Incorporation of disposed oil-contaminated soil in cement-based construction materials

Abstract

In order to realize the win-win objective of environmental conservation and waste utilization, the oil-contaminated soil was disposed in an eco-friendly way by replacing cement in cement-based materials. The effect of diesel and engine oil and the corresponding oil-contaminated soil on the cement-based materials were studied, including the heat release of cement hydration, rheological and flow properties, flexural and compressive strength, hydration products and oil leaching values. The results showed that soil and oil-contaminated soil increased the heat release hydration of unit mass cement and heat release rate of hydration of unit mass cement in acceleration period. The rheological and flow properties of cement paste and mortars were reduced by adding oil-contaminated soil. However, when the water cement ratio was 0.4 and 0.5, the flexural and compressive strength of mortars of 4% of oil-contaminated soil has increased by about 6%-10%, in standard curing 7 and 28 days. While, with the increase of the amount of oil-contaminated soil and water-cement ratio, the strengths were reduced. The leaching values of oil in the disposition satisfied the requirement of China standards. The results confirmed that, the use of appropriate oil-contaminated soil in cement-based materials improved the flexural and compressive strength and stability. This shows that the using of disposed oil-contaminated soil in cement-based materials will serve as cost-effective and environmental solution.

Keywords: Oil-contaminated soil; Cement-based materials; Cement hydration; Rheological and flow properties; Flexural and compressive Strength; Oil leaching values

1. Introduction

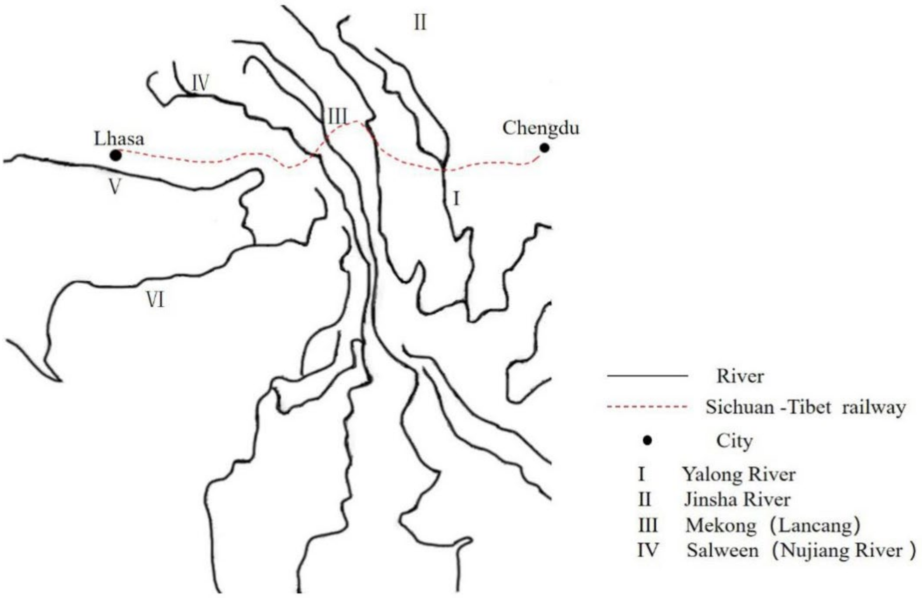
Oil and oil-contaminated soil are toxic solid wastes which can adversely affect soil-ecosystem, groundwater and surroundings, leading to environment pollutions and health problems of humans and other living beings [1-5]. Soil can be contaminated by oil from different sources, such as the illegal discharge of industrial oil, the leakage in the development of crude oil, as well as the other application processes of various types of oils.

Many disposal techniques of oil and oil-contaminated soil have been employed for environmental protection and resource utilization, which can be categorized into biological, chemical and physical methods[5-6]. Biodegradation by microorganisms and biosurfactants is an effective way being both economic and environmental friendly [8-9]. However, it can be inefficient because of the slow biodegradation rate. Furthermore, the microbial contamination and the incomplete oil elimination may occur at extremely cold temperature [9-10]. Chemical disposals utilize the chemical reagents such as surfactants, oxidation agents and acids to remove, decompose or flush the toxic in soil[11-12]. This method has the advantages of being less time consuming and more efficient, whereas the disadvantages of being expensive and the potential second pollution should be taken into accounts as well. Moreover, the optimal dosage is indeterminate[5,13]. Physical disposals include sorption, solidification, extraction, electro-kinetic separation, and burial which cannot degrade the toxic and recover the soil completely [14-16]. However, physical solidification is a widely used proposal since it is not only efficient, but also easy to implement technically [17-18].

It is essential to find the most appropriate disposal technique to deal with the oil-contaminated soil especially at construction sites. Taking Sicilian-Tibet railway as an example, the low temperature and the high altitude make it almost impossible for biodegradation to proceed in most places along the way. And chemical disposals and direct landfill of the contaminated soil are not feasible because of the low-level industrialization, high eco-friendly requirement and high environmental sensitivity, as shown in the Fig. 1. Moreover, many rivers originate from here meaning the strategical importance of this region. It is known that the cement is the most widely used construction materials, of which the solidification effect could be an effective way to dispose oil-contaminated soil alongside the construction sites of the Sichuan-Tibet railway.

(a) Environment in Tibet

(b) Oil-contaminated soil in the construction sites



(c) Rivers along the Sichuan-Tibet railway line

Fig. 1. Background of Sichuan-Tibet railway project

The mechanism of solidification can support the feasibility to dispose oil-contaminated soil in cement-based material. Due to the cement hydration, setting and hardening, oil-contaminated soil can be solidified by mechanical enclosing, physical absorption and chemical reaction[19-26]. Generally, cement can be added into soft soil to improve the performance but the solidified body is not widely used since the compressive strength and durability is not high enough to meet national standards[25-26]. Furthermore, the toxic chemicals may leach easily if the cement content is relatively low[27]. Previous studies showed that the oil can partly improve the performance of concrete. It is also reported that the appropriate dosage of cooking oil has the same effects with the superplasticizers[28], and engine oil can replace air-entraining and shrinkage-reducing agents[29-30]. Therefore, it is reasonable to solidify oil-contaminated soil by cement-based materials to achieve the waste recycling, which may have the positive influence on performance of the paste.

In order to reduce the pollution of the oil-contaminated soil in construction sites and recycle the solid wastes, this research aims to determine the feasibility and environmental impact of contaminated soil application in cement-based materials. Specific objectives are to determine the properties of cement paste and mortar incorporating diesel and engine oil contaminated soil, and the pollutant stability of disposing oil contaminated soil, in simulated construction sites.

2. Materials and methods

2.1. Materials

The cement used was ordinary Portland cement (OPC 42.5) supplied by Dujiangyan Lafarge Ryan cement factory in Chengdu, Sichuan Province, China. The monolith was from the construction site in Sichuan-Tibet railway. The chemical composition of cement and soil samples were analyzed using XRD (X-ray diffraction) and listed in Table 1. The cement was also analyzed according to China national standard Cement Mortar Strength Tests (GB/17671-1999), the results are shown in Table 3.

The soil samples were prepared by the procedure of washing drying grinding and sieving in Fig. 2, in order to filter stones and other impurities. In detail, after soaking and cleaning, the monolith was baked in the temperature of 105°C until the mass loss constantly. The soil sample was dried and grinded using a sieve of 1mm diameter. Then the soil samples were transferred into an airtight container. The performance index and particle size of the soil sample is shown in Tables 2 and 3 respectively.

The standard sand was produced by Xiamen Eseo standard sand co. LTD in China, with 2.64 $g \cdot cm^{-3}$ of apparent density, in accordance with China ISO standard. The three types of commercial oils are NO.0 diesel of China Standard, engine oils of Mobil CH-415W404L and corn oil of Arowana Brand, respectively, with their properties being shown in Table 2.

Table 1. Chemical composition (%) of OPC and soil samples

Materials	SiO_2	CaO	Al_2O_3	Fe_2O_3	MgO	SO_3	Na_2O	K_2O	TiO_2	f-CaO	C_3S	C_2S	C_3A	C_4AF
OPC.	21.20	65.66	5.43	3.87	0.87	0.91	0.56	--	--	0.95	40.91	35.49	6.84	16.76
S.	70.05	1.98	14.93	8.62	2.01	--	0.29	1.70	0.42	--	--	--	--	--

（a）monolith （b）oven-dried soil （c）ground fine soil

Fig. 2. Preparation of soil samples

Table 2. The performance index of materials

Materials	Density (g/cm^3)	water requirement of normal consistency （%）	Specific surface area m^2/kg）	Setting time （min）		Compressive strength （MPa）		Fixural value		Stability
				Initial set	Final set	3d	28d	3d	28d	
OPC.	3.1	26.3	348	155	241	25.5	44.3	6.3	8.1	qualified
Soil		--		--	--	--	--	--	--	--
--		Viscous coefficient/ （mPa·s）		Surface tension/（mN·m^{-1}）				Boiling point/（℃）		
Water	1.000	1.005		7.28				100		
Diesel	0.848	4.050		19.00				275		
Engine oil	0.868	410.010		24.50				424		

Table 3. the size percentage of soil samples (%)

size	1~0.63mm	0.63~0.315mm	0.315~0.16mm	0.16mm
samples	17.9%	32.8%	8.95%	40.2%

2.2. Experimental methods

2.2.1. Hydration heat release test and Steady-state Rheological Test

The hydration heat release of cement pastes was determined at a constant temperature of 20℃ on 72 hours on an isothermal calorimeter (Tam Air 3115 USA), with reference to the experimental method for heat of hydration of cement (the direct method) of China Standard GB2022—1980. TAM AIR has eight parallel twin-chamber measuring channels maintained at a constant temperature: one chamber contains the sample, the other contains the water with the same specific heat capacity. And test data were recorded once a minute.

Table 4. The proportion of components in cement paste

Code	Proportion	C/B	Soil	Oil	--
P	Dosage （%）	0.5	0.00%	0.00%	Control group
SP		0.5	10.00%	0.00%	Soil added separately
DP		0.5	0.00%	1.00%	Diesel added separately
SDOP		0.5	10.00%	1.00%	Diesel contaminated soil
EP		0.5	0.00%	1.00%	Engine oil
SEOP		0.5	10.00%	1.00%	Engine oil contaminated soil

The steady-state rheological curves of cement pastes under different shear protocols were determined by RHEOLAB QC rotary viscometer. The rotary viscometer was kept in the temperature of 20℃ for 30mins before starting the tests. The cement pastes were loaded into the cylinders of rotary viscometer. Then the rotary viscometer was set up in two phases symmetrical shear with shear rates in linear increase form 0s^{-1} to 100s^{-1} and linear decrease from 100s^{-1} to 0s^{-1} and the data was recorded once in a minute.

2.2.2 X-ray diffraction and Scanning electron microscopy

The paste test samples of both XRD and Scanning Electron Microscopy(SEM) were prepared according to the proportion as Table 4 and curing in standard condition(20℃, Humidity 95%) lasting 28 Days and isopropanol was used to terminated the hydration of the samples. The samples were crushed and grinded into powder(15-20 μm) for XRD and get the core of crushed samples for SEM.

Before XRD patterns collection, one side of the dehydrated slices was polished by NO.1200 sand paper and cleaned by dry compressed air. Patterns were collected at an X-ray Diffractometer (X'Pert3 Powder, Malvern Panalytical). The samples were scanned at 1.56° per minute from 5° to 65° (2-theta), with CuKα radiation generated at 40 mA and 40 kV.

Before SEM, the treated slices were mechanically polished down to 1μm and impregnated with an epoxy resin. The used microscope (QUANTA FEG250) was operated at an accelerating voltage of 15 kV.

2.2.3 Mortar preparation, slump flow and strength test

The water-cement ratios of the mortar were 0.4, 0.5, and 0.6 and the weight of sand was three times the mass of the cement. In addition, cement was replaced by the mass of 0%, 4%, 10% and 25% soil contaminated by 10% oil, which includes Diesel (D) and Engine oil (E).

The flowability and deformability of mortar mix was determinated through slump flow according to GBT 2419-94 and GBT2419-2005 of China standard. The slump flow was measured by the NLD-3 cement grit fluidity tester in 20°C and the Standard atmosphere. The slump flow (as shown in Fig. 3) was measured as the average of largest diameter (D1) of spread concrete and diameter (D2) of spread mortar at a right angle to largest diameter.

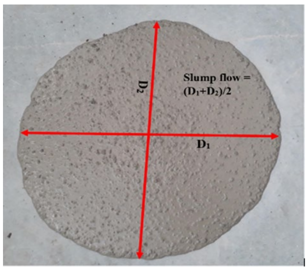


Fig. 3. Slump flow assessment for mixtures[31]

The specimens (4cm × 4 cm × 16 cm in dimension) were prepared for the mechanical properties test, including the compressive and flexural strength test. After curing to specific ages, the mechanical properties of the specimens were tested. Three samples were analyzed to calculate the mean strength. The mechanical properties for all samples were determined at 7 and 28 days respectively.

2.2.4 Leaching test of mortar determination

To identify the solidification effect of oil by cement materials, leaching test has been conducted. Test mortar samples were cured in standard condition(20℃, Humidity 95%) lasting for 7 days. Before the leaching test, the mortar samples were crushed and the core area was grinded into powder in accordance with China standard of HJ557-2010 and GB/T16488-1996, the lixiviums were prepared and oils leaching of lixiviums were measured by the means of the OIL-420 infrared spectral colorimetry spectrometer.

3 Results and discussion

3.1. Effect of contaminated soil on heat release of cement hydration

The cumulative released heat and heat flow curve of hydration of the Eight group of unit mass cement at early age (up to 72 hours) are shown in Fig. 4 and Fig. 5, respectively. It is seen that the tendency of heat release and heat flow of hydration of the mixtures are similar.

Based on the heat flow of cement hydration, the hydration of cements can be divided into four periods in Fig. 5: (1) initial reaction, (2) period of slow reaction, (3) acceleration period, (4) deceleration period[32-33]. In Fig. 4, the cumulative released heat of unit cement mass of the groups SDOP, S and SEOP show more values than others and control group P, which means that more heat released from unit cement mass as a result of the respective addition of soil and oil contaminated soil. As shown in Fig. 5, before the deceleration period, the rates of hydration heat release of groups with soil and oils contaminated soil are much higher than others, which means that hydration products are generated faster.

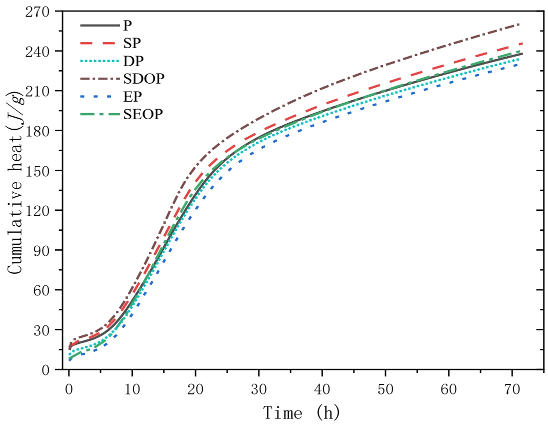


Fig. 4 The cumulative released heat of samples

(Unit cement mass)

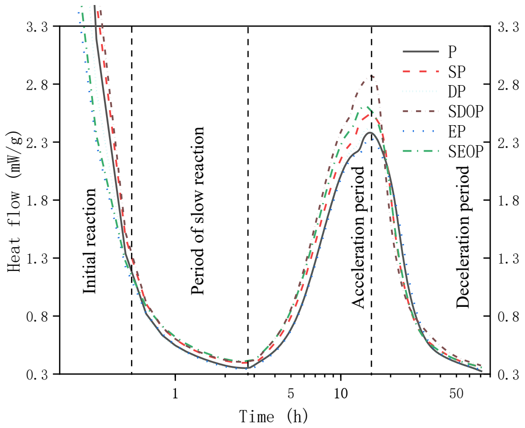


Fig. 5 the heat flow curve of samples

(Unit cement mass)

The reasons is soil may produce more nucleation sites for hydration products, similar to mineral addition and fine aggregate, which confirms with the previous studies [32-36], that means hydration production can generate in the surface of soil particles. As seen in Fig. 6, the hydration products exist in the soil particles. And for the heat released at early age of cement paste, it is because oils contaminated soil particles do not only provide a nucleation sites but also a dilution effect, which partly increased the actual ratio of water to cement. Hence in the acceleration period, the heat flow of hydration of cement with soil and oil-contaminated soil was faster.

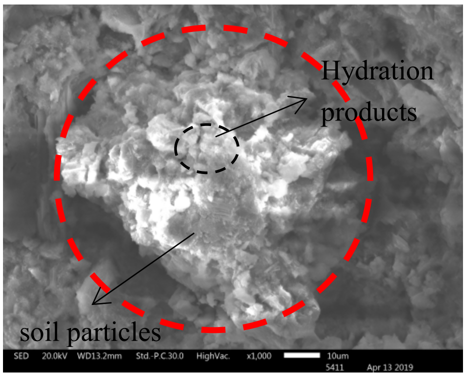
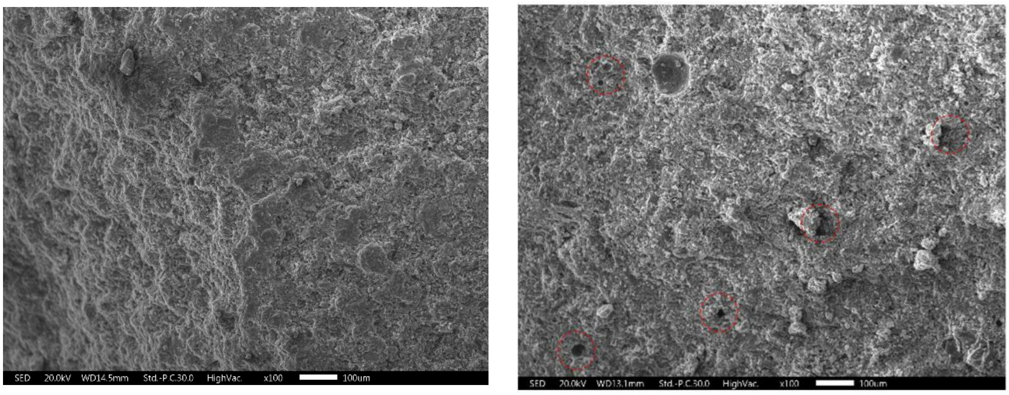


Fig. 6 SEM picture of hardened oil-contaminated soil cement paste with magnification 1000 times

But the heat flow fell quickly in the deceleration period of all the groups except the control group. At the beginning of Deceleration period, the small portion of cement particles are consumed and the large portion of cement particles begin to react, some of which are wrapped by oil that causes the reaction to slow down rapidly. Also, most of water has been consumed and then hydration reaction decreases. In the later stage of deceleration period, the water diffusion is the main controlling factor of cement hydration[32], that means the water can diffuse easily with oils and oils contaminated soil cement paste, hence that the diffusion effect of cement increases many pores and reduces the matrix density, as shown in Fig.7.



a) Control group b) Oil-contaminated soil

Fig. 7. SEM picture of hardened cement paste with magnification 100 times

3.2 Flow properties and rhelolgical of cement-based materials

Flow properties and rhelolgical reflect the workability of cement-based materials, which is very important to construction engineering. Thus, the fresh state of blended Portland cement with oils and oils contaminated soil mortars and paste were evaluated through the tests of mortar slump and rotational rheometry.

3.3. Mortar slump values

In order to analyze the effect of oil-contaminated soil of mortar flow, flow-ability percentage drop index (FPI) was introduced. Control mortars are the groups without oil and oil contaminated soil.

$$FPI = \frac{(F_x - F_c)}{F_c} \times 100\% \qquad \text{(eq. 1)}$$

F_x, F_c represent the flow values of the test mortar and control mortar, respectively.

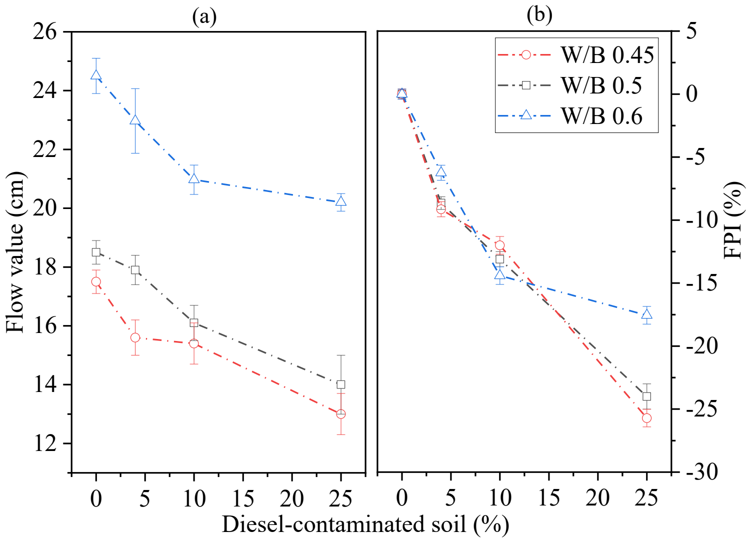


Fig. 8. Flow value and FPI of diesel-contaminated soil, with oil soil mass ratio of 0.1

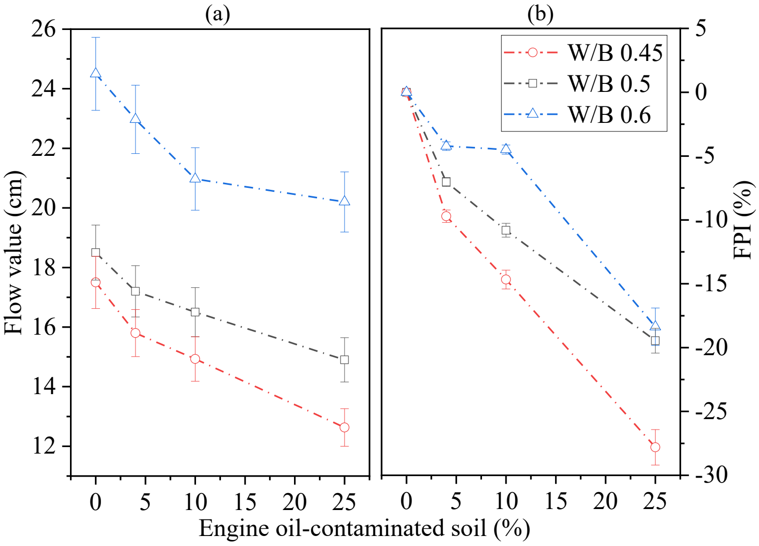


Fig. 9. Flow value and FPI of engine oil-contaminated soil, with oil soil mass ratio of 0.1

As shown in Fig. 8 and 9, (a) with water binder ration ranging from 0.45 to 0.6, the reduction of flow value of oil-contaminated soil mortar was similar and (b) with the 4%, 10%, 25% dosage of diesel-contaminated soil, FPI are respectively about 5~15%, 12.5~20% and 20~30%. With the 4%, 10%, 25%dosage of engine oil-contaminated soil, FPI are respectively about 2.5~10%, 5~10% and 17.5~30%. 4%.With the 4% ,10%,25% dosage of corn oil-contaminated soil, FPI are respectively about 7.5~10%, 12.5~15% and 17.5~25%.

According to the results, FPI of oil-contaminated soil mortar is approximately equal in all the dosage of oil-contaminated soil, which means that the addition of oil-contaminated soil which means oil-contaminated soil reduced the of the flow-ability of mortars.

3.2.2 The effect on yield stress, plastic viscosity and area of thixotropic loop of cement paste

The curves of shear stress of cement pastes with time are showed in the Fig.10. The rheological behavior of cement paste is frequently described as Bingham plastic flow[37], for which relationship between stress and rate of deformation can be expressed with two parameters, the Bingham yield stress τ_B and plastic viscosity η_B ,as

$$\tau = \tau_B + \eta_B \gamma \qquad \text{(eq. 2)}$$

where τ is shear stress and γ is the shear rate.

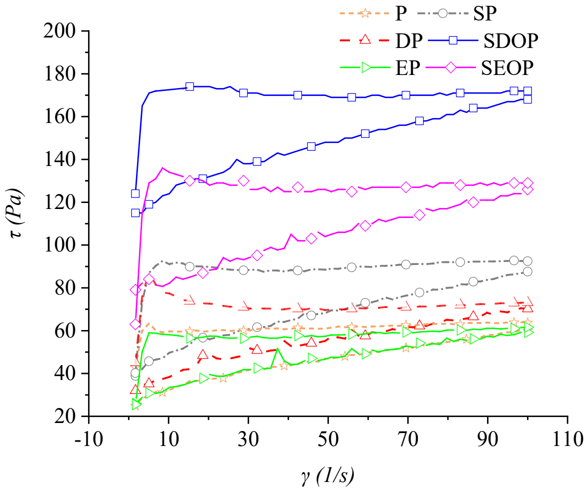


Fig. 10. Cement paste flow curves (20℃, 101kPa）

Because the flocculation network of cement paste has been broken in the falling phases of shear rate and the curves fitted with the Bingham model, the yield stress and plastic viscosity, as a function of the shear rate, were organized as shown in Fig.10. The plastic viscosity and Bingham yield stress of cement paste was calculated and as described in Fig.11.

The results showed that the addition of soil and oil-contaminated soil increased the plastic viscosity and yield stress of cement paste while the addition of different types of oils alone had a little effect on the viscosity and yield stress. Group SDOP had the greatest value of plastic viscosity and yield stress, more than 2 times of the control group P. Higher values of plastic viscosity and yield stress represent poorer rheological proprieties. However, the soil, oil and contaminated soil reduced the flow of cement paste, this is attributed to the soil improving the water demand[38]. However, it is much more difficult to break the flocculation network of the cement paste and overcome the internal friction. Meanwhile the oil had little effect on yield stress and improved the viscosity with a very lower value. That could also be the reason of the increase of high viscosity of oil than water in the cement paste.

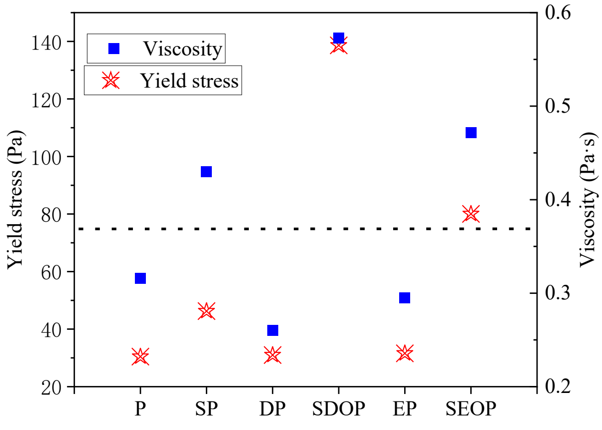


Fig. 11. The Bingham yield stress and plastic viscosity of cement paste (20℃,101Kpa）

The property of thixotropy refers that after unloading the external shear force which causes temporary increased of fluidity of cement paste, the ability to recover to the initial state. The size of the area of the thixotropic ring indicates the thixotropic properties. According to Fig.10, the area of the thixotropic ring of test pastes was calculated and result were showed in Fig.12. The results show that soil and oil-contaminated soil increased the thixotrophy of cement paste 40% to 84% than control group P. But the area of thixotropic rings of the cement paste with oil just has a little fluctuation in contrast of group P, which means that the addition of oils alone has a little effect on the thixotrophy of cement paste.

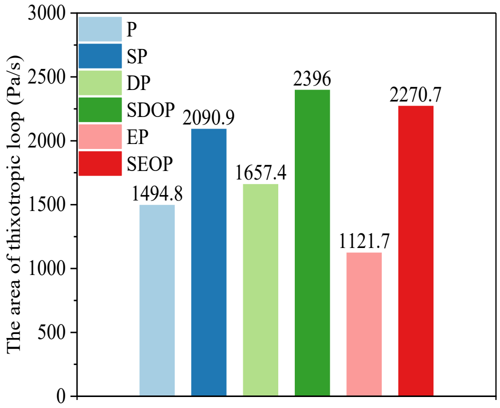


Fig. 12. The area of thixotropic loop of samples under the same shear rate time (20℃,101Kpa）

According the test results, the yield stress and plastic viscosity of cement paste increase and slump values of cement mortar decrease resulting from the addition of oil-contaminated soil. Soil adsorbing water which increased the water demand of cement paste, and oil enclosed the surface of cement and soil particles because of the absorption and hydrophobicity[38], caused the internal friction increasing and more flocculating groups forming. The two effect reduce the flow-ability of cement-based materials with oil-contaminated soil, which can be enhanced by adding water reducing agent[39].

3.3 Compressive and flexural strengths of mortar

As shown in Fig. 13, with the increasing of diesel-contaminated soil from 0% to 25%, the compressive and flexural strength of mortars of water/binder ratio 0.6 gradually reduces, but increases firstly and then decreases when water/binder ratio 0.45 and 0.5. Thus, with the addition of diesel-contaminated soil of 4%, oil/soil ratio was 0.1 and water/binder ratio were 0.45 and 0.5, with a little noticeable increase of about 2%- 6% of compressive and flexural strength of mortars.

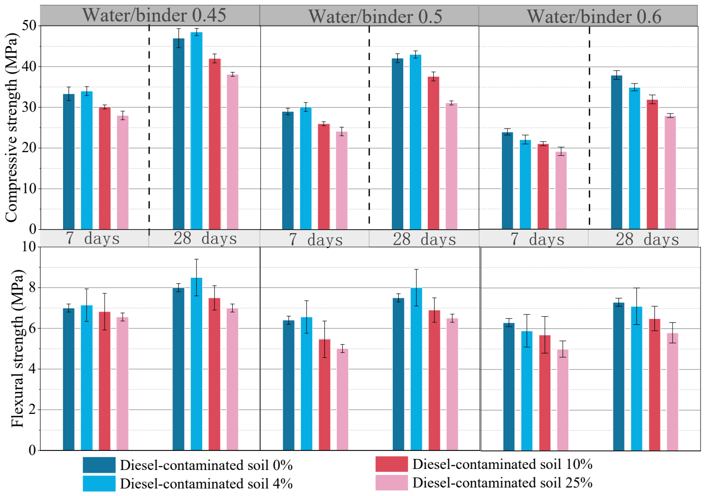


Fig. 13 Compressive and Flexural strength of diesel-contaminated soil, oil/soil ratio of 0.1

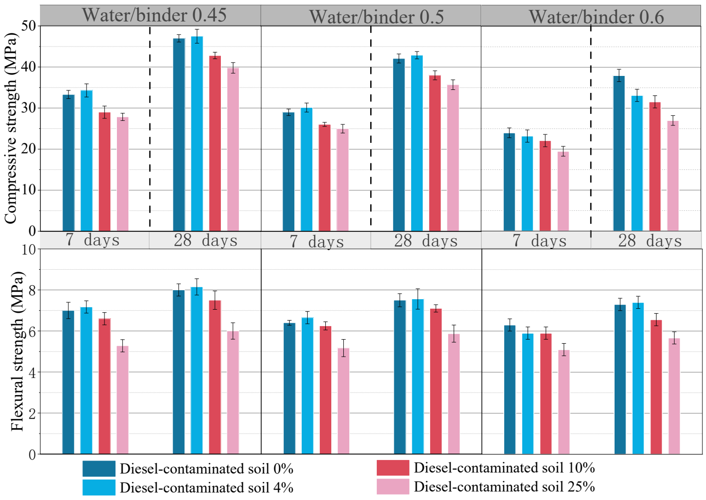


Fig. 14. Compressive and Flexural strength of engine oil-contaminated soil, oil/soil ratio of 0.1

However, the effect of engine oil-contaminated soil on the strength of mortar is similar with diesel contaminated soil. As shown in Fig. 14, the highest strength of mortar exists in which 4% engine oil-contaminated soil was added into mortar of water/binder 0.45.

The addition of 4% oil-contaminated soil showed a positive effect on the compressive strength and flexural strength of mortar. That is because oil-contaminated soil particles act as nucleation sites and a hydration production were crystallized on their surface. And after small amount of oil-contaminated soil was added into mortar, soil samples filled the vacuum of the matrix that can make the matrix denser and improve the strength of mortars. But with the increase of oil-contaminated soil, dilution of cement and many huge pores causing by oil-contaminated soil caused the matrix uncompacted and reduced the compressive and flexural strength.

3.4 The leaching values

According to the previous research reported, it is known that the larger the content of oil-contaminated soil, the worse the performance of the test pieces in all aspects. In order to evaluate the effect of cement-based materials stabilizing oil-contaminated soil, the proportion of solid leaching test was selected as Table 4, in which the content is the ratio of the admixture to cement. According to the method mentioned previously, oil leaching test of consolidation substance was carried out to test the oil content at each ratio, and the curing stability of cement-based material curing system was analyzed and studied. The specific test results are shown in the Table 4.

Table 4 The leaching values

NO.	W/B	Soil dosage	Oil dosage	Sand/binder water	Oil leaching（mg/L）	Notes.	China standard value
D1	0.5	0.00%	1.00%	3.00	0.95	Diesel	5 mg/L
D2	0.5	25.00%	1.00%	3.00	1.26	Diesel	
D3	0.5	25.00%	2.50%	3.00	2.12	Diesel	
E1	0.5	0.00%	1.00%	3.00	1.31	Engine oil	
E2	0.5	25.00%	1.00%	3.00	1.65	Engine oil	
E3	0.5	25.00%	2.50%	3.00	2.97	Engine oil	

By comparison and analysis of the test results in the Table 4, it is found that the oil leaching value of the solids in each group is lower than the standard requirement of 5mg/L, which indicates that it is feasible to use cement-based materials to solidify oil-contaminated soil.

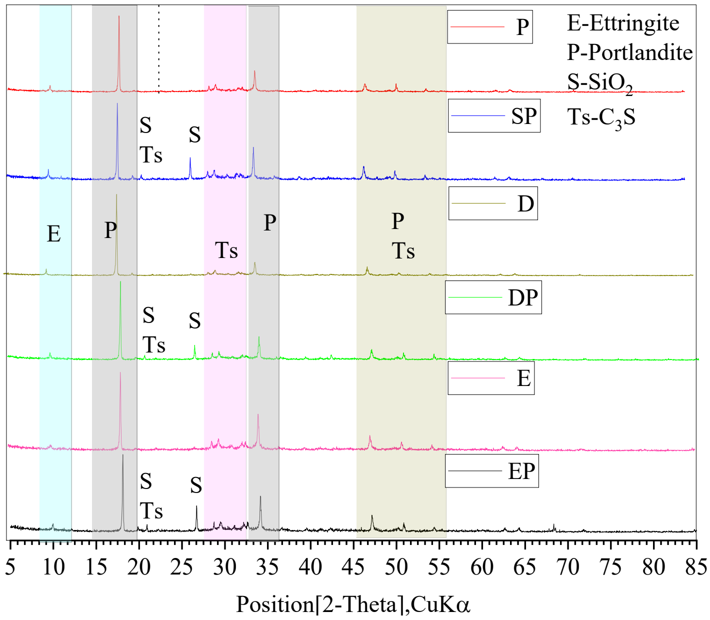


Fig. 15. The XRD pattern of cement pastes with oils and soil

According XRD pattern in the Fig.15 and the SEM in the Fig.16, it is clear that no new hydration products generate and physical sealing is the mechanism of solidify oils contaminated soil by cement-based materials, with many oil vacuole and soil particles enclosed by oils and no new product exist in the hardened cement-paste.

Fig. 16. SEM of hardened cement paste(×100）

The effect of oil-contaminated soil on the performance of cement-based materials are not always negative. Cement-based materials have a good solidification effect with oil-polluted soil. Appropriate cement content for oil-polluted soil of different degrees can not only make use of oil-polluted soil, but also solidify pollutants to realize environmental protection and economic benefits.

4. Conclusions

The effect of oil-contaminated soil on the performance of cement-based materials are not always negative. Cement-based material shows a good availability of oil contaminated soil. This study indicated that solid waste such as oil-polluted soil can be used in cement-based material to improve the performance such as consistency and strength in the appropriate methods and enhance environmental protection and benefits. Based on the results and discussion, the following conclusion can be drawn:

1) Oil-contaminated soil increased the total heat of hydration of unit mass cement and rate of heat release of unit mass of cement in the early development stage;
2) Oil-contaminated soil increased the yield stress and plastic viscosity of cement paste and decreased slump values of cement mortars;
3) Small amount of oil-contaminated soil in mortars (w/c 0.4 and 0.5) improved the compressive and flexural strength. However, when the w/c 0.6 was used, it reduced the compressive and flexural strength of mortars with the addition of oil-contaminated soil;
4) Cement-based materials have a good solidification effect on oil-polluted soil, which realized the resource utilization oil-contaminated soil. There are no new hydration products generated and physical mechanical sealing is the mechanism of solidify oils contaminated soil by cement-based materials.

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