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ORIGINAL PAPER

Effect of construction and demolition waste on the long-term geo-environmental behaviour of cemented paste backfill

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Abstract
The construction and demolition waste (CDW) could be used as backfill material to fill the voids created during the ore production in underground mining. However, there is a need to clarify the impact of CDW on groundwater pollution when it is used as backfill material in cemented paste backfill (CPB) of sulphide-rich tailings (S-rT). This study presents the influence of CDW on the long-term geo-environmental behaviour of CPB samples (CPBs) when used as replacement (10 wt.%) to S-rT. For this reason, the dynamic-tank leaching test was conducted on CPBs and pH, conductivity (Ec), sulphate (SO_4^{2-}) and heavy metals (HMs) analyses were executed for leachates over 30–360 days of leaching period. The effects of mineralogy and microstructure on the geo-environmental behaviour of CPBs were also analysed via X-ray fluorescence (XRD) and mercury intrusion porosimeter (MIP). The utilisation of CDW in CPB mixtures was found to reduce the SO_4^{2-} release (up to 13.89%), neutralise the acid generated and lower the Ec (up to 22.16%). Furthermore, the HMs releases (except As, Cu and Zn) were prevented (Ni and Cr) or reduced up to 92%, which is compatible with the improved CPB microstructure. Only the release of arsenic (As) exceeded the limit value for groundwater in CPB of CDW leachate. These findings suggest that not only the cost, strength and stability but also the impact on groundwater pollution should be considered when disposing of CDW as CPB material in underground voids.

Keywords Construction and demolition waste · Geo-environmental behaviour · Heavy metal · Groundwater quality · Microstructure · Cemented paste backfill

Introduction

In the metal mining industry, the mineral processing methods such as flotation used for the enrichment of precious metals (Cu, Pb, Zn, etc.) produce a huge amount of fine-sized (< 100 µm) sulphide-rich tailings (S-rT) (Yılmaz et al. 2020a). Cemented paste backfill (CPB), which is regarded as one of the safe disposal methods of these sulphide tailings (European Commission 2018), has been used for nearly 30 years in underground mines. CPB is an engineered product comprising of de-watered mill tailings, binder (2–9% cement by weight) and mixing water. The practice of filling the underground openings formed as a result of ore production with CPB has been increasing all over the world in recent years due to its economical, technical and environmental benefits. Yumlu (2020) stated that there are at least 400 underground CPB operations in the world today and nearly 100 facilities are in the project, construction and installation phase, while only in Europe, approximately 17 CPB plants are active and 16 are in the installation and construction stage. CPB allows the safe storage of almost 50–55% of mineral processing tailings in underground mine openings and hence reduces/eliminates the surface storage and rehabilitation costs. It exhibits significant economic benefit by lowering operating cost due to the use of a lower amount of binder and promoting the recovery of large-sized pillar ores (Fall and Samb 2009; Pokharel and Fall 2013; Ercikdi et al. 2017). The backfilling of mined out openings with CPB serves technically multiple functions such as safe working platforms for workers and mine equipment, as well as the support for ground, wall and roof by maintaining the

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stability of openings. It also assures safe exploitation with improved productivity, avoids/minimises surface subsidence and prevents caving and roof falls (Ercikdi et al. 2009, 2017; Ghirian and Fall 2014). Apart from economical and technical advantages, CPB environmentally offers substantial benefits such as the encapsulation of sulphide tailings in appropriately alkaline environment with inhibiting the generation of acid mine drainage (AMD) that promote the release of heavy metals (HMs (As, Zn, Cu, Pb, Cd, etc.)) to underground waters (MEND 2006; Coussy et al. 2011; Yang et al. 2020, Yilmaz et al. 2020b). The binder and acid-neutralizing materials (marble waste, CDW, C-class fly ash etc.) added into the CPB also increase neutralization potential and reduce porosity and permeability (10^{-4}–10^{-7} m/s) of CPB which alleviates the air and water flow around and through the CPB. All these economical, technical and environmental advantages promote the CPB technique as an integral part of underground mining activities.

During the mine extraction operations, waters coming out from the surface or underground are collected to the bottom of the underground mine pumped out of the mine which artificially lowers the ground water table in underground mines. CPB that is returned underground in the early years of mining may remain above the underground water level (UW-L) for up to thirty years. In this period, CPB, which is produced from S-rT, has the potential to generate acid and dissolved HMs at elevated concentrations when the CPB is exposed to atmospheric conditions. Moreover, some processing water containing numerous types of HMs can be delivered underground by CPB mass although most of these HMs are recovered via thickening the water that is recycling back to the processing plant. As mines are closed and pumping ceased, the water table will go back to its natural level and the CPB mass will be flooded. In this regard, the oxidation products such as acid (H^+), sulphate (SO_4^{2-}) and HMs generated and stored in the CPB mass during mine extraction stages could be released, contributing the contamination and pollution of groundwater. On the other hand, when the CPB mass is flooded, the saturated conditions within the CPB minimise the ingress of oxygen, thereby inhibiting the potential sulphide oxidation with the resultant reduction in AMD formation. The release of contaminant components (sulphate, soluble HMs, etc.) from the CPB can be significantly mitigated because the oxidation rate in the water is lower than that in the air (Verburg 2001; Chapman et al. 2003; MEND(2006)MEND Report 10.2–Paste Backfill Geochemistry–Environmental Effects of Leaching and Weathering,Mine Environment Neutral Drainage (MEND) Program, April2006,Canada., 2006; Schafer 2016; Hamberg et al. 2017a; Hamberg et al. 2018; Bull and Fall 2020; Yang et al. 2020; Yilmaz et al. 2020b).

The evaluation of the geo-environmental/geo-chemical behaviour of CPB due to the leakage risk of the harmful pollutants into groundwater has gained significant importance in recent years. For this reason, some studies were performed by implementing the dynamic-tank leaching (DTL) test to monolithic CPBs under laboratory conditions for predicting the geo-environmental properties of CPB mass remained under the UW-L (Coussy et al. 2011; Jiao et al. 2011; Hamberg et al. 2015, 2017a, 2018; Schafer 2016; Seipel et al. 2017). Coussy et al. (2011) investigated the arsenic (As)-release from CPB containing arsenopyrite tailings by utilising the DTL test for up to 64 days. The authors reported that the leachates were highly alkaline ($pH \geq 10.4$), and the high pH accelerated the release of As. Jiao et al. (2011) stated that keeping the monolithic CPBs within deionised-water and carrying out the geo-chemical tests of leachates during the long leaching periods are the most suitable test technique to elucidate the influence of CPB on groundwater quality. The researchers also analysed the change in the amount of HMs which were released from CPB by soaking the five cubic CPBs in distilled-water between 8 and 75 days. The authors put forward that the utilisation of CPB in underground mines does not pose a problem for the groundwater quality (Jiao et al. 2011). Similarly, Hamberg et al. (2015) evaluated the leachate pH of tailings and CPBs, and release of some HMs (As, Ca, Fe, S, etc.) by performing the DTL test for up to 64 days to research the environmental behaviour of CPB of OPC (CPB of ordinary Portland cement) and CPB of OPC-FA (CPB of ordinary Portland cement-fly ash) fabricated with the cyanidation tailings (CT). Compared with the CT, it was monitored that the CPBs maintained the alkaline (*7.9–10.4 c.f. 3.9–6.0 for CT*) conditions and decreased the release of sulphur. However, the leaching of As from CPB of OPC was higher than those from CT and CPB of OPC-FA. To analyse the geo-chemical behaviour of CPBs with DTL tests, Schafer (2016) fabricated the CPBs by utilising the gold tailings (sulphur content = 0.22%, pH = 7.71 and high As content) at the binder dosages ranging between 3 and 12%. He found that the increment of binder dosages from 3 to 12% reduced the total As-release by 82%. In addition, the author reported that the more appropriate HMs-release results were obtained from the DTL test performed using monolithic CPBs to evaluate the long-term geo-chemical properties of CPB when it is compared to the conventional test methods (i.e. static and kinetic tests) (Schafer 2016). In other researches (Hamberg et al. 2017a, 2018), the authors subjected the low-strength (0.2 MPa) CPBs to the DTL test at the end of the curing times of 31 (normal flooding) and 446 days (delayed flooding). The results demonstrate that the low-strength CPB failed to prevent or limit the generation of AMD; therefore, environmental problems such as the increased release of pollutants components may occur. The results also point out that the CPB mass should flood as soon as possible for the prevention or restriction of the oxidation. In the previous experimental studies performed to determine the influence of geo-environmental behaviour of the CPB on the groundwater quality (Coussy et al. 2011; Jiao et al. 2011; Hamberg et al. 2015, 2017a, 2018; Schafer 2016), the tailings materials having extremely low sulphide content (0.22–2.0%) were used, and the geo-chemical measurements were implemented over 75 days. However, in Turkey, the pyritic sulphur (S^{2-}) contents of the tailings used as CPB material in Çayeli Copper-Mine (Rize) and Kure Copper-Mine (Kastamonu) varied between 34.68–37.40% and 15.82–29.12%, respectively, depending on the tailings type (Kesimal et al. 2010, 2012).

In addition to the usage of OPC, industrial by-products (i.e. fly ash, blast furnace slag) were also utilised as an additive or partial substitution to cement or sulphide tailings in the production of CPB in the previous studies. Increasingly, large quantities of the construction and demolition waste (CDW) are generated because of the urban renewal operations (end of life or risky buildings) and natural disasters (earthquake, flood, landslide, etc.) (Tabsh and Abdelfatah 2009). Previous studies exhibited that CDW is highly alkaline (including limestone, calcium-bearing minerals and cement) and can be used as an acid-neutralization material in CPB of S-rT (Chen et al. 2017, 2018; Yılmaz et al. 2018; Liu et al. 2020a, b). Chen et al. (2017) investigated the potential use of a mixed CDW and ultrafine tailings as a backfilling aggregate in underground openings by uniaxial compressive strength and pumpability tests. They showed that the optimal proportion of CDW content in the mix should be 30% to achieve the highest strength for the CPB. They also recommended the use of superplasticiser to enhance the relatively poor flowability of CPB. Yılmaz et al. (2018) examined the effect of finely ground CDW as partial replacement (5–15 wt.%) to sulphide tailings on the strength, stability and microstructural properties of CPB. They also monitored the acid (pH) and water-extractable sulphate (SO_4^{2-}) over 360 days. They found that the utilisation of CDW enhanced the strength and microstructural properties (i.e. total porosity) of CPB while the CDW contributed limited to the resistance of CPB to acid and sulphate attack due to its low CaO content (12.23%). Similarly, Chen et al. (2018) conducted an extensive experimental study to evaluate the potential use of CDW as backfill material in CPB and they reported that the utilisation of CDW (up to 40 wt.%) produced a beneficial effect on the strength, workability and microstructural properties of CPB. They also remarked that the use of CDW in CPB is feasible from an environmental aspect by immobilising the HMs and satisfying the water quality requirements. Liu et al. (2020a, b) also explored the environmental behaviour of CPB containing CDW by performing the leaching toxicity test and stated that heavy metal (Pb, Cu, Cd, Zn and Cr) releases were not observed (except As) in the leachates obtained from CPB of CDW samples and the leachates showed alkaline properties (pH = 9.06–10.75). Although there are some attempts to utilise the CDW in CPB as a backfill material (Chen et al. 2017, 2018; Yılmaz et al. 2018; Liu et al. 2020a, b), it still requires a rigorous evaluation of the potential environmental effects of CPB of CDW because of concerns about water quality after the mines flood with groundwater.

In this study, the long-term geo-environmental properties of flooded CPB of sulphide-rich tailings (S-rT = 15.82% S^{2-}) after mine closure was studied. Furthermore, the potential use of fine-sized CDW as a replacement (10 wt.%) for S-rT was investigated to demonstrate its effect on the geo-chemical behaviour of flooded CPB. The CPBs prepared with or without replacement of S-rT using the CDW at 10 wt.% (10CDW) were cured for up to 180 days and then subjected to the laboratory-scale DTL tests for 30 to 360 days. The geo-environmental parameters (pH, conductivity, concentration of SO_4^{2-} and heavy metals (HMs) including Pb, Zn, Cu, Cd and As) of the leachates obtained from DTL tests were monitored. The release of HMs was compared to the limit values (L-Vs) set by WHO (2011) for groundwater. Furthermore, a detailed characterization of flooded CPB samples was also performed through XRD (mineralogical) and MIP (microstructural) studies.

Materials and methods

Sulphide-rich tailings and binder

The S-rT was procured from Etibakır Inc. Co. (Kure Copper-Mine) in Turkey, and the binder (OPC) requirement was met by Aşkale Cement Inc. Co. in Turkey. The S-rT was found to have a specific surface area (SSA), fines content (< 20 µm) and specific gravity (SG) of 4440 cm^2/g, 48.4% and 3.37 g/cm^3, respectively. The SSA and SG of the OPC were also determined to be 4330 cm^2/g and 3.12 g/cm^3, respectively (Table 1). The XRF and ICP-AES tests have revealed that the S-rT predominately include Fe_2O_3 and SiO_2, while the predominant minerals in the binder are CaO and SiO_2. The results of chemical analysis have also indicated that the sulphide content (S^{2-}) of S-rT is 15.82%, as confirmed by the pyrite content (FeS_2 = 29.66%) determined by XRD analysis (Table 1).

Construction and demolition waste

As a result of Urban Renewal Projects in Turkey, the CDW materials replaced with S-rT in CPB mixtures were selectively collected from the demolished buildings in Ortahisar, Trabzon. The CDW comprises the main components such as concretes, mortar-brick-concrete and concrete-brick-tile (Fig. 1a). The large-sized CDW was crushed with a

Table 1 Chemical, physical and mineralogical properties of the CDW, S-rT and OPC (Yılmaz et al. 2020a, b)

Characteristic		S-rT (%)	CDW (%)	OPC (%)
Chemical composition				
SiO_2		31.89	54.19	21.02
Al_2O_3		8.97	11.67	5.27
Fe_2O_3		33.09	5.44	3.06
MgO		4.08	3.08	2.19
CaO		3.48	12.23	62.91
Sulphide content (S^{-2}) (%)		15.82	–	–
Pyrite content (FeS_2) (%)		29.66	–	–
Physical properties				
Fines (< 20 µm) content		48.4	59.1	–
D_{10}	(µm)	2.0	1.5	–
D_{30}		8.3	4.9	–
D_{60}		31.8	20.1	–
D_{80}		70.9	52.6	–
Specific gravity (g/cm^3)		3.37	2.60	3.12
Specific surface area (cm^2/g)		4440	6600	4335
Mineralogical composition				
S-rT			CDW	
Pyrite, Albite, Quartz			Albite, Quartz	
Calcite, Chlorite			Calcite, Muscovite	

Fig. 1 Main components of CDW (**a**) and size reduction operations (crushing (**b**), grinding and mixing (**c**))

hammer (< 100 mm) and jaw crusher (< 3 mm), respectively (Fig. 1b). Thereafter, before the grinding operation, the CDW was dried in the forced-air oven (50 °C/36 h). Then, each component of the CDW was separately ground to under 100 µm by utilising a laboratory type ball mill. The granulated CDW components were equally and homogeneously mixed (Fig. 1c). The CDW was found to have a fines content (< 20 µm), SG and SSA of 59.1%, 2.60 g/cm^3 and 6600 cm^2/g, respectively (Table 1). The XRF and ICP-AES tests have shown that the CDW mostly comprises SiO_2, CaO and Al_2O_3, and the predominant mineral is quartz (Table 1).

Acid–base test (A-BT) of materials

The acid potential (AP) and neutralization potential (NP) of materials (CDW and S-rT) were calculated with the A-BT designed by Sobek et al. (1978). Within this scope, the AP value of S-rT was primarily determined with the formula of $AP = 31.25 \times S_{total}$ (total sulphur value) (Table 2). The fizz rating of materials was determined to calculate the NP values of S-rT and CDW. Then, the values of the NP and

Table 2 Acid–base accounting (ABA) test results of the S-rT and CDW (Yılmaz et al. 2020a, b)

Properties	S-rT	CDW
Sulphide (S^{2-}) content (%)	15.82	–
CaO content (%)	3.48	12.23
Fizz rating of materials	None	Slight
Acid normality (N)	0.1	0.1
Acid volume (ml)	20	40
Acid potential (AP)*	494.38	0
Neutralization potential (NP)*	8.58	90.00
Net Neutralization potential (NNP)*	− 485.80	90.00

*: kg $CaCO_3$/ton waste

net neutralization potential (NNP) of S-rT and CDW were calculated via Eqs. (1) and (2) after the A-BT (titration test) (Table 2). Further procedure for A-BT was explained in the study of Yılmaz et al. (2020a).

$$\mathrm{NP} = \left(\left(A_{\mathrm{n}} \times A_{\mathrm{v}}\right) - \left(B_{\mathrm{n}} \times B_{\mathrm{v}}\right)\right) \times 50/m \tag{1}$$

$$\mathrm{NNP} = \mathrm{NP} - \mathrm{AP} \tag{2}$$

where,

A_n and A_v are the molarity (M) and volume (mL) of the acid, respectively,

B_n and B_v are the molarity (M) and volume (mL) of the base, respectively,

m is the sample weight (g).

Preparation of CPB samples

The CPBs (Control and CPB of 10 wt.% CDW) utilised in tests were prepared at 8.5 wt.% binder dosage and 19.05 cm consistency. This is because the CPBs produced the unconfined compressive strength (UCS of ≥ 1.0 MPa at 28 days) required by mine operators at this binder dosage in the preliminary tests. The CPBs of the Control were fabricated with fully utilising S-rT (100%), while 10 wt.% CDW was replaced to S-rT in the production of CPBs containing CDW because 50–55 wt.% of tailings (S-rT) produced in underground mines can be safely stored in underground mine openings (Benzaazoua et al. 2002). Thus CPB operators do not want to reduce the amount of tailings to be transported to the underground mine stopes. The CPBs prepared for this study were coded as Control and 10CDW, and the mixture parameters for CPBs are detailly presented in Table 3. In addition, the process water in S-rT was initially found to have a sulphate content of 5616 mg/l and an acidity (pH) value of 6.85, respectively; because, the S-rT contains 73% process water.

The CPB mixtures (S-rT, CDW, OPC and tap water) were mixed by a mechanical blender having a volume of 20.8 L. The cylindrical (5 × 10 cm) moulds were utilised for the casting of the CPB mixtures which were meticulously prepared. Then, the excess water of fresh CPBs placed in the drainage stand was drained from the bottom of CPB moulds for 24 h (Fig. 2a). Thereafter, the curing process was implemented in a curing cabinet having ~ 80% humidity and 20ºC temperature for up to 180 days (Fig. 2b). The aim was to expedite the oxidation process of the CPBs by exposing the CPBs of S-rT to oxidation during 180 days of open-top curing. Right after the curing process, the chemical (metalloids concentrations) (Table 4) and mineralogical (XRD analyses) characterization of CPBs were analysed before the DTL tests.

Table 3 The mixture parameters of CPBs

Characteristics	Control	10CDW
Replacement ratio (%)	0	10
Cement dosage (wt.%)	8.5	
Solids content (wt.%)	73.70	74.15
Water to cement ratio (w/c)	4.20	4.10
Consistency value (cm)	19.05	
Strength at 180 days (MPa)	1.56	1.84

Dynamic-tank leaching (DTL) test

To examine the effect of the CPBs of S-rT on the groundwater quality in underground implementations, the long-term DTL test was performed. At the end of the 180-day curing process, the monolithic CPBs (Control and 10CDW) were demoulded and the lower and upper surfaces of CPBs

Fig. 2 CPB samples in the drainage platform (**a**) and curing cabinet (**b**)

Table 4 Major and trace metalloids concentrations of 180-day CPBs, and regulation limit values of HHMs for ground-water (WHO 2011)

Characteristics	Control	10CDW	Regulation limit value (WHO)	
Major Elements (wt.%)			mg/L	mg/m^2
Iron (Fe)	16.88	14.74	–	–
Calcium (Ca)	4.99	5.22	–	–
Aluminum (Al)	2.84	2.62	–	–
Magnesium (Mg)	1.79	1.61	–	–
Trace Elements (ppm)				
Copper (Cu)	1153	993	2.0	201
Zinc (Zn)	903	782	3.0	301
Manganese (Mn)	587	521	–	–
Cobalt (Co)	582	520	–	–
Arsenic (As)	144	124	0.01	1.0
Nickel (Ni)	54	51	0.07	7.0
Lead (Pb)	88	76	0.01	1.0
Chrome (Cr)	126	113	0.05	5.0
Cadmium (Cd)	3.1	2.8	0.03	0.3

were flattened. The length of CPBs was gauged by callipers. Thereafter, the geometric surface area (GSA) of CPBs was calculated via Eq. (3), and the leachant volumes (LV) to be utilised in the DTL tests were determined by using the Eq. (4).

$$\mathrm{GSA} = 2\pi r \times (r + h) \tag{3}$$

$$\mathrm{LV} = \mathrm{GSA} \times 9 \pm 1\ \mathrm{cm}^3 \tag{4}$$

where;

"r and h" are the radius and length of the sample, respectively,

The CPBs whose the GSA and LV were determined were soaked in closed HDPE (high-density polyethylene) containers (a CPB in each container) filled with the calculated de-ionised water (2.27–2.29 L) during the long-term leaching periods (30, 60, 90, 180, 270 and 360 days) (Fig. 3) (US EPA 1315–1 2013; ASTM C1308-08 2017). For each leaching period, the fresh de-ionised water was filled into the containers. Each DTL test was implemented in triplicate and the average values of parameters were given in the results.

At the end of each leaching period, the clear supernatants were obtained by filtering the fresh-leachate samples using a syringe-type of membrane filter (0.45 µm). Thereafter, they were expeditiously subjected to the analyses of pH (Oakton pH/Con 510, Fig. 4a), Ec (Orion Star A321, Fig. 4b) and SO_4^{2-} (Palintest 5000, Fig. 4c), while the HMs (As, Cu, Co, Zn, Cd, Ni, Pb and Cr) contents in each clear supernatant were analysed by using ICP-MS.

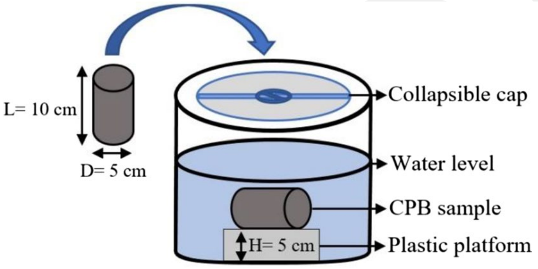

Fig. 3 The schematic scene of DTL test (US EPA 1315–1 2013)

Microstructural and mineralogical studies of CPB

Two CPBs were prepared at the same mixture properties as those utilised in the DTL test to investigate the influence of the porosity properties on the geo-environmental behaviour of CPBs. These CPBs (Control and 10CDW) not subjected to any destructive test were firstly dried in the forced-air oven (50 °C/36 h) after 180-day curing. Then, the MIP (Mercury Intrusion Porosimetry) tests were conducted on those CPBs employing MIP apparatus (Micromeritics Autopore IV 9510) (ASTM International, 2018); therefore, the technical parameters (total porosity, macro-pores, meso-pores and critical pore diameter) were determined. Furthermore, to well elucidate the relation between microstructural development and geo-chemical properties of CPBs, XRD analyses of powdered CPBs were implemented by using Bruker D8 Discover having a voltage, current and incident angles of 40 kV, 40 mA and 5° and 70° (step size = 0.02°), respectively, at AGU Central Research Facility (AGU-CRF). In the XRD analyses, Cu k-α tube (λ = 1.5406 Å) and polimetilmetakrilat (PMMA) type of standard sample holders (height = 8.5 mm and diameter = 25 mm) were utilised.

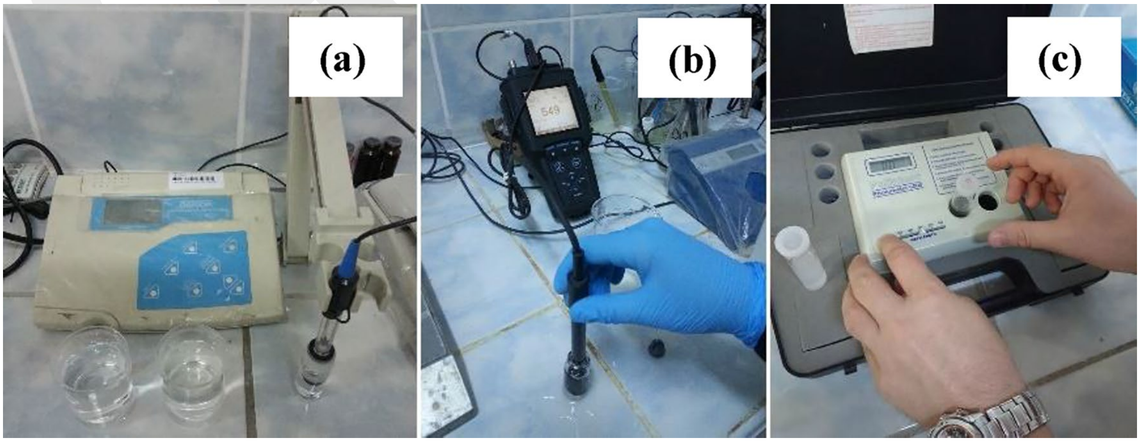

Fig. 4 pH (**a**), Ec (**b**) and SO_4^{2-} (**c**) analyses

Results and discussion

Oxidation-neutralization properties of the flooded CPB

The principal oxidation product (OP) is sulfate (SO_4^{2-}), formed by pyrite oxidation, whereas the most significant neutralization products (NPs) are Ca, Mg and Mn which are produced due to the dissolution of carbonate and silicate minerals (Hakkou et al. 2009). The variations of OP and NPs released from flooded CPB samples (Control and 10CDW) during 30–360-day DTL tests are demonstrated in Fig. 5. All flooded CPBs produced OP concentrations ranging from 63.61 to 96.33 g/m^2 at all the leaching periods. The OP concentration released from 10CDW is slightly lower (4.05–13.89%) than the Control irrespective of the leaching periods (Fig. 5). The reason may be associated with the fact that the free Ca-ions formed by the dissolution of calcite, and free OP-ions originated from pyrite oxidation formed the gypsum minerals, as confirmed by XRD profiles (Fig. 6a and b). Besides Hakkou et al. (2008) remarked that the gypsum dissolution could have been increased the released OP concentrations, in agreement with Fig. 6a. Furthermore, the partial replacement of CDW having finer particles (Table 1) to S-rT at 10 wt.% appeared to limit the OP concentration released from 10CDW by decreasing the total porosity (p_{total}) (Fig. 7). In this respect, the relatively low OP concentration in 10CDW could be referred to its lower p_{total} (*40.29% c.f. 42.72% for Control*), corresponding to a decrement of 5.69% by filling the voids between coarse S-rT particles. The findings of the current study were consistent with the previous researches (Zheng et al. 2016; Yılmaz et al. 2020a). They examined the utilisation of different granulated industrial waste materials (blast furnace slag, dolomitic limestone type-C fly ash and calcitic limestone) at 15–22.5 wt.% of replacement dosages to determine the effect of these materials on the microstructure of CPBs. It was reported that the p_{total} values were reduced from 43.67% to between 38.29% and 41.60% (Yılmaz et al. 2020a) and from 34.8% to 33.43% (Zheng et al. 2016) at 28 days, while these materials contributed the similar decline for p_{total} values (up to 12.43%) at 180 days (Yılmaz et al. 2020a). This beneficial influence was associated with the physical characteristics (fineness, pore-filling effect, etc.) of the granulated materials. Hence, the inhibition or limitation of the oxygen access into the CBP is likely the cause of the lower permeability with the resultant decrease in OP release from 10CDW (Kruse et al. 2013; Zheng et al. 2016; Yılmaz et al. 2018).

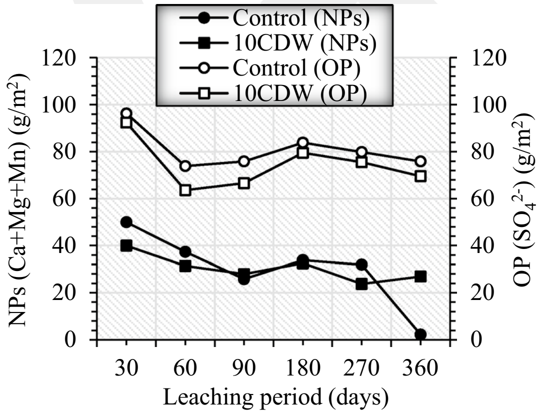

Fig. 5 OP (SO_4^{2-}) and NPs (Ca + Mg + Mn) concentrations of CPBs during the DTL test

When the variation of NPs released from the flooded CPBs (Control and 10CDW) is evaluated in Fig. 5, the higher amount (up to 34.22%) of NPs from the Control released at all the leaching periods except 90 and 360 days, compared with 10CDW. It should be noted that a significant amount of NPs comes from Ca-ions in CDW and OPC due to their high CaO concentrations (Table 1). Based on this finding, the dissolution of gypsum including the Ca-ions is likely the cause of the higher NPs released from Control, consistent with Fig. 6a (Hakkou et al. 2008). Additionally, the Control had higher macro-pores (p_{macro}) and lower meso-pores (p_{meso}) as well as its higher p_{total}, compared with 10CDW (Table 5). It was calculated that the values of p_{macro} and p_{meso} in the Control comprised of 37.49% and 5.23% of p_{total} value, whereas these values were 33.60% and 6.69% for 10CDW, respectively (Table 5). At the last leaching cycle (360 days), the release of NPs in the Control severely decreased (2.21 g/m^2), while the higher amount of NPs (26.91 g/m^2) from 10CDW was released compared to the previous leaching cycle (270 days) (Fig. 5). This is because the Control produced a higher amount of OP (SO_4^{2-}) than 10CDW at all the leaching cycles (Fig. 5) due to the higher pyrite oxidation in Control, in agreement with the XRD patterns (Fig. 6). Hence, the NPs obtained from only the binder (OPC) in the Control mixture may have depleted while neutralizing the sulphate. However, 10CDW had a higher NPs than the Control due to the presence of alkaline CDW (10 wt.%) in the mixture of 10CDW (Table 3).

Figure 8 exhibits the cumulative OP-NPs curve obtained from the values of OP and NPs released from the Control and 10CDW during the DTL test. Figure 8 also shows that 10CDW had a sufficient amount of the NPs at all the leaching cycles to neutralise the acid generated by OP, while the NPs of the Control almost depleted at leaching cycle of 360 days. However, the OP release from the Control demonstrated a slight reduction at the same leaching cycle (Figs. 5 and 8). Therefore, it can be assumed that the acid production in the Control may maintain in the on-going process, whereas 10CDW can continue to neutralise the acid produced from the principal OP due to the neutralization

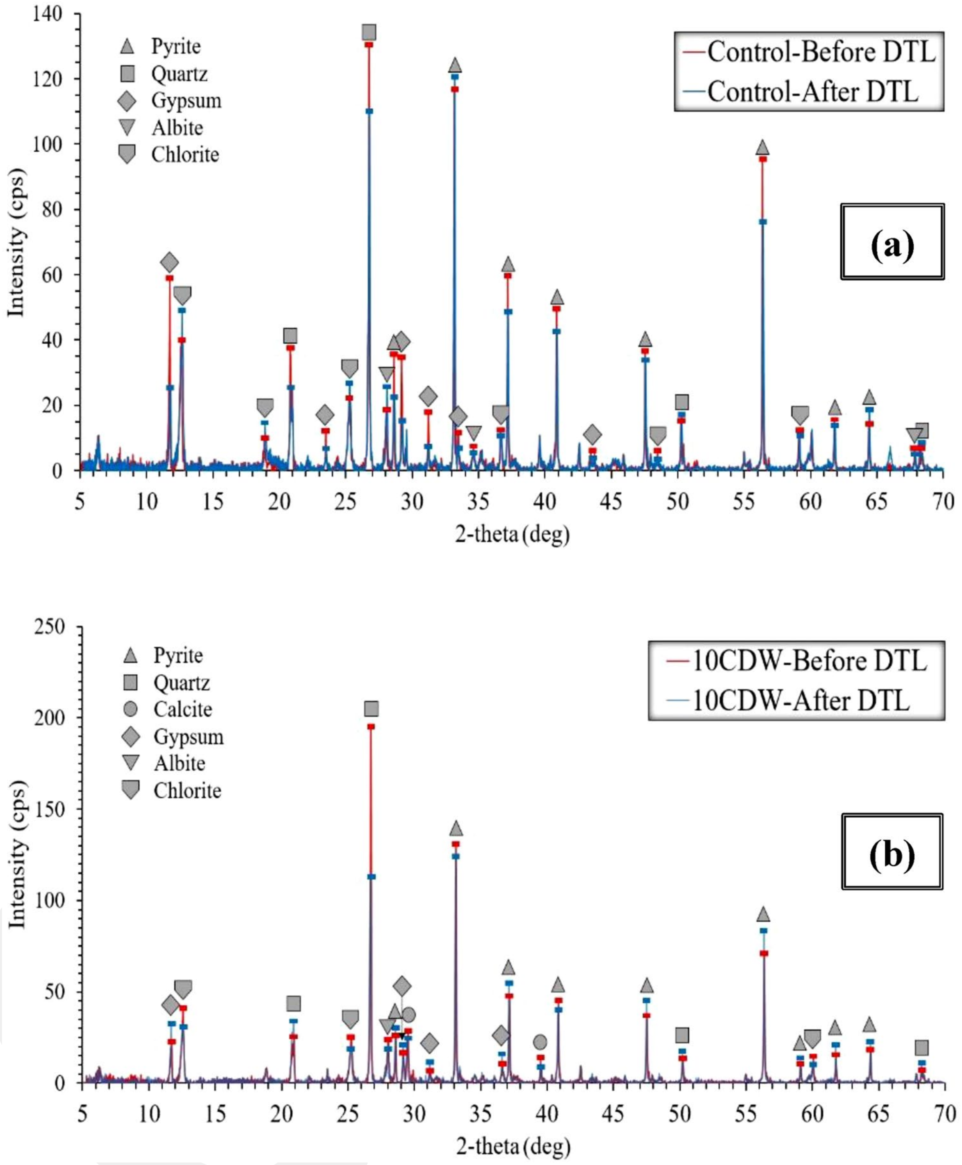

Fig. 6 XRD profiles of Control (**a**) and 10CDW (**b**) before and after the DTL tests

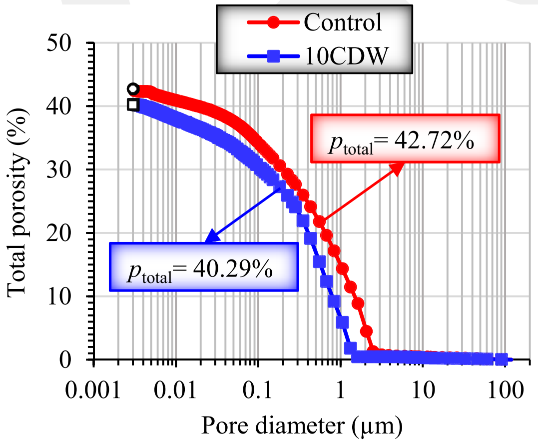

Fig. 7 The total porosity values of Control and 10CDW at 180 days

Table 5 Technical parameters of the MIP curves at 180-day CPBs

Sample name	Curing period (days)	p_{total} (%)	Cumulative porosity (%)		[c]Critical pore diameter (μm)
			[a]p_{meso}	[b]p_{macro}	
Control	180	42.72	5.23	37.49	1.63
10CDW		40.29	6.69	33.60	1.06

[a]p_{meso} (%): Meso-pores range between 0.002 and 0.05 μm

[b]p_{macro} (%): Macro-pores have pore size larger than 0.05 μm

[c]Critical pore diameter (μm): Critical pore size represents the steepest slope of the cumulative porosity curve (Yilmaz et al. 2011)

potential of CDW (*90 kg $CaCO_3$/ton waste*) (Table 2). The slopes of the curves given in Fig. 8, which can be described as the neutralization rate, also support these predictions. The

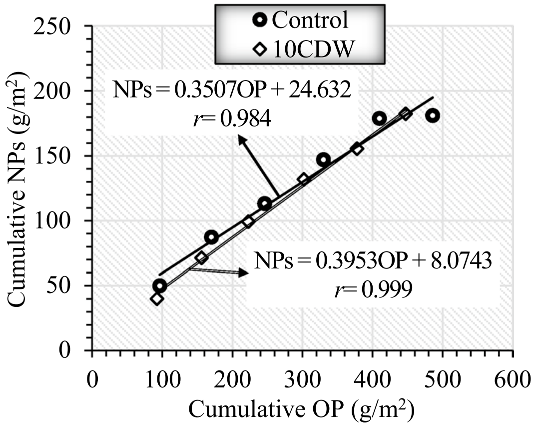

Fig. 8 OP-NPs curves of CPBs during the DTL test

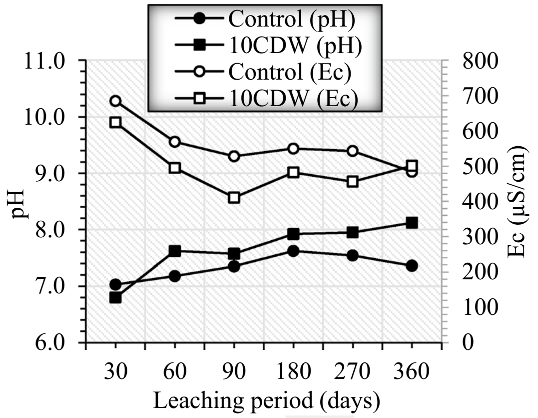

Fig. 9 pH and Ec variations of CPBs during the DTL test

slope of 10CDW was relatively higher (0.40 > 0.35) than Control. Moreover, it can be inferred from the slightly higher correlation coefficient in 10CDW (*0.999 c.f. 0.984 for Control*) that 10CDW may neutralise the acid.

pH and Ec Results of the flooded CPBs

Figure 9 presents the variations of the pH and Ec in CPB samples (Control and 10CDW) during 30–360 days of leaching periods. The results of DTL tests indicate that all monolithic CPBs generally demonstrated the pH-values above the neutral levels (6.80–8.12) irrespective of the leaching periods. Although the lower pH-value in 10CDW was measured in the first leaching period (30 days), thereafter, 10CDW produced slightly higher pH-values (up to 10.33%) than the Control. Moreover, the decline in the pH-values of the Control after 180 days was monitored, whereas pH-values of 10CDW proceeded to increase towards the alkaline level during the leaching periods (Fig. 9). The limited contribution achieved from 10CDW could be interrelated with higher neutralization capability (*90 kg $CaCO_3$/ton waste c.f. −485.80 kg $CaCO_3$/ton waste for S-rT*) and higher SSA (*6600 cm^2/g c.f. 4440 cm^2/g for S-rT*). This increases in neutralization potential of the CDW (Table 1) (Potgieter-Vermaak et al., 2006). The decrease of p_{total} and refinement of p_{macro} of 10CDW (Fig. 7 and Table 5) by filling voids among the coarse S-rT particles with the finer CDW particles is also likely the cause of the restriction of the penetration of air and moisture into the CPB pores. Thus, the oxidation rate was reduced, and the acid generation originated from pyrite oxidation was limited when it was compared to the Control. Besides these positive contributions, as can be seen from the XRD profiles (Fig. 6), the calcite dissolution in CDW limitedly increased the alkalinity of 10CDW (Rezaee and Honaker 2020; Yılmaz et al. 2020a). Moreover, the results of XRF and ICP-AES show that the CDW contained a higher amount of CaO, the primary alkaline constituent, compared to the Control (Table 1). Similarly, Jones and Cetin (2017) indicated that the dissolution of the cementitious constituents (lime: CaO, calcite: $CaCO_3$, etc.) in the recycled concrete aggregates increased the pH of AMD (Golab et al., 2006). In previous researches, considerably lower acid formation (pH-values = 10.4–11.9) from DTL tests practised up to 64 days of leaching periods was monitored (Coussy et al. 2011; Taha et al. 2019). On the contrary of this study, lower acid formation (higher pH-values) could be interrelated to the fact that the tailings and waste rocks utilised in CPB and concrete samples contained extremely lower sulphide contents (*0.47–0.84% < 15.82%*).

When the Ec-values were examined, it was seen that Ec-values quantified between 411 and 685 µS/cm at all the leaching periods (Fig. 9). All Ec-values decreased up to the end of the third leaching cycle (90 days), while Ec-values for Control and 10CDW demonstrated a more horizontal trend (456–550 µS/cm) at the following leaching periods. The descent in Ec-values of CPBs might be originated from the decrease in the quantity of water-soluble elements, consistent with the previous authors (Hakkou et al. 2009; Taha et al. 2019). They stated that the water-soluble NPs (Ca, Mg, etc.) and OP (SO_4^{2-}) values markedly affect the variation of Ec-values. Furthermore, the considerably low Ec-values (*up to 685 µS/cm c.f. up to 4200 and 11,000 µS/cm for weathered tailings*) in all CPBs were measured compared with the previous studies (Hakkou et al. 2008, 2009). The restriction of water-soluble ions from CPBs may have been achieved by stabilizing the S-rT using binder and CDW.

Assessment of heavy metals released from CPBs

The total concentrations of HMs (As, Cu, Co, Cd, Zn, Ni, Cr and Pb) released from CPBs (Control and 10CDW) over 360-day DTL tests were demonstrated in Fig. 10. It appeared that all CPBs (Control and 10CDW) produced

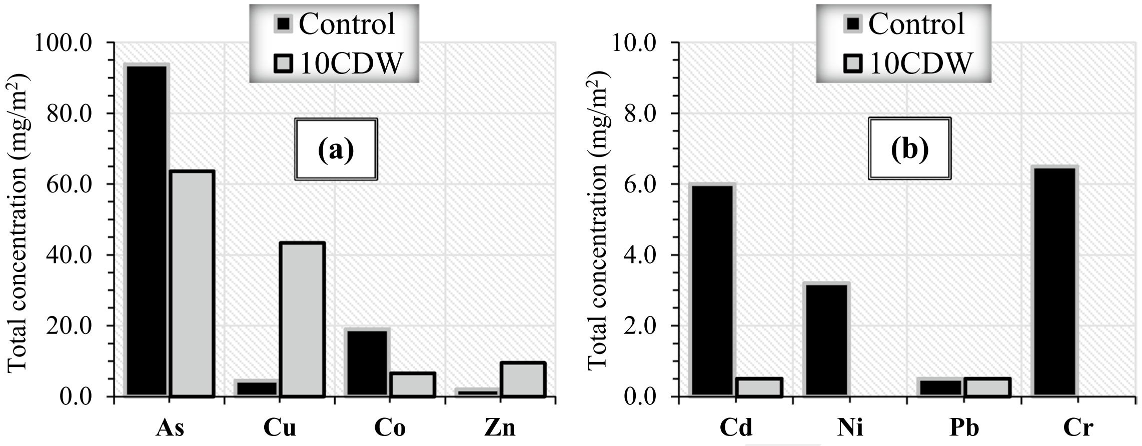

Fig. 10 Total concentrations of HMs released from CPBs during the DTL test

considerably high As-release (63.6–93.8 mg/m^2) during all leaching cycles due to the sulphide oxidation (Fig. 10a). In agreement with the findings in this study, it was reported in the past studies that the mobilization of arsenic (As) from mine tailings/wastes into the groundwater environment can be powerfully linked to the chemical process (i.e. alkaline desorption, oxidation of sulphide minerals and reductive dissolution of ferric oxyhydroxides) (Salzsauler et al. 2005; Sracek et al. 2014). In addition, Sracek et al. (2014) stated that the neutral–or alkaline-pH levels cannot prevent the transfer of As-ions desorbed from the surface of solid particles as a result of sulphide oxidation. However, the utilisation of CDW as partial replacement to S-rT in CPB mixture restricted the total release of As at the rate of 32.2% although the leachate of 10CDW had a slightly alkaline-pH level (max. 8.12) (Fig. 9). The comparatively lower As-release in 10CDW could be associated with the usage of high binder dosage (8.5 wt.%) and finer-sized CDW (Table 1) with the resultant refinement of the CPB pore structure and hence, led to the decrease in sulphide oxidation. The p_{total} (*from 42.72 to 40.29%*) and critical pore diameter (d_{cr}) (*from 1.63 to 1.06 µm*), highly effective on the fluid transfer characteristic of CPB, were reduced with the replacement of 10 wt.% CDW to S-rT in CPB mixture (Table 5) (Pokharel and Fall 2013; Cihangir and Akyol 2018). By this means of the improvement in CPB microstructure, the effective encapsulation of As may have provided due to the higher inherent strength of 10CDW (*1.84 MPa > 1.56 MPa at 180 days*) (Table 3) (Hamberg et al. 2017a; Yılmaz et al. 2018). Moreover, the absence of gypsum dissolution that damages the inherent strength of 10CDW is likely the cause of the decrease in As-release of 10CDW (Fig. 6). When the limit value (L-V) of As in groundwater was assessed, the extremely high As-release (63.6–93.8 mg/m^2 > 1.0 mg/m^2) was recorded in all CPBs (Fig. 10a and Table 4).

When the total Cu-release from CPBs was examined from Fig. 10a, it is seen that the considerably higher amount of Cu-ions (43.40 mg/m^2 > 4.50 mg/m^2) was released from 10CDW at the end of the DTL test, compared with the Control. Dayioglu et al. (2018) reported that the pH value whose Cu-solubility is the lowest is 6.5–7.0, but Cu-solubility significantly increases at higher pH levels (i.e. alkaline). Since CDW had higher neutralization potential (Table 2), the relatively higher pH level (*up to 8.12 c.f. up to 7.62 for Control*) in 10CDW is likely the cause of the high Cu-release from 10CDW (Kumpiene et al. 2008; Hamberg et al. 2018). In previous researches conducted in concrete (Gwenzi and Mupatsi 2016) and CPB (Hamberg et al. 2018), a similar amount of Cu-releases (up to 50.0 mg/m^2 and 17.4 mg/kg) occurred up to 64-day DTL tests. Furthermore, the Cu-releases in all CPBs remained below the L-V (*4.5 and 43.40 mg/m^2 < 200.8 mg/m^2*) for groundwater (WHO 2011) (Table 4).

It appeared that the Co-leaching from CPB remarkably reduced with the utilisation of CDW in CPB mixtures. That is to say that Co-release from 10CDW was 65.3% lower than that of the Control (*19.0 mg/m^2 c.f. 6.60 mg/m^2 for 10CDW*) (Fig. 10a). Because the CDW has higher CaO than S-rT (Table 1), the exchange of Co-ions with the Ca-ions in CaO bonds inside the hydration products (C–S–H, etc.), and the adhesion of Co-ions into C–S–H surfaces without damaging the C–S–H lattice structure are likely the cause of the comparatively lower Co-ions released from 10CDW (Yang et al. 2020). It was also recorded the slightly lower Co-release from the CPBs containing BFS and C-FA (1.15 and 1.20 mg/m^2), and from concretes containing WRs (≈2 mg/m^2) during the DTL tests in the previous studies (Taha et al. 2019; Yılmaz et al. 2020b). This is because the materials (BFS and C-FA) contained a higher amount of CaO *(30.62–37.02% c.f. 12.23% for 10CDW)*, whereas the higher binder dosage (340 kg/m^3) was utilised in the concretes. Furthermore, no limit value for cobalt in groundwater was set by WHO (2011).

The Zn-release from the Control and 10CDW was quantified between 2.0 and 9.5 mg/m^2 at the end of all leaching cycles. The higher Zn-release in the leachates from CPB containing 10 wt.% CDW (10CDW) was detected due to the slightly higher alkaline pH in 10CDW leachate (Fig. 10a) (Kumpiene et al. 2008; Hamberg et al. 2018). As also confirmed by current findings, Lampris et al. (2011) noted that the solubility of some metals is essentially influenced by pH and some of them (i.e. Zn and Pb) dissolve more easily at high pHs. However, the amount of Zn-leachability from samples is at the minimum level because some chemical parameters (i.e. sorption to reactive surfaces and/or secondary precipitated mineral phase) control the solubility at pH ~ 9.5 (Engelsen et al. 2010). The pH increase (to 11.0–12.0) in the concretes having WRs revealed a Zn-release of up to 10.0 mg/m^2 in the previous research (Taha et al. 2019). Compared to the L-V (301 mg/m^2) set by WHO (2011) for groundwater, the Zn-release from all CPBs was found considerably low (Table 4).

Total Cd-release from all CPBs into leachates varied from 0.5 to 6.0 mg/m^2 at the end of all leaching cycles. Compared to the Cd-release from the Control, the Cd-release from CPBs was reduced at the rate of 91.7% with the replacement of 10 wt.% CDW to S-rT (Fig. 10b). The comparatively high pH-levels (low Cd-solubility at these pH) and improvement of the microstructure (Fig. 7 and Table 5) in 10CDW may be the cause of the decrease in Cd-mobility (Hamberg et al. 2017b). Yang et al. (2020) also informed that the release of Cd-ions from CPB can be reduced with the absorption of Cd into the hydration products (C–S–H) after the ion exchange between Cd and Ca because the electric potentials of Cd- and Ca-ions show similarity. Although the substantial restriction in Cd-release from CPB into leachate was achieved by utilising the CDW, it was detected that the L-V in groundwater (0.3 mg/m^2) for Cd-release from all CPBs (Control and 10CDW) was exceeded (Table 4).

Figure 10b shows that the substitution of 10 wt.% CDW to S-rT provided the Ni-immobility from CPBs compared to that of the Control (3.2 mg/m^2). As reported by Chen et al. (2009), this in turn demonstrates the favorable influence of CDW replacement on the immobilization of Ni from CPBs. In agreement with the statements notified by Hamberg et al. (2018), the immobilization of Ni in 10CDW could be also ascribed to the physical confinement of Ni in hydration products (i.e. C–S–H) due to the higher strength of 10CDW (*1.84 > 1.56 MPa*) than that of the Control (Table 3) (Yılmaz et al. 2018). Hamberg et al. (2018) also informed that the CPBs, fabricated with the extremely low (1.0 wt.%) binder dosages at long (446 days) curing, released 8.5 mg/m^2 of Ni compared with the current study (3.2 mg/m^2) (Fig. 10b). However, increasing the binder dosages (3 wt.%) and shortening the curing time (31 days) decreased the Ni-release at the rate of 61.2%. The CPBs (Control and 10CDW) also demonstrated similar behaviour on the Cr-leachability. Compared to the Cr-leachability from Control (6.5 mg/m^2), CDW significantly contributed to the Cr-immobility (no-release) from CPBs (Fig. 10b). These beneficial contributions could be attributed to the improvement of the physicochemical bond of the Cr-element with the C–S–H (Taha et al. 2019). Thus, this can lead to the encapsulation of metal-bearing phases with the C–S–H, which are denser in the high-strength CPB of 10CDW (Fig. 7) (Table 3 and 5) (Zheng et al. 2016; Hamberg et al. 2018; Yılmaz et al. 2018, 2020a). Notwithstanding this, Hamberg et al. (2017b) indicated that the higher Cr-release (up to ≈0.39 mg/m^2) from high-strength CPBs (3 wt.% binder dosages with FA) subjected to the DTL test was recorded. When the amounts of Ni- and Cr-release from CPBs were examined according to L-V in groundwater, the Cr-release from only the Control was detected to be above (*6.5 > 5.0 mg/m*^{2}) the L-V (Fig. 10b and Table 4).

When the Pb-release from the CPBs (Control and 10CDW) was evaluated in Fig. 10b, the same amount of the Pb-release (0.5 mg/m^2) from the CPBs was detected. However, all CPBs produced lower Pb-release than L-V (0.5 < 1.0 mg/m^2) set by WHO (2011) (Fig. 10b and Table 4). In previous researches (Taha et al. 2019; Gwenzi and Mupatsi 2016), the monolithic concrete samples were fabricated to examine the environmental characteristics (i.e. heavy metal leaching) of concretes by using the DTL tests over 63–64 days of leaching periods. The substantially higher Pb-leachability from the concretes was also detected, compared to the CPBs in this study (Table 6).

Conclusion

The effect of using fine-sized CDW (10 wt.%) as a replacement on the long-term geo-environmental behaviour of flooded CPB was evaluated through laboratory-scale DTL tests. The CPBs (Control and 10CDW) cured as open-top for up to 180 days were subjected to the DTL tests for up to 360 days. Various parameters including the acidity (pH), conductivity (Ec), oxidation-neutralization products (OP: SO_4^{2-} and NPs: Ca + Mg + Mn, respectively) and the concentration of heavy metals (HMs) in the leachates generated were monitored over the test period. The MIP (for microstructure) and XRD (for mineralogy) examination of CPBs were also conducted to provide further insight into their behaviour under test conditions. Following conclusions can be drawn from these studies:

a. The use of CDW as replacement appeared to reduce (by up to 13.89%) OP (SO_4^{2-}) leachability of CPB compared

Table 6 Nomenclature of mathematical symbols used in the equations

Equations	Mathematical symbols	Nomenclature
$*AP = 31.25 \times S_{total}$	$*AP$	Acid potential (kg $CaCO_3$/ton waste)
	S_{total}	Total sulphur value in S-rT
$*NP = ((A_n \times A_v) - (B_n \times B_v)) \times 50/m$	$*NP$	Neutralization potential (kg $CaCO_3$/ton waste)
	A_n	Molarity (M) of acid
	A_v	Volume (mL) of acid
	B_n	Molarity (M) of base
	B_v	Volume (mL) of base
	m	Sample weight (g)
$NNP = *NP - *AP$	NNP	Net Neutralization potential (kg $CaCO_3$/ton waste)
$*GSA = 2\pi r \times (r + h)$	$*GSA$	Geometric surface area (cm^3) of sample
	r	Radius (cm) of sample
	h	Length (cm) of sample
$LV = *GSA \times 9 \pm 1\ cm^3$	LV	Leachant volume (LV) used in DTL test

with the Control irrespective of the leaching periods. This can be attributed to the reduced porosity of CPB due to the use of fine-sized CDW, decreasing the ingress of air (oxygen) into CPB.

b. A sharp decrease in NPs after a leaching period of 270 days as shown in the plot of OP-NPs indicated a limited neutralization potential of the Control. The addition of CDW appeared to improve the neutralization potential and hence buffering capacity of CPB for the generation of acid by the oxidation of pyritic sulphide present in the tailings.
c. A reverse trend of decreasing pH in leachates of the Control was observed following the 180-day leaching cycle. In comparison, pH in the leachates obtained from 10CDW was consistently high with tendency to remain at the alkaline levels over leaching periods.
d. The decrease of OP and NPs released from all CPBs decreased the Ec-values of leachates for up to third leaching cycle (90 days). Thereafter, more stable ripple in Ec-values was monitored irrespective of the mixture types.
e. The microstructure (total porosity, macro-pores, meso-pores and critical pore diameter) of the CPB was found to improve by utilising the CDW as replacement. This resulted in the reduced release of hazardous components.
f. The release of metals (except Zn and Cu) from those CPB that contained CDW was observed to be substantially mitigated (up to 92%). The release of As, Cr and Cd from the Control was determined to exceed the limit values (L-Vs) set by WHO for groundwater. On the other hand, only the As-release from 10CDW could not be reduced below the L-V.

It can be deduced from these findings that the utilisation of CDW in CPB mixtures can considerably enhance the quality (i.e. strength and durability, microstructure) of CPB and mitigate the leachability of the hazardous pollutants (except As) from the CPB. In this respect, it may also provide the safely storage and hence environmentally sound management of CDW.

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Author contributions T Y contributed to conceptualization; investigation; methodology; resources; validation; visualization; roles/writing—original draft; B E contributed to supervision; writing—review and editing; funding acquisition; project administration.

Declarations

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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