A review of the mechanical and leaching performance of stabilised/solidified contaminated soils

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Abstract

Stabilisation/solidification (S/S) technology, which basically involves chemical fixation and immobilisation of contaminants, mainly metals, in the matrix of cementitious binders, is widely used for treatment of contaminated soils. This paper presents a critical review of the performance of commonly used blended binder systems in S/S technology. The binders considered are Portland cement, and cement-fly ash, cement-slag, lime-slag and lime-fly ash blends. This work compares and evaluates the performance of contaminated soils treated by the binders in terms of commonly used mechanical and leaching properties, including unconfined compressive strength (UCS), bulk density, hydraulic conductivity and leachability. The long-term performance of S/S treated soils is also reviewed. It was observed that the inclusion of slag in a binder blend gave superior performance compared to fly-ash. Generally, the leachability of common contaminants in soil can be reduced to acceptable levels with about 20 - 35% dosage of the different binders. The UCS was observed to be optimum around the optimum water content for compaction. The hydraulic conductivity generally fluctuated around 10^{-9} m/s over time. Long-term performance of treated soils showed consistent effectiveness over a period of 5 - 14 years with the occurrence of fluctuations in mechanical and leaching behaviour owing to the complex nature and variability of S/S treated soils.

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24	The UCS was observed to be optimum around the optimum water content for compaction. The
25	hydraulic conductivity generally fluctuated around 10 ⁻⁹ m/s over time. Long-term performance of
26	treated soils showed consistent effectiveness over a period of 5 - 14 years with the occurrence of
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28	S/S treated soils.

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30 Keywords: cement, compressive strength, fly ash, leachability, hydraulic conductivity, slag.

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32 1. Introduction

Stabilisation/solidification (S/S), which usually employs the addition of cementitious binders to contaminated soils in order to immobilise the contaminants present, has emerged as a cost effective and efficient remedial measure for contaminated soils (<u>Al-Tabbaa and Perera 2005a</u>). S/S treatment entails chemical fixation and physical encapsulation of contaminants. The process is aimed at minimising the rate of contaminant migration into the environment or reducing the toxicity. Contaminant migration is restricted by vastly decreasing the surface area exposed to leaching and/or by isolating the wastes/soils within an impervious capsule.

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The combined process of stabilisation and solidification usually results in increasing the strength, and decreasing the leachability, compressibility and hydraulic conductivity of the treated material. However, decrease in leachability is the most important factor, since from an environmental point of view; S/S does not make sense when there is no decrease in leachability (Kogbara et al. 2011). S/S is most suitable for the immobilisation of metals, and to a lesser extent for organic contaminants because of the detrimental effects on the hydration and structural formation of the materials (Young 1972). Due to the high pH of cement, the metals are retained
in the form of insoluble hydroxide or carbonate salts within the hardened structure. Details on
terminology, history, design criteria, binders and contaminant stabilisation mechanisms can be
found elsewhere (Wiles 1987; Conner 1990; Glasser 1997; Conner 1998; Conner and Hoeffner
1998; LaGrega et al. 2001; Bone et al. 2004; Shi and Spence 2004; Spence and Shi 2005; Paria
and Yuet 2006; Du et al. 2010).

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Although there are many reviews on S/S technology, very few have considered in depth, or 54 brought together, the mechanical and leaching performance of contaminated soils treated by 55 different cementitious binders from different studies. Hence, this paper seeks to fill that gap in 56 the literature by providing a critical review of the mechanical and leaching performance of 57 commonly used blended binder systems in S/S technology. Moreover, this work combines 58 information on the key factors that influence S/S treatment of contaminated soils, which are 59 ordinarily the subjects of entire books. This would be invaluable to remediation experts and 60 environmental professionals as it would help in making informed decisions on the application of 61 one binder or another. 62

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64 2. Overview of S/S binders and stabilisation mechanisms

S/S binders can be divided into two groups, primary stabilising agents and secondary stabilising agents. Primary stabilising agents are those stabilising agents that can be used alone to bring about the stabilising action required. Portland cement (CEMI) and lime are the most common. While secondary stabilising agents includes pozzolanic materials (i.e. materials that react with lime or cement in the presence of water to produce a cementitious compound) like pulverized 70 fuel ash (PFA) also known as fly-ash and ground granulated blast furnace slag (GGBS) (LaGrega et al. 2001), that are not very effective on their own but can be usefully used in conjunction with 71 lime or cement (Bone et al. 2004). The above mentioned binders are the most commonly used 72 although there are several other binder materials for S/S works including natural bentonite clays, 73 organophilic clays, bitumen, cement kiln dust, silica fume and some proprietary binders like 74 Geodur, EnvirOceM, etc. Details on the basic principles of S/S binders, research and applications 75 have been reviewed in state of practice reports (Al-Tabbaa and Perera 2005a, b; Al-Tabbaa and 76 Perera 2005c). 77

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Generally, binders are mixed with wastes or soils containing contaminants with the aim of 79 stabilising and/or solidifying the contaminants by way of immobilising them within the binders. 80 Many contaminated soils are characterised by the concomitant presence of organic and inorganic 81 contaminants. Immobilisation of inorganic contaminants in soils involves both stabilisation and 82 solidification, while that of organics mainly involves solidification only, as chemical bond(s) 83 may not be formed (Wiles 1987). The following mechanisms have been identified as fixation 84 mechanisms involved in the interaction of inorganic contaminants with soils and/or binders. 85 They are: pH-dependent precipitation, redox-controlled precipitation of insoluble compounds, 86 sorption potential and incorporation into crystalline components of the cement matrix (Bone et 87 al. 2004). On the other hand, although organic contaminants are not essentially stabilised, 88 cement-based systems operating at ambient temperatures and pressures in aqueous environments 89 are involved in a few organic reactions. These include: hydrolysis, oxidation, reduction and the 90 formation of organic salts (Conner 1990). Further, organic matter such as humus can retard the 91

- hydration of cement due to the action of fulvic and carbonic acids. This can also have a negative
 influence on characteristics of the cement matrix (<u>de Korte and Brouwers 2009a</u>).
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As mentioned earlier, CEMI, PFA, GGBS and lime are the most commonly used binders, either 95 singly or in blended binder systems in the literature. Hence, this paper will focus on studies that 96 have deployed binder blends involving the above-named binder materials for treatment of 97 contaminated soils. It appears that the most common combinations of the above materials in the 98 literature are blends of CEMI-PFA, CEMI-GGBS, lime-PFA and lime-GGBS. CEMI is normally 99 used alone and it is the most commonly used binder for S/S of contaminated soils and have been 100 applied to a greater variety of wastes than any other binder has. Therefore, the section(s) on 101 performance of S/S treated soils will deal with the deployment of the different blended binder 102 systems mentioned above in previous S/S works. 103

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105 **3.** Test methods for S/S treated soils

S/S treatment of a contaminated soil is usually designed to satisfy some criteria, which are 106 mainly leachability and strength, depending on the end use of the treated material. In addition to 107 leachability and strength, a range of other properties that could be specified depending on the end 108 use include hydraulic conductivity, bulk density, porosity, compaction, freeze-thaw durability, 109 compressibility, California bearing ratio (CBR), Moisture Condition Value (MCV), etc. Clearly, 110 the leachability of the stabilised/solidified (S/S) soil is the most important design parameter. Two 111 leaching tests in common use are the batch leaching test, BS EN 12457 (BSI 2002), and the tank 112 leaching test, NEN 7375 (Environment Agency 2004). The batch leaching is considered a worst-113 114 case scenario since the material is crushed prior to testing hence maximising the leaching potential of contaminants, while the tank-leaching test assesses the leaching potential due to diffusion processes which is likely to be a more realistic scenario in practice. The acid and base neutralisation capacity (ANC/BNC), DD CEN/TS15364 (BSI 2006), and analysis of contaminants in the leachate to assess their availability at pH values of interest is sometimes used.

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Hydraulic conductivity, sometimes used interchangeably with permeability, indicates the rate at 121 which water can flow through a material, which is a key variable in environmental behaviour. 122 S/S materials often rely on a reduction of the ingress and egress of water in and out of a 123 monolithic mass of material to reduce leaching potential. Determining the likely permeability of 124 the treated material is therefore especially important in regards to determining the potential for 125 126 the transport of leachate bearing contaminants to move through the treated material into underlying strata and eventually into groundwater. Thus, hydraulic conductivity is closely related 127 to leachability. The UCS is used as a measure of the ability of a monolithic S/S material to resist 128 mechanical stresses. It relates to the progress of hydration reactions in the product, and durability 129 of a monolithic S/S material, and is therefore a key variable. Bulk density is the mass per unit 130 volume of the material. It can be used together with moisture content and specific gravity to 131 calculate S/S material porosity and degree of saturation. Bulk density can also be used together 132 with mass change factor to calculate volume increase due to S/S treatment (Perera et al. 2005b). 133 134 It can also be used to determine the volume of wastes to be treated, shipped off site, or returned to the site (Lin et al. 1996). 135

The properties described above have been reported to be the most commonly used for 137 performance tests. The relevance of other tests, including those above, in the assessment of the 138 effectiveness of S/S processes has been reviewed (Bone et al. 2004; Perera et al. 2005b). Table 1 139 140 summarises some available regulatory limits for the most commonly used performance tests. Leaching thresholds are provided for only five metals, Cd, Cu, Pb, Ni and Zn, which are amongst 141 those commonly found in contaminated soils, alongside total petroleum hydrocarbons (TPH) 142 (Kabata-Pendias and Mukherjee 2007). The above-mentioned metals and TPH are the major 143 contaminants of interest in this work. 144

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Table 1. Regulatory limits for mechanical and leaching behaviour

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Adapted from (Kogbara and Al-Tabbaa 2011)

Performance criteria	UCS	Hydraulic conductivity	Cd	Ni	Zn	Cu	Pb
Environment Canada WTC: Proposed UCS before immersion	440	N/A	N/A	N/A	N/A	N/A	N/A
for controlled utilisation (kPa)							
UK Environment Agency: 28 d	1,000	N/A	N/A	N/A	N/A	N/A	N/A
UCS limit for disposal of S/S							
treated wastes in landfills (kPa)							
UK and USEPA hydraulic	N/A	< 10 ⁻⁹	N/A	N/A	N/A	N/A	N/A
conductivity limit for in-ground							
treatment and landfill disposal,							
respectively (m/s)		0					
Environment Canada WTC:	N/A	$< 10^{-8}$	N/A	N/A	N/A	N/A	N/A
Proposed permeability limit for							
landfill disposal scenarios (m/s)							
Environmental Quality Standard	N/A	N/A	0.0045	0.02	N/A	N/A	7.2
for inland surface waters (mg/l)							
Hazardous waste landfill WAC for	N/A	N/A	5	40	200	100	50
granular leachability (mg/kg)							
Stable non-reactive hazardous	N/A	N/A	1	10	50	50	10
waste in non-hazardous landfill							
WAC (granular leaching) (mg/kg)							
Inert waste landfill WAC for	N/A	N/A	0.04	0.4	4	2	0.5
granular leaching (mg/kg)							

149 **4 Performance parameters of S/S treated soils**

150 **4.1 Overview**

This section reviews the deployment of the different blended binder systems considered in this 151 work in previous S/S works. As mentioned in section 2, the binders considered are CEMI, 152 CEMI-PFA, CEMI-GGBS, lime-PFA and lime-GGBS blends. The performance of soils treated 153 by the cement and lime-based binders is evaluated in terms of selected performance parameters, 154 which include UCS, leachability, hydraulic conductivity and bulk density. Consequently, sub-155 sections 4.2.1 - 4.2.5 deals with all four performance parameters for each of the above-156 mentioned binders. In addition, the variation of these performance parameters in the long-term 157 are also considered. Furthermore, sub-section 4.2.6 provides a comparison of the different 158 binders in terms of the afore-mentioned performance parameters. 159

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161 Generally, majority of previous studies deploying the afore-mentioned cement and lime-based binders focused on UCS and leachability. This is because both performance parameters are 162 necessary for successful stabilisation and solidification. Few studies include hydraulic 163 conductivity and bulk density among the performance parameters used for evaluating S/S treated 164 soils. Hence, the tables on performance parameters in section 4.2 contain more information on 165 UCS and leachability compared to hydraulic conductivity and bulk density. Furthermore, CEMI 166 has been widely used in S/S treatment of contaminated soils and other hazardous wastes than any 167 other binder (Spence and Shi 2005). As sequel, extensive discussion on the performance 168 properties of S/S treated soils is made in the section on CEMI, and reference to such made while 169 discussing the same properties in the sections on the other binders. 170

172 4.2.1 CEMI S/S treated contaminated soils

The details of the soil and binder characteristics, and mix composition and curing age from ten studies, which dealt with S/S treatment of contaminated soil using CEMI, are summarised in Table 2a. Table 2b shows the performance characteristics of the S/S treated soils detailed in Table 2a in terms of the four parameters of interest in this work, namely, UCS, bulk density, hydraulic conductivity and leachability.

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After soil particles and contaminants have been wetted by cement grout, the addition of more 179 binder increases the binding force of the particles; hence, UCS increases with binder dosage 180 (Tables 2a and 2b). It is well known that in CEMI, the formation of calcium silicate hydrates 181 (C-S-H) is principally responsible for strength development, and more C-S-H is formed as the 182 binder dosage increases. Most studies on S/S of contaminated soil normally focus on strength 183 and other performance parameters at a standard curing age of 28 days. However, cement 184 hydration reactions continue beyond the standard curing age. These provide the reasons why 185 UCS increases with increase in binder dosage and curing age (Bone et al. 2004; Paria and Yuet 186 2006). However, although UCS increases with curing age, over a long time as cement hydration 187 approaches completion, the UCS reaches a plateau. This was observed in a study in Table 2b 188 (Al-Tabbaa and Evans 2000; Al-Tabbaa and Boes 2002) where the UCS after 5 years was 189 slightly less than the UCS at 28 months. 190

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Furthermore, the strength level achieved by a stabilised soil depends on the water content of the soil-cement mixture as it is for concrete mixtures. However, there is a dearth of literature on the variation of UCS with water content in CEMI S/S contaminated soil, although there are a few 195 studies on that for uncontaminated soils. It has been reported that the 28-day UCS and other mechanical properties of CEMI-treated contaminated sandy soil was optimum around the 196 optimum moisture content (OMC) for compaction of the S/S treated soil during sample 197 preparation (Kogbara et al. 2010). Moreover, the UCS varies with the soil type, nature, and 198 amounts of contaminants present. A comparison of the soil and binder characteristics and the 199 UCS of three studies in Tables 2a and 2b (Lin et al. 1996; Yilmaz et al. 2003; Kogbara et al. 200 2010), which employed 20% binder dosage illustrates this. The UCS values in the last two 201 studies (Yilmaz et al. 2003; Kogbara et al. 2010) were close at 2.52 and 2.24 MPa, respectively, 202 while that of the other study (Lin et al. 1996) (8.74 MPa) was markedly different. With sands and 203 gravel, the UCS is higher than with silt and clay. This is due to the effect of particle size, which 204 is also visible in concrete mixtures. However, the presence of fresh hydrocarbon pollution leads 205 206 to lower UCS values (Al-Sanad and Ismael 1997) as observed in the third study above (Kogbara et al. 2010) which had more gravel content. All the same, it can be deduced from the above that 207 the 1 MPa UCS criterion in Table 1 can be met with 20% CEMI dosage for different soil types 208 and contamination scenarios. The data in Table 2b even suggests that around 10% CEMI dosage 209 could achieve that. 210

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Contaminated soils generally achieve higher strengths after S/S treatment. Without the binder, the soils will usually have lower strength, as they cannot cope with internal tensile forces. However, contaminants in the soil may interfere with the cement hydration process and lead to a more complicated strength development than in uncontaminated cemented soils. The type of metal, the metal concentration, and the cement content are major factors that affect cement hydration and strength (Chen et al. 2010). The interferences of a few contaminants on cement

- hydration and in turn strength (<u>Trussell and Spence 1994</u>; <u>Tremblay et al. 2002</u>; <u>Bone et al. 2004</u>;
- 219 <u>Paria and Yuet 2006</u>) are summarised as follows:
- 220
- Cd, Cr and Zn have been associated with increased formation of ettringite, which under some circumstances causes expansion and cracking of cement.
- Pb retards cement hydration by precipitating onto the surface of the Ca and Al silicates as
 insoluble Pb sulphates and carbonates forming impermeable coating, hence high
 concentrations may cause a weak S/S product.
- Zn effectively prevents appreciable hydration of cement, possibly because of a chemical,
 rather than physical mechanism.
- Oil and grease and other organic compounds are also known to decrease strength in
 cement mixtures. This is because hydrocarbons tend to coat cement particles, which
 delays their hydration and setting time.
- 231

In contrast to the above, a different trend in the UCS of CEMI S/S contaminated soil has been 232 233 reported (Lin et al. 1996). The 7-day UCS of an oil-spiked soil (4% oil content) containing Pb (see Table 2a) was found to be higher than that of the same soil not spiked with oil. The 234 possibility of the presence of Pb leading to a stronger structure in clay-fly ash mixtures has been 235 236 reported (van Jarsveld and van Deventer 1999). Therefore, it is possible that in the presence of certain concentrations of metals, relatively low levels of hydrocarbon contamination would not 237 238 cause detrimental effects on the UCS. In other words, although Pb and oil individually reduce the 239 UCS, depending on the concentration, together they may cause an increase in UCS.

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Reference	Soil type and composition (including natural pH and other details if stated)	Initial amount of prime contaminants (mg/kg)	Binder dosage (%)	W/S ratio	Curing age (days)
(Lin et al. 1996)	100% sand	Pb: 1,366	13	0.14	7
		TPH: 40,000	16.7		
			20		
			23.1		
(<u>Day et al. 1997</u>)	Relatively dense sand and gravel	Cd: 130	35	0.21	28
	(other details not specified) pH - 7.75		45		
(Al-Tabbaa and	Made-ground	Cd: 8.7	9.3	0.05	Testing at
Evans 2000)*	consisting mainly of clayey sand and sandy clay	Cu: 1,264	8.5% CEMI,		day:
and		Pb: 2,801	0.8% bentonite		
(Al-Tabbaa and		Ni: 105			56
<u>Boes 2002</u>)*		Zn: 1,589			
		Coal tar: 1,400			784 (2.3 yrs)
		Mineral oil: 566			
		Toluene extract: 1,700			1826 (5 yrs)
(Sanchez et al. 2000)	Sandy loam	Pb - 49,935	30	0.22	28
	68% sand, 26% silt, 6% clay		33	0.29	
(Sanchez et al. 2002)	Purely Sand	As: 3,050	40	0.13	28
		Cd: 3,100			
		Cu: 2,920			
		Pb: 2,700			
		Zn: 3,220			
(<u>Yilmaz et al. 2003</u>)	Silt,	Cd: 970	10	0.15	28
	27% sand, 18% clay, 55% silt	Cu: 3,640	20		
		Pb: 4,380			
		Zn: 3,760			
		Cr: 1,410			

Table 2a. Soil and binder characteristics of CEMI S/S treated contaminated soils

_	Reference	Soil type and composition (including natural pH and other details if stated)	Initial amount of prime contaminants (mg/kg)	Binder dosage (%)	W/S ratio	Curing age (days)
	(Shawabkeh 2005)	Model soil	Cd	50	0.30	14
		50% sand, 50% clay	3,000			
			9,000			
	() (1 2010)		18,000	5 20	0.50	
	(<u>Moon et al. 2010</u>)	55.7% sand, $33.8%$ silt, $10.3%$ clay	Zn 4.072	5-30	0.50	/
		pH - 8.31	4,975	at 5% intervals		28
_	(Voglar and Lestan	Soils from 40 sampling points in Cinkarna	Cd: 146 ± 68	15	0.25	28
	<u>2010</u>)	brownfield, Slovenia	Cu: 1,111 ± 1,997		to 0.45	
		(other details not specified)	Pb: $26,400 \pm 20,140$			
			Ni: 46 ± 16			
			$Zn: 9,979 \pm 11,910$			
	(Kogbara et al.	Clayey silty sandy gravel	Cd: 3467 ± 153	5 - 20	0.13 to	28
	<u>2010</u>)*;	65% gravel, 29% sand, 2.8% sand, 3.2% silt	Cu: $3,167 \pm 231$	at 5% intervals	0.19	84
	(<u>Kogbara 2011</u>)*	Spiked with a mixture of metals and hydrocarbons	Pb: $3,733 \pm 208$			
	and	pH of spiked contaminated soil - 9.83	N1: $3,567 \pm 153$			
	(<u>Kogbara et al.</u>	Organic matter content – 0.22%	$Zn: 4,233 \pm 289$			
_	<u>2012</u>)*	S: water-to-solids ratio TPH: Total netroleum hydroca	$\frac{1PH: 6312 \pm 1482}{\text{*these studies w}}$	vere carried out on the	ame soil over tim	
246	•••		these studies w	vere earlied out on the s	same som över tin	
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 Table 2a (continued). Soil and binder characteristics of CEMI S/S treated contaminated soils

Reference	UCS	Bulk	Hydraulic		Leachal	bility
	(MPa)	density (Mg/m ³)	conductivity (m/s)	Major leaching test(s)	Final leachate pH	Comments on leaching behaviour
(<u>Lin et al. 1996</u>)	4.78 (13% dosage) 7.47 (16.7% dosage) 8.74 (20% dosage) 10.0 (23.1% dosage)	2.11 2.11 2.11 2.11 2.11	2.6 x 10 ⁻⁹	TCLP	Not specified	 All samples passed the TCLP Pb leaching criteria of 5 mg/l. Four (4) wt% TPH had little effect on Pb leachability. TPH leachability not studied.
(<u>Day et al. 1997</u>)	4.70 (35% dosage) 5.20 (45% dosage)	Not determined	Not determined	TCLP	Not specified	0.8 and 8.8 mg/kg of Cd leached in 35 and 45% dosage mixes, respectively.
(<u>Sanchez et al. 2000</u>)	Not determined	Not determined	Not determined	ANC	2 – 13	 Pb leaching as a function of pH exhibited characteristic amphoteric behaviour with solubility minima at pH 9. leachability of treated soils less than that of untreated soil by an order of magnitude within the pH range, 9 – 11. For pH 5 – 8 and >12, Pb solubility was similar in treated and untreated soils.
(<u>Al-Tabbaa and Evans</u> <u>2000</u>) and (<u>Al-Tabbaa and Boes</u> <u>2002</u>)	1.30 (56-d) 3.25 (784-d) 2.97 (1826-d)	Not determined	0.70 x 10 ⁻⁹ 0.15 x 10 ⁻⁹ 0.31 x 10 ⁻⁹	TCLP NRA leaching test*	10.6 (56-d) 7.5 (784-d) 1826-d not determined	 NRA leaching test after 1,826 days showed: 4.9 mg/kg of Cu, <0.05 mg/kg of Zn, 1.2 mg/kg of Ni and 0.16 mg/kg of mineral oil. Cd and Pb were below detection levels. TCLP and data from earlier curing ages not determined.

Table 2b. Performance characteristics of CEMI S/S treated contaminated soils

Reference	UCS	Bulk density	Hydraulic	Leachability		
	(MPa)	(Mg/m^3)	conductivity	Major leaching	Final	Comments on leaching
			(m/s)	test(s)	leachate pH	behaviour
Sanchez et al. 2002)	Not determined	Not	Not	Modified ANC	4 - 12.5	- Metal release was influenced
		determined	determined			by changes in pH and speciation.
				L/S = 5		Pb and As illustrated the impact
						of re-speciation due to
						carbonation.
						- At pH > 11, Cd solubility
						increased with pH, and for pH $<$
						11, it decreased with increasing
						pH.
						- For $pH > 9$, Pb solubility
						increased with increase in pH.
<u>Yilmaz et al. 2003</u>)	1.15 (10% dosage)	Not	Not	TCLP	6.1 – 6.8	- In all cases, there was $> 90\%$
	2.52 (20% dosage)	determined	determined		in TCLP	retention of metals in the
				Batch leaching		solidified mass.
				L/S = 10	8.1 – 9.5	- 10% binder dosage was
					in batch	inadequate to reduce the
					leaching	leaching of Cd to acceptable
						levels.
(<u>Shawabkeh 2005</u>)	11	Not	Not	TCLP	Not	- Amount of Cd leached varied
		determined	determined		specified	with the initial concentration.
						- 240, 700 and 1,300 mg/kg
						were leached in increasing order
						of the initial amount of Cd.

Table 2b (continued). Performance characteristics of CEMI S/S treated contaminated soils

Reference	UCS	Bulk	Hydraulic		Leacha	bility
	(MPa)	density	conductivity	Major leaching	Final	Comments on leaching
		(Mg/m^3)	(m /s)	test(s)	leachate pH	behaviour
(<u>Moon et al. 2010</u>)	Not determined	Not	Not	TCLP	5.5	- No significant difference in
		determined	determined		6.5	leachability of samples cured
					7.1	for 7 and 28-days.
					8.0	- 892 mg/kg Zn was leached out
					9.8	of untreated soil with pH of 4.5.
					10.9	- 440 mg/kg was leached out in
						the 5% dosage mix, while 4
					pН	mg/kg was leached in $5 - 15\%$
					increased	dosage mixes.
					with binder	- No leachable Zn was detected
					dosage	at 25 and 30% dosages.
Voglar and Lestan	2.15	Not	Not	TCLP	Not	- The batch and TCLP leaching
<u>2010</u>)		determined	determined		specified	tests showed that Cd, Pb, N
				Batch leaching		and Zn leachability were
				T 1 .		significantly reduced or below
				Tank test		detection limit.
						- Cu leachability increased after
						S/S treatment in the batch
						leaching test but decreased after
						S/S treatment in the TCLP test.
						- The predominant release
						mechanism in the tank test was
C_{ord} and C_{ord} and C_{ord} and C_{ord} and C_{ord}	$28(84) dav^{\dagger}$	28(84) day	28(84) day	ANC	62 128	Water content showed no
$(K_{ogbara} \simeq 2011)$	0.33(0.4)(5% dosage)	20(04)- $uuy1 79(1 79)$	$9.7(17) \times 10^{-9}$	- And 2 mea/a	0.2 - 12.0	significant effect on leachability
(<u>Rogoara 2011</u>) and	1.68(2.0)(10% dosage)	1.79(1.79) 1.81(1.91)	$9.7(17) \times 10^{-9}$	HNO ₂ addition		- 20% dosage satisfied most
Coghara et al. 2012)	1.00 (2.0) (1070 dosage)	1.01 (1.71)	$1.5(14) \times 10^{-9}$	- Tank leaching		leaching criteria except for Ph
<u>(0g0uru et ur. 2012</u>)	1.83 (15% dosage)	1.8/	4.5 X 10			- predominant leaching
	2.24 (20% dosage)	1.74	3.5×10^{-9}			mechanism: surface wash-off
ANC. Acid neutralisatio	on capacity (BSI 2006) TCLP ⁻	Foxicity characteri	stic leaching procedu	re(IISEPA 1986) ND	Not determined	L/S: Liquid-to-solid ratio

Table 2b (continued). Performance characteristics of CEMI S/S treated contaminated soils

264 Similarly, there was no observable effect on the bulk density of the oil-spiked soil with increase in binder dosage (Lin et al. 1996) (Table 2b), as was the case with the same soil without oil 265 contamination. However, a different trend was observed in a soil with much lower (1%) oil 266 content. Bulk density increased with binder dosage between 5% and 15% dosage (Kogbara et al. 267 2012) (Table 2b). Thus, oil contamination may impede increase in bulk density with increasing 268 binder dosage depending on the oil concentration. Nevertheless, the bulk density of cement-269 stabilised contaminated soils generally increases with increase in binder dosage as the cement 270 grout easily wets the particles and contaminants. Hence, the more the binder is added, the more 271 void spaces are filled, leading to increase in bulk density with increasing binder dosage. 272 Moreover, the bulk density depends on the particle size distribution of the soil, and the specific 273 density of cement, which is higher than that of soil. Therefore, the bulk density of the mixture will 274 275 increase with cement content.

276

The hydraulic conductivity of most S/S treated soils generally fluctuates around 10^{-9} m/s over time 277 (Table 2b). There are not many studies that report hydraulic conductivity results alongside UCS 278 and leachability, since as mentioned earlier, successful S/S treatment is usually assessed by both 279 parameters. Hydraulic conductivity values around 10⁻⁹ m/s are considered sufficient for recycling 280 of the stabilised contaminated soil, for instance, as a sub-base course in road pavement (Lin et al. 281 1996). There seem to be conflicting findings as regards hydraulic conductivity changes over time. 282 In one study, the hydraulic conductivity of CEMI-treated soils increased between 28 and 84 days 283 (Kogbara et al. 2012) (Table 2b). While in another, the hydraulic conductivity of cored made-284 ground samples decreased between 2 and 28 months, and increased between 28 months and 5 years 285 (Al-Tabbaa and Evans 2000; Al-Tabbaa and Boes 2002) (Table 2b). This varied response of the 286

hydraulic conductivity over time has been attributed to a combination of the following factors. The continued hydration of the cementitious constituents causes the hydraulic conductivity to decrease. Further, the long-term interaction between the contaminants and the soil–grout matrix superseding the effect of the continued hydration of the cementitious material, causing an increase in the hydraulic conductivity (Al-Tabbaa and Evans 2000; Al-Tabbaa and Boes 2002). Overall, there appears to be increase in hydraulic conductivity of S/S treated materials over time though not in a manner that is detrimental to recycling them for other uses.

294

295 CEMI has been widely used for the treatment of metals in soils as summarised in Table 2. The studies selected focus on the earlier-mentioned most common metals in soils (Table 2a). The 296 studies on leaching behaviour indicate that the amount of contaminant leached from an S/S 297 treated soil depends on the initial concentration present (Table 2b). There is no generally 298 accepted binder dosage limit for the reduction of average levels of contaminant concentrations 299 found in soils. Different studies used different binder dosages and water contents for S/S 300 treatment, depending on the nature and level of the contamination, and the judgement of the S/S 301 treatment designer. Moreover, different leaching tests are also used in the assessment of leaching 302 behaviour, with the results being scenario-specific. Most of previous studies evaluated 303 leachability using the Toxicity Characteristic Leaching Procedure (TCLP) (USEPA 1986). 304 The test was originally designed to simulate leaching from wastes co-disposed with municipal solid 305 306 wastes in a landfill. However, it has been used for assessment of leaching from contaminated soils, which has little or no relationship with the test's original plan. Especially, as there is 307 increasing inclination towards re-use of stabilised contaminated soil as filler for construction 308 309 purposes rather than disposal to landfill (Shawabkeh 2005; Voglar and Lestan 2010).

310 In spite of the above, it can be deduced from the leaching studies (Table 2) that between 15% to 20% binder dosage would be required to reduce the leachability of average metal concentrations 311 found in soils to acceptable levels. There seems to be no significant effect of water content on the 312 leachability of contaminants within the water content range that allows for workability of the 313 soil-cement mixture (Kogbara et al. 2012). In some cases, depending on the pH attained, the 314 leachability of Cu was higher in treated soils than untreated soils (Table 2b) (Voglar and Lestan 315 2010). Similarly, Pb leachability was found to be the same in both treated and untreated soils at 316 pHs between 5 and 8 and greater than 12 (Sanchez et al. 2000). Similarly, the solubility of Cd 317 and Pb had a minimum around pH 11 and 9, respectively. The leachability of both metals 318 increased with increasing pH beyond the afore-stated pH values (Sanchez et al. 2002) (Table 2b). 319 Hence, it has been suggested that CEMI may not be suitable for soils with high Pb 320 concentrations, depending on the management scenario for the treated contaminated soil. For 321 instance, Pb leachability in S/S treated soil with $\geq 10\%$ CEMI dosage was found to exceed the 322 limit for stable non-reactive hazardous and inert waste landfills (Kogbara et al. 2012). These 323 observations are due to the well-known solubility behaviour of metals as a function of pH 324 (Figure 1). The solubility of these metals decreases with pH up to a value of about 10 or more. 325 Above this pH, the metal solubility increases with pH as the metal cations form complex soluble 326 anions with excess hydroxide anions (Shi and Spence 2004). 327

328



331Figure 1. Cd, Pb, Ni and Zn hydroxide solubility at 25°C in dilute solution332as a function of pH (<u>Stegemann and Zhou 2009</u>)

333

330

It is well known that release of metals from S/S materials is influenced by changes in pH and 334 metal speciation. It has been shown that Cd illustrated the impact of reduced pH without re-335 speciation, resulting in increased release at lower pHs, while Pb illustrated the impact of re-336 speciation due to carbonation, resulting in reduced release as a function of pH (Sanchez et al. 337 338 <u>2002</u>). Thus, stabilisation of Pb within the S/S matrix occurs by re-speciation with cement constituents. Its release during leaching is governed by solubilisation phenomenon at the 339 interface between the matrix and the leaching solution. This solubilisation phenomenon is 340 341 controlled by the release of hydroxides, mainly from calcium hydroxide (Sanchez et al. 2000). Similarly, X-ray powder diffraction (XPRD) results have shown that Zn-substituted ettringite 342 and Zn₆Al₂(OH)₁₆CO₃.4H₂O were possible phases responsible for Zn immobilisation in cement-343 344 treated soils (Moon et al. 2010).

Very few studies have considered the long-term leaching behaviour of cement-treated 346 contaminated soils. Since cement hydration continues after 28 days, there may be changes in 347 release rates of contaminants from the treated material beyond this time and this must be 348 considered when evaluating leaching data (Bone et al. 2004). Furthermore, acidic influences in 349 the environment, for instance, carbonation by CO₂ uptake, and natural leachants like rainwater or 350 landfill leachate with slightly acidic pH, will progressively lower the pH of S/S soils over time 351 leading to release of contaminants. As a result, there are concerns regarding the long-term 352 effectiveness of S/S treatment due to uncertainties in a number of areas like test methods, 353 observed deficiencies in the process application, observed lack of chemical binding in crushed 354 samples of treated wastes, and uncertainties of performance arising from anticipated behavioural 355 degradation of the material over time (Perera et al. 2005a). Consequently, recent studies have 356 also considered combining S/S and biodegradation in order to achieve some form of (organic) 357 contaminant attenuation over time (Kogbara 2013). 358

359

The long-term leaching behaviour of an S/S treated soil was considered in a fairly recent study 360 (Antemir et al. 2010). The study evaluated the field performance of a 4-year old S/S treated 361 contaminated soil at the former Astra military explosives Fireworks site in SE England. A 362 hotspot of metals contamination, containing up to 96,000 mg/kg Cu, 81,000 mg/kg Zn and 750 363 mg/kg Pb was treated with 20 wt% dosage of EnvirOceMTM, a superfine sulphate-resisting 364 Portland cement at a 0.2 - 0.3 water/cement ratio. The results are summarised in Table 3. The 365 pH-dependent leaching of the metals in the untreated and 4-year old S/S soil is shown in Figure 366 2. 367

369 Table 3. Average concentrations of contaminants in untreated and S/S treated soils after remediation and 4 years later



Adapted from (Antemir et al. 2010)

Figure 2. pH-dependent leaching of (a) Cu (b) Pb and (c) Zn in untreated and S/S treated soil (<u>Antemir et al. 2010</u>)

380

379

381 The leachability of the metals remained at low levels after 4 years of S/S treatment. It was observed that the shape of the pH-dependent leaching curves changed dramatically upon S/S 382 treatment, providing a strong indication of different chemical phenomena governing the release 383 of contaminants from the treated material. However, irrespective of this, metal leaching was 384 reduced by one to two orders of magnitude in comparison to the untreated soil, particularly in the 385 alkaline pH range. Metal leaching data in the S/S soil, correlated with the major element 386 concentrations (Ca, Al and Si) in solution, indicating either encapsulation or incorporation in the 387 alumino-silicate hydration phases (Antemir et al. 2010). The observed long-term leaching 388 389 behaviour of the treated soils was corroborated by recent studies on pH-dependent leaching behaviour of contaminants over an 84-day period (Kogbara et al. 2012). Such studies simulate 390 long-term behaviour of S/S treated soils after they are subjected to acidic influences in the 391 environment. Further work on pH-dependent leaching behaviour of S/S treated soils cured for 392 longer periods is necessary to provide more information on the durability of the treated soils. 393

394

395 4.2.2 CEMI-PFA S/S treated contaminated soils

As in the case of CEMI S/S treated soils, the details of the soil and binder characteristics, of 396 some studies, which dealt with contaminated soil treatment using CEMI-PFA, are 397 summarised in Table 4a. Table 4b shows the performance characteristics of the treated soils 398 detailed in Table 4a. There is a dearth of literature on the optimum ratio of CEMI-PFA mixes 399 400 for maximum strength in stabilised contaminated soils. However, with uncontaminated soils it is documented that the optimum proportion of PFA in the mix would depend on the chemical, 401 physical and mineralogical properties of the PFA used (Naik et al. 1991). Table 4a shows that 402 403 despite uncertainties in the optimum mix ratio of CEMI and PFA for effective stabilisation, the

404 choice of mix ratio was between equal proportions of CEMI and PFA (CEMI:PFA=1:1) and 1 part CEMI to 4 parts PFA (CEMI:PFA = 1:4) in majority of the studies. Such choices were made 405 based on the experience of a previous study. There have not been concerted efforts to evaluate 406 the optimum mix ratio before using the binder for S/S of contaminated soil due to the volume of 407 experimental work required. The binder formulations chosen still resulted in acceptable 408 mechanical and leaching properties. Thus, the optimum mix ratio is likely to fall within the 409 afore-stated mix ratios. This is because, generally, without cement, most fly ashes shows very 410 little self-hardening property with curing time due to low free CaO content (Kaniraj and 411 412 Havanagi 1999) and significant quantities of cement would be required in a mix for optimal performance. 413

414

Similarly, there is a dearth of literature on the water content of compaction for maximum 415 strength of CEMI-PFA stabilised contaminated soil. However, in one case, it was observed that 416 the UCS of contaminated sandy gravel treated with the binder (CEMI:PFA=1:4) increased with 417 increasing water content within the range OMC-2 to OMC+5 (Kogbara et al. 2013). This differed 418 from the observation with uncontaminated soil where maximum UCS was obtained on the dry 419 side of OMC for sandy soil stabilised with CEMI:PFA=1:5 binder (Arora and Aydilek 2005). 420 However, it has been shown that generally, the best mechanical and leaching behaviour is 421 obtained around the OMC for different binder systems (Kogbara 2011). Hence, it is for this 422 reason that only OMC values are shown in studies where different water contents were employed 423 in Table 4, for simplicity of the table. 424

425

427 The UCS of CEMI-PFA treated soils was generally less than 1 MPa at 28 days in virtually all the studies in Table 4b, although exact UCS values were not stated in a study (Akhter et al. 1990) 428 where equal proportions of CEMI and PFA was used. This is because PFA addition does not 429 result in high strength in this time frame and strength levels largely depend on the quantity of 430 cement present (Kogbara et al. 2013). However, strength levels increased above the 1 MPa mark 431 at 56 days and beyond but this depended on the nature of contamination in the soil. Soils with 432 high hydrocarbon content had much lower values even after 3 months (Perera 2005; Perera and 433 Al-Tabbaa 2005; Kogbara et al. 2013) (Table 4a and 4b). Especially, where the contaminants 434 435 were artificially spiked on the soil and did not have sufficient time to interact with the soil. Strength levels were also found to increase with the binder dosage (Kogbara et al. 2013). 436

437

As regards long-term strength behaviour, which was considered by a study (Al-Tabbaa and 438 Evans 2000; Al-Tabbaa and Boes 2002) in Table 4, this varied with the mix ratio, specifically 439 the cement content of the two mixes studied. The mix with higher cement content (CEMI:PFA = 440 3:8) showed a relative increase over the 5-year period, while the strength behaviour of the other 441 mix (CEMI:PFA=1:4) suggest that the strength is reaching a plateau at 5 years (Table 4b). This 442 implies that numerous factors that affect the strength development of the mixes come into play 443 over time, such as interaction with contaminants, and in situ curing conditions. The 5-year 444 strength was between three and six times greater than that at 2 months, which is perhaps an 445 446 indication of the continual in situ long-term hydration of cementitious materials in the presence of contamination (Al-Tabbaa and Boes 2002). 447

- 448
- 449

Reference	Soil type and composition (including natural pH and other details if stated)	Initial amount of prime contaminants (mg/kg)	Binder dosage (%)	W/S ratio	Curing age (days)
(Akhter et al. 1990)	Loess with 2% organic content	12,200 each of	30	0.29	28
	(composition and natural pH details not specified)	As and Cr	C:PFA = 1:1		
		Leachable As: 8,400			
		Cr: 8,820			
(Al-Tabbaa and	Made-ground	Cd: 8.7	12.5	0.15	28
<u>Evans 2000</u>)	consisting mainly of clayey sand and sandy clay	Cu: 1,264			
and		Pb: 2,801	Two mixes:		56
(Al-Tabbaa and		Ni: 105	C:PFA = 1:4		
<u>Boes 2002</u>)		Zn: 1,589	C:PFA = 3:8		784 (2.3 yrs)
		Coal tar: 1,400			
		Mineral oil: 566			1,826 (5 yrs)
		Toluene extract: 1,700	10.5	0.1.5	• •
(<u>Chitambira 2004</u>)	Model soil	Cd – 8.7	12.5	0.15	28
	49% gravel, 37% sand, 7% silt and 7% clay	Cu - 1,264	C:PFA = 3:8		90
		Pb - 2,801			180
		$N_1 - 105$			
		Zn = 1,589			
		$\frac{\text{Mineral oil} - 566}{\text{N} + 1.729}$	167		5 112
(<u>Antemir 2005</u>)	Pepper steel factory site in Florida	Pb - 1,728	10.7	Not	5,113
	(soil type not specified)	AS = 23.2	C.PFA = 3.2	specified	(14 years)
(Danana and Al	Madalasil		10.5	0.15	20
(Perera and AI-		Cu = 8.7	12.3	0.15	28
<u>Tabbaa 2005</u>)	49% gravel, 37% sand, 7% silt and 7% clay	Cu = 1,204	C.FFA = 3.8		00
and		Ni 105			90
(<u>Perera 2005</u>)		7n = 1589			
		211 - 1,307 Paraffin oil - 8 700			
		1 aranını 011 – 0,700			

Table 4a. Soil and binder characteristics of CEMI-PFA S/S treated contaminated soils

_	Reference	Soil type and composition (including natural pH and other details if stated)	Initial amount of prime contaminants (mg/kg)	Binder dosage (%)	W/S ratio	Curing age (days)
	(Moon et al. 2010)	55.7% sand, 33.8% silt,	Zn	20	0.50	7
		10.3% clay	4,973	Three mixes:		28
		Organic matter content -0.6%		C:PFA = 1:3		
		Natural pH – 8.31		C:PFA = 1:1		
_				C:PFA = 3:1		
	(<u>Kogbara et al.</u>	Clayey silty sandy gravel	Cd: 3467 ± 153	5, 10 and 20	0.14 to	28
	<u>2013</u>)	65% gravel, 29% sand, 2.8% sand, 3.2% silt	Cu: $3,167 \pm 231$	C:PFA = 1:4	0.21	84
		Spiked with a mixture of metals and hydrocarbons	Pb: $3,733 \pm 208$		OMC: 0.16	
		pH of spiked contaminated soil - 9.83	N1: $3,56 / \pm 153$		for 5 &	
		Organic matter content – 0.22%	$2n: 4,233 \pm 289$		10%	
			$1PH: 0312 \pm 1482$		dosage,	
					20%	
					2070 dosage	
	C: CEMI	PCB: Polychlorobiphenyls TF	PH: total petroleum hydrocarbons	OMC: Opt	timum moisture c	ontent
454						
155						
455						
456						
450						
457						
-						
458						
459						
460						
461						

Table 4a (continued). Soil and binder characteristics of CEMI-PFA S/S treated contaminated soils

Reference	UCS	Bulk	Hydraulic		Leachal	bility
	(MPa)	density	conductivity	Major leaching	Final	Comments on leaching
		(Mg/m^3)	(m/s)	test(s)	leachate pH	behaviour
(Akhter et al. 1990)	$\geq 0.35*$	Not	Not	Modified TCLP	> 5.2	- 1,014 and 1,170 mg/kg of As
	for all mixes	determined	determined	L/S = 10	(exact pH not	and Cr, respectively leached
					stated)	out.
						- Binder not very effective
						given the high dosage used.
(Al-Tabbaa and Evans	C:PFA = 1:4	1.58	C:PFA = 1:4	TCLP	C:PFA = 3:8	- The leachate concentrations of
<u>1998</u>),	0.36 (28-d)	for both	0.72×10^{-9}		10.1 (56-d)	Cu, Pb and Zn after 5 years
(Al-Tabbaa and Evans	1.00 (56-d)	mixes at 28	3.60×10^{-9}		7.2 (784-d)	increased by up to 3, 82 and
<u>2000</u>)	3.15 (784-d)	days. Data	$0.90 \ge 10^{-9}$		7.4 (1,826-d)	104 times, respectively, above
and	2.37 (1,826-d)	for other	0.86 x 10 ⁻⁹		same details	their 2-month values.
(Al-Tabbaa and Boes	C:PFA = 3:8	time points	C:PFA = 3:8		not provided	- Leachate pH decreased over
<u>2002</u>)	0.51 (28-d)	not	1.31×10^{-9}		for the	time due to carbonation.
	1.30 (56-d)	available.	$0.69 \ge 10^{-9}$		C:PFA = 1:4	- The reduction in pH over time
	3.50 (784-d)		0.78×10^{-9}		mix	caused increase in metal
	5.41 (1,826-d)		1.25 x 10 ⁻⁹			solubility.
(Chitambira 2004)	0.4 (28-d)	2.23 (28-d)	0.85×10^{-9}	TCLP	~ 7.3 for	- Metal retention improved with
	2.0 (90-d)	2.09 (90-d)	0.95 x 10 ⁻⁹	Batch leaching	TCLP	curing age, especially for Cu,
	3.8 (180-d)		180-d value			Pb and Zn.
			not		~ 11.7 for	- Oil leachability was lower in
			determined		Batch	stronger mixes
			0		leaching	
(<u>Antemir 2005</u>)	2.22	Not	2.50×10^{-8}	TCLP	Not specified	- As levels was below detection
	Average of 5 cores	determined	Range:	Multiple Extraction		limits in both leaching tests.
	Range: 0.11 – 4.69		1×10^{-7} -	Procedure (MEP)		- Pb leachability decreased
			3.6 x 10 ⁻⁹			from 1728 to 4 mg/kg in TCLP.
(Perera and Al-Tabbaa	0.4 (28-d)	1.45 (28-d)	Not	Batch leaching	~ 12	- Only leachability of Pb was
<u>2005</u>)	0.7 (90-d)		determined			investigated.
and	2.0 (180-d)	1.40 (90 - d)				- Pb leachability decreased with
(Perera 2005)						time from 140 to 50 mg/kg
(<u> </u>						between 28 and 90 days

Table 4b. Performance characteristics of CEMI-PFA S/S treated contaminated soils

	Reference	UCS	Bulk	Hydraulic		Leacha	bility
		(MPa)	density	conductivity	Major leaching	Final	Comments on leaching
			(Mg/m^{3})	(m/s)	test(s)	leachate pH	behaviour
	(Moon et al. 2010)	Not determined	Not	Not	TCLP	5.3 (1:3)	- Zn leachability decreased
			determined	determined		6.0 (1:1)	between 7 and 28 days.
						7.9 (3:1)	- 260, 50 and 50 mg/kg were
						Mix ratio in	leached out of C:PFA = $1:3, 1:1$
						parenthesis	and 3:1, respectively, at 28 days
((<u>Kogbara et al. 2013</u>)	28-day OMC values	28-day	28-day OMC	- ANC	5.4 - 11.5	- Water content showed no
		0.09 (5%)	OMC values	values	at 0, 1 and 2 meq/g		significant effect on leachability
		0.10 (10%)	1.68 (5%)	Not available	HNO ₃ addition		- 10% and 20% binder dosage
		0.45 (20%)	1.71 (10%)	1.58×10^{-9}	- Tank leaching		reduced the leachability of
		84-day	1.64 (20%)	4.69×10^{-5}			metals in the treated soil below
		0.30 (10%)	84 - day	84 - day			that of the untreated soil, but
		5% & 20% dosage data	1.82 (10%)	4.9/ x 10 ⁻			5% dosage did not.
		not available	5% & 20%	5% & 20%			- The binder was quite effective
			dosage data	dosage data			The binder was the least
							- The billion was the least suitable for TPH
							immobilisation among those
							studied Leachability increased
							significantly over time
							- The predominant leaching
							mechanism was surface wash-
							off in the tank test.
465	C: CEMI OM	C: Optimum moisture content	TCLP: Toxicity	characteristic leachi	ing procedure (<u>USEPA 1986</u>) ANC: Acid n	eutralisation capacity (BSI 2006)
-05	* Testing only carried or	it on mixes with UCS > 0.35 MP	a exact UCS value	es not provided			
466	Testing only current of		u, exact o e o vulue	s not provided			
467							
468							

Table 4b (continued). Performance characteristics of CEMI-PFA S/S treated contaminated soils

470 The bulk densities of the studies were different, even those that used the same binder dosage and water content (Al-Tabbaa and Evans 2000; Al-Tabbaa and Boes 2002; Chitambira 2004; Perera 471 2005; Perera and Al-Tabbaa 2005) were markedly different (Table 4b). The difference in bulk 472 density was probably due to differences in the degree of compaction in the studies. The bulk 473 density was observed to decrease between 28 and 90 days in some of the afore-mentioned 474 studies, although other study (Kogbara et al. 2013) recorded an increase in bulk density within 475 the said time-frame (Table 4b). The exact trend for bulk density even with increasing binder 476 dosage (Kogbara et al. 2013) is unclear. 477

478

The hydraulic conductivity was generally around 10⁻⁹ m/s and was similar to those of soils 479 stabilised with CEMI. Its evolution over time was compared between two mixes (Al-Tabbaa and 480 Evans 2000; Al-Tabbaa and Boes 2002). The average hydraulic conductivity of both made 481 ground mixes at 5 years were similar (Table 4b). There was a varied response of the hydraulic 482 conductivity of the mixes over time as it increased between 28 and 56 days in one mix, while it 483 decreased in another. However, it appears that the mix with greater PFA content (CEMI:PFA 484 =1:4) had a more stable evolution over time (apparent decrease beyond 56 days) unlike the other 485 mix whose evolution over time was unclear. Its (CEMI:PFA = 3:8) hydraulic conductivity 486 decreased between 28 and 56 days and increased subsequently, although the 5-year value was 487 quite similar to the 28-day value. The likely reasons for the varied response have been provided 488 489 in section 4.2.1.

490

Although, combinations of CEMI and PFA have been used to treat metal sludges, very fewstudies have deployed the binder for treatment of metals in contaminated soils. A few studies

493 have used PFA alone alongside combining it with lime – e.g. (Dermatas and Meng 2003). PFA addition was found to increase the immobilisation pH region for Pb and Cr. The findings of 494 previous studies (Table 4) show that metal leachability decreases with curing age in 495 contaminated soils treated by the binder. The binder dosage required for effective leachability 496 reduction was between 10 and 20%. However, in a particular case, it was observed that even with 497 30% dosage, the binder was not very effective for stabilising As and Cr (Akhter et al. 1990). 498 Equal proportions of CEMI and PFA in the mix was found to be more effective in Zn 499 stabilisation than higher proportion of PFA in the mix (Moon et al. 2010) (Table 4b). The binder 500 501 is also not suitable for TPH immobilisation as TPH leachability increased significantly over time probably due to the binder's low buffering capacity to pH changes (Kogbara et al. 2013). 502

503

504

4.2.3 CEMI-GGBS S/S contaminated soils

Table 5 shows the details of a few studies in which CEMI-GGBS blends were used to treat 505 contaminated soils. Very few published studies have actually deployed CEMI-GGBS blends for 506 treatment of contaminated soils, although it has been shown to be effective in ground 507 508 improvement, and has been used for other hazardous waste streams. One study (de Korte and Brouwers 2009b) in which CEMI-GGBS was used in combination with lime for contaminated 509 soil treatment is also shown in Table 5. 510

511

Bulk density was determined on only two of the studies in Table 5b. It looks like the bulk density 512 increases with binder dosage although there is contradictory evidence between 10 and 20% 513 binder dosage in one of the studies (Kogbara 2011) (Table 5b). All the same, it can be observed 514 that the bulk density increases by only a little amount even with large increases in binder dosage. 515

ReferenceSoil type and composition(including natural pH and other details if stated)		Initial amount of prime contaminants (mg/kg)	Binder dosage (%)	W/S ratio	Curing age (days)
Akhter et al. 1990)	Loess with 2% organic content	As: 12,200	9	0.37	28
	(composition and natural pH details not specified)	Cd: 10,000	17	0.34	
		Cr: 12,200	30	0.29	
		Pb: 10,900	C:GGBS = 1:1		
Allan and Kukacka	Alluvial with silty to gravelly sand	Cr ³⁺ : 200 and 1,000	17	0.23	28
1995)	(composition not specified)		33	0.24	
,	Natural pH – 8.4	Cr ⁶⁺ : 200, 500 and 1,000	50	0.24	
	*		C:GGBS =1:4*		
			C:GGBS =2:3*		
			C:GGBS =3:2*		
(de Korte and	Sandy soil, containing clay and poor in humus	Cd – 20	13.6	0.21	28
Brouwers 2009b)	(natural pH not specified)	Cr - 28	21.9		
/		Cu – 27	C:GGBS: lime =		
		Pb – 140	2:7:1		
		Ni – 22			
		Zn - 150			
		Mineral oil - 49			
(<u>Kogbara 2011</u>)	Clayey silty sandy gravel	Cd: 3467 ± 153	5	0.13 - 0.20	28
and	65% gravel, 29% sand, 2.8% sand, 3.2% silt	Cu: $3,167 \pm 231$	10	OMC:	84
(Kogbara and Al-	Spiked with a mixture of metals and hydrocarbons	Pb: 3,733 ± 208	20	0.16 for	
Tabbaa 2011)	pH of spiked contaminated soil - 9.83	Ni: 3,567 ± 153	C:GGBS = 1:9	5%, 0.17	
<u>140044 2011</u>)	Organic matter content -0.22%	Zn: $4,233 \pm 289$		for 10% &	
		TPH: 6312 ± 1482		0.15 for	
				20%	
				dosages.	

anta an laaahi	Leachability				UCS	Reference
behaviour	Final leachate pH	Major leaching test(s)	conductivity (m/s)	density (Mg/m ³)	(MPa)	
6 and 0.4 mg/kg 2,100, 5.6 and 3 Pb leached in 9, binder conten y. MI-GGBS blend w to be more effecti CEMI alone.	> 5.2 - (exact pH not C stated) m ar re - ol fo	Modified TCLP L/S = 10	Not determined	Not determined	\geq 0.35 for all mixes	(<u>Akhter et al. 1990</u>)
rations up to 1,00 pilised to give TCI procentration less the all cases. resistance improv- sing GGBS contents caused part of Cr $^{6+}$ to Cr $^{3+}$. ility of both Cr(I T) decreased wi GGBS content.	7.5 - 10.7 - (17% dosage) m 9.2 - 10.4 le (33% dosage) 5 9.7 - 11.3 - (50% dosage) w - re - ar in	TCLP Tank test	$C:GGBS=1:4 \\ 1.1 \times 10^{-7} \\ 9.0 \times 10^{-8} \\ 1.0 \times 10^{-10} \\ C:GGBS=2:3 \\ 4.0 \times 10^{-7} \\ 9.0 \times 10^{-8} \\ 7.0 \times 10^{-11} \\ C:GGBS=3:2 \\ 2.0 \times 10^{-7} \\ 7.0 \times 10^{-11} \\ 1.5 \times 10^{-10} \\ \end{bmatrix}$	Not determined	C:GGBS=1:4 8.5 (17% dosage) 12.5 (33% dosage) 24 (50% dosage) C:GGBS=2:3 8 (17% dosage) 15 (33% dosage) 29 (50% dosage) C:GGBS=3:2 6 (17% dosage) 20 (33% dosage) 35 (50% dosage)	(<u>Allan and Kukacka</u> <u>1995</u>)
G	in		7.0 x 10 ⁻¹¹ 1.5 x 10 ⁻¹⁰		20 (33% dosage) 35 (50% dosage)	

	Reference	UCS	Bulk	Hydraulic		Leachal	oility
		(MPa) density conductivity Major	Major leaching	Major leaching Final Comments			
			(Mg/m^3)	(m/s)	test(s)	leachate pH	behaviour
	(de Korte and	18.6 (13.6% dosage)	2.09	Not	Tank test	Not specified	- There was no significant
	Brouwers 2009b)	30.3 (21.9% dosage)	2.21	determined		_	difference in contaminant
							emission among the two binder
							dosages used.
							- 64d emission less than 1
							mg/m^2 for all contaminants.
	(<u>Kogbara 2011</u>)	28 (84)-day	28 (84)-day	28-day	- ANC	5.8 - 11.2	- Water content showed no
	and	OMC values	OMC values	OMC values	at 0, 1 and 2 meq/g		significant effect on leachability
	(Kogbara and Al-	0.1 (0.13) (5% dosage)	1.79 (1.92)	-	HNO ₃ addition		- Leachability of metals was
	Tabbaa 2011)	0.5 (0.8) (10% dosage)	1.82 (1.93)	1.09×10^{-8}	- Tank leaching		reduced to meet relevant
		0.44 (20% dosage) -	1.69 –	4.66 x 10 ⁻⁹			criteria with up to 20% dosage.
		No 84-day data for the	No 84-day	84-day			- The binder was quite effective
		20% binder dosage.	data for	2.14 x 10 ⁻⁸			for Pb immobilisation.
			20% binder	28 & 84-day			- pH-dependent leachability of
			dosage.	data for 5%			the metals studied was found to
				dosage and			decrease over time.
				84-day for			- The predominant leaching
				20% dosage			mechanism was surface wash-
	C: CEMI	MC: Ontimum moisture content	TCL D: Toxicity	not available.	ing procedure (USEDA 1086		off in the tank test.
528	C. CEMII C	SMC. Optimum moisture content	ICLF. Toxicity	characteristic leach	ing procedure (<u>OSEFA 1980</u>) ANC. ACI	(BSI 2000)
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550							
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532							
533							

Table 5b (continued). Performance characteristics of CEMI-GGBS S/S treated contaminated soils

534 The UCS behaviour in Table 5b shows that the higher the replacement levels of GGBS in the blend, the lower the strength. This has been attributed to non-optimum gypsum contents 535 particularly at replacement levels in excess of 50% (Cook et al. 1986). However, depending on 536 the total binder dosage, the UCS may decrease with high slag content at higher binder dosages 537 and increase with increasing slag content at lower dosages (Allan and Kukacka 1995). The 538 optimum proportion of GGBS for maximum strength appears to lie between 50 - 60% of the total 539 binder dosage (Allan and Kukacka 1995). Furthermore, the presence of high concentrations of 540 contaminants, especially high hydrocarbon content in the soil leads to very low strength. Hence, 541 542 the very large differences in strengths between two studies (de Korte and Brouwers 2009b; Kogbara and Al-Tabbaa 2011) (Table 5b) with similar binder dosages. 543

544

There are discrepancies between the two studies (Allan and Kukacka 1995; Kogbara and Al-545 Tabbaa 2011) in Table 5b containing hydraulic conductivity data on the binder. Hydraulic 546 conductivities are higher in the first study (Allan and Kukacka 1995) by more than one order of 547 magnitude, even though it used higher binder dosages. This may probably be due to difference in 548 test facilities as a rigid wall permeameter was used in the first study and flexible wall 549 permeameter used in the other study. This sometimes causes enormous difference in test results 550 (Christopher et al. 2006). Nevertheless, the hydraulic conductivities obtained with flexible wall 551 permeameters, which are recommended for low permeability soils (Christopher et al. 2006), was 552 within acceptable limits $(10^{-8} \text{ to } 10^{-9} \text{ m/s}, \text{ see Table 1}).$ 553

554

555 The findings of the studies show that the binder could effectively reduce the leachability of most 556 of the common metals in soils and was quite effective for Pb, which poses problems during

immobilisation in CEMI. The binder also has the potential to partially reduce Cr^{6+} to Cr^{3+} when 557 immobilising the contaminant. A few other studies have reported that GGBS activated by other 558 alkaline materials showed good immobilisation of metals. In one study (Deja 2002), sodium 559 carbonate was used as activator and the binder was doped with up to 2% of Cd, Cr, Pb and Zn, 560 and tank-leaching tests showed immobilisation rates exceeding 99.9%. Another study (Qian et al. 561 2003) employed sodium silicate-sodium hydroxide solution as activator and the binder was 562 doped with up to 2% Zn. It was observed that chemical fixation mechanisms like the formation 563 of insoluble calcium zincate precipitate and the incorporation of Zn^{2+} into the C-S-H lattice was 564 responsible for the effective immobilisation of Zn^{2+} in the binder matrix. 565

566

567 4.2.4 Lime-GGBS S/S contaminated soils

Most of previous studies utilising lime-GGBS binder for contaminated soil treatment have 568 focussed on the use of lime, since as mentioned in section 2, it is a primary stabilising agent just 569 like cement. Moreover, conventional stabilisation of contaminated soils has been based on lime 570 rather than GGBS. Hence, there are few studies combining both binder materials for 571 contaminated soil treatment. Table 6 summarises the details of some previous studies in this 572 direction. Table 6a indicates that there is a paucity of specific literature on lime-GGBS mixes. 573 The binder has been mostly used for stabilisation of uncontaminated soils (i.e. in ground 574 improvement). With uncontaminated soils, it has been observed that higher UCS is achieved 575 576 with more GGBS in the mix than with more lime. Previous works on uncontaminated soils suggest that the optimum mix for maximum strength is about one part lime to four parts GGBS 577 (Kogbara 2011). However, the precise relationship between strength and binder components is 578 579 complex due to interactive effects between the binder components.

Reference	Soil type and composition	Initial amount of prime	Binder dosage	W/S ratio	Curing age
	(including natural pH and other details if stated)	contaminants (mg/kg)	(%)		(days)
(Akhter et al. 1990)	Loess with 2% organic content	As: 12,200	18	0.37	28
	(composition and natural pH details not specified)	Cd: 10,000	L:GGBS =1:35		
		Cr: 12,200	18.5	0.37	
		Pb: 10,900	L:GGBS =1:17		
			30	0.34	
			L:GGBS =1:34		
(Dermatas 1994a;	Montmorillonite sand	Pb: 7,000	5, 10 and 15	OMC	90
Dermatas 1994b)	Kaolinite sand		Lime	exact value	
				not	
				specified	
(Yukselen and	19% sand, 56% Silt, and 20% clay	Cu: 510	3.85, 4.76 and	Not	Not
<u>Alpaslan 2001</u>)	3% organic matter	Pb:153	6.25	specified	specified
	Natural pH -2.73	Leachable:	Lime		
	Water content – 15.57%	Cu: 70 mg/l			
(<u>Alpaslan and</u>	19% sand, 56% Silt, and 20% clay	Pb: 7,700	1,1.3,2,	Not	Not
Yukselen 2002)	3% organic matter	Leachable Pb: 170 mg/l	2.4,4.8,9.1 and	specified	specified
	Natural pH – 2.73		16.7		
	Water content – 15.57%		Lime		
(Shah et al. 2003)	Loamy silt	Fuel oil: 100,000	5, 10 and 20	OMC	7
	Natural moisture content – 10.5%	Leachable: 380 mg/l	Lime	value not	
				specified	
(Moon and Dermatas	Montmorillonite sand	Cr ³⁺ : 4,000	10	Not	Not
<u>2005</u>)	Kaolinite sand		Lime	specified	specified
(Schifano et al.	London clay, pH 11.40	TPH	5, 10, 20	Not	Not
<u>2005</u>)	Kaolinite sand, $pH \sim 5.5$	London clay: 250	Lime	specified	specified
		Kaolinite sand: 2,370			

Table 6a. Soil and binder characteristics of Lime-GGBS S/S treated contaminated soils

	Reference	Soil type and composition (including natural pH and other details if stated)	Initial amount of prime contaminants (mg/kg)	Binder dosage (%)	W/S ratio	Curing age (days)
	(Korac et al. 2007)	NS	Total:	6.25	Not	Not
		Natural pH - 2	Cu: 1,200	Lime alone	specified	specified
			Pb: 700	UFS alone		
			Zn: 170	L:UFS = 3:1		
			Leachable:			
			Cu: 11.3 mg/l			
			Pb: <1 mg/l			
			Zn: 5.7 mg/l			
	(<u>Kogbara 2011</u>)	Clayey silty sandy gravel	Cd: 3467 ± 153	5	0.13 - 0.20	28
	and	65% gravel, 29% sand, 2.8% sand, 3.2% silt	Cu: $3,167 \pm 231$	10	OMC:	84
	(Kogbara et al.	Spiked with a mixture of metals and hydrocarbons	Pb: $3,733 \pm 208$	20	0.18 for	
	2011)	pH of spiked contaminated soil - 9.83	Ni: 3,567 ± 153	L:GGBS = 1:4	5%, 0.15	
	<u> </u>	Organic matter content -0.22%	Zn: $4,233 \pm 289$		for 10% &	
			TPH: 6312 ± 1482		0.14 for	
					20%	
					dosages.	
585	L: Lime	UFS – used foundry sand (by-product of iron and ste	el industry just like GGBS)	OMC: Opt	imum moisture co	ontent
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500						
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500						
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Table 6a (continued). Soil and binder characteristics of Lime-GGBS S/S treated contaminated soils

593	Table 6b. Performance characteristics of Lime-GGBS S/S treated contaminated soils						
Reference	UCS	Bulk	Hydraulic		Leachal	oility	
	(MPa)	density	conductivity	Major leaching	Final	Comments on leaching	
		(Mg/m^3)	(m/s)	test(s)	leachate pH	behaviour	
(Akhter et al. 1990)	$\geq 0.35*$	Not	Not	Modified TCLP	> 5.2	- 1,640, 1,850 and 1.6 mg/kg of	
	for all mixes	determined	determined	L/S = 10	(exact pH not stated)	Cd leached from the 18, 18.5 and 30% dosage mixes, respectively. - 70, 40 and 6 mg/kg of Pb leached from 18, 18.5 and 30% dosage mixes respectively	
						- The binder was quite less effective for Cd immobilisation at lower dosages.	
(Dermatas 1994a;	Not determined	Not	1×10^{-5} to 4 x	TCLP	5 - 12	- Effective binder dosage:	
Dermatas 1994b)		determined	10^{-7} for mont-	Tank test		$\geq 10\%$.	
			morillonite			- TCLP Pb leachability < 5	
			sand 1×10^{-5} to			mg/l. Negligible Pb leaching in	
			1×10^{-5} for			Dh leachability was influenced	
			kaolinite			by clay mineral: nH controlled	
(Yukselen and	Not determined	Not	Not	TCLP	5-6	- Effective binder dosage	
Alpaslan 2001)		determined	determined	1021	for 3.85 to	6.25%.	
/					6.25% lime	- 94% reduction in Cu	
					dosages.	leachability.	
						- Pb concentration in leachate	
						too low.	
(<u>Alpaslan and</u>	Not determined	Not	Not	TCLP	12.5 - 13	- Effective binder dosage: \geq	
Yukselen 2002)		determined	determined		for 4.8, 9.1 &	4.8%.	
					16.7% lime	-82-93% reduction in Pb	
					dosages.	leachability.	
					5 - 6	- Precipitation as $Pb(OH)_2$ and	
					10F 1 to 2.4%	encapsulation controlled	
					nme dosages.	leachadility.	

Table 6b. Performance characteristics of Lime-GGBS S/S treated contaminated soils

		· · · · · ·						
	Reference	erence UCS	Bulk Hydraulic		Leachability			
		(MPa)	density	conductivity	Major leaching	Final	Comments on leaching	
			(Mg/m^3)	(m/s)	test(s)	leachate pH	behaviour	
	(Shah et al. 2003)	0.08 (5% dosage)	Not	Not	Flow through	Not specified	Treatment with 10% lime	
		0.11 (10% dosage)	determined	determined			dosage caused 87% reduction in	
		0.12 (20% dosage)					leachable oil concentration.	
	(Moon and Dermatas	Not determined	Not	Not	Tank test	Not specified	- 94% reduction in Cr^{3+}	
	<u>2005</u>)		determined	determined			leachability.	
							- Cr^{3+} leachability not	
							influenced by clay mineral after	
							lime treatment.	
	(Schifano et al. 2005)	Not determined	Not	Not	Batch leaching	NS	- 87% reduction in TPH	
			determined	determined			concentration for London clay.	
							- 80% TPH reduction for	
							kaolinite sand.	
							- TPH reduction was	
_							independent of binder dosage.	
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600								
500	,							

Table 6b (continued). Performance characteristics of Lime-GGBS S/S treated contaminated soils

Reference	UCS	Bulk density	Hydraulic		Leacha	ability
	(MPa)	(Mg/m^3)	conductivity (m/s)	Major leaching test(s)	Final leachate pH	Comments on leaching behaviour
(Korac et al. 2007)	Not determined	Not determined	Not determined	Column test	12.5 lime-treated, 4 UFS-treated, 11 lime-UFS treated	 Lime treatment yielded 98% Cu and 72% Zn reduction, whil lime-UFS treatment gave 78% Zn reduction, Cu leachabilit was below detection limit. Leachability of Cu and Z higher in UFS-treated soil tha in untreated soil.
(<u>Kogbara 2011</u>) and (<u>Kogbara et al. 2011</u>)	28 (84)-day OMC values 0.04 (0.2) (5% dosage) 0.4 (0.8) (10% dosage) 0.8 (20% dosage)**	28 (84)-day OMC values 1.72 (1.74) 1.75 (1.79) 1.68**	28-day <i>OMC values</i> 1.22 x 10 ⁻⁸ 4.05 x 10 ⁻⁸ 5.42 x 10 ⁻⁸ 84-day** 1.45 x 10 ⁻⁷ (5% dosage) 7.31 x 10 ⁻⁸ (10% dosage)	- ANC at 0, 1 and 2 meq/g HNO ₃ addition - Tank leaching	5.8 - 12.2	 Water content showed n significant effect on leachability The binder showed th potential to reduce TPI leachability to some extent. The binder had problems wit Pb immobilisation due to its hig pH regime, similar to CEMI. Differences in leachability of contaminants over time were no statistically significant

Table 6b (continued). Performance characteristics of Lime-GGBS S/S treated contaminated soils

** Testing only carried out on mixes with UCS > 0.35 MPa, exact UCS values not provided

Table 6b shows that lime and lime-GGBS blend gives relatively low strength, especially when used for treatment of soils with large amounts of hydrocarbons (Shah et al. 2003; Kogbara et al. 2011). UCS, bulk density and hydraulic conductivity were not determined in majority of the studies in Table 6. Further, there is generally a paucity of data on the mechanical performance of lime or lime-GGBS treated contaminated soils. This is probably because the studies, which mostly utilised lime alone, focused on reducing granular leachability in soil and were not so concerned about mechanical performance of the treated soils.

615

It can be seen from two studies (Dermatas 1994a; Kogbara et al. 2011) (Table 6b) that 616 considered the hydraulic conductivities of lime and lime-GGBS treated soils that the binder 617 results in high hydraulic conductivity compared to cement-based binders. In one case (Kogbara 618 619 et al. 2011), the hydraulic conductivity appears to increase with binder dosage and curing age. Such increased hydraulic conductivity is associated with the presence of lime. The reaction of 620 lime with soil particles, especially clays, leads to agglomeration and flocculation of clay particles 621 with a consequent reduction in the plasticity and an increase in shear strength of soils. This in 622 turn leads to increase in permeability with lime addition (Kogbara 2011). However, there is little 623 information on the effect of lime addition on sandy and gravelly soils that was studied in the 624 above work. In a related study on ground improvement, it was observed that lime addition 625 reduced the permeability of poorly graded river sand and increased that of sandy silty clay (El-626 627 Rawi and Awad 1981). Further, there existed a moulding water content at which the permeability of the lime-sand mixture reached a minimum. Thus, more work is required in this area to fully 628 elucidate the effect of lime-GGBS binder on hydraulic conductivity in different soil types. 629

631 The major contaminants frequently treated by lime are Pb and Cu, and about 10% lime dosage would be effective in significantly reducing their leachability. Lead (Pb) was found to present 632 problems during immobilisation with lime-GGBS (Kogbara 2011). Low binder dosage may 633 satisfy certain leaching criteria but higher dosage may not, especially where the pH attained by 634 higher binder dosages corresponded to the zone for increased Pb leachability (see Figure 1). 635 However, it has been shown that the mechanism of Pb immobilisation in lime-stabilised soil is 636 not only through precipitation as Pb(OH)₂ at alkaline pH but also encapsulation within the crystal 637 structure of the cementitious compounds formed. Hence, significant reduction in Pb leachability 638 could still be achieved at pH values greater than the pH (9.5–11) range, where Pb(OH)₂ is least 639 soluble (Rha et al. 2000; Alpaslan and Yukselen 2002). The type of clay mineral present in the 640 soil also controls Pb leachability (Dermatas 1994a; Dermatas 1994b). Lime has also shown 641 potential for reducing TPH leachability to some extent, although the reduction was found to be 642 independent of binder dosage (Schifano et al. 2005). 643

644

645 4.3.5 Lime-PFA S/S contaminated soils

Table 7 summarises the details of some contaminated soils treated by lime-PFA blends. It looks 646 like lime-PFA blends have been deployed more for S/S treatment of contaminated soils than 647 lime-GGBS blends. Most of the studies on lime-PFA binder deployed it for treatment of As, Cr 648 and Pb contamination. Thus, although this review does not focus on As and Cr, they will be 649 650 briefly discussed in this section. There are very few studies dealing with the other most common metals, Cd, Cu, Ni and Zn. One of such studies (Feigl et al. 2010) employed the binder for about 651 99% reduction of the leachability of very low levels of Cd (0.4 mg/l), Cu (1.5 mg/l) and Zn (89 652 653 mg/l), coupled with phytostabilisation.

Reference	Soil type and composition (including natural pH and other details if stated)	Initial amount of prime contaminants (mg/kg)	Binder dosage (%)	W/S ratio	Curing ag (days)
(Akhter et al. 1990)	Loess with 2% organic content	As: 12,200	30	0.34	28
	(composition and natural pH details not specified)	Cr: 12,200	L:PFA = 1:5		
			34	0.34	
			L:PFA = 1:5		
Dermatas and Meng	Kaolinite sand	Pb: 7,000	35	OMC	28
<u>2003</u>)	(composed of clay and fine quartz sand)	Total Cr: 1,945	L:PFA = 1:2.5	value not specified	
(Shah et al. 2003)	Loamy silt	Fuel oil: 100,000	20	OMC	7
	Natural moisture content – 10.5%	Leachable: 380 mg/l	L:PFA = 1:1	value not	
		-	L:PFA = 3:1	specified	
(Dermatas et al.	Kaolinite sand	As: 124	35	OMC	90
2004)	(composed of clay and fine quartz sand)		L:PFA = 1:2.5	value not	
	pH: 4 – 6.5			specified	
(Jing et al. 2006)	Soil from Cr-contaminated industrial waste site	Cr: 1,330	25	OMC	28
(<u> </u>	(details not specified)	,	L:PFA = 1:4	value not	
				specified	
Moon and Dermatas	Kaolinite sand	Pb: 7,000	35	OMC	28
2006)	(composed of clay and fine quartz sand)		L:PFA = 1:2.5	value not	
,				specified	
	L: Lime C	OMC: Optimum moisture content			

Table 7a. Soil and binder characteristics of Lime-PFA S/S treated contaminated soils

661	Table 7b. Performance characteristics of Lime-PFA S/S treated contaminated soils						
Reference	UCS	Bulk	Hydraulic		Leachal	bility	
	(MPa)	density	conductivity	Major leaching	Final	Comments on leaching	
		(Mg/m ')	(m/s)	test(s)	leachate pH	behaviour	
(<u>Akhter et al. 1990</u>)	$\geq 0.35*$	Not	Not	Modified TCLP	> 5.2	- 4,020 mg/kg of As leached	
	for all mixes	determined	determined	L/S = 10	(exact pH not	from the 30% dosage mix.	
					stated)	- 5,300 mg/kg of Cr leached	
						from the 34% dosage mix.	
						- The binder showed poorer As	
						& Cr leachability performance	
(Demoster and Mana	(((NT - 4	N - 4		2 12	compared to others tested.	
(Dermatas and Meng	0.00	NOL	NOL	ICLP	3 - 13	- The binder reduced the	
<u>2005</u>)		determined	determined		evaluated	TCL P regulatory banchmark of	
					evaluated	5 mg/l Ph immobilisation was	
					above nH	ensured if the treatment TCLP	
					range	pH was kept between 8 and 1	
					Tunge.	- Adsorption was predominant	
						Pb immobilisation mechanism	
						at $pH > 9$. The binder widens	
						Pb immobilisation range from 5	
						to 13.	
						- Total Cr leachability was	
						reduced by 99.7% of the initial	
						amount.	
(<u>Shah et al. 2003</u>)	0.11 (L:PFA = 1:1)	Not	Not	Flow through	Not specified	- Leachability was not	
	0.12 (L:PFA = 3:1)	determined	determined			evaluated for lime-PFA mixes	
						as it was done for only selected	
						mixes. A combination of	
						lime:PFA:cement = $2:1:1$ was	
						round to give better leachability	
						lime alone	
						nine alone.	

Reference	UCS	Bulk	Hydraulic		Leachal	bility
	(MPa)	density (Mg/m ³)	conductivity (m/s)	Major leaching test(s)	Final leachate pH	Comments on leaching behaviour
Dermatas et al. 2004)	Not determined	Not determined	Not determined	Semi-dynamic leaching test (<u>ANS 1986</u>)	10.5	 There was no significant effect on reduction of As emission even with 35% of the binder. Precipitation as low soluble Ca-As compounds was the dominant release mechanism.
(<u>Jing et al. 2006</u>)	1.43	Not determined	Not determined	TCLP ANC	8.74	 Release of Cr(III) controlled by adsorption on Fe oxides at pH<10.5, and precipitation of Ca₂CrO₅.6H₂O at pH > 10.5. There was 60% reduction in TCLP Cr concentration from 104 mg/kg to 42 mg/kg.
Moon and Dermatas 2006)	Not determined	Not determined	Not determined	Semi-dynamic leaching test (<u>ANS 1986</u>)		 The controlling mechanism of Pb immobilization appeared to be precipitation. The formation of Pb₂SiO₄ (a very insoluble compound) was observed. The controlling leaching mechanism of Pb was diffusion.

Table 7b (continued). Performance characteristics of Lime-PFA S/S treated contaminated soils

665 TPH: Total petroleum hydrocarbons

666 It has been suggested that a mix ratio of one part lime to two and half parts PFA (lime:PFA = 1:2.5) is the optimum dose for treatment of hexavalent Cr-contaminated soil (Kostarelos et al. 667 2006). This may reasonably apply to other metals since majority of the mix ratios in Table 7a 668 contain no more than five parts PFA to one part lime. There are very few 'easily accessible' 669 studies on the mechanical behavior of soils treated with the binder. This can be seen in Table 7b, 670 as it contains no information on the bulk density and hydraulic conductivity of the treated soils. 671 All the same, Table 7b shows a reasonable UCS level of 1.43 MPa at 28 days (Jing et al. 2006) 672 with 20% dosage. A much higher value (6.66 MPa) (Dermatas and Meng 2003) was even 673 674 obtained with the afore-mentioned optimum mix for Cr-leachability reduction. These compare favourably with the UCS values for CEMI-PFA mixes in Table 4b. 675

676

With respect to leachability reduction, it appears that the mix ratio of the binder constituents 677 significantly affects the leaching results, especially for Cr. With comparable (~35%) dosages of 678 the binder, it showed poorer (57%) TCLP Cr leachability reduction in one study (Akhter et al. 679 1990), which employed a mix ratio of lime: PFA = 1:5. This compares with the 60% reduction 680 with a lime: PFA = 1:4 mix obtained elsewhere (Jing et al. 2006), albeit with a lesser (25%) 681 binder dosage. While in another study (Dermatas and Meng 2003), a 99.7% TCLP Cr 682 leachability was obtained with a lime:PFA = 2.5 mix ratio. However, this differences may also 683 depend on the initial contaminant concentration as the contaminant concentrations in the two 684 685 studies with similar binder dosages (Akhter et al. 1990; Dermatas and Meng 2003) was largely different. 686

688 Lime-PFA blends do not seem to be very good for As immobilisation as they showed no significant difference to untreated soils even at 35% dosage addition (Akhter et al. 1990; 689 Dermatas et al. 2004). The binder even mobilised As concentrations to higher levels than in 690 untreated soil (Feigl et al. 2010). Conversely, the binder is quite effective for Pb immobilisation 691 as it even widens the immobilisation pH range. This is due to its fly ash content, which forms 692 pozzolanic products that either adsorb Pb on to fresh surfaces or incorporate Pb by means of 693 chemical inclusions. Additional pozzolanic product formation with increasing curing age further 694 increases the amount of non-extractable Pb (Dermatas et al. 2006). 695

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697 **4.3.6** Comparisons between binders

This section synthesizes information in the previous sections to provide a comparison of the 698 699 effectiveness of the five different binder systems in terms of the key mechanical and leaching properties considered. Table 8 summarises the comparisons between the binders and provides 700 useful information to help in the choice of one binder over another, depending on the 701 contaminated soil management scenario. Bulk density is not included in the table, as it is not 702 considered to be of utmost importance in assessment of the effectiveness of S/S treated soils, 703 compared to UCS and hydraulic conductivity. Hence, it was not determined in most of the 704 studies in the tables on performance characteristics of S/S treated soils. 705

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- 707

Binder	Strength behaviour	Hydraulic conductivity	Leaching behaviour
system		behaviour	
CEMI	 - 28-day UCS can range from 300 kPa for 5% binder dosage to around 11 MPa for 50% binder dosage. - As with all binders, the UCS depends on 	- Hydraulic conductivity values at 28 days are in the 10^{-8} to 10^{-9} m/s range. This would satisfy criteria for	 It is a very versatile and dependable binder that can be used for reducing the leachability of a wide range of metals. It usually presents problems with Pb immobilisation, such
	the type of soil and would be significantly reduced if treating soil with fresh hydrocarbon contamination.	recycling options, especially in construction works.The hydraulic conductivity	that higher binder dosages increase Pb leachability beyond acceptable levels. Hence, it is not suitable for soils with high Pb concentrations.
	- Can easily meet the 1 MPa UCS criteria with around 10% binder dosage. Further, there is significant strength gain with time.	may increase by about half to one order of magnitude over time.	 It also has fair stabilisation performance for Cu and TPH. It can reduce TPH leachability to some extent. It can maintain leachability levels below acceptable limits with appropriate binder dosage.
CEMI-PFA	 Strength depends on cement content of the mix. Typical recorded 28-day UCS ranges from 90 kPa for 5% binder dosage to 500 kPa for 12.5% binder dosage. Strength usually builds up over time since pozzolanic reactions take time to complete. Typical recorded UCS can reach 2 MPa at 90 days. 	Hydraulic conductivity is similar to that of CEMI treated soils in the 10 ⁻⁸ to 10 ⁻⁹ m/s range.	 It can be used for reducing the leachability of many metals in contaminated soils. However, it is very suitable for Cu and Pb-contaminated soils unlike CEMI. Especially, PFA content increases the immobilization pH range for Pb. It is not suitable for TPH immobilisation as concentrations were found to increase over time especially as the binder has a relatively lower buffering capacity in acidic environments.
CEMI-GGBS	 GGBS replacement levels in excess of 50% leads to significant reduction in strength. UCS could be comparable to those of CEMI depending on mix ratio. Typical recorded 28-day UCS ranges from 100 kPa with 5% binder dosage to 35 MPa with 50% binder dosage, depending on nature of contamination. UCS could even be higher than CEMI for similar binder dosages. GGBS could typically replace more cement than PFA for the same strength. 	 The binder shows similar hydraulic conductivities (in the 10⁻⁸ to 10⁻⁹ m/s range) to CEMI and CEMI-PFA treated soils. Lower values in the 10⁻¹⁰ m/s range have been recorded with 50% binder dosage. 	 It offers better immobilisation of Cu than CEMI and is comparable to CEMI-PFA in reducing Cu leachability. Similarly, it also shows good leachability reduction for Pb just like CEMI-PFA. Its good immobilisation potential for Cr has also been recorded. It also shows good immobilisation potential for Cd, Ni and Zn, albeit with lesser capacity compared to CEMI.

Table 8 (continued)	. Comparisons of	he performance of	characteristics of	f different binder systems
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Binder system	Strength behaviour	Hydraulic conductivity behaviour	Leaching behaviour
Lime-GGBS	 Typical 28-day UCS in the literature ranges from 40 kPa with 5% binder dosage to around 800 kPa for 20% binder dosage. The presence of contaminants in the soil has a greater deleterious effect on the strength of lime-GGBS than on CEMI and CEMI-GGBS. 	 The binder has much higher hydraulic conductivity compared to other binders due to its lime content. Increased hydraulic conductivity with lime content is more severe with clay soils. 	 Just like CEMI, it offers good immobilisation potential for Cd, Ni and Zn, but it is not very suitable for immobilisation of Cu and Pb due to its high pH regime. It has a very high buffering capacity similar to CEMI; hence, it is suitable for acidic environments. It appears to be marginally better than the other binders in reducing TPH leachability.
Lime-PFA	 Typical recorded 28-day UCS values ranges from 1.43 MPa for 25% binder dosage to 6.66 MPa for 35% binder dosage. Gains strength over time due to pozzolanic reaction. The ultimate strength may be comparable to that of CEMI-treated soil depending on the mix ratio. 	 There are very few easily accessible studies on contaminated soil treatment with recorded hydraulic conductivity values. Further work is required in this area. However, lime-PFA concrete is known to have decreased hydraulic conductivity due to decreased water content and production of additional cementitious compounds, which in turn reduces pore interconnectivity. Values of the order, 10-9 m/s have been recorded for uncontaminated soils. 	 The binder has mostly been used for treatment of As, Cr and Pb. A lime:PFA = 1:2.5 mix has been suggested as optimum for effective reduction of Cr leachability. It is not very effective for As immobilisation as it showed no difference from untreated soils in some leaching studies. It is very effective for Pb immobilisation as it widens the immobilisation pH range and adsorbs Pb unto fresh pozzolanic products or incorporates it through chemical inclusions.

714 Cement generally shows superior performance on strength behavior compared to the other binders, especially for soils with high organic content (Jegandan et al. 2010). However, 715 depending on the mix proportions, contaminated soil treated with CEMI-GGBS binder could 716 even demonstrate higher strength values than CEMI-treated soil at the same binder dosage, 717 especially as curing age increases. This is because the pozzolanic reaction is slow and the 718 formation of calcium hydroxide requires time (Oner and Akyuz 2007). One important aspect of 719 strength development not mentioned earlier is the UCS after immersion in water. The test is used 720 to assess whether the stabilised material has hardened chemically and is not susceptible to 721 deleterious swelling reactions. It has been shown that GGBS-based binders, especially CEMI-722 GGBS, show superior performance to the other binders in this regard (Kogbara 2011). 723

724

The binders generally show hydraulic conductivities in the 10^{-8} to 10^{-9} m/s range. Lime-GGBS binders have been found to yield increased hydraulic conductivity values compared to the other binders, which can fall in the 10^{-7} m/s range over time. Especially, as the hydraulic conductivity of S/S treated soils generally appears to increase with curing age.

729

Leachability studies showed that CEMI and CEMI-PFA were effective for Cd, but at lower dosages, lime-GGBS was observed to be significantly less effective for the metal. It is well known that Pb presents problems with both rate of setting and leachability in CEMI. However, the PFA and GGBS-based binders were notably effective for Pb immobilization. Nevertheless, it was observed that inclusion of GGBS in a binder blend generally offered superior performance compared to PFA. Further, the pH-dependent leachability of metals in CEMI-GGBS treated soils have been found to decrease as curing age increases over an 84-day period due to continuation of pozolanic reactions (Kogbara and Al-Tabbaa 2011). Overall, CEMI is a very versatile and
dependable binder compared to the other binders in this work, for most the metals focused on. In
every case, inclusion of CEMI resulted in leachate concentrations as low as or lower than the
corresponding mixture without CEMI (Akhter et al. 1990).

741

Although, there was evidence of reduced strength and increased hydraulic conductivity and 742 leachability in some cases, available information on long-term tests shows that S/S treatment was 743 generally still effective at 5 to 14 years, with continued hydration still taking place without 744 serious sign of deterioration due to ageing. There were cases of fluctuations in mechanical and 745 leaching properties over time owing to the complex nature and variability of S/S treated soils. 746 The effect of the long-term interaction between contaminants and soil-grout materials seems to 747 be dominant over those of small differences in grout constituents over a long period (Al-Tabbaa 748 and Boes 2002). 749

750

751 **5** Conclusions

This work reviewed the performance of S/S treated soils utilising blends of CEMI, CEMI-PFA, 752 CEMI-GGBS, lime-GGBS and lime-PFA in terms of the UCS, bulk density, hydraulic 753 conductivity and leachability. The UCS was observed to be optimum around the OMC and it 754 increased with binder dosage for all binders. Acceptable UCS and hydraulic conductivity levels 755 756 for recycling in construction works, and leachability of most metals can be reduced to acceptable levels, with about 20 - 35% of the binders studied. However, more binder dosage does not 757 always lead to a better stabilised/solidified product. Some binders were more suitable for certain 758 759 contaminants than others were. This work helps provide useful information on scenarios to

choose one binder over another depending on the end use of the S/S treated soil. Long-term performance of S/S treated soils showed consistent effectiveness over a period of 5 to 14 years with the occurrence of fluctuations in mechanical and leaching behaviour owing to the complex nature and variability of S/S treated soils. Further work on pH-dependent leaching behaviour of S/S treated soils cured for long periods is necessary to provide more information on the durability of S/S treated soils.

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