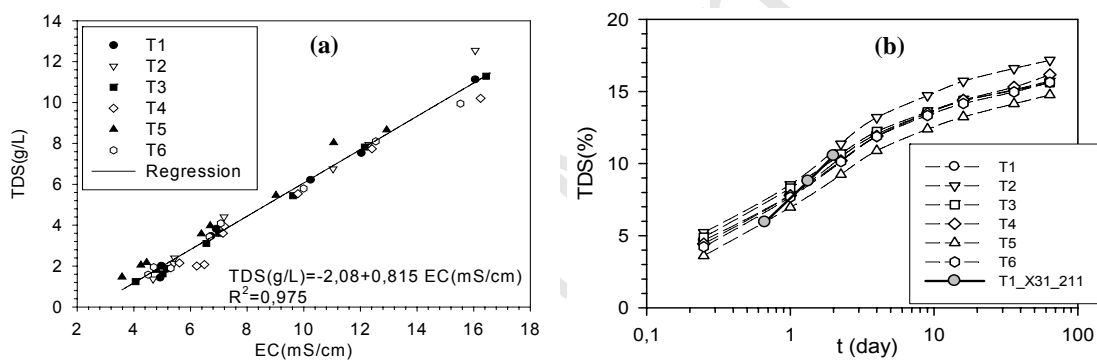


Fig. 9- a)- Relationship between TDS and EC; b)- Evolution of TDS over time according to NEN 7345 for T1 to T6 and to NF X31-211 for T1.

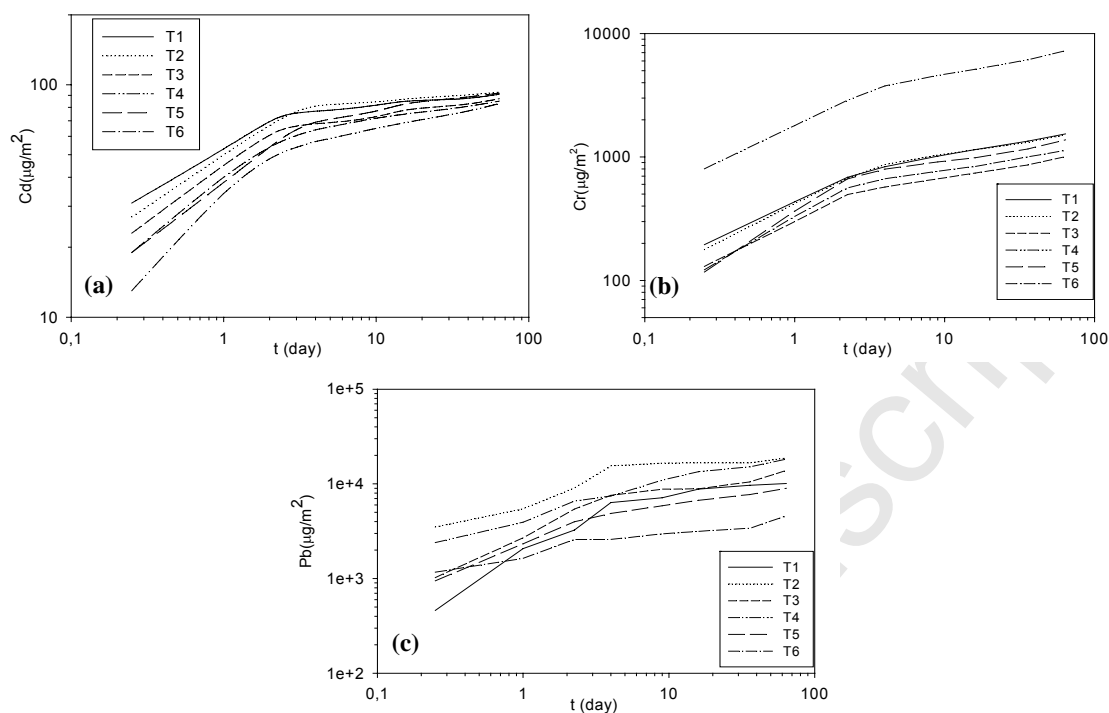


Fig.10- Cumulative leaching for T1 to T6 products according NEN 7345 of a)- Cd; b)- Cr; c)- Pb.

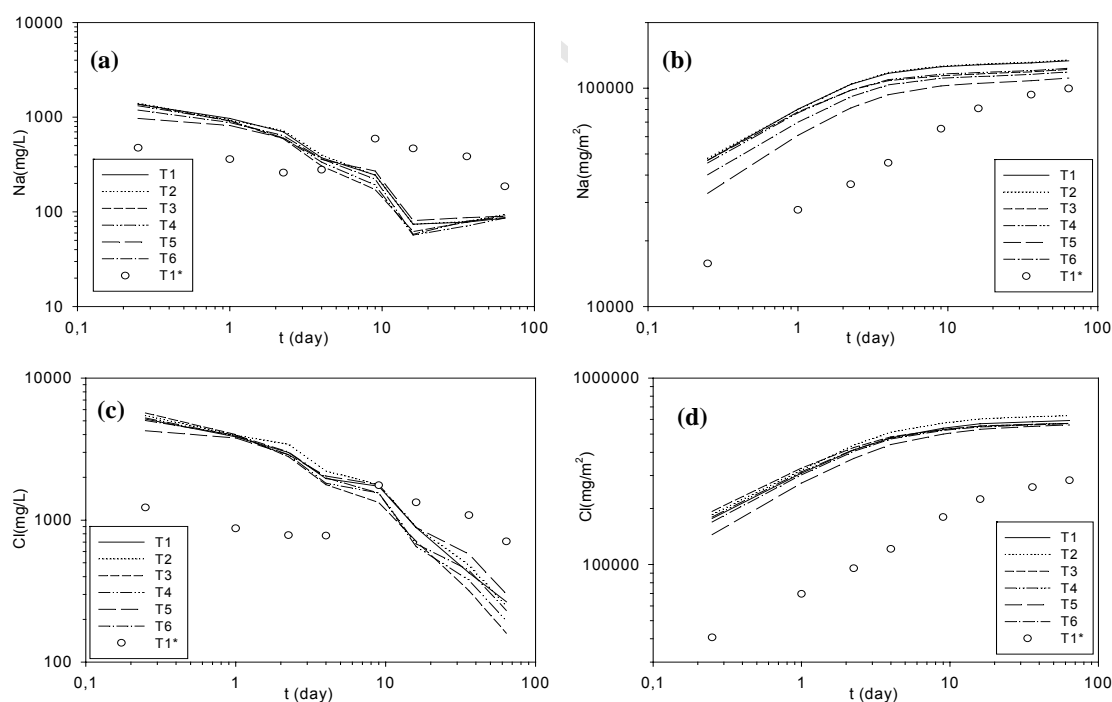


Fig. 11- Concentration and cumulative leaching for T1 to T6 products and for monoliths T1 casted out only after 464 days (T1\*) tested according NEN 7345 over time for a)-b) Na; c)-d) Cl<sup>-</sup>.

## TABLES

Table 1 – Formulations T1-T6 tested in the S/S treatment of APC residues.

	T1	T2	T3	T4	T5	T6
APC residues (g)	1000	1000	1000	1000	1000	1000
Binder (g)	620	496	620	620	620	620
Water (mL)	780	660	780	780	720	700
LR (mL) <sup>(*)</sup>	18	18	18	18	18	18
Type of binder	CEM IV	CEM IV	CEM I+PFA	CEM II	CEM IV	CEM IV
Type of additive	-	-	-	-	H <sub>3</sub> PO <sub>4</sub>	Na <sub>2</sub> O.SiO <sub>2</sub>
Additive added (mL)	-	-	-	-	60	80
Cement/liquid (kg/L)	0.78	0.73	0.78	0.78	0.78	0.78
Cement/APC residues (kg/kg)	0.62	0.50	0.62	0.62	0.62	0.62

(\*) LR- liquid reagent (aqueous solution of calcium nitrate and citric acid)

Table 2- Total content and total availability for APC residues (A4), CEM I, CEM II, CEM IV and PFA.

	<i>Total elemental content</i>					<i>Total availability</i>				
	APC (A4)	CEM I	CEM II	CEM IV	PFA	APC (A4)	CEM I	CEM II	CEM IV	PFA
		[52]	II [41]	[54]	[51]			II[41]	IV	
Pb (mg kg <sup>-1</sup> )	2408	14	43	43	129	216	32	0.21	26	1.5
Cd (mg kg <sup>-1</sup> )	87	0.2	2	0.7	2	86	<DL	0.25	<DL	<DL
Zn (mg kg <sup>-1</sup> )	6367	52	63	144	173	4360	46	30	61	6.8
Cr (mg kg <sup>-1</sup> )	188	55	29	46	137	28	47	10	40	9.4
Ni (mg kg <sup>-1</sup> )	87	23	ni	31	98	26	35	ni	35	2.6
Cu (mg kg <sup>-1</sup> )	600	35	ni	28	90	288	15	ni	12	3.3
Hg (mg kg <sup>-1</sup> )	15	0.08 <sup>a</sup>	ni	0.06 <sup>a</sup>	0.49 <sup>a</sup>	nd	nd	ni	nd	nd
K (g kg <sup>-1</sup> )	24.9	4.98 <sup>b</sup>	4.9	16.3 <sup>c</sup>	32.4 <sup>b</sup>	28.6	5.2	3.1	5.7	1.4
Na (g kg <sup>-1</sup> )	27.8	2.22 <sup>b</sup>	ni	9.9 <sup>c</sup>	5.9 <sup>b</sup>	28.4	0.9	ni	1.1	0.18
Ca (g kg <sup>-1</sup> )	350	424 <sup>b</sup>	83.9	329 <sup>c</sup>	20.0 <sup>b</sup>	nd	412	81	367	57
Fe (g kg <sup>-1</sup> )	9.94	18.3	ni	29.4 <sup>c</sup>	52.5 <sup>b</sup>	0.05	<DL	ni	<DL	<DL
Al (g kg <sup>-1</sup> )	36.5	23.8 <sup>b</sup>	19.6	38.8 <sup>c</sup>	135 <sup>b</sup>	1.19	0.48	0.25	0.15	0.13
Si (g kg <sup>-1</sup> )	83.1	102 <sup>b</sup>	352	140 <sup>c</sup>	239 <sup>b</sup>	12.2	11.6	4.12	8.8	1.3
Cl <sup>-</sup> (g kg <sup>-1</sup> )	nd	0.2	ni	<DL	ni	132	2.0	<0.05	1.8	<DL

<sup>a</sup> -Measured in our study; <sup>b</sup> - Calculated from the oxides composition; <sup>c</sup> - Calculated from the oxides composition from [53] ni- not indicated; nd- not determined; DL- detection limit;

Table 3- Total availability, TA (dry basis), and percentage of leaching against total elemental content (TA / TC).

	<b>T1</b>		<b>T3</b>		<b>T4</b>		<b>T5</b>		<b>T6</b>	
	TA	TA/TC (%)	TA	TA/TC (%)	TA	TA/TC (%)	TA	TA/TC (%)	TA	TA/TC (%)
Pb (mg kg <sup>-1</sup> )	56.4	4	331	23	198	14	4.7	0.33	269	19
Cd (mg kg <sup>-1</sup> )	48.5	99	50.4	99	49.7	97	44.1	88	44.2	88
Cr (mg kg <sup>-1</sup> )	32.4	32	23.1	19	47.4	47	23.5	23	20.6	20
K (g kg <sup>-1</sup> )	19.6	118	19.2	112	14.9	89	16.4	98	16.2	96
Na (g kg <sup>-1</sup> )	17.1	112	15.8	103	15.3	99	14.3	94	17.9	115
Cl <sup>-</sup> (g kg <sup>-1</sup> )	74.4	98	72.9	95	70.7	93	70.8	93	64.2	84

Table 4- Measured mean cumulative leaching over 64 days of Pb, Cd, Cr, K, Na, Ca, Cl<sup>-</sup> and TDS for T1- T6 according NEN 7345.

	<b>T1</b>	<b>T2</b>	<b>T3</b>	<b>T4</b>	<b>T5</b>	<b>T6</b>
Pb (µg/m <sup>2</sup> )	10099	18793	13841	18229	8978	4596
Cd (µg/m <sup>2</sup> )	91	93	87	83	92	85
Cr (µg/m <sup>2</sup> )	1537	1515	1006	7245	1376	1129
K (g/m <sup>2</sup> )	159	163	154	141	152	150
Na (g/m <sup>2</sup> )	134	135	122	123	111	119
Ca (g/m <sup>2</sup> )	532	562	493	560	445	424
Cl <sup>-</sup> (g/m <sup>2</sup> )	592	629	570	571	561	569
TDS (%)	15.7	17.2	15.6	16.2	14.8	15.6



### Highlights

Six formulations of S/S were tested for APC residues from MSW incineration;  
The formulations included different binders, soluble phosphates and silicates;  
The setting time was significantly reduced when soluble phosphates were used;  
Most of the treatments fail the limit of solubilisation due to leaching processes.  
The industrial recipe can be improved by using soluble silicates or phosphates.