

Article

Investigation of Environmental Leaching Behavior of an Innovative Method for Landfilling of Waste Incineration Air Pollution Control Residues

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Abstract: Waste incineration air pollution control (APC) residues require pretreatment before landfilling because these types of residues encompass pollutants from an incineration gas stream. The environmental concerns of APC residues consist of a risk of leaching and subsequent release of potentially harmful substances that occur under environmental exposure. The stabilization/solidification (S/S) method of incineration residues is one of the most applied technologies for hazardous incineration residues. Portland cement is commonly used as a binder material in S/S for pollutant encapsulation, in order to change the hydrological characteristics of the landfilled material. Based on previous research, an innovative S/S method for APC residues is investigated, meant to replace Portland cement with cement-like material made from lignite fly ash (FA). To do this, a lab-scale landfill was created through the promoted S/S method and exposed to the environment for 12 months. Thus, this article assesses the lab-scale leaching behavior of a landfill disposal material exposed to environmental conditions and attempts to prove the promoted innovative S/S method. The results show that the replacement of Portland cement with a substitute material for utilization in the S/S method can mitigate energy consumption in the industrial cement subsector.

Keywords: incineration residues; heavy metal stabilization; cement substitute; energy



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1. Introduction

In recent decades, municipal solid waste incineration (MSWI) residues have been produced in significant quantities worldwide, being generated by waste incineration plants. Under the framework of integrated waste management, incineration is used as a treatment for a wide range of wastes. Waste is generally a highly heterogeneous material, consisting essentially of organic substances, minerals, metals and water.

Depending on waste composition and on operating incineration conditions, smaller amounts of CO, HCl, HF, HBr, HI, NO_x, SO₂, VOCs, PCDD/F, PCBs and heavy metal compounds (among others) are formed or resident [1,2]. These pollutants are transferred from waste input to the air pollution control (APC) residues, which are formed by the sorbent material used in the flue gas treatment process. Thus, from an environmental perspective, APC residues are normally classified as hazardous waste [3–5].

From a technological point of view, the gas cleaning systems commonly used are dry, semidry and wet scrubber. The first two systems inject lime or other alkaline reactants into the exhaust gas stream to remove pollutants and heavy metal compounds. The byproducts are removed as APC residues in bag filters or electrostatic precipitators, like a powder state material. The wet scrubber systems use significant amounts of water in the cleaning process compared to the others; two types of APC residues are generated, namely waste

water and fly ash. No matter which kind of cleaning system is applied, APC residues are disposed of in landfills [6,7].

The environmental concerns of landfill disposal are leaching and subsequent release of potentially harmful substances that occur under environmental exposure. Thus, APC residues should be pretreated to prevent environmental contamination that occurs due to the landfill disposal leaching phenomenon [7–9].

As far as landfill disposal is concerned, water is the main transport medium of pollutants, so the overall water balance will determine the actual amount of water reaching the application site. Therefore, several methods for APC residue treatment have been developed to reduce the leaching of pollutants. These include extraction, stabilization/solidification (S/S), vitrification and chemical stabilization [10].

Cement-based solidification is probably the most widespread technique for treating APC residues; this is due to its simplified process, which involves mixing the residues, cement, water and other additives to develop the strength of a binder material for encapsulation of APC pollutants [10].

In the current context of “Rethinking Energy: Earth System Science Approaches 2020”, several research studies are focused on finding solutions to reduce or substitute the amounts of ordinary Portland cement (OPC) with other binder materials, due to the fact that technological processes of OPC production require high energy consumption and emit significant amounts of CO₂ [11–13].

This paper emphasizes the S/S method, the main purpose of which is to encapsulate the hazardous residues into a binder material in order to mitigate the leaching of heavy metals after final disposal. Through the S/S method, the heavy metal compounds found in residues are subjected to chemical stabilization and immobilization reactions in the hydraulic binder [11,14].

The binder material used in this research consists of cement-like material made from lignite incineration residue fly ash (FA) and flue gas desulfurization (FGD) byproducts. Lignite FA has pozzolanic properties, and FGD byproducts are used as alkali activators [15,16]. Residues or their specific constituents should always be utilized or recovered as raw materials if technically possible and environmentally beneficial.

Based on this environmental approach, the OPC substitute materials used in these experiments require no cost or energy consumption for production because they are produced as a residue from coal energy recovery processes.

2. Materials and Methods

Two types of incineration residues were used in the experiments—residues from lignite combustion as FA and semidry FGD byproducts used as binding material and as catalysts and residues from hazardous waste incineration as APC residues subjected to S/S treatment. The materials were sampled from a coal-fired power plant, CET South Timișoara (<http://www.colterm.ro>, accessed on 14 December 2020), and from a hazardous waste incineration facility, Demeco SRL Bacău (<https://demeco.ro/>, accessed on 14 December 2020). Solid residues used in the experiments were found in a powder state, with a particle size distribution under 700 μm.

Based upon previous research, a cement-like material was created by intense hydraulic mixing of the investigated solid materials and water in a controlled ratio until a homogeneous mixture was achieved, with the aim of activating geopolymerization/cementing reactions of pozzolanic elements contained in FA.

Applying this process, a slurry liquid similar to mud was obtained, which was land-filled into a mold with a base size of 20 m² and height of 1.2 m. After 24 h of environmental exposure, a monolith of 24 m³ was obtained. The experimental landfill material (LFM) mixture consisted of tap water (W), lignite fly ash (FA), FGD byproducts and APC residues in the following mass ratio: LFM = 60% W + 24% FA + 8% FGD + 8% APC [9].

During the experiments, a hydraulic mixer was used to activate cementing reactions of solid particles in the presence of water and to achieve a homogenous liquid slurry

with pipeline hydraulic characteristics. The main hydraulic parameters of interest are viscosity and density of the resulting slurry. For a solid/liquid mixing ratio of 1.0/1.5 used during experiments, a slurry liquid consistency with a viscosity of 1.35 cP and a density of 1.29 kg/L was obtained. The hydraulic mixer device used during experiments and the construction of the landfill disposal material are presented in Figure 1.

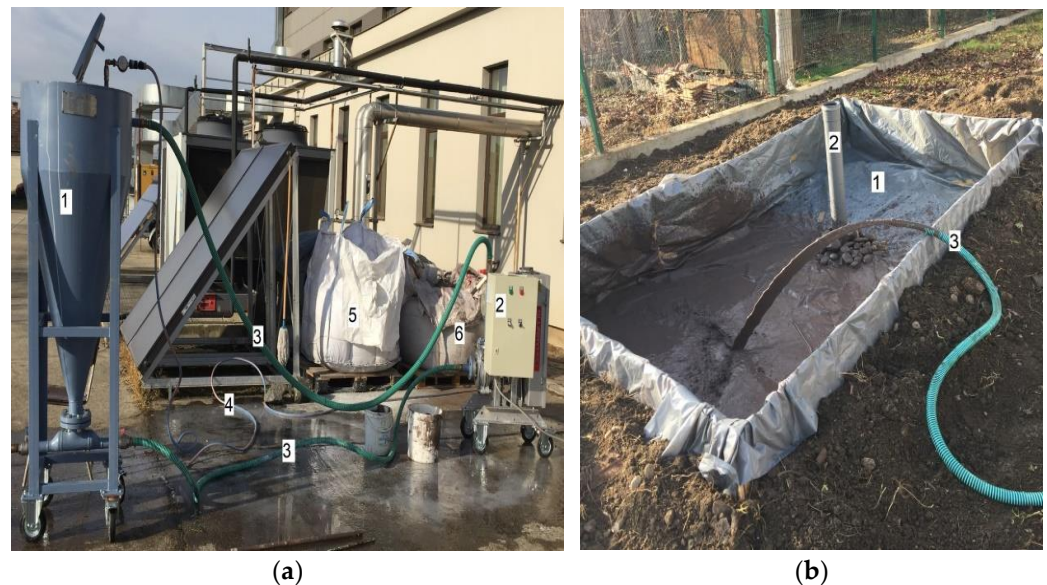


Figure 1. Landfill disposal material construction: (a) hydraulic mixing device: 1—tank, 2—centrifugal pump, 3—recirculation hose, 4—water feeding hose, 5—APC residues, 6—FA and FGD residues; (b) landfill mold: 1—plastic membrane, 2—drain system, 3—slurry discharge hose.

The created landfill disposal material was exposed to the environment for 12 months, with the aim of investigating leaching behavior of landfill material under varying seasonal temperatures in the environment and exposure to sun radiation, frosting, defrosting, wind, rain, snow and a combination of these.

After exposure, the resulting landfill leachate was analyzed to track the heavy metal elements that might migrate via infiltrating water into the environment, with the aim of investigating the chemical stability of the landfill material.

To assess the chemical stability of the binder material under the influence of water, samples of landfill material were investigated by measuring the dissolution rate of the landfill material immersed in distilled water media with pH 6.2 for 28 days. The pH of the solution was measured using a digital pH meter (Type Mettler Toledo). Periodically (1, 7, 14, 21 and 28 days), samples (100 mL) were filtered to remove any suspensions and analyzed for certain ionic species (Cu, Cr, Cd, Ni and Pb).

Metal ion contents were determined by atomic absorption spectrometry using the Varian AAS 280FS atomic absorption spectrometer.

3. Results

3.1. Prescreening of Experimental Materials

To assess the stabilization of pollutants in cement-like binder materials, a prescreening of experimental materials was conducted to identify the types and concentrations of the pollutants that could be released into the environment, if no treatment of S/S was applied. The values are indicated in Table 1. The pollutant analysis was conducted through atomic absorption spectrometry of the filtrate resulting from the mineralization of samples using an aqua regia acidic mixture according to ISO 1107/1998 standard protocols.

Table 1. Pollutant species concentrations in investigated residues [9].

Element.	MAV ¹ (mg/kg _{d.m.}) ²	Composition (mg/kg _{d.m.}) ²		APC
		FA	FGD	
Cd	5	3	<0.01	<0.01
Cr	70	220	<0.01	211
Cu	100	141	570	451
Mo	30	12	5	30
Ni	40	166	141	29
Pb	50	31	380	1881
Sb	5	6	33	703
Zn	200	171	0.29	11,100

¹ MAV—maximum allowable value according to SR-EN 12457-2003 for hazardous waste materials. ² d.m.—dry matter.

As results from Table 1 show, the investigated incineration materials FA, FGD and APC are hazardous materials with a high potential for environmental contamination, so pretreatment before landfilling is mandatory. Residue pollutant species such as Cd, Cr, Cu, Ni, Pb and Zn are above the maximum allowable values imposed by legislation, especially for the APC residues, which contain very high values for Cu (451 mg/kg d.m.), Pb (1881 mg/kg d.m.), Sb (703 mg/kg d.m.) and Zn (11,100 mg/kg d.m.).

3.2. Chemical Stability of Landfill Material and Investigation of Landfill Leachate

By using hand tools (pickaxe, chisel and hammer), several blocks of landfill material were extracted. Then, 15 small samples of 3 cm³ with a weight of 30 g per sample were cut with a hand saw. Each sample was fully immersed in 100 mL of distilled water. At intervals of 1, 3, 7, 14 and 28 days, sets of triplicate samples were analyzed to track the pollutants that might be released into the water. The results are presented in Table 2.

Table 2. Pollutant species concentrations.

Element	MAV ¹ (mg/L)	Sample (mg/L)					Landfill Leachate (mg/L)
		Day1	Day 7	Day 14	Day 21	Day 28	12 Months
Cd	0.04	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01
Cr	0.5	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Cu	2.0	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4
Ni	0.4	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
Pb	0.39	0.09	0.09	0.09	0.09	0.09	0.09
Zn	4.0	<0.4	<0.4	<0.4	<0.4	<0.4	<0.4
Cl ⁻	1000	448	483	517	552	587	589
SO ₄ ²⁻	800	328	383	675	753	765	778

¹ MAV—maximum allowable value according to SR-EN 12457-2003 for nonhazardous waste materials.

A sample from a leachate pit collection of the landfill disposal material was also analyzed. The landfill disposal material was foreseen as a leachate collection system, which was designed to collect only the infiltrating water that passes through the landfill material into a pit [9]. The obtained results are presented in the last column of Table 2.

According to the values from Table 2, it is observed that the Cl⁻ and SO₄²⁻ anions are found in considerable quantities. Lead ions are found in traces, and the rest of the cations are below the detection limit of the device. The presence of Cl⁻ and SO₄²⁻ in the leachate is supported by the presence of this anion in the composition of the raw materials.

The values of results for pollutant concentrations given by the chemical stability tests and landfill leachate indicate that the heavy metals were chemically bound into the cement-like material and were water-insoluble.

Concerning the pollution limits, it can be concluded that tracked pollutant species concentrations found in leachate are under the MAV values and are under permitted values according to actual legislation limits.

3.3. Investigation of the pH of the Lecheate Samples

The intensity of leaching due to soft water (4–8 °D) is influenced by the following factors: water hardness, stagnant or flowing water, the amount of water that leaks under pressure, soft water temperature, type of cement, density of the concrete, quality and condition of the concrete surface and dimensions and age of the concrete. Thus, the change in the pH of the leachate during stability tests is an important parameter to know [17]. The leachate pH variation behavior is illustrated in Figure 2.

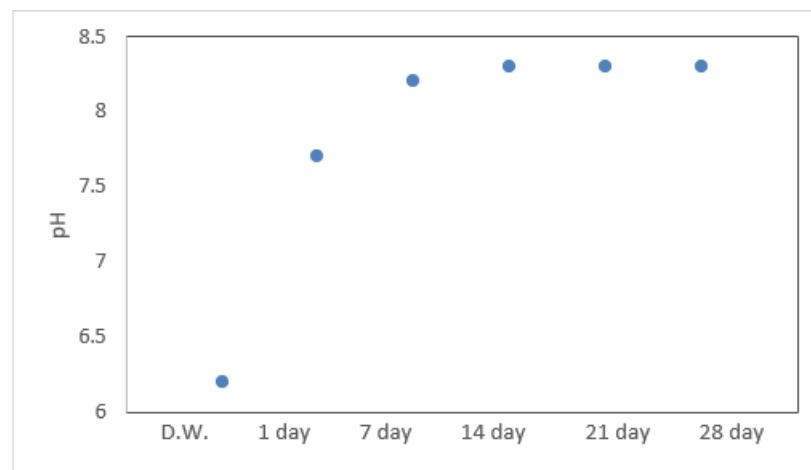


Figure 2. Leaching variation over 28 days.

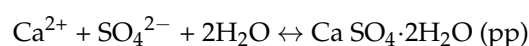
As can be observed from Figure 1, there was a small increase in pH; this is due to the high alkalinity of the material (pH 12–13.5). It is known that water without hardness or with very low hardness, such as rainwater, melting snow or even river water (soft water), is the main factor influencing the appearance of surface corrosion of the material. This water dissolves some of the hydration products of cement, such as calcium hydroxide ($\text{Ca}(\text{OH})_2$). During the stabilization process, a slight increase in the pH values could be observed, which is related to the light alkalinity of the FA and the presence of chemical compounds with basic hydrolysis. These compounds are formed by calcium and magnesium salts present in the composition of raw materials used in the experiments [9,18]. After 7 days, the material stabilized, so that the pH remained constant (pH 8.3).

4. Discussion

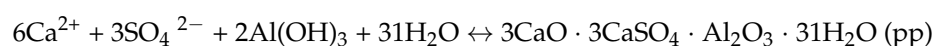
According to experimental results, it can be concluded that tracked heavy metal pollutants were encapsulated in the investigated cement-like material. As indicated in the previous research work, the main binder material is represented by the high content of FA pozzolanic compounds SiO_2 (58.77%), Al_2O_3 (15.41%) and Fe_2O_3 (7.22%), which are 81.4% by mass. By definition, “a pozzolan is a siliceous or siliceous and aluminous material, which in itself possesses little or no cementitious value but that will, in finely divided form and in the presence of moisture, chemically react with calcium hydroxide (lime) at ordinary temperatures to form compounds having cementitious properties” [12].

The geopolymerization/cementing reactions of pozzolanic elements that lead to cement-like material are developed as follows [9]:

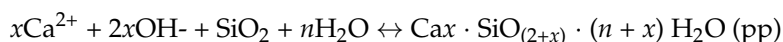
Hydrated calcium sulfate (gypsum), fast reactions:



Ash rock formation (at the beginning it is a fast reaction):



Calcium silicate hydrate (slow reaction) formation:



The high content of calcium hydroxide found in the FGD byproduct used in the experiments reacts with pozzolanic compounds and develops lime-bearing phases like calcium silicate, calcium aluminate and calcium aluminosilicate, which are subsequently hydrated in the presence of water. During the cementing process, new unsolved mineral compounds arise. Newly formed mineral crystals are gradually deposited by joining other particles in the slurry mass, which encompass them. Due to the adsorption phenomenon, the formed ash rock matrix encapsulates pollutants in its interior. The reaction between fly ash and lime can be considered mostly controlled interface. The formation of complex phases like calcium aluminosilicate, calcium iron and aluminum silicate cannot be ruled out [13].

According to the experimental results, it can be concluded that the promoted S/S method has accomplished the stabilization of APC pollutants into a cement-like material made from coal incineration residues that could be used for OPC replacement. Based on the high content of pozzolanic compounds found in the substitute material activated in the presence of an alkali FGD byproduct, a binder matrix is created that binds and encompasses the APC residues into crystallization structures and mitigates the migrations of heavy metals into the environment.

By substituting OPC with FA and FGD byproducts, energy savings could be achieved in industrial cement subsectors. The quantities of APC residues produced from waste incineration plants may vary in ranges of 10–50 or 15–60 kg per tons of incinerated waste, depending on the flue gas cleaning system, as presented in Table 3 [19].

Table 3. Typical data of the residues arising from waste incineration plants [19].

Type of Residue	Specific Amount (Dry) (kg/t of Waste)
Bottom ash/slag	150–350
Boiler ash	40–40 ¹
Wet scrubber APC as fly ash	15–40
Semiwet APC	20–50
Dry APC	15–60
Sludge from waste water treatment	1–15

¹ Fluidized-bed furnaces produce higher amounts of boiler ash.

According to the promoted S/S method and the experimental mixing ratio by mass, 60% W and 40% solid materials were used, of which 24% was FA, 8% was FGD and 8% was APC residues. Transforming this into mass units means that for stabilizing 1 kg of APC, 3 kg of FA and 1 kg of FGD from lignite combustion are needed, with a mass percentage of 20% APC and 80% cement substitute materials.

From previous research studies, common quantities of cement required for S/S applied to MSWI APC pretreatment range from 5% to 95% by mass of the mixing ratio [11,20]. It can be concluded that, by mass, the mixing ratio between hazardous and binder materials used in the present experiments is in the same range.

Based on this approach, it can be affirmed that for the investigated S/S method used in these experiments, a quantity of 4 kg of cement (3 kg FA + 1 kg FGD) has been substituted for stabilizing 1 kg of hazardous APC residue. Typical electrical energy consumption of a modern cement plant ranges from 110 to 120 kWh per ton of cement [21]. Considering the costs, the total amount of electrical energy savings that could be achieved by using the promoted S/S method for stabilization of 1 ton of APC residues ranges from 440 to 480 kWh.

This fact is beneficial from an environmental point of view. A life cycle assessment (LCA) can be used to highlight the environmental impact of the OPC production process. LCA is a technique used to assess the environmental impacts of a product during its entire

life, from cradle to grave: extraction of raw material (cradle), transportation to the factory, processing and handling of materials, product fabrication, storage (gate), distribution, use, end of life treatment, recycling and disposal (grave). Various LCA approaches may be formulated, depending on if they are applied to a commercial product, process or service.

In this particular case, it is appropriate to apply a LCA cradle-to-gate approach because a cement substitute is proposed to be used through the promoted S/S method. The cradle-to-gate approach is an assessment of a partial product life cycle from resource extraction (cradle) to the factory gate (cement for our case). LCA studies pursue the following main steps: definition of goal and scope, life cycle inventory (functional unit, system boundary), impact assessment and interpretation (results and discussions).

Following LCA streamlines, our goal was to assess the environmental impacts related to energy consumption and emissions in the fabrication of 1 ton of OPC, in order to underline the environmental benefits associated with S/S treatment of hazardous APC, by substituting OPC with cement-like materials produced as byproduct materials from coal-fired power plants. Due to the fact that the investigated substitute materials, namely FA and FGD, are byproducts utilized as recovered raw materials, their environmental impacts are excluded from the scope of LCA.

The LCA environmental indicators for OPC fabrication are energy consumption and CO₂ emissions. In order to calculate these indicators, it is necessary to consider the life cycle inventory (LCI) that quantifies the environmental indicators from the extraction of raw materials to the gate of the OPC factory. The LCI requires precise operating data inputs from an existing OPC factory, which means that LCI must be tailored individually for each particular case.

Since there are different industrial processes for OPC fabrication around the world and various assumptions that have to be taken into account regarding cement-type specifications to fit our particular goal and scope, the OPC type CEM I 42.5 N-SR3 MH/LA produced in Cementa AB's Degerhamn factory from Sweden has been chosen. Cementa AB within Heidelberg Cement, one of the cement manufacturing companies in Europe with highly developed technologies, has evaluated and improved cement's impacts on the environment [22].

The results of energy consumption related to CEM I production developed in the manufacturing process at the investigated cement factory are given in Tables 4 and 5.

Table 4. Electrical energy consumption per ton of ordinary Portland cement (OPC) [22].

Process	Amount		Percentage
	(MJ)	(kWh)	(%)
Cement mill	146.89	40.80	37
Clinker kiln	115.13	31.98	29
Raw material preparation	115.13	31.98	29
Other processes	19.85	5.52	5
Total	397	110.28	100

Table 5. Thermal energy fuel consumption per ton of OPC [22].

Fuel Type	Amount		Percentage
	(MJ)	(kWh)	(%)
Hard coal	3351	930.83	78.5
Gasoline	6	1.66	0.1
Converted fuel oil	440	122.22	10.3
Plastics	302	83.89	7.1
Solvent wastes	171	47.50	4
Total	4270	1186.11	100

As it can be observed from the data presented in Tables 4 and 5, total energy consumption for 1 ton of OPC is 1296.39 kWh (4667 MJ), of which 110.28 kWh (397 MJ) is electrical energy demand and 1186.11 kWh (4270 MJ) is heat energy demand as fuel consumption. It can be noticed that the major share of fuel demand belongs to fossil fuels (78.6%), and only 21.4% represents alternative fuels. Therefore, fossil fuels are still in the pole position for the cement industry.

According to the cement fabrication process, heat consumption is related to the clinker kiln, where fuel is combusted at a high temperature of 1450 °C, as required by the fabrication process [21,22]. Based on this aspect, the second environmental indicator of CO₂ emissions is related to fuel consumption. The results of CO₂ emissions into the atmosphere related to the production in the manufacturing process at the investigated cement factory are given in Tables 6 and 7.

Table 6. Amounts and sources of fossil fuel CO₂ emissions per ton of OPC [22].

Process	Amount of CO ₂ (kg)	Percentage (%)
Combustion fuels in the kiln	382	45.8
Calcination of limestone	438	52.5
Electric power	3	0.4
Transportation	5	0.6
Operation of diesel-driven vehicles	4	0.5
Production of gas oil	1	0.1
Others	1	0.1
Total	834	100

Table 7. Amounts and sources of alternative fuel CO₂ emissions per ton of OPC [22].

Type of Fuel	Amount of CO ₂ (kg)	Percentage (%)
Converted fuel oil	1.1	7
Plastic pellets	12.3	77
Solvent waste	0.6	4
Electric power	2.0	12
Total	16	100

As presented in Tables 6 and 7, the total CO₂ emissions into the atmosphere are 840 kg for 1 ton of OPC production. By comparing the mass streams, it can be noticed that the CO₂ emissions almost reach the cement production; expressed as mass ratio, CO₂/OPC is 0.84. Therefore, cement production has a significant negative impact on the environment, concerning GHG emissions.

Based on previous LCA results and interpretations, the importance of cement substitution with cement-like materials for S/S treatment of hazardous APC residues is highlighted.

5. Conclusions

The promoted S/S method for hazardous APC incineration residues has accomplished the main goal of leaching reduction through the investigated landfill disposal, according to the experimental results.

The cement-like materials used in the experiments involve no production costs and no energy consumption due to the fact that they are generated as residues from a coal-fired plant. From this point of view, the investigated pozzolanic material, lignite FA, is a serious contender for cement replacement.

Besides the reduction of energy consumption in the cement industry production subsectors, substituting cement in S/S pretreatment for hazardous APC residues will have a positive impact on other environmental issues like CO₂ reduction and conservation of natural resources.

Both investigated solid residues were generated in a powder state at high temperature values, being unstable under environmental moisture exposure. If no other options are available, both types of residues require a similar pretreatment before landfilling into final disposal.

6. Patents

This work is related to patent RO131486A0 process for treating residues from domestic waste incineration by stabilization/solidification in ash rock, based on which the experimental landfill disposal has been accomplished.

Author Contributions: Conceptualization, M.R.W. and D.D.; methodology, M.R.W. and I.I.; validation, A.N.; formal analysis, A.N.; investigation, M.R.W. and A.N.; resources, M.R.W. and A.N.; data curation, A.N.; writing—original draft preparation, M.R.W.; writing—review and editing, D.D.; visualization, I.I.; supervision, M.R.W.; project administration, M.R.W.; funding acquisition, M.R.W. All authors have read and agreed to the published version of the manuscript.

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Conflicts of Interest: The authors declare no conflict of interest.

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