Indian Journal of Engineering & Materials Sciences
Vol. 19, April 2012, pp. 79-86

Copper tailings as a potential additive in concrete: consistency, strength and toxic metal immobilization properties

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Received 27 December 2011; accepted 23 March 2012

This paper presents the results of a study investigating the consistency, hardened and toxic metal immobilization properties of concretes containing copper tailings as an additive. To compare the effects of copper tailings on strength and strength related properties across two concrete classes, two series of concretes with 0.57 and 0.50 water-to-binder ratios are used. For each series, three mixtures incorporating copper tailings at 0%, 5%, and 10% addition levels by mass are prepared. Copper tailings have a slight negative impact on the slump, setting time and porosity of mixtures. However, improved mechanical strengths and abrasion resistance, and reduced chloride penetration compared to the control specimens are observed in mixtures incorporating copper tailings. Toxicity characteristic leaching test results revealed that the release of heavy metals from mixtures containing copper tailings is considerably lower than the United States Code of Regulations limits. Overall, it seems that there is a potential for the use of copper tailings as a zero-cost, environmentally-friendly additive in concrete, especially at a 5% addition level.

Keywords: Copper tailings, Concrete, Mechanical strength, Water absorption, Chloride resistance, Leaching

The disposals of waste materials arising from increased industrial activities are causing a serious waste management problem all over the world. Thus far, the construction industry, a major contributor to this predicament has been playing a role in the quest to recycle these waste materials efficiently. Several research studies have comprehensively shown that industrial by-products like silica fume, coal fly ash, and ground granulated blast furnace slag can be used in producing durable concretes[1,2]. Utilization of these industrial by-products as aggregates and cement replacement materials in concrete presents important fiscal and environmental benefits such as savings in disposal cost, the use of a zero-cost raw material and a cleaner environment.

Copper is one metal with wide applications in several industries. Hence, with the increasing demands for copper and copper allied products, it is expected that more ores will be processed to meet supply. Unfortunately, this implies that additional millions of tons of copper processing wastes will be added to the existing deposits in most major copper producing countries of the world. The massive waste generation going on in the copper industry, was highlighted by Boger[3] when he submitted that about 230,000 tons of dry copper tailings is produced daily at the Escondida copper mine in Chile. These copper tailings contain toxic heavy metals which pollute the environment. Water and soil contamination at abandoned copper mines and waste sites were reported by Benvenuti *et al.*[4] and Yukselen[5].

Over the years, severe environmental impacts have also been associated with copper tailings dam failures. According to Grimalt *et al.*[6], approximately 2 million m³ of mud containing heavy metals were spread over 4286 ha of land and surface water during the 1998 Aznalcollar tailings pond failure in Spain. Similarly, Destouni[7] suggested that about 1.6 million m³ of tailings impoundment water was released to surrounding waterways during the year 2000 Aitik copper tailings dam failure in Sweden. Lungu[8] reported that the 2006, tailings spillage at the Nchanga copper processing plant, Zambia, released high concentrations of heavy metals into the nearby surface water, thereby contaminating the local source of water supply. Loss of lives has also been recorded during severe tailings dam failures. The death of 54 people during the 1928 Barahona copper tailings dam failure in Chile was reported by Harder and Stewart[9]. Likewise, Barrerra *et al.*[10] were of the opinion that about 300 lives were also lost in Chile, during the 1965, El Cobre Dam failure. The death of 89 persons

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during the 1970 Mufulira copper tailings dam collapse in Zambia, was highlighted by Blight and Fourie[11]. Hence, given these aforementioned menaces associated with the disposal of copper processing wastes, devising methods for the utilization of these by-products as valuable and useful materials in concrete is necessary.

Research findings from studies on the recycling of one of the copper processing wastes, copper slag, have shown that there is a significant potential for its utilization as a cementitious material. Increases in compressive strengths were recorded by Tixier *et al.*[12] in mortar samples containing copper slag. In concrete mixtures, Al-Jabri *et al.*[13] noticed that a 5% partial replacement of cement with copper slag yielded a marginally lower but similar performance as the control concrete mixture. Similarly, Moura *et al.*[14] observed mechanical strength enhancement in concretes containing copper slag as a concrete additive. Concerns about the fate of toxic metals contained in copper processing wastes have also been expressed. However, various studies have shown that heavy metals can be effectively stabilized and solidified in cement-based materials. Park[15] investigated the stabilization of heavy metals in OPC, clinker kiln dust (CKD) modified OPC, and quick setting agent (QSA) modified OPC. He observed lower leaching of heavy metals in the blended mixtures. Studies by Giergiczny and Krol[16] showed that OPC, fly ash and GGBFS blended mortar mixtures can successfully immobilize heavy metals. Shi and Kan[17] submitted that the leaching of heavy metals from cement paste samples containing MSWI fly ash was below the Chinese regulatory limits. Choi *et al.*[18] observed effective immobilization of heavy metals in mortars containing tungsten tailings, especially in mixtures containing binary blend of the waste and GGBFS. Similar results were also obtained in related studies on the leachability of heavy metals from copper slag blended mortar mixtures. Heavy metal concentrations in leachates lower than those of the regulatory limits were recorded in mortars containing copper slag by Zain *et al.*[19] and Mesci *et al.*[20] In a related study, Alp *et al.*[21] reported that the leaching of heavy metals from mortar samples produced with copper slag clinker cement was below regulatory limits.

This paper investigated the possibility of using copper tailings as an additive in concrete by considering consistency, mechanical strength, porosity and leaching behaviour of concrete mixtures incorporating it at 0%, 5% and 10% addition levels.

Experimental Procedure

Materials

The binders used in preparing paste and concrete mixtures in this study were Portland slag cement CEM III/A (Class 32.5N) and copper tailings obtained at various depths from deposits at an abandoned processing facility at Lefke, Cyprus. Tailings samples were air dried and sieved with a 600 µm sieve in order to remove agglomerated particles before usage. The fine and coarse aggregates were sourced from locally crushed limestone rock.

Physical and chemical properties of constituent materials

Physical properties such as the specific gravities of the cement and copper tailings were determined according to the ASTM C 188[22] specification. The specific surface areas were obtained using ASTM C 204[23] and Blaine's air permeability apparatus. The particle size distribution of the waste material was also determined using sieve analysis and the hydrometer method. Finally, an oxide analysis of the copper tailings was performed.

The specific gravities of the fine, coarse aggre gates and copper tailings were 2.70, 2.54 and 4.29, respectively. The high specific gravity of the copper tailings was attributed to the high concentration of iron (III) oxide in it. Similarly, the water absorption property of the tailings (13.8 %) was much higher than the 0.13-0.55 % recorded in the literature for copper slag by *Shi et al.*[24]. It is suspected that prolonged exposure to weathering might have also contributed to the increased porosity of the tailings. The particle size distribution curve of the copper tailings is shown in Fig. 1. The curve indicates that approximately 50% of the tailing particles are finer than 0.1 mm. Some of the physical and chemical properties of cement and copper tailings are also shown in Table 1.

Mixture proportions

The water-to-binder (w/b) ratios used for the setting time pastes were dependent on the findings from the consistency tests. To compare the effects of copper tailings at 0%, 5%, and 10% addition levels by mass of cement on strength and strength related properties across two concrete classes, 0.57 and 0.50 water-to-binder (w/b) ratio mixtures were also prepared. These mixtures were identified as C0 for control and C5 and C10 for mixtures containing copper tailings. For each mixture, six 150 mm cubes and six 100 mm × 200 mm cylinders were cast for the determination of compressive and splitting tensile strengths at 28 days and 90 days. Moreover, an additional four 100 mm × 200 mm cylinders and six 100 mm × 100 mm × 500 mm prisms were cast for each mixture. On the 90th day, four 100 mm ×52 mm cylindrical specimens for the porosity tests were cut from the middle of these four 100 mm diameter × 200 mm long cylinders, while twelve 50 mm cubes were also sawn from the prisms for abrasion and impact resistance tests. On completion of the casting operations, specimens were kept in the curing room for 24 h before they were then de-moulded and left in the room at a temperature of 23°C ± 2°C and humidity of 85% ± 5% until the time of testing. The mixture proportions are summarized in Table 2.

Table 1—Chemical and physical properties of cement and copper tailings

Component	Cement	Copper tailing
Chemical composition (%)		
SiO_2	29.15	11.20
Al_2O_3	7.34	-
Fe_2O_3	2.42	85.30
CaO	50.04	-
MgO	3.99	-
SO_3	1.97	-
Cl	0.01	-
Loss on ignition	1.65	-
Insoluble residue	0.27	-
Heavy metal content (mg/kg)		
Cu	-	2284
Zn	-	402
Pb	-	60
Cr	-	12
Cd	-	0.86
Physical properties		
Specific gravity	2.96	4.29
Blaine specific surface area (cm^2/g)	3440	537
Absorption (%)	-	13.82

Consistency

The slump of each mixture was determined according to the ASTM C 143[25] guideline, while the setting times of cement pastes incorporating different percentages of copper tailings were evaluated using the BS EN 196-3[26] specification.

Volume of permeable voids

The total permeable voids in concrete samples were determined after 90 days of curing and according to the specifications of ASTM C 642[27].

Compressive strength of concrete

Compressive strengths of concrete mixtures were determined after 28 days and 90 days of storage in the curing room using BS EN 12390-3[28] as a guide.

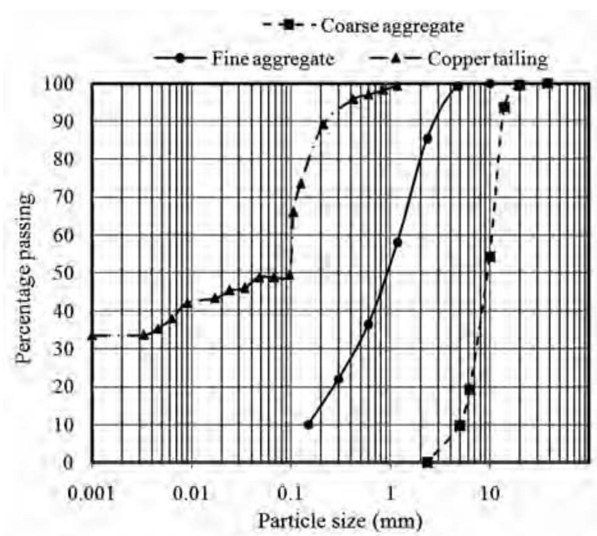

Fig. 1 – Particle size distribution of copper tailings

Table 2—Mixture Proportions

W/B ratio	Mixture name	Tailings (% by mass)	Quantities (kg) Water	Cement	Tailings	Fine	Coarse	Slump (mm)
0.57	C0	0	225	395	0	810	950	150
	C5	5	225	395	20	810	950	80
	C10	10	225	395	40	810	950	50
0.50	C0	0	225	450	0	818	887	140
	C5	5	225	450	22	818	887	75
	C10	10	225	450	45	818	887	50

Splitting tensile strength of concrete

Splitting tensile strengths of all the concrete mixtures were determined after 28 days and 90 days of storage in the curing room according to the ASTM C 496[29] guidelines.

Modified abrasion and impact resistance of concrete

The abrasion and impact resistance of concretes were evaluated using the Los Angeles abrasion apparatus, according to the ASTM C 535[30] specification. However, instead of the standard 1000 revolutions, test specimens and twelve 50 mm diameter steel balls were subjected to 500 rotations and the mass losses were determined.

Chloride penetration test

Actual penetration of chloride ions into concrete specimens was evaluated by immersing four 50 × 50 × 50 mm cubic specimens coated on all but one side in a 3% NaCl solution for 28 days. Thereafter, the specimens were split and sprayed with a 0.1N silver nitrate solution as suggested by Otsuki *et al.*[31] to determine the chloride penetration depths. These depths were identified as points within the samples where free chlorides above 0.15% by mass of cement reacted with the 0.1N silver nitrate ($AgNO_3$) solution to form a white precipitate of silver chloride (AgCl). The absence or limited availability of free chloride was signified by a brown precipitate of silver oxide (AgO) produced from the reaction of $AgNO_3$ solution and hydroxides in the concrete samples.

Leaching tests

The toxicity characteristic leaching procedure (TCLP) developed by the United States Environmental Protection Agency (US EPA) was used in evaluating the leaching of heavy metals from crushed concrete samples. The test was performed according to the US EPA Method 1311[32] whereby crushed concrete samples less than 9.5 mm in size, glacial acetic acid, 20:1 liquid-solid ratio and 30 rpm agitation for 18 h were used. Furthermore, the same test process was repeated using deionised water in lieu of glacial acetic acid. After the extraction and filtration of the leachates, heavy metal ions concentrations therein were determined by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS).

Results and Discussion

Consistency

Slump values in Table 2 shows that for the two w/b ratios, concrete slump decreased as the copper tailings content in the mixtures increased. This behaviour contrasts with the findings by Li and Wu[33] and Ferraris *et al.*[34] that the presence of ultra-fine particles of cement additives like fly ash in mixtures improve consistency. It equally contradicts the submissions by Al-Jabri *et al.*[35] that copper slag improves the consistency of concrete mixtures. The reduced slump witnessed in the blended mixtures were attributed to the coarse particles of the tailings and their high water absorption property (Table 2) which adversely affected consistency by partially absorbing available mixing water. Clearly, the 0.13-0.55% water absorption capacity of copper slag reported by *Shi et al.*[24] compared to the 13.8% of copper tailings is a major reason why the consistency of mixtures containing both materials are different. Furthermore, the effect of coarse particles on mixture consistency was buttressed by the submission of McCarthy and Dhir[36] that coarse fly ash reduces mixture workability. Similar opinion was expressed by Pan *et al.*[37] when they suggested that the reduced workability of mortar mixtures containing sewage sludge ash (SSA) is traceable to the porous and irregular morphology of SSA.

The setting time results shown in Fig. 2 highlighted the delay in setting time of pastes as the substitution level of copper tailings increased. This occurrence is partly due to delayed hydration induced by the heavy metal ions contained in the copper tailings. This postulation is supported by the assertion by Hashem *et al.*[38] that the presence of Cu (II) ions retard cement hydration. Zain *et al.*[19] were also of the opinion that the presence of Cu, Pb and Zn compounds in copper slag increased cement paste setting times. The poor fineness of the copper tailings as shown in Table 1 and Figure 1 could also have contributed in decreasing the overall reactivity of cementitious particles.

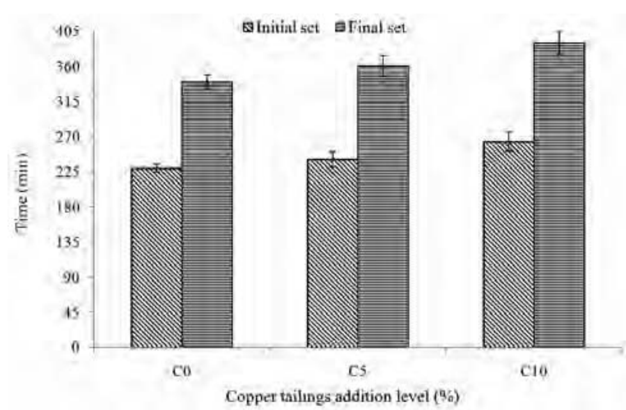

Fig. 2 – Setting time of pastes

Volume of permeable voids

The percentage volumes of permeable voids of concretes at the age of 90 days are shown in Fig. 3. For the 0.57 w/b ratio concretes, the volumes of permeable voids were 15.7% for C0, 16.4% for C5 and 18.1% for C10. Similar but slightly reduced porosity were equally observed for the 0.50 w/b ratio concretes, where the values were 15.5% for C0, 15.3% for C5, 15.7% for C10. The results from these two concrete series showed that these mixtures became slightly more porous as the copper tailings content increased. The increased void content of these mixtures incorporating copper tailings is attributed to the coarse and porous tailings particles. However, there is likelihood that this higher volume of voids in these concretes containing copper tailings might not have increased sample permeability, given that the fine particles of the tailings may have reduced overall pore interconnectivity.

Compressive strength

Figure 4 presents the relationship between compressive strength of the concrete mixtures and copper tailings addition levels. Figure 4a indicated that at a w/b ratio of 0.57, all the copper tailings concrete mixtures yielded higher compressive strengths at 28 days. The percentage compressive strengths relative to the control at 28 days were 107.1% for C5 and 104.1% for C10. Similarly, the relative strengths to the control at the 90th day were 112.3% for C5 and 109.2% for C10. Figure 4b highlighted similar but reduced strength increases for concrete mixtures with a w/b ratio of 0.50. The observed percentage compressive strengths relative to the control at the 28th day were 100.3% for C5 and 103.3% for C10, while those for the 90th day were also 100% for C5 and 100.9% for C10. Moura *et al.*[14] equally observed similar trend whereby copper slag addition enhanced concrete compressive strength and the rate of strength increments were higher for mixtures with high w/c ratios compared to those with lower ones. The observed increases in strengths of the 0.57 w/b mixtures could be partly traced to the slight reduction of the w/b ratio by water absorption by the porous tailings particles. It is equally suspected that the filler effect of fine particles of the tailings contributed to the strength enhancement witnessed. However, compared to the 0.57 mixtures, the impact of copper tailings on the compressive strengths of the 0.50 w/b ratio concretes was minimal. This was apparently a result of the reduced porosity of these mixtures which consequently, required less filling.

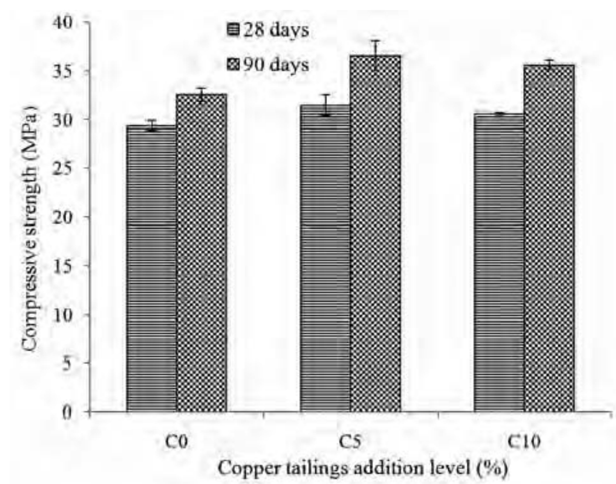

Fig. 4a – Compressive strengths of 0.57 w/b ratio concretes

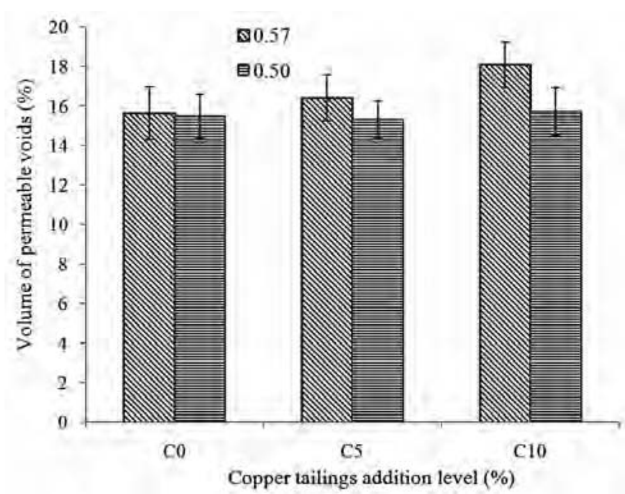

Fig. 3 – Volume of permeable voids in concretes

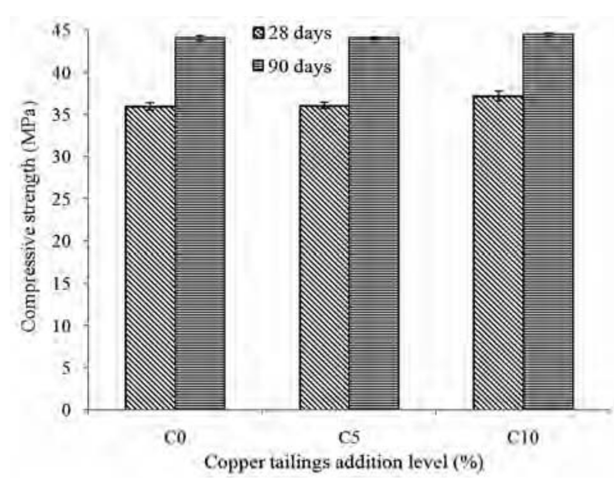

Fig. 4b – Compressive strengths of 0.50 w/b ratio concretes

Splitting tensile strength

Figure 5 shows the splitting tensile strength of mixtures prepared using the two w/b ratios. The 28 days and 90 days average tensile strength values of mixtures with a w/b ratio of 0.57 are presented in Fig. 5a. Higher splitting tensile strengths were recorded for concrete mixtures containing copper tailings as an additive. At the 90th day, the increases in splitting tensile strengths compared to the control samples were 22.5% for C5 and 16.2% for C10 samples. In contrast to the 0.57 w/b mixtures, Figure 5b indicated reduced strength developments for 0.50 w/b ratio mixtures. However, these strength values were still comparable to those of the control samples. The 90 day increases in splitting tensile strengths compared to the control specimens were 7.7% for C5 and 0% for C10 samples. It is suspected that the increased tensile strength observed in the copper tailings blended concretes could be as a result of enhanced bonding between aggregates and the cement paste engendered by the porous and small particles of the tailings. Improved tensile strength of concrete mixtures at low fly ash content and slightly reduced strength at higher volume of utilization of fly ash was reported by Lam *et al.*[39] They suggested that appropriate amount of fly ash enhances tensile strength of mixtures, through improved interfacial bond between paste and aggregates.

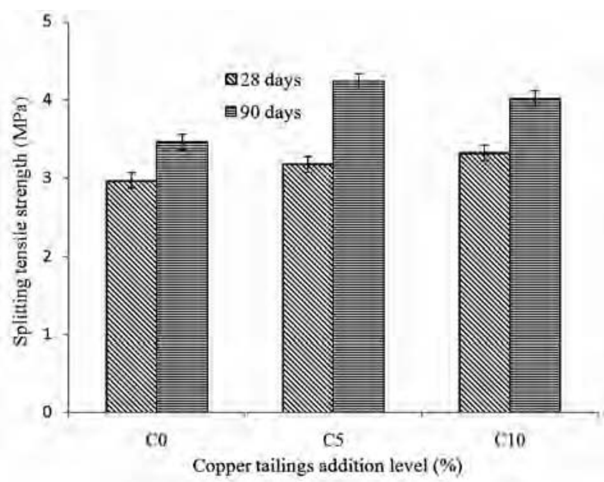

Fig. 5a – Splitting tensile strengths of 0.57 w/b ratio concretes

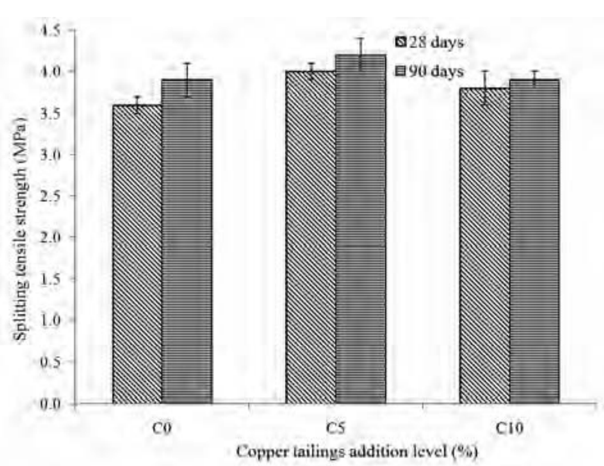

Fig. 5b – Splitting tensile strengths of 0.50 w/b ratio concretes

Modified abrasion and impact resistance of concrete

Abrasion and impact resistance of each mixture was assessed by the mass loss of 50 mm cubic concrete specimens after 500 revolutions in the Los Angeles Abrasion machine. Figure 6 showed that at a w/b ratio of 0.57, the C5 samples recorded the highest resistance to impact and abrasion, while for the 0.50 w/b ratio, the maximum resistance was obtained from the C10 samples. The mass losses of the 0.57 w/b ratio concrete samples were 76.6% for C0, 67% for C5 and 75.8% for C10. A similar trend was also observed in the 0.50 w/b ratio concrete samples where the mass losses were 70.2% for C0, 46.5% for C5 and 37.1% for C10. A more enhanced resistance to degradation was obtained from the 0.50 w/b ratio mixtures containing copper tailings. The improved resistance of mixtures containing copper tailings could be as a result of the higher compressive strengths of these samples compared to those of the control samples. Some studies suggested that abrasion resistance is strongly influenced by concrete compressive strength[40,41].

Chloride penetration test

The chloride penetration depths calculated for concrete specimens after 28 days of immersion in a 5% NaCl solution are presented in Fig. 7. These results showed that concrete mixtures containing

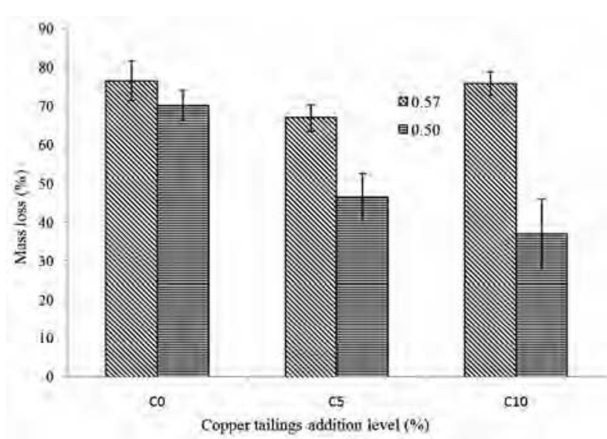

Fig. 6 – Abrasion and impact resistance of concretes

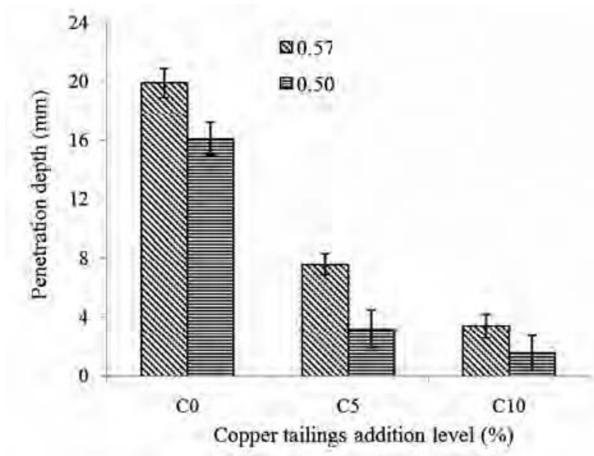

Fig. 7 – Chloride penetration depths in samples

copper tailings had enhanced chloride penetration resistance compared to the control samples. It was observed that the depth of chloride penetration progressively decreased with an increase in addition level. For the 0.57 w/b ratio mixtures, depths of penetration were 19.9 mm for C0, 7.6 mm for C5 and 3.4 mm for C10 samples. Similarly, the depths of penetration for the 0.50 w/b ratio mixtures were 16.1 mm for C0, 3.2 mm for C5 and 1.6 mm for C10 samples. The reduced depths of chloride penetration witnessed in these copper tailings blended samples were attributed to reduction of w/b ratios due to the absorption of mix water, and filler effects of the fine particles of the tailings which induced pore refinement in samples. Hence, it is suspected that the discontinuous pore structure formed, significantly reduced the ingress of chlorides in samples. We also suspect that chemical reaction between chloride and Pb contained in these tailings may have also influenced test outcomes.

Heavy metals leachability

Some of the US Code of Federal Regulations CFR[42] leachate quality criteria are shown in Table 3, while the concentrations of heavy metals in leachates are presented in Table 4. In comparison to deionised water, the use of acetic acid as an extractant yielded higher concentrations of heavy metals in leachates. Test results also suggest that w/b ratio has a considerable impact on the release of heavy metals from concrete. Thus, of the two w/b ratios evaluated, lower concentrations of heavy metals in leachates were obtained from concrete mixtures with a 0.50 w/b ratio. This occurrence could be attributed to the denser microstructure of the 0.50 w/b mixtures, which reduced the permeability of samples. Moreover, it also appears that the release of heavy metals from samples on exposure to the two leachants decreased with increase in copper tailings content of concrete. This is probably as a result of improved pore refinement and the consequent reduction in concrete permeability induced by fine particles of the tailings. Overall, these leaching test results indicated clearly that the leaching of selected heavy metal ions from concretes containing copper tailings were very low and did not exceed the CFR limits shown in Table 3. Similar observations were made in related studies by Zain *et al.*[19] and Alp *et al.* [21].

Table 3—U S CFR limits for some heavy metals

Element	TCLP (mg/L)
Cr	5
Cd	1
Pb	5

Table 4—Concentration of heavy metals in leachates

Leaching medium	W/B ratio	Mix name	Concentration in leachate (mg/L)		
			Cr	Cd	Pb
Deionized water	0.57	C5	0.114	< 0.00004	0.0003
		C10	0.082	< 0.00004	0.0002
	0.50	C5	0.120	0.00005	0.0033
		C10	0.103	< 0.00004	< 0.0002
Acetic acid	0.57	C5	0.263	< 0.00004	0.0008
		C10	0.174	< 0.00004	0.0003
	0.50	C5	0.123	0.00005	0.0038
		C10	0.109	< 0.00004	0.0007

Conclusions

In this paper, the possibility of using copper tailings containing toxic heavy metals as a concrete additive was explored. Based on the experimental results the following conclusions are drawn:

The use of copper tailings as an additive in pastes led to a minor increase in setting times. It equally reduced concrete slump.

It was also observed that the volume of permeable voids increased slightly as the copper tailings content of concrete mixtures became higher.

In both concrete w/b ratios investigated, the addition of copper tailings to concrete enhanced its abrasion resistance, compressive and tensile strengths.

The enhancements were more noticeable at higher w/b ratio and at the 5% addition level.

Chloride penetration depths significantly lower than that of the control specimens were obtained from concrete samples containing copper tailings.

For the two leachants used in this study, the concentration of heavy metals in leachates decreased as the copper tailings content of the concretes increased.

The leachate concentrations obtained from the use of deionised water and acetic acid as leaching media for concretes were significantly lower than the US CFR limits.

Considering consistency, mechanical strengths, chloride penetration and leaching tests results, it seems that there is a potential for the use of copper tailings as a concrete additive, especially at 5% addition level per mass of cement in concrete.

Acknowledgements

The author would like to acknowledge the financial supports of the Rector's Office, Eastern Mediterranean University, Mersin, Turkey. The authors would also want to express thanks to Mr Ogün Kılıç of the Materials of Construction Laboratory, Department of Civil Engineering, EMU, for his support during the experimental testing program.

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