MgO-GGBS binder stabilized/solidified PAEs (Dimethyl Phthalates)
contaminated soil: Strength and leachability in early stage

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Abstract

The MgO-GGBS (granulated ground blast furnace slag) binder, as a novel binder, has gained increasing attention in recent decades in S/S (stabilization/solidification) to treat contaminated soils. In this study, the physical strength and leaching performance of a MgO-GGBS (MG) binder was examined and compared with PC (Portland cement) and PF (PC and fly ash) regarding the S/S treatment of PAEs (dimethyl phthalates) contaminated soil after a 7-day curing. PC, GGBS, FA (fly ash) and MgO were used as binders and the effect of different MG dosage was discussed. The results show that the order of early-stage strength after a 7-day curing was: $PF < MG < PC$, and PAE leachate concentrations was in the order of $MG < PC \approx PF$. The increase in MG dosage can improve the strength of binders and immobilize PAEs, while the increase of initial PAE concentrations (3000-10000 mg/kg) can reduce the strength and increase the PAEs leaching. Therefore, the MG binder showed a better performance for the S/S of PAEs contaminated soil than PC and PF binders. The relation between leachability and strength was further explored and is expected to provide guidance for the early-stage strength estimation after the S/S of PAEs contaminated clay soil with the MG binder.

Key words: Solidification/Stabilization; novel binder; GGBS; organic pollutants; MgO

1. Introduction

PAEs (dimethyl phthalates) have been widely used as plasticizers in industrial products or as non-polymeric products in cosmetics, fragrances, lubricants and paints (Li et al. 2016; Paluselli et al. 2018). They are physically bound to the polymeric matrix by the hydrogen bond and van der Waal force and can be easily released during production, use and disposal (Lü et al. 2018). Therefore, the extensive production (~300 million tonnes/year) (Garcia and Robertson 2017) and application of plastics have made PAEs one of the most ubiquitous organic pollutants in the environmental media (Zhang et al. 2015). They have also been listed as priority pollutants by environmental agencies of various countries including US, Canada, China and European Union due to their endocrine disrupting potential (Li et al. 2006; Bui et al. 2017; Lü et al. 2018). PAEs can be exposed to human via air inhalation, skin contact with dust and soil and the food chain with the human exposure to PAEs of ~70 µg/kg/day (Net et al. 2015). The PAE concentrations in soil generally fall in the range of 0.001 to 30 mg/kg (Cai et al. 2008; Zeng et al. 2009) and higher concentrations can be observed in contaminated sites. The elevated PAE concentrations in soils pose a high risk to human health via skin contact and intake of food products (i.e., vegetables or crops) grown on contaminated soils. Therefore, it is important and necessary to remediate PAEs contaminated soil.

S/S (Stabilisation/solidification) is a commonly used low-cost remedial technique to improve the physical characteristics of contaminated soil and immobilize various contaminants based on various cementitious materials, such as PC (Portland cement) and slags, due to their high strength and low permeability. However, the production of cement needs intensive energy and contributes to anthropogenic carbon emission

(Hrvoje et. al. 2016), and therefore the partial replacement of cement with industrial by-products, such as GGBS (granulated ground blast furnace slag) and FA (fly ash), has attracted much attention in recent decades in order to reduce energy requirement and carbon footprint (Chen et al. 2019; Wang et al. 2019; Du et al. 2020). The blends of PC with FA or GGBS have been regarded as promising alternatives as S/S binders because the unburned content in FA can adsorb organics (Perera 2005) and the slow hydration rate of GGBS can help improve the strength at medium and later ages (Kogbara 2011). The addition of novel additives, such as MgO, can provide added benefits in S/S performance and has attracted increasing in recent decades, especially for the S/S of soils contaminated by metals (Chen et al. 2009; Cubukcuoglu and Ouki 2012; Jin and Al-Tabbaa 2014; Rahman et al. 2016). This is due to the fact that MgO has a pH neutralisation range (9-11) to precipitate metals, good ion-exchange ability and strong complexation ability to metal ions with O-H groups (García et al. 2004; Jin et al. 2016; Wang et al. 2018a). However, MgO-based binder has been rarely applied in organics decontamination. It was reported that the presence of organic compounds can hinder the hydration of cementitious materials, and it is hard to convert organics into hydration products (Leonard and Stegemann 2010a). Therefore, the immobilization of organic contaminants mainly relies on physical entrapment and adsorption. Insoluble (non-polar) compounds tend to be retained in the solid matrix, while soluble (polar) contaminants are still leachable (Leonard and Stegemann 2010a). For example, the solidified/stabilized petroleum drill cuttings by the PF (PC and FA) binder satisfied the UK non-hazardous landfill acceptance criteria except the chloride and oil contents (Leonard and Stegemann 2010a). In comparison, Al-Ansary and Al-Tabbaa (2007) reported that PC, GGBS-PC and lime-PC outperformed microsilica-PC, FA-MgO-PC in the

S/S of synthetic drill cuttings. The 30% GGBS-PC binder immobilised the oil concentrations in the low oil content mix to inert waste, and the 20% GGBS-PC and 30% lime-PC binders managed to convert drill cuttings to a stable non-reactive hazardous waste. Our previous field study (Wang et al. 2015a) suggested that the MG (MgO-GGBS) binder was efficient in immobilizing heavy metals, SVOCs (semi volatile organic compounds) as well as TPHs (total petroleum hydrocarbons) in spite of low strength and relative high organics leaching at high initial organics concentrations. Therefore, MgO-based binder holds huge potential in S/S to eliminate organic pollutants in soils. However, to our best knowledge, S/S has scarcely been used in PAEs decontamination. Therefore, a laboratory-scale investigation into PC-based and MgO-based binders treated PAEs contaminated soil was carried out in this study to examine their physical strength and leachability and further provide guidance for the application of MgO-based binder in S/S of PAEs contaminated soil.

The objectives of this study were: (1) to investigate the early-stage strength and the leachability of stabilized/solidified PAEs contaminated soil; (2) to compare the effectiveness of the MG binder with PC-based binders in remediating PAEs contaminated soil; and (3) to evaluate the effects of initial PAE concentrations and MG dosage on the S/S effectiveness of the MG binder. This applicable research is expected to provide new insights in the binder selection and the in-situ S/S application for clay soil remediation.

2. Materials and Methods

2.1 Materials

The soil samples used in this study are clean clay soils without contamination, and collected from a construction site in Liuyang, Hunan Province, China. By weight, its mineral composition is 28.83%-70.49%-1.22% concentration of clay-silt-sand. The moisture content is 26.5 wt%; pH is 6.89; the liquid limit and plastic limit are 34.3 $\omega\%$ and 23.1 $\omega\%$, respectively, with a plastic index of 11.2. PC32.5, GGBS, FA and MgO were used as binders in this study. PC, FA and GGBS was produced by the Shanghai Conch Cement CO., LTD, and MgO was purchased from the Hao Sheng New Material company, China. FA and MgO were used in this study due to their good adsorption ability towards organics and heavy metals. As shown in Table 1, the compositions of PC and GGBS were provided by the manufacturer and those of MgO were measured using XRF (X-ray Fluorescence Spectrometer). The ratio of GGBS and MgO was 9:1 with detailed physiochemical properties listed in Table 2 (Wang et al. 2020). DCM (dichloromethane, CH_2Cl_2) and HCl (hydrochloric acid) were purchased from the Nanjing wide field instrument and equipment CO., LTD, and they were used for extraction.

Table 1 Compositions of binders used in this study (wt%)

	CaO	SiO ₂	Al ₂ O ₃	MgO	Fe ₂ O ₃	SO ₃	MnO	TiO ₂	K ₂ O
PC	46.6	27	12.6	1.02	3.29	3.33	0.06	0.51	1.13
GGBS	36.6	33.9	16.1	6.82	0.88	2.01	0.28	1.04	0.66
MgO	0.84	0.99	0.41	95.8	0.19	0.28	0.02	-	0.01

Table 2 Physiochemical properties of GGBS, MgO and PC

	Density	Surface area	Reactive MgO	Reactivity	pH
GGBS	-	0.28 m ² /g	-	1.3	11.22 ^a
MgO	3.58 g/cm ³	27.83 m ² /g	79.2%	40.9	10.69 ^b
PC	3.15 g/cm ³	0.35 m ² /g	-	4.6	

^awater:GGBS=1:1; ^bwater:MgO=5:1

The clay soil was dried in air and ground to pass a 2 mm sieve in order to remove impurities from the soil. PAEs were added into dry soil samples to produce artificial organic pollution with the concentrations of 3000 mg/kg, 5000 mg/kg and 10000 mg/kg. In other words, samples contain 0.3%, 0.5% and 1% of PAEs, respectively. The binders and water were evenly mixed with dry soil samples and then the mixture was placed in split PVC cylinders (50 mm inner diameter and 100 mm high). After cured at 20 ± 2°C with a humidity of 95% for 7 days, the samples were demolded and subjected to UCS (unconfined compressive strength) and batch leaching tests. The preparation and initial curing operations strictly followed the standard ASTM D1632-17 (2017) (ASTM International 2017). The compositions of soil, binder and PAEs of each group are shown in Table 3. Mixes were named in the following order: binder, percentage of binder used, the percentage of PAEs added. Group 1 was designed to discuss the effectiveness of MG10-0.3 (MgO and GGBS) compared with conventional binders, PC10-0.3 (control group) and PF10-0.3 (PC:FA=1:2 (Wang et al. 2015a)). Group 2 was to study the effect of MG dosages (7.5%, 10% and 15%). Group 3 was to evaluate the effect of PAE concentrations (0 mg/kg, 5000 mg/kg and 10000 mg/kg). MG7.5-0.5, MG7.5-1, MG10-0.5 and MG10-1 were also conducted to explore the relationship between leached PAE concentrations and mechanical strength. The combination of each sample, curing temperature, water content, curing time and binder dosages were selected based on Wang et al. (2018).

Group	Sample	Soil	GGBS	MgO	PC	FA	Water	PAEs
Group 1	PC10-0.3	67.19	0	0	10	0	22.51	0.3
	PF10-0.3	67.19	0	0	3.33	6.67	22.51	0.3
	MG10-0.3	67.19	1	9	0	0	22.51	0.3
Group 2	MG7.5-0.3	70.16	0.75	6.75	0	0	22.04	0.3
	MG10-0.3	67.19	1	9	0	0	22.51	0.3
	MG15-0.3	61.26	1.5	13.5	0	0	26.44	0.3
Group 3	MG15-0	61.26	1.5	13.5	0	0	26.74	0
	MG15-0.3	61.26	1.5	13.5	0	0	26.44	0.3
	MG15-0.5	61.26	1.5	13.5	0	0	26.24	0.5
	MG15-1	61.26	1.5	13.5	0	0	25.74	1

2.2 Testing methods

In order to describe the mechanical property of stabilized/solidified soil and examine the S/S effectiveness, the UCS tests were conducted in triplicate following the standard ASTM D4219-08 (ASTM International 2008). The axial strain rate was set at 1%/min, and the data of the axial compressive force was collected per second. The crushed samples with particle sizes between 1 and 4 mm were then subjected to the TCLP (toxicity characteristic leaching procedure) test (USEPA 1990). 12.5 g specimens were added into 250 mL diluted sodium acetate solution (initial pH 4.8). The leachate solutions were agitated for 18 h. It should be noted that the control TCLP test indicated that total organic matter in clean soil as well as after the addition of MG binder (i.e., MG7.5-0, MG10-0, MG15-0 samples) is lower than the detection limit. After agitation and 24 h's standing, the liquid from the TCLP test was

transferred into a 500 mL plug-contained conical flask for organics extraction. In order to speed the extraction reaction as well as acting as a pH buffer, 5 mL of 12 M hydrochloric acid was added into the flask. 15 mL of DCM was then added to the supernatant as extractants for liquid-liquid extraction and hand-shaking for 2 min in order to mix PAEs with extractants well for adequate extraction (Wang et al. 2015c). After repeating for 3 times, the total extracted sample was then poured into a weight recorded container and placed in a fume cupboard for DCM evaporation. The extracted volume poured into a container and the waste volume of the liquid left in the flask were also recorded before the evaporation. After 48 h's evaporation, the mass of the residual was recorded, and the concentration of organics was calculated by the mass balance method. The UCS tests were carried out in triplicate while the leaching tests were carried out in duplicate.

3. Results and discussion

3.1 Physical strength

3.1.1 Effect of different binders

Figure 1 gives the UCS values of 3000 mg/kg PAEs contaminated soil treated by different binders (Group 1), i.e., PC, PF and MG, after the curing period of 7 days at 20°C at a humidity of 95%. In spite of triplicate UCS tests, the error bar for the PF group was zero because one sample was broken and cannot be subjected to the UCS test, and the UCS data of another two samples were the same. Although the average UCS value of MG10-0.3 (86.6 kPa) was only 63% of that of PC10-0.3 (137.6 kPa), it was 30% higher than that of PF treated soil (61.1 kPa) at this early stage (7 days' curing). This is due to the fact that under the same pollution conditions, the early-stage strength of binders is affected by the reactivity of binding materials which has an effect on their hydration reaction rate. According to Wainwright and Rey (2000), the reactivity of binder materials can be determined by the chemical modulus ((CaO+MgO)/SiO₂). They claimed that the higher the value, the more

reactive the binders are. As shown in Table 2, the calculated reactivity of PC and MgO is 4.6 and 40.9, respectively, much higher than that of GGBS (1.3) and FA (0.17) (Wang et al. 2018a). Therefore, the presence of GGBS and FA with low reactivity will hinder the hydration of PC at early age. After 7 days' curing, the strength development of PC, PF and MG treated PAEs contaminated soil is in the order of PC>MG>PF. This finding is consistent with the results obtained in Wang et al. (2018) that the strength of binders was in the order of PC>MG>PF for treating metal-contaminated soil after 28 days' curing. In addition, Liu (2013) found that the early stage UCS value of MG stabilized/solidified uncontaminated soil was only 30%-40% of that of PC treated soil, which is similar to our results, and the strength increased significantly after 7 days' curing and still saw an upward trend after 90 days. Jin et al. (2016) also demonstrated that the strength of MG blends was weak at the beginning and improved after 3 years' treatment in an S/S field trial with the contamination by both heavy metals and organics. The physical strength of binders after longer curing time will be further addressed in future.

Figure 1 UCS values of PAEs contaminated soil treated with 10% of PC, PF and MG binders (PAEs concentration=3000 mg/kg)

In order to have a general understanding of the physical strength of different binders, Table 4 presents a comparison of UCS values of different mixes in application to stabilized/solidified contaminated soil in some studies. It is notable that the UCS

value of PC is the highest in the early stage compared with binders based on other materials, such as FA, MgO, CaO, lime, etc. This is particularly true in early stage (7 days' curing), but the difference becomes narrow or reverse (MG>PC) with a longer curing time as can be found in Table 4. This is consistent with the reactivity of GGBS and PFA mentioned above, indicating a longer time requirement to accomplish the full hydration.

Table 4 A comparison of UCS values of mixes applied in S/S in different studies

Mixes	Curing period	UCS (MPa)	Pollutants	Reference
PC, CaO, FA, etc.	28 days	PC>CaO+FA	Naphthalene, pyrene, motor oil	(Dermatas et al. 2003)
PC, PF	7, 28, 56 days	PC>PF (7-56 days)	Drill cutting	(Leonard and Stegemann 2010b)
PC, PF, MG, etc.	1.5 years	MG>PF>PC	Zn, As, Cr, Cu, Ni and Pb and total organics	(Wang et al. 2015a)
PC, MG, etc.	40 days, 1 year, 3 years	PC>MG (40 days) PC=MG (3 years)	Zn, As, Cr, Cu, Ni and Pb and total organics	(Wang et al. 2016)
PC, PF, MG	7 days	PC>MG>PF	PAEs	This study

3.1.2 Effect of MG dosage

The effect of MG dosage (7.5%, 10% and 15%) on the strength development of stabilized/solidified PAEs contaminated soil was evaluated by UCS tests. As shown in Figure 2, the UCS values in the early stage increased by around 19% from 81.5

kPa to 96.8 kPa as MG dosages increased from 7.5% to 15%. The increasing trend is consistent with our previous study (Wang et al. 2016) that the average UCS values of MG2.5, MG5 and MG10 increased with increasing dosages (2.5%, 5% and 10%) in the case of the in-situ S/S of soil contaminated by heavy metals and total organics after three years' curing. This indicates that the increase in MG dosage can improve the strength of stabilized/solidified soil contaminated by heavy metals or/and organics.

Figure 2 UCS values of PAEs contaminated soil treated with 7.5%, 10% and 15% of the MG binder (PAEs concentration=3000 mg/kg)

3.1.3 Effect of initial PAE concentrations

MG15 binders were used to assess the effect of different PAE concentrations on the strength of stabilized/solidified PAEs contaminated soil. The average UCS value of MG15-0 after 7 days' curing was approximately 100 kPa. As shown in Figure 3, the strength decreased very slightly at a concentration of 3000 mg/kg (0.3%), then decreased by 33% when the concentration increased to 5000 mg/kg (0.5%).

However, the increase in PAE concentrations from 5000 mg/kg to 10000 mg/kg made no further significant difference to UCS values. That is, the most obvious reduction of physical strength was seen with the increase in PAE concentrations from 3000 mg/kg to 5000 mg/kg. The negative effect of PAEs on physical strength of stabilized/solidified soil in early stage can be explained by two reasons: 1) PAEs, as a kind of organic matter, decrease soil strength by reducing the friction component of strength (Ekwue et al. 2014); 2) PAEs are highly lipophilic and insoluble in water, and therefore their coverage on the binder surface can reduce the contact of binders with water and further retard the hydration reaction (Wang et al. 2015b). Further tests will be conducted to explore the relation between physical strength and initial organics concentrations.

Figure 3 UCS values of MG15 treated soil contaminated with different PAE concentrations of 0, 3000, 5000 and 10000 mg/kg

3.2 Leachability

3.2.1 Effect of different binders

Approximately 3000 mg/kg of PAEs and 10% of the binder dosage were applied to assess the effect of MG, PF and PC binders on the PAEs leachability. As shown in Figure 4, MG acted as a more efficient binder in immobilizing PAEs in soil in early stage, with the leachate concentration of 581.3 ± 23.4 mg/L, than PC and PF with their leachate concentrations of 831.1 ± 150.2 mg/L and 831.6 ± 209.2 mg/L, respectively. Their order of immobilization ability towards PAEs in early stage ($MG > PF \approx PC$) is similar to that towards heavy metals and total organics after 1.5 years' curing ($MG > PF > PC$) (Wang et al. 2015a), demonstrating the outstanding ability of MG to immobilize organics besides heavy metals in soil. This can be attributed to the brucite structure produced by MgO and Ht (hydrotalcite) produced by MG (Wang et al. 2015a, 2015b), which can help immobilize pollutants by containing various exterior ions and trap organics by hydrogen bonds (Gillman 2005). In addition, the high surface area of MgO may contribute to the adsorption of PAEs. As for the PF binder, the addition of FA was supposed to be able to immobilize organic pollutants due to the adsorption ability of its unburned carbon content (Perera 2005). Leonard and Stegemann (2010b) also found that the addition of a high carbon fly ash in PC (1:1 by mass) can improve the immobilization of hydrocarbons and chloride in petroleum drill cuttings after the curing periods of both 7 and 56 days. However, it was observed that the PF binder performed similarly to PC in PAEs immobilization in early stage. This may be explained by the very low reactivity of FA at 0.17 compared with that of PC (4.6) (Wang et al. 2018a), producing a lower hydration degree of PF than PC after 7 days' curing.

Figure 4 PAEs leachability in contaminated soil treated with different binders

3.2.2 Effect of the MG dosage

The effect of the MG dosage on the leaching performance of stabilized/solidified PAEs contaminated soil was evaluated using 3000 mg/kg of PAEs and 5%, 10% and 15% of MG, and the results are shown in Figure 5. With the increasing MG dosages from 7.5% to 10%, the leachate concentrations decreased by about 16% from 689.0 ± 25.3 mg/L to 581.3 ± 23.4 mg/L, and then decreased slightly to 558.4 ± 58.4 mg/L at a further higher MG dosage of 15%. This suggests that the increase in MG dosage can immobilize more PAEs in treated soil. From a practical standpoint, MG10 should be a cost-effective dosage for the immobilization of about 3000mg/kg PAEs in contaminated soil.

Figure 5 PAEs leachability in contaminated soil treated with different MG dosages
(initial PAEs concentration=3000 mg/kg)

3.2.3 Effect of initial PAE concentrations

The effect of initial PAE concentrations on leachability was studied with 3000 mg/kg, 5000 mg/kg and 10000 mg/kg of PAEs and MG15. As shown in Figure 6, the leachate concentration doubled from 558.4 ± 58.4 mg/L to 1130.7 ± 87.2 mg/L when the concentration of PAEs increased by 1.67 times from 3000 mg/kg to 5000 mg/kg. Similarly, the leachate concentration increased further by 2.21 times to 2497.8 ± 114.8 mg/L at a doubled concentration of 10000 mg/kg. That is, more PAEs tended to be leached in stabilized/solidified soil with severer pollution due to lower immobilization effectiveness. In comparison with the strength data in section 3.1.3, in early stage, higher PAE levels had a more negative impact on the leaching performance of MG treated soil than on their strength developments. Therefore, pretreatment options, such as chemical oxidation and biodegradation, are suggested prior to S/S treatment when treating soil with severe organic pollution.

Figure 6 PAEs leachability in contaminated soil at different initial PAE concentrations treated by the MG15 binder

Considering the superior S/S effectiveness of the MG binder over the PC and PF binders, it is a need to further explore the relation of early stage strength and leachability of PAEs contaminated soil stabilized/solidified by the MG binder. Figure 7 shows the distribution of UCS values as a function of PAEs leachate concentrations for each individual test after a 7-day curing. The strengths varied from 66.24 kPa to 320.54 kPa at the leachate concentration range of 26.92 to 2498 mg/L. The UCS values in early stage decreased sharply when the leachate concentration increased from around 26.92 mg/L to around 1200 mg/L and then leveled off with the further increase in leachate concentrations to about 2500 mg/L. This trend agrees well with that of S/S soil contaminated by heavy metals and total organics after a 3-year curing (Wang et al. 2015b). The decrease in physical strength with increasing leachate concentrations of PAEs is due to the fact that the existence of PAEs can wrap the binder grains and isolated the binder from water, making the binder surface

hydrophobic and hard to react with water (Pollard et al. 1991; Wang et al. 2015b). In this case, the hydration reaction of binders was hindered, producing lower UCS values.

Figure 7 The relation between UCS values and the leachate concentrations of PAEs contaminated soil stabilized/solidified by the MG binder in early stage

This relation can also provide guidance for the early-stage strength prediction in practice when using the MG binder to stabilize/solidify soil with PAEs pollution or even organic pollution. For example, when the leachate concentrations of PAEs were over 1000 mg/L, the physical strength of MG stabilized/solidified soil reduced to approximately 70 kPa under the conditions of this study. This may suggest that the strength of MG treated soil is less likely to be higher than 70 kPa when the PAEs leachate concentration is more than 800 mg/L in early stage. However, the water/solid ratio was reported to probably make an impact on the S/S performance of contaminated soil (Wang et al. 2018b), which has not been studied in this study.

Therefore, further research is needed to illuminate the relationship among the water/solid ratio and various soil types and the binder dosage.

4 Conclusions

This study presents the strength and leaching performance of the MG binder stabilized/solidified PAEs contaminated soil in early stage (7-day curing), and provides a comparison with two conventional binders (PC-based binders). The following conclusions can be drawn:

- 1) The average UCS value of MG10-0.3 was only 63% of that of PC10-0.3 but 30% higher than that of PF treated soil after 7 days' curing.
- 2) The increase in MG dosage can improve the strength of stabilized/solidified PAEs contaminated soil, while the increase in initial PAE concentrations in the range of 3000 mg/kg to 10000 mg/kg can reduce the strength.
- 3) MG is more efficient in reducing the leachate concentration of PAEs in stabilizing/solidifying PAEs contaminated soil compared with PC based binders even in early stage. The average PAE leachate concentrations of MG were only 69.9% of those of PC and PF.
- 4) The increase in MG dosage is beneficial to immobilize PAEs in contaminated soil, while lower immobilization effectiveness was observed with severer PAEs pollution.
- 5) The strength decreased sharply first and then levelled off with the increase in PAEs leachate concentrations for the MG binder, which may be useful for the early-age strength estimation of PAEs contaminated clay soil after the S/S treatment by the MG binder.

Data Availability Statement

All data, models, or code that support the findings of this study are available from the corresponding author upon reasonable request.

Acknowledgments

This study is supported by the National Key R&D Program of China (2018YFC1803100), the National Natural Science Foundation of China (No. 51978157) and the Fundamental Research Funds for the Central Universities. The third author would like to thank the Killam Trusts of Canada for kindly providing the Izaak Walton Killam Memorial Postdoctoral Fellowship.

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