SSRG International Journal of Civil Engineering Volume 6 Issue 4, 39-45, April 2019
ISSN: 2348 – 8352 /doi:10.14445/23488352/IJCE-V6I4P108 © 2019 Seventh Sense Research Group®

Investigation of the Influence of Light Crude Oil on the Performance of Cement Mortar

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Abstract

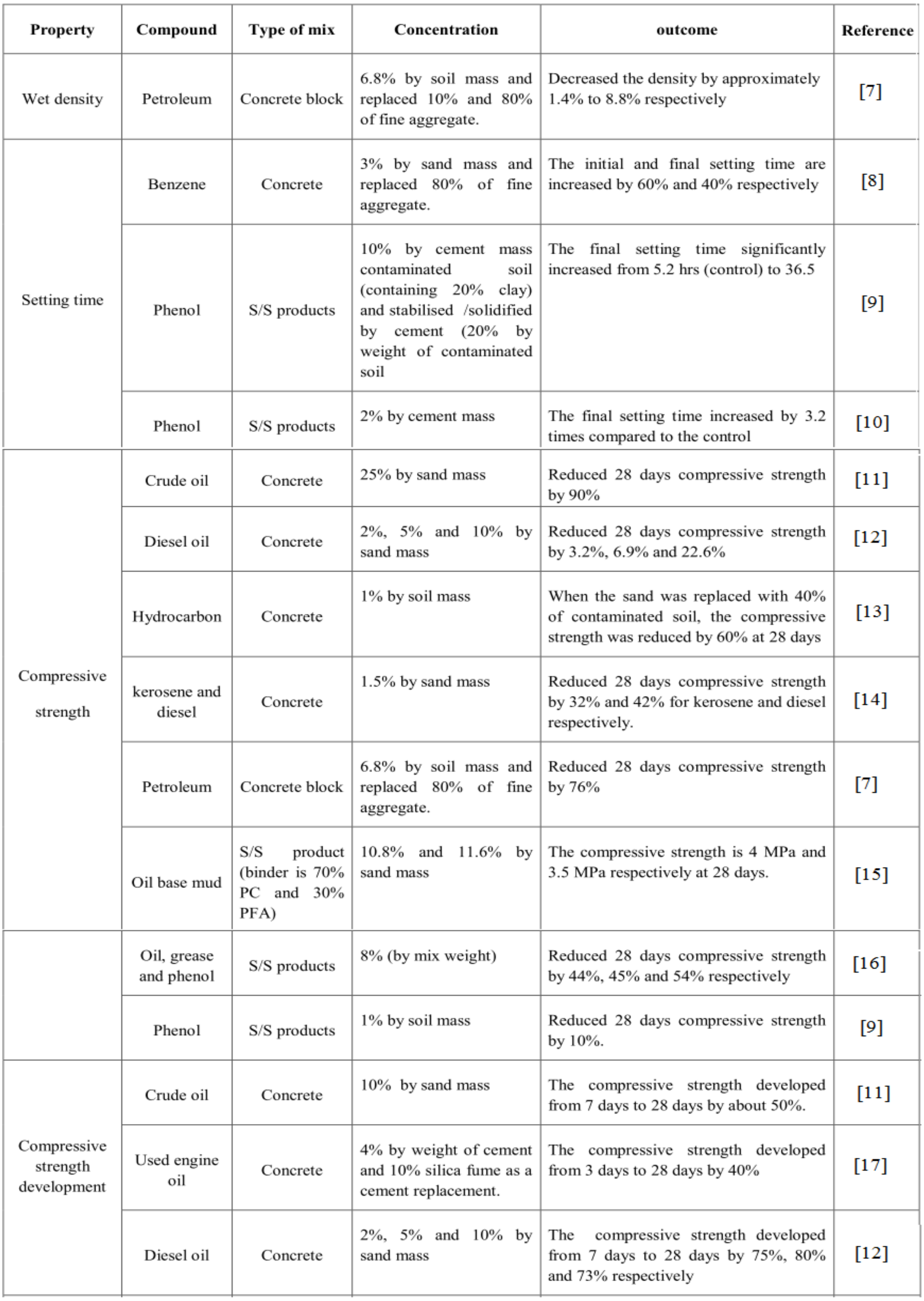
Spillage and leakage of hydrocarbon from petroleum activities have continually increased and are recognized as important contamination sources. The management of hydrocarbon contaminated fine-grained material is a major environmental concern. These hazardous materials have been regulated by legislation and have become one of the pressing needs confronting the petroleum industry. Current treatment technologies are either cost-prohibitive and/, or the treated products have to be sent to the landfill without any potential end-user, thereby rendering these solutions unsustainable. Cement-based stabilization/solidification (S/S) is an emerging technology; however, there is currently quite limited knowledge on the Effect of oil on the cement solidification process and its consequent effect on the performance of the resultant cementitious mix. The appropriate end-use scenario for cement-based mortar is examined when incorporating light crude oil and 10% aggregate mass. Calorimeter study indicated that the introduction of light crude oil into cement mortar has an impact but no discernible dramatic change. It caused some hydration inhibition but little retardation. Increased light crude oil fraction in the cement mortar increased setting time and decreased air content. The compressive strength decreased by ~50 % compared to the control at 28 days age. However, since the compressive strength covered a wide range of values, various feasible end-use scenarios for oil-contaminated mortar exist.

Keyword - *Light crude oil, mortar, compressive strength, stabilization, solidification.*

I. INTRODUCTION

The oil industry provides essential products that can be major contributors to enhance the quality of our life. However, oil contamination is a major environmental concern worldwide. The volume of oil-contaminated materials generated at a particular site has continually increased, and in some places, is not consistently monitored and may not be monitored at all. Hydrocarbon is common contamination associated with the petroleum industry, and it can arise due to oil spill, leaking of petroleum From above and ground storage tanks, drilling and production operations, and pipeline failure. Recent environmental legislation has imposed restrictions on the release or discharge of these toxic materials. As a result, effective management ways are needed [1]. Although there are several treatments and management techniques for oily wastes (e.g., thermal methods, biological treatment, and subsurface disposal), most of these techniques have either drawbacks in terms of financial cost or their Effect on the environment. Furthermore, the final application for treated products is most often not intended for re-use [2]. Cement-based stabilization/solidification has emerged as a viable alternative technology due to its improvement of petroleum wastes' physical and mechanical properties by binding the oily materials in a structure formed by cementing pozzolanic materials chemically and physically stable and mechanically handle products [3]. The ability to withstand mechanical stress in compressive strength is one of the most important aspects of stabilized/solidified products. Compressive strength is linked to hydration reactions in the mortar and durability of the monolithic S/S material. It can be used as an indirect method to determine the extent to which the waste has been chemically transformed into a monolith. There is also a link between compressive strength and leach-ability due to air content and permeability. Lower compressive strength will lead to greater permeability and, therefore, greater leaching potential. Higher air content will decrease compressive strength and increase leaching [4], [5]. The cement-based stabilization/solidification process's effectiveness has been highly questionable, and its applicability to organic waste has been controversial [6]. This is mainly because little is known about the mechanism responsible for the immobilization of organic contaminants in S/S materials and the subsequent strength development and leachability. There are scattered studies into the Effect of organic compounds on the properties and behavior of cementitious materials. Table I summarises the different outcomes from some of these studies, which use concentrations equivalent to those used in this study.

Table I: Past observation on the Effect of organic materials on different properties of cementitious materials



Property	Compound	Type of mix	Concentration	outcome	Reference
Wet density	Petroleum	Concrete block	6.8% by soil mass and replaced 10% and 80% of fine aggregate.	Decreased the density by approximately 1.4% to 8.8% respectively	[7]
Setting time	Benzene	Concrete	3% by sand mass and replaced 80% of fine aggregate.	The initial and final setting time are increased by 60% and 40% respectively	[8]
	Phenol	S/S products	10% by cement mass contaminated soil (containing 20% clay) and stabilised /solidified by cement (20% by weight of contaminated soil	The final setting time significantly increased from 5.2 hrs (control) to 36.5	[9]
	Phenol	S/S products	2% by cement mass	The final setting time increased by 3.2 times compared to the control	[10]
Compressive strength	Crude oil	Concrete	25% by sand mass	Reduced 28 days compressive strength by 90%	[11]
	Diesel oil	Concrete	2%, 5% and 10% by sand mass	Reduced 28 days compressive strength by 3.2%, 6.9% and 22.6%	[12]
	Hydrocarbon	Concrete	1% by soil mass	When the sand was replaced with 40% of contaminated soil, the compressive strength was reduced by 60% at 28 days	[13]
	kerosene and diesel	Concrete	1.5% by sand mass	Reduced 28 days compressive strength by 32% and 42% for kerosene and diesel respectively.	[14]
	Petroleum	Concrete block	6.8% by soil mass and replaced 80% of fine aggregate.	Reduced 28 days compressive strength by 76%	[7]
	Oil base mud	S/S product (binder is 70% PC and 30% PFA)	10.8% and 11.6% by sand mass	The compressive strength is 4 MPa and 3.5 MPa respectively at 28 days.	[15]
	Oil, grease and phenol	S/S products	8% (by mix weight)	Reduced 28 days compressive strength by 44%, 45% and 54% respectively	[16]
	Phenol	S/S products	1% by soil mass	Reduced 28 days compressive strength by 10%.	[9]
Compressive strength development	Crude oil	Concrete	10% by sand mass	The compressive strength developed from 7 days to 28 days by about 50%.	[11]
	Used engine oil	Concrete	4% by weight of cement and 10% silica fume as a cement replacement.	The compressive strength developed from 3 days to 28 days by 40%	[17]
	Diesel oil	Concrete	2%, 5% and 10% by sand mass	The compressive strength developed from 7 days to 28 days by 75%, 80% and 73% respectively	[12]

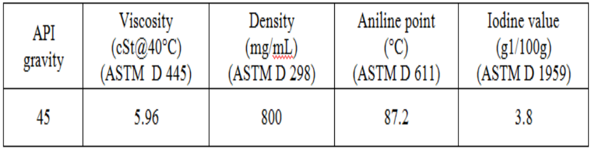
The presence and levels of oil contaminants in the cementitious materials are among the most important factors affecting the compressive strength [18]. The compressive strength that can be obtained in S/S material will determine its suitability for various end-uses. A minimum compressive strength of 350 Pa is considered adequate for bonding [19]. The recommended compressive strength at 28 days for materials that are to be disposed of in a landfill is 0.35 MPa under a provision listed in the United States Environmental Protection Agency (USEPA) guidelines and 1.0 MPa in the France and Netherlands [20]. However, a higher compressive strength value of 3.5 MPa in a sanitary landfill according to Wastewater Technology Centre (WTC), Canada [21]. A minimum compressive strength of 5.2 MPa at 28 days curing is described by International Masonry Institutes, the USA, for masonry's mortar [22]. Furthermore, under the UK department of transport, a minimum of 7 days cube compressive strength ranging between 5 and 15 MPa is required [23]. According to Ontario provincial standards, a minimum of 30 MPa at 28 days of curing is required to construct sidewalks [24]. This research aims first to understand the impact of incorporated light crude oil (LCO) generated from petroleum processes on the cement solidification process and secondly, to establish its effect on the resultant mortar's properties. This knowledge is needed to determine the appropriate end-use of these materials.

II. MATERIALS AND METHODS

A. *Materials*

ASTM type 1 cement (Cement Australia), which meets general purpose (GP) requirements [25], was used. The fine aggregate used was that of Calga double washed sand (Rocla Quarry Products Pty Ltd) with an absorption capacity of 0.65%, a specific gravity of 2.57, and a median particle size of 0.5mm. The water source for use as drinking water standard (pH 7.4; 2.29 µS/cm: Total dissolve salt 131 mg/L). Glenium, a polycarboxylate Ether polymer-based high-range water-reducing admixture (HWR) (BASF Construction Chemicals Pty Ltd), was used. The light crude oil (LCO) was supplied by Wildcat Chemical Australia Pty Ltd at Imdex Group from Kenmore oil field, Queensland, Australia, and analyzed at Oil Check Laboratory service, Sydney, Australia. The metals in the LCO were only Sulphur (202 mg/kg), sodium (4 mg/kg), and Nickel (2 mg/kg). The properties of LCO are illustrated in Table II.

Table II: Properties of Light crude oil



API gravity	Viscosity (cSt@40°C) (ASTM D 445)	Density (mg/mL) (ASTM D 298)	Aniline point (°C) (ASTM D 611)	Iodine value (g1/100g) (ASTM D 1959)
45	5.96	800	87.2	3.8

B. *Mix Proportions*

The composition of the mortar was in accordance with [26], with the mix proportions being 1 part of cement and 3 parts of sand (by mass) at a fixed water/cement ratio (w/c) of 0.50. Each mortar batch comprised cement (225 g), fine aggregate (675 g), water (112.4 g), HWR (0.1mL) with between 0 to 67.5g of added LCO, which has been reported as % by sand mass.

C. *Preparing, Casting, and Curing of Test Specimens*

The mixing methodology followed the procedure outlined in ASTM C 305 [27] using the Hobart mixer (model N-50 G) except when adding LCO. LCO was weighed (% by sand mass), added to the fine aggregate in a plastic bowl, and then premixed thoroughly using a spatula for 5 to 7 minutes before being added to the other ingredients. All laboratory work was conducted at 22 ± 2 °C. HWR has been used with all mixes to give reproducible flow (60 ± 10%) proven to be most suitable for proper consolidation of specimens by hand. The protocol for molding the mortar ASTM C109/C109M [28] was adopted and modified to minimize any impact of the protocol on any subsequent leaching tests. No mold release agents were used. Instead, the mold was lined with a non-stick tape. The cube molds were sealed using zip-lock plastic bags to prevent water from evaporating and stored in a moist atmosphere for 24 h utilizing a large plastic box. Demoulding took place after that, and specimens are having 50 x 50 x 50mm dimensions were sealed in zip lock plastic bags and thereafter placed in a curing tank filled with water for up to 28 days at a temperature of 22.0 ± 0.5 °C.

D. *Test Methods*

Both a semi-adiabatic calorimeter (F-Cal 4000) and an isothermal calorimeter (I-Cal 4000) from Calmetrix Incorporation (USA) were used for monitoring the temperature and heat evolution of the mortar mixes following the procedures outlined in the instrument manual. The reported calorimetric data was an average of triplicate samples taken from a single batch with an inter-sample standard deviation of less than 1%. The fresh mortars were tested in accordance to ASTM C 1437 [29] for flow, ASTM c 138 [30] for wet density, air content (TESTING Bluhm & Feuerherdt GmbH, ASTM C 231 [31] and ASTM C 807 [32] for setting times (H-3085 Humboldt Vicat Tester). Compressive strength was tested using ASTM C109/C109M [28] test method requirements and a compliant compression testing machine. A vertical load rate of 1.5 kN.s^{-1} was applied to each specimen until failure. The maximum loads recorded for three individual test specimens were taken to represent the average compressive strength of each set of mortars tested.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. *Chemical Characteristics of LCO*

The aniline point is used to provide an estimate of the aromatic hydrocarbon content of mixtures. Aromatic hydrocarbons exhibit the lowest aniline point values, while cycloparaffins and olefins have intermediate values and paraffins the highest values. The iodine value of oil is a measure of the saturation of the fatty acids. As the value increases, there are more double bonds and consequently more unsaturated hydrocarbons. LCO has a significant amount of saturated bonds, as evidenced by a high aniline point and low iodine value (Table II).

B. Effect of LCO on Hydration Process

The LCO shows peak temperature to be relatively uniform but small with a small increased time to peak (at the content of 10%). Therefore, cement hydration is both inhibited and retarded but not significantly (Fig.1a). Several trends may be seen from the LCO heat evolution data (Fig.1b). The mixes containing 0% and 2% LCO have similar second peak (S) times and values. However, their third peak (T) times and values differ. This shows C_3S hydration is not impacted at this concentration, but C_3A is impacted. The mixes containing 6% and 10% LCO have both inhibition and accelerated C_3S (peak S) and inhibited and retarded C_3A (peak T). This shows that the low concentration of LCO initially impacts the only C_3A, but at a higher concentration, C_3S is also impacted. However, the cumulative heat evolution (Fig.1c) shows that although there are differences between the different LCO fractions, the cement inhibition effects are relatively small.

C. Effect of LCO on Flow and Wet Density

The control mix without HWR was below the requirement flow value. It is inferred from the results that the addition of LCO shows a gradual increase in flow (Fig.2). This increase in flow for mixes containing oil is similar to other studies reported in the literature [33],[34],[35]. Wet densities were found to decrease with increasing LCO addition levels (Fig.2). This reduction can be attributed to the oil-free mortar (2270 kg/m^3) being replaced by lower density LCO (800 kg/m^3) when it is placed in a mould of a fixed volume. The Effect of density decreases has been noted in another study [34].

D. Effect of LCO on Air Content

LCO can alter the air content of fresh mortar as plotted in Fig.3. As a general trend, the percentage of air content decreases with increased LCO content. The air content may decrease with the increasing LCO content because of mixing related phenomena. However, this is still speculative and requires further work. For the control mortar (0% LCO), the air content obtained was 6.4 %. This value is higher than in the literature. Other studies have shown that the typical air content obtainable for non-air entrained mortar is lower than 3% [36-38]. This difference in the air content values can be attributed to the methods used for measuring air [18]. An air meter gives a higher value for air content than a calculated one [39].

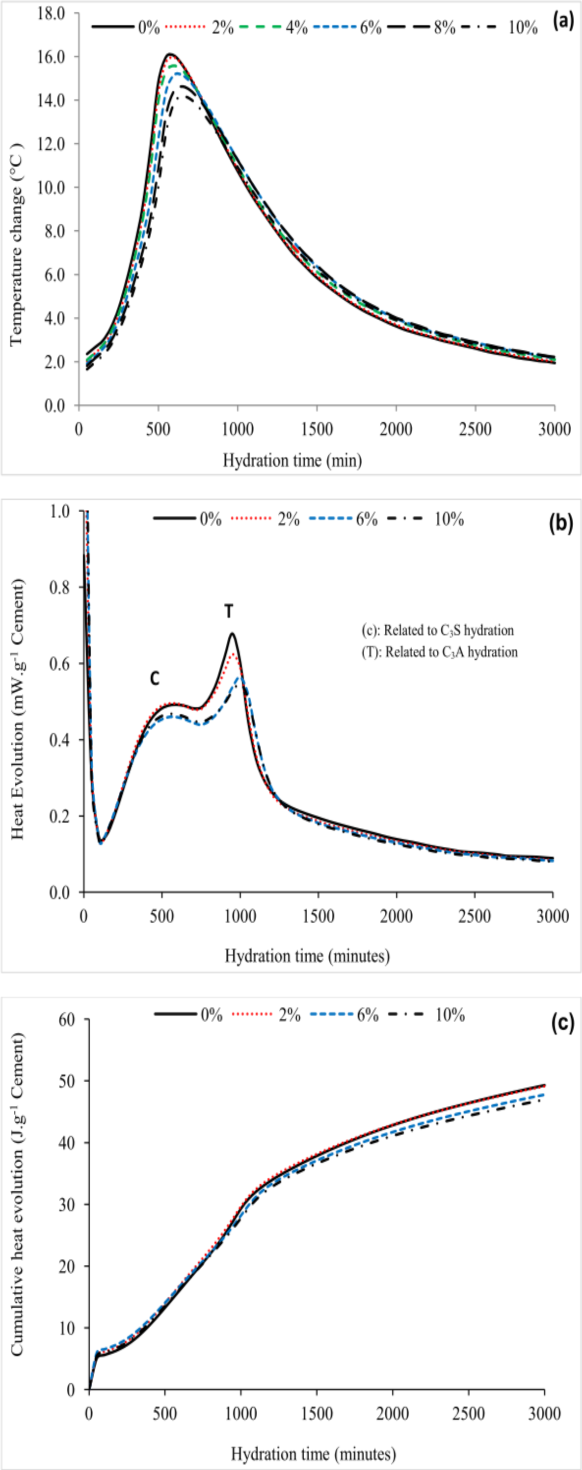


Fig 1: Thermal profile for the hydration of mortar containing LCO, a) Temperature evolution by the semi-adiabatic calorimeter, b) Heat evolution by the isothermal calorimeter, c) Cumulative heat evolution by the isothermal calorimeter

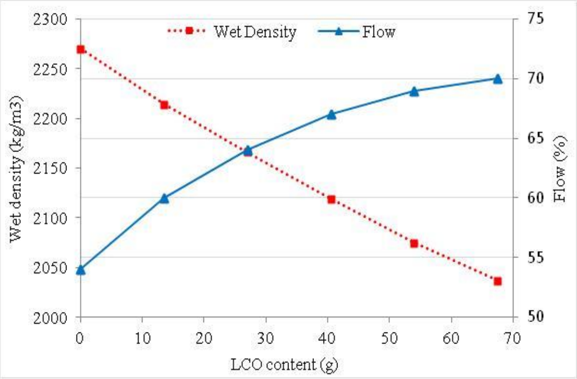


Fig 2: Effect of LCO on flow and wet density

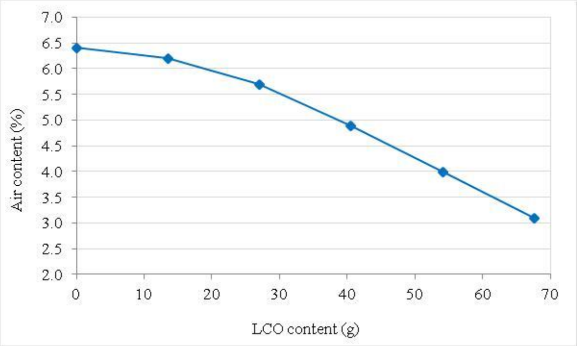


Fig 3: Effect of LCO on-air content

E. *Effect of LCO on Setting Time*

In general, the mortars' setting time was observed to increase with the increase in LCO contents (Fig. 4). This increase may be attributed to LCO coating the cement particles and retarding their hydration by hindering the formation of cement hydration phases and therefore delaying the setting of the mortar. Other studies have also demonstrated that setting time has increased when incorporating oily materials [40]. This previous study reported that the effect of organic compounds on cement hydration is variable and highly concentration-dependent.

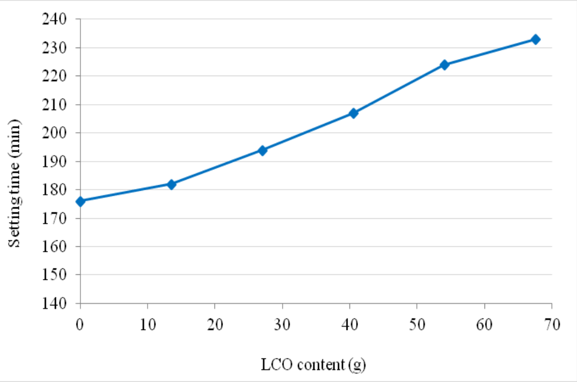


Fig 4: Effect of LCO on setting time

F. *Effect of LCO on Compressive Strength*

The compressive strength behavior of the specimens incorporating LCO at 1, 7, and 28 days curing time was quite different from that without LCO (Fig.5). From the results, it was observed that all mixes follow a similar trend where higher LCO contents in mortars resulted in a reduction in compressive strength. The percentage reduction in compressive strength of mortar incorporating LCO in relation to the control is illustrated in Fig.6. The compressive strength decreased by 38.8%, 58.1%, and 49.9% at 1, 7, and 28 days ages, respectively, compared to the control when mortar is incorporated 10% of LCO. Reduction in compressive strength of cementitious-based materials due to the incorporation of oily materials has previously been reported[11],[12]. Reference [18] stated that oil nature makes a significant difference to the strength achieved in the mortars and the mechanisms causing strength loss.

Further knowledge is needed about these strength loss mechanisms and, in particular, oil characterization methods that can identify these critical chemical properties. The development rate of all mixes is relatively high up to 7 days, followed by a slower rate between 7 and 28 days (Fig.5 and Fig.7). The little strength development from 7 to 28 days suggested that all strength development after 7 days was mainly due to the strength forming phases present at 7 days and that the mechanisms causing CSH inhibition at 7 days were still occurring at 28 days [18]. From the point of view of stability, results suggest that the solidification of LCO yields adequate compressive strength for many end-uses. Although with the addition of 2% of LCO, the compressive strength at 28 days reduced to 38.4 MPa, this value remains higher than the minimum value specified for use in the construction of sidewalks given by Ontario provincial standards for roads and public roads. Even with 10% LCO content, the compressive strength noted at 7 days (18 MPa) still complies with use in sub-base and base materials. Indeed, the incorporation of 10% of LCO leads to a compressive strength at 28 days of 23.1 MPa. Even though this strength is 50% of the control mix, it remains acceptable for oil-contaminated landfills and used in masonry rendering systems.

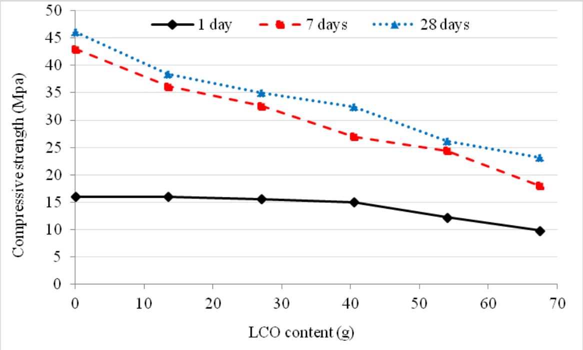


Fig 5: Effect of LCO on compressive strength

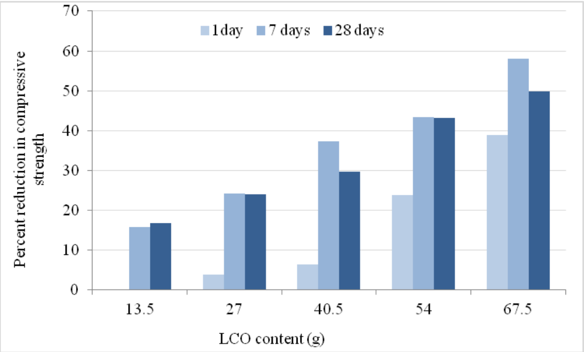


Fig 6: Percent reduction in compressive strength

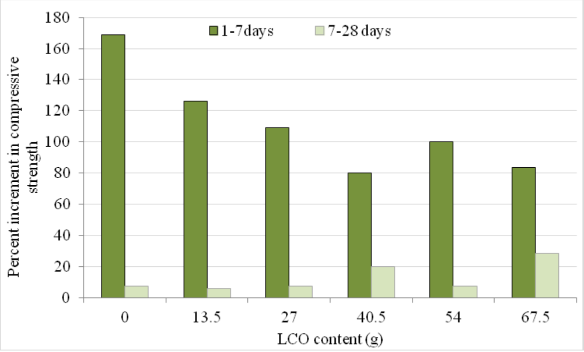


Fig 7: Percent increment in compressive strength

IV. CONCLUSIONS

Increased LCO content in mortar led to increased reaction inhibition (lower peaks) and delayed reaction but not significantly. There were changes to the hydration of various hydration phases (C_3S, C_3A, and C_2S). As a result, there were changes to the compressive strength and setting time. In general, the mortars' setting time was observed to increase with the increase in LCO content. The incorporation of LCO into mortars was found to decrease compressive strength. The reduction in compressive strength is a function of LCO concentration, where the higher concentration, the higher the strength reduction. However, the observed compressive strengths covered a wide range of values indicating various feasible end-use scenarios were still possible for LCO impacted mortar. Therefore, while stabilization of oil-contaminated material resulting from the oil industry or contaminated sites appear to be a viable method of treating oil-contaminated materials, fundamental knowledge is still needed to establish relationships between oil content and mortar performance. This is necessary for the development of construction materials with suitable characteristics for re-use rather than disposal. Further investigation is needed on both the permeability, leach-ability behavior of stabilized mortars to understand better the causes of decreased strength and the potential release of oil from such mortars.

ACKNOWLEDGMENT

The authors gratefully acknowledge the Libyan Embassy's financial support in Australia for this research to proceed. The authors also acknowledge the support and cooperation provided by Mr. Rami Hadad, Manager, Civil laboratories at the University of Technology, Sydney. The authors would also like to extend their special gratitude to Mr. Neil Mahoney, Sales Manager at Wildcat Chemicals, and Mr. Shaun Wilson, Senior Operations Coordinator at AMC Oil & Gas (Imdex Group), for supplying the crude oil required for this study.

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