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Published in: Journal of Hazardous Materials

Link to article, DOI: 10.1016/j.jhazmat.2021.125220

Publication date: 2021

Document Version Peer reviewed version

Link back to DTU Orbit

Citation (APA):

Zhan, X., & Kírkelund, G. M. (2021). Electrodialytic remediation of municipal solid waste incineration fly ash as pre-treatment before geopolymerisation with coal fly ash. *Journal of Hazardous Materials*, *412*, Article 125220. https://doi.org/10.1016/j.jhazmat.2021.125220

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Title page:

Electrodialytic remediation of municipal solid waste incineration fly ash as pre-treatment before geopolymerisation with coal fly ash

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Contributions: Xinyuan Zhan: Conceptualization, Methodology, Validation, Writing- Original draft preparation, Writing-Reviewing and Editing. Gunvor Marie Kirkelund: Conceptualization, Supervision, Writing-Reviewing and Editing.

Acknowledgment: Xinyuan Zhan gratefully thanks the China Scholarship Council for the grant (CSC201906050008). Laboratory technicians Ebba Cederberg Schnell, Natasja Dueholm and PhD student Benjamin Alexander Regaard Ebert, Department of Civil Engineering, Technical University of Denmark are thanked for experimental and analytical help. Aase og Einar Danielsens Fond is acknowledged for funding the experimental work.

Electrodialytic remediation of municipal solid waste incineration fly ash as pre-treatment before geopolymerisation with coal fly ash

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7 Abstract

Municipal solid waste incineration (MSWI) fly ash is classified as hazardous waste and needs 8 to be disposed of according to strict regulations. By disposal, valuable resources in the MSWI fly 9 ash is lost, and other solutions are sought for. The effect of electrodialytic remediation (ED) as a 10 pre-treatment for removing heavy metals from MSWI fly ash before using the treated ash in 11 geopolymerization with coal fly ash was explored. ED pre-treatment for MSWI fly ash increased 12 the Si reactivity and the Si/Al ratio. The mixture of 80% coal fly ash and 20% ED treated fly ash 13 with 8M NaOH (L/S 0.37 mL/g) was found optimal, with a resulting compressive strength of 15.3 14 MPa, which was higher than the reference coal fly ash geopolymer. The leaching concentrations 15 of Pb, Zn, Cr, Cu and Ni were below 0.02 mg/L with Mn and Cd at 0.023 and 0.027 mg/L, 16 respectively. The enhanced mechanism for ED treated MSWI fly ash in geopolymer was 17 confirmed by FTIR analysis and SEM images. The resistance against extreme leaching 18 environments for treated fly ash geopolymer was stronger than raw fly ash geopolymer, and 19 physical encapsulation of geopolymeric gels contributed to the heavy metal immobilization. 20

Keywords: MSWI fly ash; electrodialytic remediation; heavy metals; geopolymers;
immobilization.

23

24 1. Introduction

25 Municipal solid waste incineration (MSWI) has become the mainstream method in waste management of household waste due to three advantages, namely energy recovery from waste, 26 mass and volume reduction, and removal of pathogens. Accordingly, the by-product, MSWI fly 27 ash (FA), is produced in large amounts. About 64 and 106.8 million tons of MSW are incinerated 28 in EU by 2020 (CEWEP, 2013) and China by 2018 (China Statistical Yearbook, 2019). About 3% 29 30 (Hjelmar et al., 2011) of the mass of incinerated MSW ends up as fly ash, corresponding to 1.92 and 3.06 million tons fly ash in EU and China. Meanwhile, the MSWI fly ash is classified as 31 hazardous waste due to its considerable content in heavy metals, soluble salts and polychlorinated 32 33 dibenzo-p-dioxins/furans (PCDD/Fs). Therefore, the FA must be disposed of before or further processed before alternative use. The European Commission encourages the circular economy, 34 and MSWI fly ash as a secondary material plays a crucial role in promoting circular economy 35 (Quina et al., 2018), but it needs to be pretreated. There are three main approaches for treatment 36 before disposal or potential reuse of FA: recycling valuable elements by extraction (Quina et al., 37 2008, Luo et al., 2019), stabilization/solidification (S/S) (Zhan et al., 2019) and thermal treatment 38 (Hwang et al., 2012). 39

The S/S method includes chemical agents immobilization, cement stabilization and other cementitious materials encapsulation (Shiddique, 2010; Wang et al., 2015; Mu et al., 2018; Atanes et al., 2019). Geopolymer, as a novel cementitious material, has received more attention in recent years for immobilizing heavy metals than other S/S approaches. FA can be utilized with red mud

(Ye et al., 2016), metakaolin (Jin et al., 2016), coal fly ash, mineral residues or other industrial 44 solid waste containing aluminium and silicon resources which are necessary for geopolymerisation. 45 The dominant S/S mechanism of heavy metals in FA is the gel mesh encapsulation in geopolymers 46 (Li et al., 2019). The proportion of FA for preparing geopolymers was proposed at 5-15%, which 47 cannot decrease the compressive strength and the durability of heavy metals in geopolymers 48 (Lancellotti et al., 2010; Zhao et al., 2019). However, when the proportion of FA in geopolymers 49 increased, the compressive strength of geopolymer containing FA decreased over time due to the 50 high salts content (chlorides and sulfates)(Gunasekara et al., 2019), which could be problematic 51 52 for the durability and mechanical performance of the geopolymers. Furthermore, FA has a low level of aluminium and silicon resources (Zheng et al., 2011) combined with a high content of 53 soluble heavy metals, which indicates that it is not a proper material for geopolymerisation in its 54 raw state. Therefore, FA must be pretreated to improve its properties when used in 55 geopolymerization and following enhancing the performance of MSWI fly ash-based geopolymers. 56 Furthermore, the recycling proportion of FA after pretreatment in geopolymers can be increased. 57 Electrodialytic remediation is an extraction and separation method that can remove toxic 58 metals from contaminated materials (Kirkelund et al., 2019). The contaminated material is 59 60 suspended in distilled water in a suspension compartment which is separated from the electrolyte compartments by ion-exchange membranes. The metals are extracted from the material due to 61 acidification processes in the electrodialytic cell, and ionic metal species are separated from the 62 63 suspenions compartment to the electrolytes by electromigration in an applied electric field. The total amounts of SiO₂ and Al₂O₃ for a Greenlandic FA increased from 8.1% to 17.1% by 64 electrodialytic remediation (Kirkelund et al., 2018), and it reduced the chloride and soluble heavy 65

metals content (Belmonte et al., 2018). Thus, the result showed that electrodialytic remediation isa potential pre-treatment method for FA.

In this work, electrodialytic remediation was used as a pre-treatment method to improve the chemical properties of FA before the utilization in coal fly ash geopolymerisation. The experimental outcome of the electrodialytic remediation was analyzed, especially for the heavy metals and chlorides. The difference between the pretreated and raw FA used in geopolymerisation was studies for mechanical and environmental properties. It was finding that the electrodialytic remediation for FA increased by 33.3% in the recycling proportion in geopolymers.

74

75 2. Materials and methods

76 2.1 MSWI fly ash and coal fly ash collection

MSWI fly ash (FA) in this study was from Amager Bakke MSWI plant of Copenhagen,
Denmark. Coal fly ash (CFA) was collected from a coal power plant in Chongqing, China and
classified as ASTM Class F fly ash.

80 2.2 Electrodialytic remediation experiments

The electrodialytic remediation (ED) apparatus was built with three compartments, as shown in Fig.1. The ED apparatus was made up of two 5 cm long cylinders with an inner diameter of 8 cm (electrolyte chambers I and III) and a 10 cm long cylinder in between (suspension chamber II). The anion exchange membrane (AN) and cation exchange membrane (CAT) were placed between the electrode (chamber I and III) and the middle chamber (II), respectively. Chambers I and III were the anode and cathode compartments and filled with 500 mL circulating electrolytes of a 0.01 M NaNO₃ solution with a pH of 2 adjusted by HNO₃. The ash suspension with 100 g FA and 350

mL distilled water in chamber II was automatically stirred. Some details concerning the ED 88 experiment were as follows: 89

Power supply: A DC power supply (Hewlett Packard E3612A) kept a constant current of 50 90 91 mA and monitored the voltage variation between the cathode and anode. Electrodes: The electrodes were obtained from Permascand and made of platinum-coated 92 93 titanium wires with a diameter of 3 mm. Electrolyte circulation in chamber I and III: A peristaltic pump was applied to circulate 94 electrolyte of the electrode compartment. Water flux (Lima et al., 2008) occurred over ED 95

experiment and reduced by adjusting the pump rate. 96 Exchange membrane: The ion exchange membranes were obtained from Ionics (anion 97 exchange membrane 20 SZRA B02249C and cation exchange membrane CR67 HUY 98 N12116B).

Stirring: A flexible plastic flap fastened on a glass rod was put in chamber II. The glass stick 100 was fixed into an overhead stirrer. 101

99

Control and measurement during the ED experiment: The pH and electrical conductivity of 102 • ash suspension and voltage drop between electrodes were determined and recorded daily. The 103 catholyte and anolyte were kept the pH at 1-2 by adjusting with 6M NaOH solution in the 104 anolyte and 8M HNO₃ solution in the catholyte. The whole ED experiment lasted 28 days and 105 was conducted at ambient temperature. 106

107 After ED experiment: the ash suspension was filtered through a 45 µm filter and the treated material dried at 105°C before further digestion and heavy metal analysis. The membranes 108 and stirrer were placed in 1 M HNO₃ and the electrodes in 5 M HNO₃. All liquid samples, 109

including suspension liquid and electrolytes were kept for heavy metal analysis and the EDtreated fly ash (TFA) was used for geopolymer samples.

112

113 2.3 Geopolymer preparation

The geopolymer mixes are shown in Table 1. The raw materials were blended for 5 min to acquire homogeneity and then stirred manually with 8 M NaOH solution (L/S ratio of 0.37 mL/g) for 10 min. The mixture was poured into rubber moulds (20*20*20 mm) and entrapped air bubbles were removed through vibration (5 min). The moulds were kept in a curing box at 80 °C and cured for 24 hours. The samples were demoulded after 24 hours and cured for 7 and 28 days at ambient temperature and humidity. Leaching tests, compressive strength, XRD, FTIR and SEM analysis of S/S solids were conducted.

121 2.4 Analytical tests

122 Based on Technical specification for pollution control of fly-ash from municipal solid waste incineration (HJ 1134-2020), leaching method of HJ 557-2010 was used to assess the leaching 123 behavior of heavy metals in raw materials and geopolymers samples (Table 1) and performed for 124 three repetitions. Four grams of fly ash sample was mixed with 40 mL distilled water (L/S = 10125 mL/g), and the samples were horizontally shaken at 110 ± 10 rpm/min and an amplitude of 40 126 mm for 8 h at ambient temperature; after that, the samples were kept stationary for 16 h and the 127 128 pH of suspensions was measured, and then the solutions were filtered before Cd, Cu, Cr, Zn and 129 Pb were analysed by ICP-OES (Varian 720-ES, USA). Cl anion was determined on the leaching solution by ion chromatography (Dionex ICS-1100, Thermo Scientific, USA). Each leaching test 130 was characterized by mean and deviation values. The acid-alkaline against experiments of FA, 131 132 CFA20FA and CFA20TFA geopolymers were conducted in accordance with the method of HJ

133 557-2010 for two blinds, but the leaching solutions were 0.01, 0.05, 0.1, 0.3, 0.5, 1 M HNO₃, 0.5 134 and 1 M NaOH solutions, respectively. The reactivity of Al and Si in the raw materials was 135 determined based on the methods reported by Panagiotopoulou et al. (2007) and conducted in three 136 replicates. One g sample was dissolved in 40ml 10M NaOH solution and the mix was shaken for 137 24 hours at 20 and 80 °C, whereafter 2 ml leachate was diluted to 20 ml, filtrated and acidified 138 with 2 ml concentrated HNO₃. The samples were stored at 4°C before Al and Si were analysed by 139 ICP-OES.

Mineral phases of raw materials and the composite geopolymers were assessed by use of 140 XRD (X'Pert Pro, PANalytical, Netherlands) with CuKa radiation in the 20 range from 10° to 80° 141 142 at a scanning rate of 0.02°/s for 2h. The data of XRD was analyzed by Jade 5.0 and the crystallinity 143 of samples was calculated based on built-in silicon data of Highscore Plus. Raw materials and 144 geopolymers were measured between 4000 and 600 cm-1 by use of FTIR (Spotlight 400, PerkinElmer, USA). The micro-morphology of raw materials and geopolymers were tested on 145 powder samples by use of SEM (Quanta 250, FEI, USA). The major elements of the raw materials 146 147 were determined by the use of XRF (XL3t, Thermo Scientific Niton Co., US). Chinese Standard GB/T 17671-1999 (GB/T 17671, 1999) were used as a guideline to test the compressive strength 148 of samples and an Instron 6025 was used for this purpose on three replicates of the mixtures seen 149 in Table 1. 150

152 3. Results and discussion

153 3.1 Electrodialytic experiment

The pH and electrical conductivity of the FA suspension, along with the voltage drop over the 154 ED cell are presented in Fig. 2. The pH of the fly ash suspension decreased during the ED 155 experiment due to the acidification by the water splitting at the anion exchange membrane. The 156 electrical conductivity of the suspensions first decreased and then increased. The former was 157 explained by that the hydroxyl anion is the most conductive and when it is neutralized by protons 158 the conductivity decreases, and the latter increase was due to the constitutes of FA dissolving 159 during the acidification of the suspension. The voltage drop of the ED system was almost kept 160 constant at 4V, indicating that the energy this pre-treatment method needed was at a low level 161 owing to the low consuming power (UI). The final distribution of heavy metals in the ED system 162 163 is shown in Fig. 3 and the details in the Table S1. The removal rates of Cd, Cu, Mn and Zn (the amount of the metal except what remains in treated ash) were 98%, 80%, 78% and 84%, 164 respectively, higher than that of Cr (36%), Ni (45%) and Pb (12%). For Cd, Cu, Mn and Zn, the 165 major amounts were in the cathode side of the electrodialytic cell, which means that these metals 166 were mainly removed as cations (Cd^{2+} , Cu^{2+} , Mn^{2+} and Zn^{2+}). In contrast, the Pb and Cr mainly 167 remained in the TFA, which indicates that the major type of Pb and Cr in FA belongs to acid-168 insoluble fractions. For Pb, Pb^{2+} ions dissolving from FA can react with the SO_4^{2-} to the insoluble 169 PbSO₄. The proportion of Pb found in the anolyte was higher than other heavy metals, which 170 means that Pb also formed anions, such as PbO₃²⁻, Pb₂Cl₅⁻ and PbCl₄²⁻ (Jiao et al., 2016). The pH 171 of the ash suspension at the end of the electrodialytic experiment was 1.63. The metal proportion 172 found in the ash suspension at the end of the experiment that had not electromigrated to the 173 174 electrode chamber indicates that the heavy metals in the filtrate from ash suspension were in the

form of neutral metal complexes or may be in non-soluble solid phases. For instance, 18 % Cr was
found in ash suspension, suggesting uncharged Cr metal complexes such as non-soluble chromate
or dichromate compounds (Šcancar and Milacic, 2014).

178 3.2 Characteristics of CFA, FA and TFA.

The chemical composition and leaching characteristics of CFA, FA and TFA are shown in 179 180 Table 2. The CFA contained large amounts of aluminium-silicate resources, which indicate that 181 the CFA is an eligible geopolymer material. In contrast, the FA was mainly composed of chlorides, sulfates and Ca-bearing compounds, in which chlorides prevent FA from being a high-grade 182 material for geopolymerization. After ED pretreatment for FA, silicon in TFA increased and 183 chloride decreased, which was beneficial to the geopolymeric reaction due to more formation of 184 Si[OH]₄⁻ (Li et al., 2019). The FA had large quantities of heavy metals like Zn, Pb, Cd, Cu, Cr, 185 Mn and Ni compared with coal fly ash, as well as for Cl⁻ ions. Based on the leaching test, it was 186 found that the leaching concentrations of heavy metals in FA were low except the Pb and Zn. This 187 can be explained by that the alkaline property of FA caused the heavy metals to form hydroxide 188 compounds and precipitate on the FA particles. However, the Pb leaching concentration exceeded 189 1 mg/L, limit value of GB 8978-1996. The heavy metal concentrations for TFA were lower than 190 191 for raw MSWI FA except for Pb. The most significant decrease in concentrations was seen for Cd, 192 Cu, Mn and Zn. Based on Fig. 3, the soluble metals in the fly ash were dissolved into the suspension and the majority of the metal ions migrated to the cathode. Thus, the increase of Pb 193 194 concentration for TFA is explained by that 44 % of the ash dissolved in the ED experiment and 195 this dissolution was mainly due to the soluble salts. The leaching concentrations of heavy metal in 196 treated fly ash were higher than that of the raw fly ash, and exceeded the limit value of GB 8978-197 1996 for Cd, Cu, Ni, Pb and Zn. The main factor causing the leaching increase was the final pH of the leaching solution which was for treated fly ash which was acidic (pH 4.7) compared to the alkaline (pH 11.9) for the untreated fly ash. However, the leaching concentration of Cr and Pb decreased for the TFA due to the formation of their non-soluble compounds.

The Al and Si reactivity of the raw materials used for geopolymerisation is shown in Table 3 201 and all materials contained reactive Al and Si. The reactive Al and Si of the FA and TFA was 202 similar or even higher than of the CFA at ambient temperatures. The reactive Al and Si 203 concentration increased with higher temperature, which has also been found for metakaolin 204 (Granizo et al., 2014) and gold mine tailing (Falayi, 2019). Note that the increase in Si reactivity 205 for CFA at 25 and 80 °C was two times the reactivity of Al, which means that the reactive Si/Al 206 ratio under high temperature increases and this could favor the development of compressive 207 208 strength for CFA geopolymers (Tzanakos et al., 2014). TFA contained more reactive Si and less reactive Al compared to FA, so that the ratio of reactive Si/Al increased after electrodialytic 209 treatment of the fly ash. 210

The crystalline phases of CFA, FA and TFA were determined by XRD and are shown in Fig. 211 4. CFA contained the crystal minerals of mullite (PDF#15-0776), quartz (PDF#46-1045), calcite 212 (PDF#05-0586) and hematite (PDF#33-0664) with low total crystallinity of 5%. The low 213 crystallinity combined with the chemical composition of coal fly ash, indicated that most 214 aluminium silicate resources of CFA were amorphous. In contrast, the crystalline phases of the FA 215 were mainly anhydrite (PDF#37-1496), halite (PDF#05-628), quartz, calcite and hematite 216 (PDF#33-0664) and of the TFA were mainly anhydrite, bassanite (PDF#41-0224) and quartz. A 217 significant difference between FA and TFA, was that NaCl was removed due to the electrodialytic 218 treatment and this is confirmed by the reduced chloride leaching in Table 2. It can be expected that 219

alkaline Ca-bearing compounds in FA such as anhydrite, bassanite and calcite can promote thegeopolymeric reaction of CFA geopolymers (Zhan et al., 2018).

222 3.3 CFA based geopolymer with FA and TFA

223 The compressive strength of geopolymers with FA and TFA is shown in Fig. 5. When the 224 proportion of FA in the geopolymer was 5 - 15% and cured for 7 days, there was no obvious effect on the compressive strength. However, the increase of FA deteriorated the compressive strength 225 226 of geopolymers at 28 days of curing. This result indicates that the FA can be a potential substitute 227 for CFA at low proportion and consistent with the result reported by Tian et al. (2020). Meanwhile, 228 with 20% FA in the geopolymer, the compressive strength of CFA-FA geopolymer decreased and further decreased when using 25% FA. When using the TFA, a proportion of 15 and 20% TFA in 229 230 the geopolymer resulted in similar or higher compressive strengths as the CFA and CFA-FA geopolymers with up to 15% FA. This difference between the FA and TFA geopolymers can be 231 explained by that the FA contains considerable amounts of soluble salts and less aluminum-silicate 232 resources while the Si reactivity for the TFA increased and the reactive Al decreased (Table 233 3). Furtheremore, the reactive Al concentration of TFA decreased and it was observed that fewer 234 bubbles emerged during the preparation of geopolymers with TFA than with FA, which could be 235 due to that metallic aluminum of FA decreased after ED pre-treatment. Metallic aluminum in 236 FA is not normally measured, due to difficulties in measurements. However, Aubert et al. (2004) 237 238 reported the FA has about 0.2% metallic aluminum, and metallic aluminum is detrimental in geopolymer preparation and metallic aluminum can result in more pores in the geopolymers due 239 to H₂ released (Tian et al., 2020). Therefore, the soluble salts, low content in aluminum-silicate 240 241 and metallic Al contributed to the difference of CFA-FA and CFA-TFA geopolymers. The

CFA20TFA was the optimal for preparing geopolymers, reaching 15.3 MPa for curing 28 daysand similar as CFA geopolymer.

244 3.4 Characterization of geopolymers

The XRD diffractograms of the geopolymers are shown in Fig. 6. The low crystallinity (5-245 6 %) in all geopolymer samples indicate that the geopolymer reaction has occurred. In GP-CFA, 246 247 the peak of mullite decreased and the peak of zeolite (Na₈(AlSiO₄)₆CO₃) appeared, along with the decrease of geopolymer crystallinity compared to the diffractogram of the CFA itself (Fig. 5). The 248 zeolite was considered as resulting from the geopolymer process and can co-exist with geopolymer 249 250 products (Rozek et al., 2019). However, the mineral phases of CFA20FA geopolymer were complex and composed of zeolite, mullite, thenardite, quartz, calcium silicate hydrate, thaumasite, 251 hematite, halite and dellaite, which can be explained by the NaOH reacting with FA, that consists 252 253 of considerable impurities. In contrast, the diffraction peak of thenardite (Na₂SO₄) for CFA20TFA 254 geopolymer increased without other sulfate-bearing phases, and TFA contained considerable 255 amounts of gypsum (CaSO₄), thus this phenomenon indicates more calcium (Ca) involved in the geopolymerization. The original zeolite (Na₈(AlSiO₄)₆CO₃) derived from CFA geopolymer 256 disappeared in CFA20TFA and a new zeolite (Na₈(Al₆Si₆O₂₄)S₂O₃ • 3H₂O) was formed, which 257 means that TFA can inhibit the form of original zeolite and more amorphous geopolymeric gels 258 are formed. 259

Fig. 7 shows the FTIR spectra of raw materials (CFA, FA and TFA) and geopolymers (GP-CFA, CFA20FA and CFA20TFA). In Fig. 6, one broad peak at 1096 cm⁻¹ of CFA FTIR spectrum was ascribed to stretching vibrations of T-O (T = Al or Si) bonds (Fernández-Jiménez and Palomo, 2005). The bands at 1412 cm⁻¹ and 875 cm⁻¹ of FA were attributed to symmetric stretching vibrations of O-C-O bond indicating the existence of carbonates (CO₃²⁻) (Jin et al., 2016). The

band frequencies at 1730 cm⁻¹ and 3606 cm⁻¹ in TFA were assigned to asymmetric O-H stretching 265 vibration. An intense and broad band of S-O at 1110 cm⁻¹ with a minor band at 659 cm⁻¹ due to 266 SO₄²⁻ (Lanzon and Garcia-Ruiz, 2012) were also observed in the TFA infrared spectra, which 267 shows the existence of gypsum, consistent with the results of XRD. The bond frequencies of T-O 268 in geopolymers exhibited a bathochromic shift (lower wavenumbers) of about 136 cm⁻¹ from 1096 269 to 960 cm⁻¹ compared to the raw materials, revealing that the vitreous component of raw materials 270 was reacting with the alkali activator and formed the alkaline aluminosilicate gel (Fernández-271 Jiménez and Palomo, 2005). Furthermore, the characteristic asymmetric stretching vibration of Al-272 O and Si-O for reported geopolymers (Jin et al., 2016; Gunasekara et al., 2019) occurred at the 273 peaks in the range of 900 cm⁻¹ and 1000 cm⁻¹, which is also consistent with what was seen in this 274 study. However, the stretching band (962 cm⁻¹) of Si-O-T (T = Al or Si) was affected by the 275 276 addition of MSWI fly ash in the geopolymer. With the addition of FA and TFA, it shifted from 962 cm⁻¹ to 960 cm⁻¹ (CFA20FA) and 967 cm⁻¹ (CFA20TFA). The blue-shift (higher wavenumbers) 277 of the stretching bond of Si-O-Al (Si) in CFA20TFA compared with GP-CFA indicates that the 278 heavy metals influenced the molecular structure of geopolymers and incorporated in the gels 279 (Huang et al., 2018). 280

To further explore the geopolymeric mechanism of geopolymers and the difference on the raw and treated fly ash geopolymers, SEM pictures of the raw materials and geopolymers (cured for 28 d) are presented in Fig. 8. The microstructure of FA consisted of massive flocculus, while the TFA demonstrated large amounts of crystal minerals and this is inagreement with the high crystallinity result of the XRD. The micro-morphology of CFA was made up of smooth round particles and some impurities (like unburn carbon). By contrast, there were some rough round particle in GP-CFA geopolymer, which was reacted by the NaOH to form geopolymer gels (Jin et al., 2016). Note that the reacted round particle of CFA20FA had severe reaction with smooth
surface and its geopolymer gel was layered with micro particles around the geopolymer gel.
Compared with CFA20FA geopolymer, the geopolymer gel of CFA20TFA tightly stuck on the
reacted rough round particles (Jiang et al., 2020), and the microstructure was denser than
CFA20FA, which further indicate why the compressive strength of CFA20TFA geopolymer was
higher.

294

295 3.5 pH-desorption of heavy metals in FA and geopolymers

The leaching from the geopolymers is shown in Table 4. The metal leaching from all samples 296 was generally low and not detected ($\leq 20\mu g/l$) and no clear trends were seen in metal leaching with 297 increased fly ash proportion in the geopolymers. The pH in leaching solutions for geopolymers 298 was highly alkaline and higher than the pH in the leaching solution of the FA and TFA. The 299 leaching pH decreased in the 28 days cured samples compared to 7 days, which indicates that the 300 301 geopolymerisation process continued and the aluminum-silicate, consumed OH⁻ ions to form geopolymeric gels in the form of Si[OH]₄-/Al[OH]₄-. The final pH of leaching solution for CFA-302 TFA geopolymers was slightly lower for all samples except for the sample CFA25TFA cured for 303 304 28 days, where pH was 12.4. This indicates that the lower pH of the TFA also influenced the pH of the geopolymers. The Cd, Ni, Zn, Cu, Mn and Cr leaching concentrations for CFA-25FA 305 geopolymers increased over time, which indicate that the highest FA addition reduced the 306 stabilizing capacity of heavy metals in the geopolymers. Contrarily, the Cd, Ni and Mn leaching 307 behaviours of CFA-TFA geopolymers showed reduced leaching from 7 to 28 days, indicating that 308 the ED pre-treatment for FA can enhance the stabilizing capacity of heavy metals for the 309 geopolymers. The Cl leaching concentration of CFA-TFA geopolymers was lower than that of 310

311 CFA-FA geopolymers. Based on the leaching results, the proportion of 20% TFA for geopolymers seems optimal. The leaching concentration of Pb, Zn, Cu and Mn for CFA20TFA was lower than 312 that of CFA20FA, and all heavy metals leaching concentration met the heavy metals control 313 standard of GB 8978-1996. The leaching concentrations of Pb, Zn, Cr, Cu, Ni and Mn in 314 CFA20TFA were below the detection limit with Mn and Cd below the control limit (GB8978-315 1996). The leaching of heavy metals for geopolymers would mainly result from the FA and TFA, 316 rather than the CFA. Therefore, the mobility index (Kirkelund et al., 2020) of the geopolymers 317 was calculated in Table 5. If mobility index is below 1, indicating an immobilization of heavy 318 319 metals in geopolymers, and above 1 a mobilization. The mobility index of Cr, Zn and Pb for CFA-FA geopolymers were below 1 at curing 28 days. In comparison with that, the mobility index of 320 Mn, Cd, Zn, Pb, Ni and Cu for CFA-TFA geopolymers were below 1. Therefore, more heavy 321 metals can be incorporated in CFA-TFA geopolymers. Note that the mobility index of Cr, Cd, Mn, 322 Zn, Pb, Ni and Cu for CFA20TFA geopolymer was the lowest among geopolymers and reduced 323 along with curing times, indicating that its stabilizing capacity of heavy metals was strong and 324 stable. 325

Fig. 9 shows the leaching behavior of heavy metals for FA, CFA20FA and CFA20TFA 326 geopolymers under alkaline and acidic conditions. From Fig. 8a, it can be seen that the Pb, Zn and 327 Cu showed a similar V-shaped desorption trend. The leaching behaviors of Pb and Zn for various 328 pHs were due to their amphoteric properties, while that of Cu was due to the form of CuO_2^{2-} 329 /[Cu(OH₄)]²⁻ at alkaline pH. Cd, Mn and Ni mainly leached at acidic pH. In contrast, the Cr 330 leaching behavior had the least fluctuation range, especially between the pH of 0.5 and 14. The 331 metal leaching behavior from the geopolymers was different than the FA. The leaching 332 333 concentration of heavy metals for CFA20FA geopolymer (Fig. 9 b) showed a plateau with limited 334 leaching between pH 1 and 7 and demonstrated a U-shape trend, especially for Zn, Pb and Cu. This indicates a strong stabilizing capacity of heavy metals in the acidic environment for the 335 geopolymer, than for the FA, which started leaching of metals at pH 2. This is probably due to a 336 higher acid-base buffering capacity in the geopolymers. The leaching behaviors of heavy metals 337 in CFA20FA geopolymer was similar to that of FA, but the leaching concentrations of heavy 338 metals in CFA20FA geopolymer were lower than that of FA, as expected, since the total amount 339 of heavy metals was lower in the geopolymers. For CFA20TFA geopolymer (Fig. 9c), the leaching 340 concentration of heavy metals is lower than for CFA20FA (Fig. 9b) and especially at the strong 341 342 acidic environment, which suggests that the durability in an extreme acidic environment for CFA20TFA is stronger than for CFA20FA, and this can be explained by the heavy metals could 343 be incorporated in the geopolymeric gels as the blue-shift of the stretching bond of Si-O-Al(Si) 344 (Fig. 7). 345

346 3.6 Economic analysis

In this work, FA was pretreated by ED system and put in CFA geopolymer. In order to further apply this method, the cost analysis of this method was assessed. The costs of human resource, labor, materials transportion and equipment depreciation were not taken into consideration, only for energy consumption and chemical reagents cost. The energy consumption can be calculated as (Oliveira et al., 2019):

$$E = \int V \times I dt$$

where E is the energy consumption (Wh), V is the voltage variation between electrodes (V), I is the current (A) and t is duration (h). Therefore, the energy consumption of ED system was 1.41 KWh for 1 kg MSWI fly ash. Based on the average price in Chinese market, the commercial price of NaOH is \$ 257.1/t and the electricity cost is \$ 0.07/KWh. According to above results, 200 kg TFA and 800 kg CFA needs 160 kg NaOH, thus the cost of producing geopolymers needs \$ 52.5/t.
But, the more energy consumption was due to the stirrer in ash suspension (Oliveira et al., 2019).
Due to the first time to use this method and lacking of pilot experiment, the energy consumption of stirrer need to further consider. Therefore, the reduction of energy consumption for stirrer and duration should be further researched, like reducing duration time and using pulse stirrer next.

362 4. Conclusion

The removal rates of Cd, Cu, Mn and Zn for TFA were higher than that of Cr, Ni and Pb. ED 363 remediation alone for FA increased the leaching concentrations of Cd, Cu, Mn, Ni and Zn, even 364 though the total heavy metal concentration was reduced. The leaching concentrations of heavy 365 metals from the FA were low, except the Pb and Zn. The ED pre-treatment for FA improves the Si 366 reactivity, not just the Si total content, along with an increase in the Si/Al ratio. The compressive 367 strength of CFA20TFA was higher than that of CFA20FA geopolymer and reached 15.3 MPa for 368 369 curing 28 days, which revealed that the ED pre-treatment for FA could upgrade its quality as precursor for geopolymer and increase its recycling potential. The leaching concentrations of Pb, 370 Zn, Cr, Cu, Ni and Mn in CFA20TFA were below the detection limit with Mn and Cd below the 371 372 control limit. The original zeolite (Na₈(AlSiO₄)₆CO₃) derived from CFA geopolymer disappeared in CFA20TFA, meaning that TFA can inhibit the formation of original zeolite and form more 373 amorphous geopolymeric gels. The blue-shift (higher wavenumbers) of the stretching bond of Si-374 O-Al (Si) in CFA20TFA compared with GP-CFA indicates that the heavy metals had influenced 375 the molecular structure of geopolymers and incorporated in the gels. Compared with CFA20FA 376 geopolymer, the geopolymer gel SEM of CFA20TFA tightly sticked on the round particles, and 377 the microstructure was denser than CFA20FA. The durability against extreme environment of 378 CFA20TFA was stronger than CFA20FA and physical encapsulation of geopolymeric gels 379

contributed to the heavy metal immobilization. The electrodialytic remediation for MSWI fly ashincreased by 33.3% in the recycling amount in coal fly ash geopolymers.

382

383 Acknowledgements

Xinyuan Zhan gratefully thanks the China Scholarship Council for the grant (CSC201906050008). Laboratory technicians Ebba Cederberg Schnell, Natasja Dueholm and PhD student Benjamin Alexander Regaard Ebert, Department of Civil Engineering, Technical University of Denmark are thanked for experimental and analytical help. Aase og Einar Danielsens Fond is acknowledged for funding the experimental work.

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solid at 0.37 mL/g (Coal fly ash, C	FA; MS WI IIY ash, FA; II	eated wis willing ash,	1гА).
Geopolymers	CFA (wt%)	FA(wt%)	TFA(wt%)
GP-CFA	100	0	0
CFA5FA	95	5	0
CFA15FA	85	15	0
CFA20FA	80	20	0
CFA25FA	75	25	0
CFA15TFA	85	0	15
CFA20TFA	80	0	20
CFA25TFA	75	0	25

Table 1 The test mixtures for MSWI fly ash and coal fly ash geopolymers with the ratio of NaOH (8M) to solid at 0.37 mL/g (Coal fly ash, CFA; MSWI fly ash, FA; Treated MSWI fly ash, TFA).

499

Chemical composition (wt%)												
Samp le	SiO ₂	Al_2O_3	CaO	Fe ₂ O ₃	Na ₂ O	K ₂ O	TiO ₂	SO_3	Cl			
CFA	34.0	21.5	4.34	14.7	0.67	1.20	2.84	1.40	0.02			
FA	5.78	2.27	22.0	1.10	9.17	12.2	0.78	21.0	8.5			
TFA	16.9	2.64	29.0	2.00	1.48	0.57	2.00	29.7	0.1			
	Heavy metals (mg/kg)											
Samp le	Cd	Cr	Cu	Mn	Ni	Pb	Zn					
CFA	<2	15.2±2.0	18.3±1 .3	125±2. 0	6.1±1.1	40.4±2.0	8.5±1.1					
FA	279±6.0	125±2.0	1080± 19	472±8. 9	35.3±1.2	6080±25 8	34460±15 20					
TFA	9.5±0.3	96±3.0	330±4. 0	163±5. 4	22.1±1.0	6900±26 5	6710±439					
		L	eaching c	oncentrati	ons of heav	y metals (mg	g/L)					
Samp le	Cd	Cr	Cu	Mn	Ni	Pb	Zn	Cl-	Final pH			
CFA	< 0.02	$0.04{\pm}0.0$ 02	< 0.02	< 0.02	< 0.02	$0.02{\pm}0.0$ 03	< 0.02	4.0±0.1 5	13.0±0. 02			
FA	< 0.02	0.57±0.0 2	< 0.02	< 0.02	< 0.02	3.9±0.51	1.8±0.13	7310±1 25	12.0±0. 01			
TFA	$0.35{\pm}0.0$ 03	$0.05{\pm}0.0$ 1	6.7±0. 06	1.2±0. 08	0.13±0.0 06	1.1±0.02	54±0.62	41±3.4	4.7±0.0 1			
LVa	0.1	1.5	5.0	2.0	1.0	1.0	2.0	-	-			

501 Table 2 Physicochemical properties of coal fly ash (CFA), MSWI fly ash (FA) and treated FA (TFA).

^a Limit value in GB8978-1996: Integrated wastewater discharge standard.

Table 3 The reactivity of Al and Si in raw materials for geopolymers under different temperatures (Coal flyash, CFA; MSWI fly ash, FA; treated MSWI fly ash, TFA).

elements	Temperature (°C)	CFA	FA	TFA				
$\Lambda 1 (m \alpha/L)$	25	62.4±1.7	180.7±2	69.0±1.12				
AI (IIIg/L)	80	694±12.3	320±1.6	138±2.1				
\mathbf{S} : $(\mathbf{m}_{\mathbf{z}}/\mathbf{I})$	25	116±3.9	296±1.3	1050±11.5				
SI(mg/L)	80	2500±8.6	738±8.1	1300±5.3				
Si/Al monal natio	25	1.79	1.58	14.7				
SI/AI morai fatio	80	3.47	2.22	9.08				

Curing	Leach	GP-	CFA5F	CFA15F	CFA20F	CFA25F	CFA15	CFA20	CFA25T
time	ing	CFA	А	А	А	А	TFA	TFA	FA
	Cr	*	*	0.10±0. 002	*	*	0.093±0 .002	*	*
	Mn	*	*	0.04±0. 002	0.035±0 .002	*	$0.04{\pm}0.007$	0.031±0 .001	*
	Cd	*	*	*	0.03±0. 001	*	*	0.042±0 .002	0.034±0. 0025
	Zn	*	0.023±0 .003	0.025±0 .004	*	*	*	*	*
/ days	Pb	*	*	*	*	*	*	*	*
	Ni	*	*	0.432±0 .013	*	*	$0.46{\pm}0.08$	*	*
	Cu	*	*	*	*	*	*	*	*
	Cl-	7.9±0.3	184±15	849±11	1020±1 8	1500±3 1	8.1±0.0 3	9.5±0.2 1	8.8±0.12
	pН	12.6±0. 05	12.5±0. 03	12.4±0. 02	12.1±0. 02	12.2±0. 04	11.9±0. 04	11.8±0. 03	11.9±0.0 2
	Cr	*	0.026±0 .002	*	*	*	*	*	0.08±0.0 06
	Mn	0.023±0 .002	0.026±0 .002	0.04±0. 002	0.025±0 .005	0.029±0 .001	0.031±0 .001	0.023±0 .001	*
	Cd	*	*	0.025±0 .001	*	0.035±0 .001	0.03±0. 002	0.027±0 .002	0.043±0. 001
20.1	Zn	*	0.028±0 .003	0.022±0 .002	0.022±0 .004	*	*	*	0.024±0. 002
28 days	Pb	*	*	*	*	*	*	*	0.097±0. 01
	Ni	*	*	*	*	*	*	*	*
	Cu	*	*	*	*	*	*	*	*
	Cl	6.2±1.7	173±10	850±16. 5	1070±0. 5	1330±2 2	6.7±0.3	8.2±0.1 1	8.2±0.06
	pН	12.2±0. 04	12.2±0. 03	12.1±0. 01	12.1±0. 05	12.0 ± 0.03	11.7±0. 02	11.8±0. 01	12.4±0.0 4

Table 4 The leaching test (mg/L) of coal fly ash geopolymers with raw and pretreated MSWI fly ash at 7 and
 28 days (FA, MSWI fly ash; TFA, treated MSWI fly ash; the middle number is the proportion of fly ash)

510 * represents the value was below 0.02 mg/L.

-	5		5	0 1 7		0	2	
Curing	Leachin	CFA5F	CFA15F	CFA20F	CFA25F	CFA15TF	CFA20TF	CFA25TF
time	g	А	А	А	А	А	А	А
	Cr	0	1.17	0	0	11.7	0	0
	Mn	*	*	*	*	0.22	0.13	0
	Cd	*	*	*	*	0	0.60	0.39
7 days	Zn	0.26	0.09	0	0	0	0	0
	Pb	0	0	0	0	0	0	0
	Ni	*	*	*	*	23.6	0	0
	Cu	*	*	*	*	0	0	0
	Cr	0.91	0	0	0	0	0	6.04
	Mn	*	*	*	*	0.17	0.10	0
	Cd	*	*	*	*	0.57	0.38	0.49
28 days	Zn	0.32	0.08	0.06	0	0	0	0.002
	Pb	0	0	0	0	0	0	0.36
	Ni	*	*	*	*	0	0	0
	Cu	*	*	*	*	0	0	0

Table 5 Mobility index of heavy metal in geopolymers at curing 7 and 28 days.

⁵¹³ * represents that the leaching concentrations of raw materials were below the detect limit and this

514 calculation was meaningless.



517 Fig. 1. The schematic figure of three-compartment electrodialytic remediation set-up (chamber I, anode chamber; AN, anion exchange membrane; chamber II, suspension chamber; CAT, cation
520 exchange membrane; chamber III, cathode chamber).





Fig. 2. The process parameters during electrodialytic pre-treatment for MSWI fly ash.





Fig. 3. The distribution of heavy metals in electrodialytic setup after electrodialytic remediation
 experiment.

 $\label{eq:Halite(NaCl, PDF#05-0628)} \begin{array}{l} Q=Quartz(SiO_2, PDF#46-1045) \ I=Hematite(Fe_2O_3, PDF#33-0664) \\ C=Calcite(CaCO_3, PDF#05-0586) \ A=Anhydrite(CaSO_4, PDF#37-1496) \\ B=Bassanite(CaSO_4 \cdot 0.5H_2O, PDF#41-0224) \ M=Mullite(Al_6Si_2O_{13}, PDF#15-0776) \\ \end{array}$



Fig. 4. XRD graphics of coal fly ash, raw and pretreated MSWI fly ash (MSWI fly ash, FA; Treated MSWI fly ash, TFA; coal fly ash, CFA).



Fig. 5. The compressive strength of coal fly ash geopolymers with different initial and pretreated MSWI fly
 ash at 7 and 28 days.

$$\begin{split} & \mathsf{M}=\mathsf{Mullite}(\mathsf{Al}_{6}\mathsf{Si}_{2}\mathsf{O}_{13},\mathsf{PDF\#15-0776}) \quad \mathsf{Q}=\mathsf{Quartz}(\mathsf{SiO}_{2},\mathsf{PDF\#46-1045}) \; \mathsf{C}=\mathsf{Calcite}(\mathsf{CaCO}_{3},\mathsf{PDF\#05-0586}) \\ & \mathsf{H}=\mathsf{Hematite}(\mathsf{Fe}_{2}\mathsf{O}_{3},\mathsf{PDF\#33-0664}) \; \mathsf{F}=\mathsf{Zeolite}(\mathsf{Na}_{8}(\mathsf{AlSiO}_{4})_{6}\mathsf{CO}_{3},\mathsf{PDF\#42-0214}) \; \mathsf{A}=\mathsf{Anhydrite}(\mathsf{CaSO}_{4},\mathsf{PDF\#37-1496}) \\ & \mathsf{G}=\mathsf{Gonnardite}((\mathsf{Ca},\mathsf{Na})_{2}(\mathsf{Si},\mathsf{Al})_{5}\mathsf{O}_{10}\cdot\mathsf{3H}_{2}\mathsf{O},\mathsf{PDF\#45-1324}) \; \mathsf{D}=\mathsf{Dellaite}(\mathsf{Ca}_{6}(\mathsf{SiO}_{4})(\mathsf{Si}_{2}\mathsf{O}_{7})(\mathsf{OH})_{2},\mathsf{PDF\#29-0376}) \\ & \mathsf{B}=\mathsf{Thaumasite}(\mathsf{Ca}_{3}\mathsf{Si}(\mathsf{SO}_{4})_{2}(\mathsf{OH})_{6}\cdot\mathsf{9H}_{2}\mathsf{O},\mathsf{PDF\#44-1423}) \; \mathsf{E}=\mathsf{Calcium}\;\mathsf{Silicate}\; \mathsf{Hydrate}(\mathsf{Ca}_{2}\mathsf{SiO}_{4}\cdot\mathsf{H}_{2}\mathsf{O},\mathsf{PDF\#29-0373}) \\ & \mathsf{T}=\mathsf{Thenardite}(\mathsf{Na}_{2}\mathsf{SO}_{4},\mathsf{PDF\#37-1465}) \; \; \mathsf{Z}=\mathsf{Zeolite}(\mathsf{Na}_{8}(\mathsf{Al}_{6}\mathsf{Si}_{6}\mathsf{O}_{2})\mathsf{S}_{2}\mathsf{O}_{3}\cdot\mathsf{3H}_{2}\mathsf{O},\mathsf{PDF\#38-0514}) \end{split}$$



Fig. 6. XRD graphics of raw materials and geopolymers (MSWI fly ash, FA; coal fly ash, CFA; treated MSWI
fly ash, TFA; the middle number was the proportion of ash).



548 Fig. 7. FTIR graphics of raw materials and geopolymers (MSWI fly ash, FA; coal fly ash, CFA; treated MSWI
549 fly ash, TFA; the middle number was the proportion of ash).



fly ash, TFA; the middle number was the proportion of ash).



558 Fig. 9. The plot of pH-desorption for metals in (a) MSWI fly ash, (b) CFA20FA and (c) CFA20TFA .