



## Leaching Behavior of Construction and Demolition Waste (Concrete and Gypsum)

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### A B S T R A C T

Recycling of Construction and Demolition Waste (CDW) aims to minimize the generation of waste and reduce the dependency on natural resources. The aims of the research are to characterize inorganic element and to determine the leaching behavior of CDW (concrete and gypsum) by means of the leaching test. The analyzed results were compared with the European Union (EU) Landfill Directive to assess their acceptance criteria. Both wastes were found to have elements of Ca, Mg, Fe, Zn, Mn, Pb, Cu, Cd, As, Cr, Se, Ni, Cl and  $\text{SO}_4^{2-}$ . The highest concentration and variety of inorganic element found in waste gypsum (WG) were  $\text{SO}_4^{2-} > \text{Ca} > \text{Cl} > \text{Mg} > \text{Zn} > \text{Cu} > \text{Fe}$ . X-ray diffractometric (XRD) analysis proved that the WCo was dominated by quartz, calcite, ettringite, cordierite, diopside and the WG was only dominated by gypsum. The leaching behavior of WG demonstrated pH dependent particularly for the elements of Ca, Mg, Fe, Zn, Cu and Mn but only the elements of Ca and Cr in WCo were shown to be pH dependent in the leaching test. The element of  $\text{SO}_4^{2-}$  from the WG indicated a higher reading than WCo without the influence of pH. Noticeably, the concentration of  $\text{SO}_4^{2-}$  within the WG strongly require regulation and control before it can be utilized as part of raw materials in the production of environmental friendly recycled building materials.

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## INTRODUCTION

The Malaysian Construction Industry Master Plan 2006-2015 is a program developed in the effort to achieve sustainable development in the construction industry by promoting the use of the Industrialized Building System (IBS) [1]. This program aims to maintain sustainability in the construction industry through the efficient use of natural resources, improvement in the quality of construction materials and minimization of generated waste. Globally, the annual generation of Construction and Demolition Waste (CDW) increased the amount of waste in the landfill. In the US, 323 million tons of CDW has been generated whereas in Australia about 16-40% of total solid wastes that were dumped in the landfill were CDW [2, 3]. In China, in the year 2007 [4] 3,158 tons of CDW were dumped into the landfill per day and approximately 40% of the total municipal solid wastes

were CDW [5]. A case study in Malaysia reported that a total of 175,000 tons and more than 100,000 tons of CDW were generated annually in Kuching and Samarahan, respectively [6]. The dumping of CDW is on or nearby a municipal solid waste landfill [7] has become one of the most common waste disposal practices due to the lack of proper management by the responsible agencies. Low pH ( $< \text{pH } 6$ ) of leachate released from young municipal solid waste landfill [8] in Malaysia has possibly changed the properties of CDW due to the leaching behavior of chemical elements from such waste. Leaching of chemical elements from leachate is highly related to pH, for an example, maximally ammonium were leached out from leachate at pH 9.5 [9]. Furthermore, direct leaching of heavy metal from the non-sanitary municipal solid waste landfill with an impact on water had resulted in the decomposition in the soil [10], which subsequently affected the groundwater. In recent times, public awareness on the effect of leachate on groundwater contamination has caused serious concerns [11].

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With the intention of achieving the target of IBS through the efficient use of natural resources and improvement in the quality of construction materials, many studies have reported that substitution of natural resources with CDW in the production of recycled building materials have the potential of being utilized in various applications either as a structural or non-structural concrete. However, the drawback in the properties of recycled concretes made from CDW can be attributed to the changes in their physicochemical properties such as density, water absorption, and sulfate content, which needed to be overcome [12]. Thus, the study of the mechanical structure features of recycled concrete has become increasingly crucial [13-15]. Considerable efforts have been made by researchers to achieve recycled concrete with superior mechanical strength through the examination of the role of percentage substitution of CDW with natural resources, comparison of the effects of fine and coarse aggregates uptake and the addition of agro-waste. Apart from achieving a high mechanical strength of the recycled products, the direct or indirect environmental impacts of these products after the application on building structures require evaluation due to the high possibility of causing the leaching of hazardous elements into the environment when they come in contact with rain water, surface water or groundwater [16].

Thus, the lack of comprehensive environmental assessments on CDW needs to be addressed besides focusing on the production of good mechanical features of recycled building material. The evaluation of chemical - mineralogical characteristics and the release behavior of chemical element from CDW are required before these materials can be used in construction and building material. The pH dependent leaching behavior of CDW is one of the most relevant methods to evaluate the effects of environment condition on the properties of construction material where the amount of potential pollutant might leach out into the soil; surface and groundwater should be taken into account. The effective evaluation of the environmental impact should not rely on the total content of heavy metal, but the amount of pollutant leached from a material should be referred to instead [17,18].

This paper focuses on the characteristics of the inorganic elements and also to determine the leaching behavior of CDW through leaching test. The concentration of each inorganic element obtained was compared to the European Union (EU) Landfill Directive (Inert Waste) [19].

## MATERIALS AND METHODS

### Collection and Preparation of Samples

Two different types of Construction and Demolition Waste (CDW) were prepared: waste concrete (WCo) and waste gypsum (WG). A total of 50 kg of the samples were collected thrice with a weekly interval as triplicate at the nearby Papan Landfill located at Batu Gajah, Ipoh, Malaysia. The selected CDW were segregated from other types of waste. Ready-made concrete (NC) and gypsum (NG) were obtained commercially as a controls. Both CDW were homogenized and reduced using the quartering method according to the British Standard: Testing aggregate, Part 102: Methods for sampling (BS 812-102:1989) [20]. A preliminary assessment indicated 0.3 mm of the fine aggregate has the highest content of sulfate compound which need to be resolved before being utilized with the aim of complying with the concrete standard. 0.3 mm of the fine aggregate from each sample was prepared according to the British Standard: Testing aggregate, Part 103: Sieve Test (BS 812-103.1:1985) [21].

### Characterization of Inorganic Element

The composition of inorganic elements of WCo and WG were examined using the X-ray fluorescence (XRF) spectrometry, whereas the mineralogical constituent was examined using the X-ray diffractometric (XRD) techniques with diffraction patterns in the  $2\theta$  angular ranging from  $2^\circ$  to  $90^\circ$ . The entire test samples are manually grounded into the size of below  $75\ \mu\text{m}$  before being homogenized for the analysis of XRF and XRD.

$\text{SiO}_2$ ,  $\text{CaO}$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{MgO}$ ,  $\text{Fe}_2\text{O}_3$ ,  $\text{SO}_3$ ,  $\text{P}_2\text{O}_5$ ,  $\text{K}_2\text{O}$ ,  $\text{TiO}_2$ ,  $\text{MnO}$ ,  $\text{Na}_2\text{O}$ ,  $\text{Rb}_2\text{O}$ ,  $\text{SrO}$ ,  $\text{ZrO}_2$ ,  $\text{Ag}_2\text{O}$  and  $\text{Cr}_2\text{O}_3$  were identified through XRF, while, XRD were found on quartz ( $\text{SiO}_2$ ), calcite ( $\text{CaCO}_3$ ), ettringite ( $\text{Ca}_6\text{Al}_2(\text{SO}_4)(\text{OH})_{12}\cdot 26\text{H}_2\text{O}$ ), cordierite ( $\text{Mg,Fe}_2\text{Al}_4\text{Si}_5\text{O}_{18}$ ), diopside ( $\text{MgCaSi}_2\text{O}_6$ ), albite ( $\text{NaAlSi}_3\text{O}_8$ ), hematite ( $\text{Fe}_3\text{O}_4$ ), portlandite ( $\text{Ca}(\text{OH})_2$ ) and gypsum ( $\text{CaSO}_4\cdot 2\text{H}_2\text{O}$ ).

### Leaching Test

The leaching test (CEN/TS 14429) was to assess the leaching behavior of inorganic elements at different pH values [22]. The samples were agitated for a period of 48 h using an end-over-end mechanical roller followed by an overnight sedimentation to achieve an equilibrium concentration. The suspensions were prepared with deionized water ( $>18.2\ \text{M}\Omega\ \text{cm}$ ) in acid cleaned HDPE vessels with a concentration of  $10\pm 0.3\ \text{mL/mg}$ .  $\text{HNO}_3$  and  $\text{NaOH}$  were added to achieve the end-pH values in regular intervals between 2 to 13. The material pH was determined before the suspensions were filtrated through the  $0.45\ \mu\text{m}$  membrane filters. Subsequently, chemical analysis were performed using inductively coupled plasma - optical emission spectrometry (ICP-OES) to examine the availability of inorganic element,

while anions (sulfate and chloride) were analyzed using ion chromatography spectrometry.

## RESULTS AND DISCUSSION

### Characterizations of Inorganic Element

Table 1 demonstrates the chemical compositions of WCo and WG obtained using the XRF analysis. The major oxides existing in the WC were CaO with a total of 70.88%, followed by SiO<sub>2</sub> (20.68%). However, NC was observed to have a higher percentage of the compound of SiO<sub>2</sub> (43.20%) compared to CaO (38.41%). WG and NG differed in major oxide contents, which were SO<sub>3</sub> and CaO. Evidently, WG appeared to have a higher percentage of SO<sub>3</sub> (41.51%) than CaO (31.2%). However, NG showed a higher percentage of CaO (54.68%) than SO<sub>3</sub> (42.57%). In fact, there were only slight differences in the percentages of sulfur trioxide between NG and WG, but the reduction of CaO in WG might be due to leaching activity from the media. Mashitah et. al. also detected a higher amount of CaO in WCo and the presence of such compound was closely related to the surface attached cement paste onto the surface of concrete based waste materials [23].

Cementitious materials from Portland cement contained three main compounds (calcium silicate hydrates (CSH), CaO.SiO<sub>2</sub>.H<sub>2</sub>O as well as aluminates and the consequence of the reaction between these three main compounds have led to the elevated amount of

CaO [24]. Moreover, oxyanions such as Cr<sub>2</sub>O<sub>3</sub>, Nb<sub>2</sub>O<sub>5</sub> and Ag<sub>2</sub>O were only found in the NC. Hence, these results may definitely be closely related to the transport behavior of concrete aggregate during their service life.

For the chemical composition of SO<sub>3</sub>, NC recorded a higher amount than WCo with a total of 1.93% and 0.61%, respectively. The SO<sub>3</sub> content of WCo exhibited close to the limit of the Spanish Structural Concrete Code (EHE-08, 2008) [25] and also the European standard EN-12620 (2008) [26]. In contrast with NC, the SO<sub>3</sub> content in both WG and NG has exceeded the limits of both standards. Since the concentration of sulfate in WCo falls within the standard limits in both EN-12620 (2008) and EHE-08 (2008) (below 1%), it has the potential to be recycled as a secondary product.

Mineralogical characteristic analysis of WCo shows major compound of quartz (SiO<sub>2</sub>), calcite (CaCO<sub>3</sub>), ettringite (Ca<sub>6</sub>Al<sub>2</sub>(SO<sub>4</sub>)(OH)<sub>12</sub>•26H<sub>2</sub>O), cordierite (Mg,Fe)<sub>2</sub>Al<sub>4</sub>Si<sub>5</sub>O<sub>18</sub>), diopside (MgCaSi<sub>2</sub>O<sub>6</sub>), while, the minor peaks come from albite (NaAlSi<sub>3</sub>O<sub>8</sub>), hematite (Fe<sub>3</sub>O<sub>4</sub>) and portlandite (Ca(OH)<sub>2</sub>) (Figure 1(a)). On the contrary, WG was observed to show a high peak of gypsum, but minor peaks of quartz, calcite and ettringite, (Figure 1(b)). For qualitative analysis of XRD and XRF, the results reveal a peak of ettringite, which could possibly be the outcome of the carbonation of WCo. As reported by previous research, the increment of Ca content in WCo may be due to the reaction between ettringite and cement [27, 28].

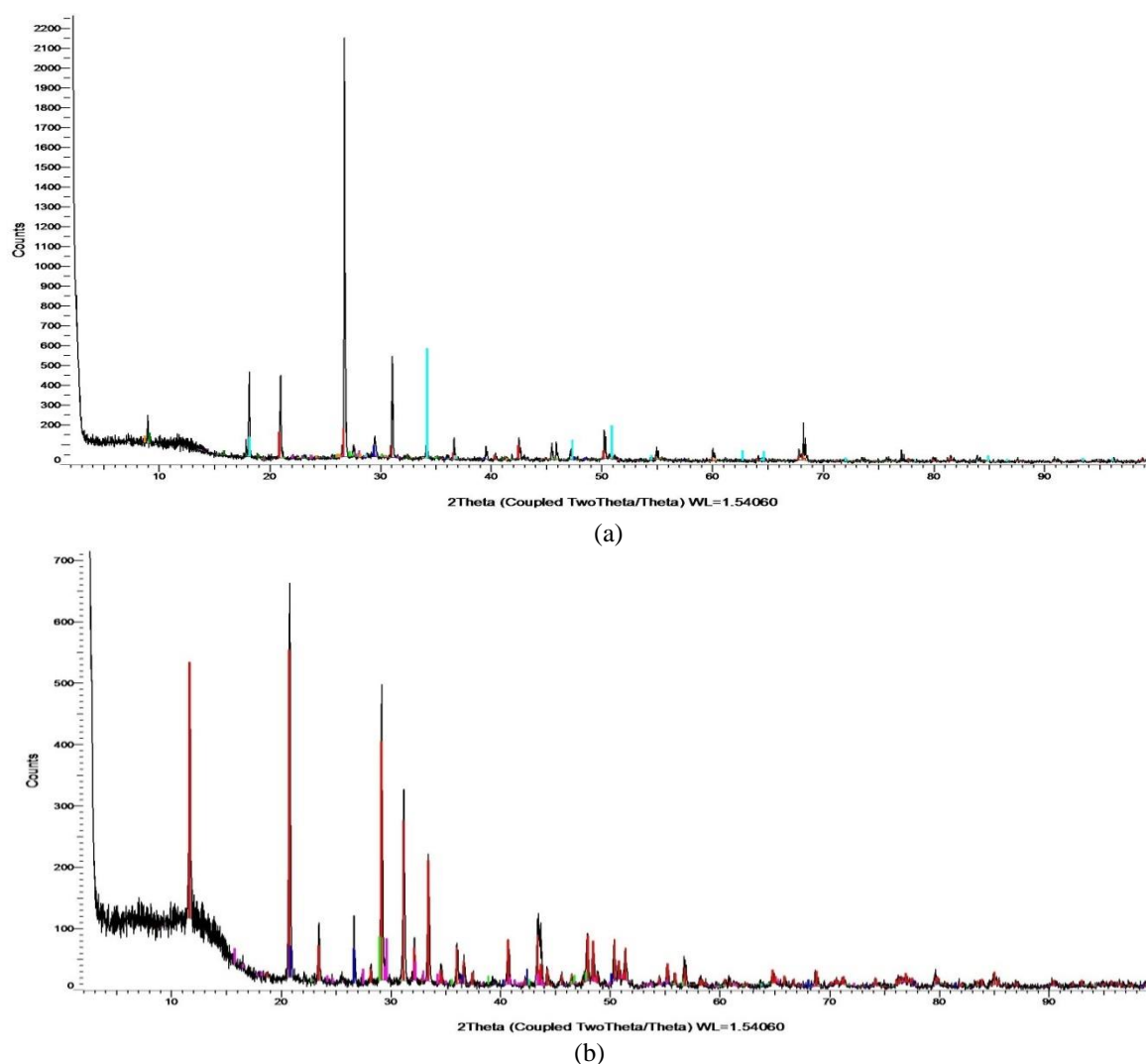
**TABLE 1.** List of inorganic element presents in four different types of CDW via XRF.

Element	Types of CDW (wt %)			
	WCo	NC	WG	NG
SiO <sub>2</sub>	20.68 ±6.47*	42.15 ±1.20*	-	-
CaO	70.88 ±9.22**	38.42 ±2.42**	31.60 ±0.57	53.34 ±1.90
Al <sub>2</sub> O <sub>3</sub>	3.43 ±1.52	8.01 ±0.43	0.36 ±0.01	0.14 ±0.07
MgO	1.99 ±0.19	3.61 ±0.55	0.06 ±0.01	1.04 ±0.21
Fe <sub>2</sub> O <sub>3</sub>	1.38 ±0.73	2.78 ±0.66	0.18 ±0.01	0.05 ±0.02
SO <sub>3</sub>	0.61 ±0.47	1.93 ±0.01	41.21 ±0.43***	42.24 ±0.47***
P <sub>2</sub> O <sub>5</sub>	0.06 ±0.02	0.06 ±0.18	0.01 ±0	0.01 ±0.003
K <sub>2</sub> O	0.67 ±0.13	1.25 ±0.18	0.07 ±0	0.01 ±0
TiO <sub>2</sub>	0.11 ±0.04	0.29 ±0.06	0.02 ±0.01	0.002±0.001
MnO	0.06 ±0.55	0.09 ±0	0.01 ±0	0.02 ±0.01
Na <sub>2</sub> O	0.06 ±0.01	0.10 ±0	-	-
Rb <sub>2</sub> O	0.01 ±0	0.02 ±0.01	-	-
SrO	0.04 ±0.01	0.02 ±0.01	-	-
ZrO <sub>2</sub>	0.02 ±0.01	0.08 ±0.01	-	-
Ag <sub>2</sub> O	-	0.05 ±0.01	-	-
Cr <sub>2</sub> O <sub>3</sub>	-	0.04 ±0.01	-	-

\* Significantly different at  $p \leq 0.05$ ,  $F_{(1,2)} = 21.24$ ,  $p = 0.044$

\*\* Significantly different at  $p \leq 0.05$ ,  $F_{(1,2)} = 21.76$ ,  $p = 0.043$

\*\*\* Not significantly different at  $p \geq 0.05$   $F_{(1,2)} = 0.1$ ,  $p = 0.783$

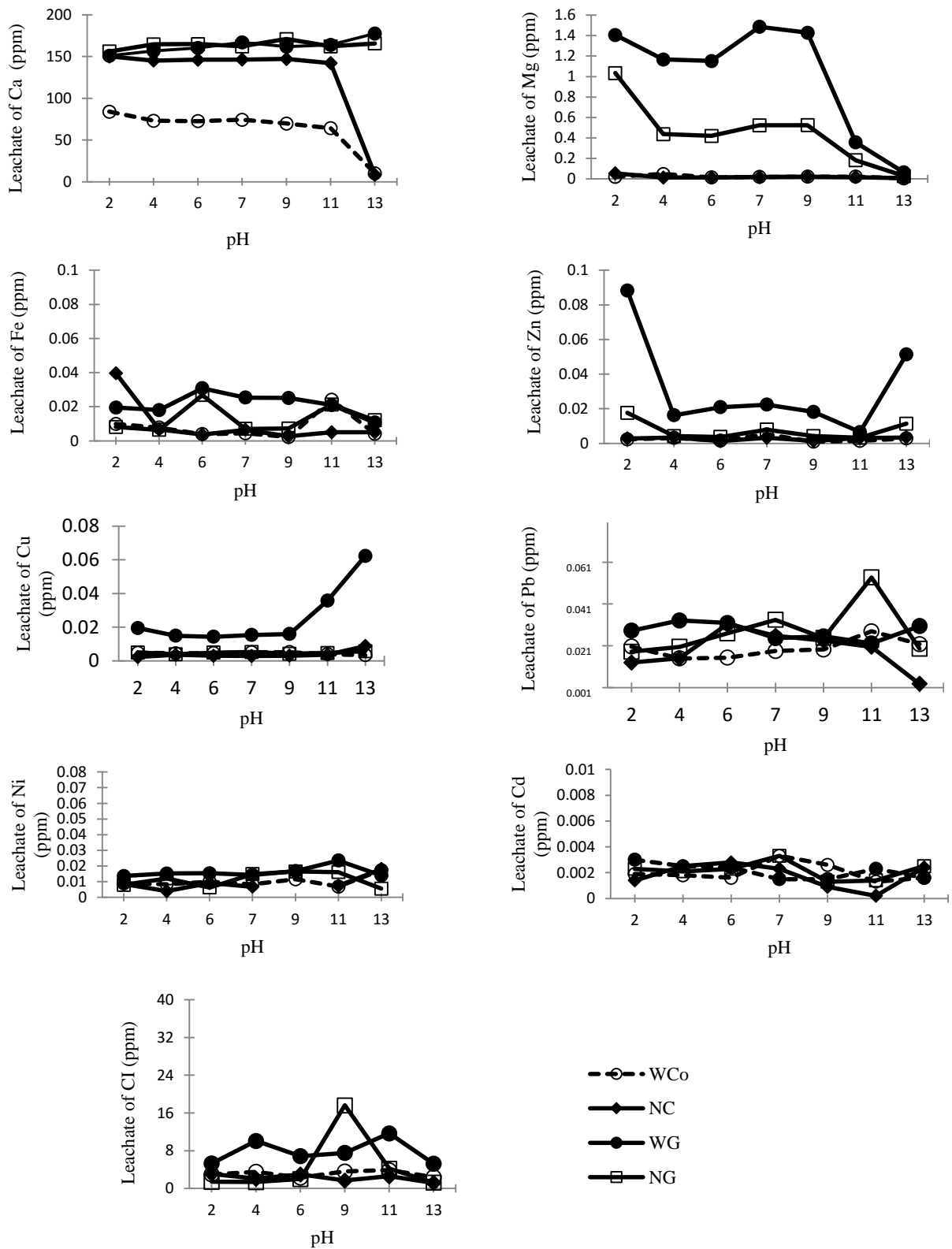


**Figure 1.** (a) X-Ray Diffraction pattern for the presence of chemical compound in WCo. (b) X-Ray Diffraction pattern for the presence of chemical compound in WG

### Leaching Behavior of Inorganic Elements

CDW is commonly disposed of in Malaysia by dumping it in the vicinity of municipal solid waste landfills [7]. The situation is worsened especially when the wastes were dumped onto young landfill leachate with a lower pH (< 6.5) [8, 29, 30] or acidic field site which can possibly result in progressive changes in the strong alkaline properties of CDW. Therefore, the leaching behavior of a wide range of pH value is required to determine the possibility of inorganic elements being leached out into the environment. The effective evaluation of environmental impact does not solely rely on the total content of heavy metal, but the total amount of contaminant that can be leached out, infiltrated into the

soil and also dissolved in water were referred instead [18,31]. Under the leaching test, the elements present in the leachate were Ca, Mg, Fe, Zn, Mn, Pb, Cu, Cd, As, Cr, Se, Ni,  $\text{SO}_4^{2-}$  and Cl. As demonstrated in Figure 2, Ca is highly immobilized in the leachate for all the samples but gypsum products (WG and NG) have the tendencies to contain the highest amount of Ca compared to WCo and NC. In the comparison between fresh and aged cement-based materials, the concentrations of Ca in WCo are lower than NC. Thus, three differently released patterns can be observed for both materials (i) highly carbonated and aged concrete, (ii) fresh concrete and (iii) gypsum.



**Figure 2.** The effect of pH toward leached of inorganic elements in WCo and WG. Each waste was compared with Natural Concrete Aggregate (NC) and Natural Gypsum (NG) as control.

These patterns are slightly similar to the findings reported by Engelsen and his co-worker where they claimed that the leaching behavior of cement-based material was related to the degree of carbonation [32, 33]. In line with the data generated from the XRD analysis, WG manifested a high peak of calcium compound, which came from a compound of gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) and the peak appeared to be higher than the calcium compound of portlandite in WCo. Comparisons between WCo and WG clearly showed different leaching patterns of Mg and Zn, in which WG released the elements in more than one order of magnitude compared to WCo. Although the results of the XRF analysis proved that the elements of Ca, Mg, and Zn were highly detected in WCo compared to WG, these elements tend to leach higher in WG than WCo. Hence, the leaching patterns of Ca, Mg, and Zn were not affected by the total content of chemical in each material.

In the determination of pH dependent leaching behavior of inorganic elements, the precipitations of meta at high alkaline condition were different between concrete (WCo and NC) and gypsum (WG and NG). At a high pH (pH13), Ca was insoluble in concrete (WCo and NC). However, in such condition, the gypsum product was not affected. The unstable phase of calcite in low pH condition caused the release of Ca from cementitious material [34]. In spite of that, in an alkaline region, Mg demonstrated a declination in the leaching trend for both materials. High leaching amount of Ca and Mg at high pH in aged concrete (WCo) were in accordance with the findings of Solpuker et.al. [34]. In contrast, the acidic eluent showed less solubility of Cu in aged WCo and WG. At high acidic and alkaline pH (pH 2 and pH 13), Zn was highly leached from the WG. Noticeably, aged WCo showed a high concentration of Cr particularly at a high pH (pH 11 – pH 13) condition. Although a previous study reported the leaching behavior of Cr was close to the regulatory limit of the European Landfill (EU landfill) [35], but the highest concentration of Cr for WCo at pH 11 and pH 13 are still within the requirement range. Past studies reported that the leaching of As, Se and Cr from WCo were related to the degradation of hydrated attached mortar on the surface, where the presence of anionic species in the form of  $\text{AsO}_4^{3-}$ ,  $\text{CrO}_4^{2-}$  and  $\text{SeO}_4^{2-}$  may result in the substitution of  $\text{SO}_4^{2-}$  and  $\text{SiO}_4$  in their structures after they achieved a certain level [32, 36].

Such elements (As, Se and Cr) tend to have sorption with calcium silicate hydrate (C-S-H), portlandite, ettringite, monosulfate cement phase [36-38] calcium sulfoaluminate phase (AFt) and calcium aluminate hydrates (AFm) [39, 40]. This process frequently occurs in the presence of carbonation, Internal Sulfate Attack (ISA) or dissolution of ettringite during their service life [27, 41]. Carbonation takes place when the cement matrices are partially dried or during the existence of a relative humidity gradient which can possibly cause the diffusion of  $\text{CO}_2$  (from atmosphere) into the pore structures and consequently react with cations and aqueous hydroxide to form carbonates as the neutralization of solid matrix such as calcite ( $\text{CaCO}_3$ ) [42].

The existence of carbonate in concrete aggregate imposed carbonation process is caused by the cement hydrate phases [33]. Furthermore, in conjunction with the depletion of aqueous hydroxide such as portlandite ( $\text{Ca(OH)}_2$ ), the process of decalcification and polymerization of C-S-H occurs to leach Ca ion in aged WCo [43]. The formation of ettringite and portlandite was probably due to the adsorption on the surface by a retention mechanism of  $\text{AsO}_2^-$  [44],  $\text{AsO}_4^{3-}$  [45] and  $\text{SeO}_3^{2-}$  [46].

Based on the XRF analysis, WG consisted of a high amount of  $\text{SO}_4^{2-}$  (41.5%) but WCo tends to be high in  $\text{SiO}_2$  (20.7%). Therefore, the substitution of sulfate and silicate ion with the elements of As, Cr, Se, Mn mostly occurred in both CDW. Since the percentage of  $\text{SO}_4^{2-}$  in WG was higher than the concentration of  $\text{SiO}_2$  (WCo), the sorption of inorganic elements (As, Se, Mn) was also highly inclined towards WG and leached at selected pH after achieving the equilibrium phase. As a result, the concentration of  $\text{SO}_4^{2-}$  leached out from WG was higher than WCo due to the maximum sulfur content ( $41.21 \pm 0.43\%$ ) as shown in XRF analysis. On the other hand,  $\text{SO}_4^{2-}$  in WG has totally exceeded the standard limit of EU Landfill, which must be below 100ppm (Figure 4).

Markedly, WG from CDW definitely requires proper disposal management due to its potential of leaching high concentration of sulfate to the environment. The detrimental effects of sulfate to the environment have been one of the main concerns of many researchers in the effort of fulfilling the acceptable criteria for concrete production.

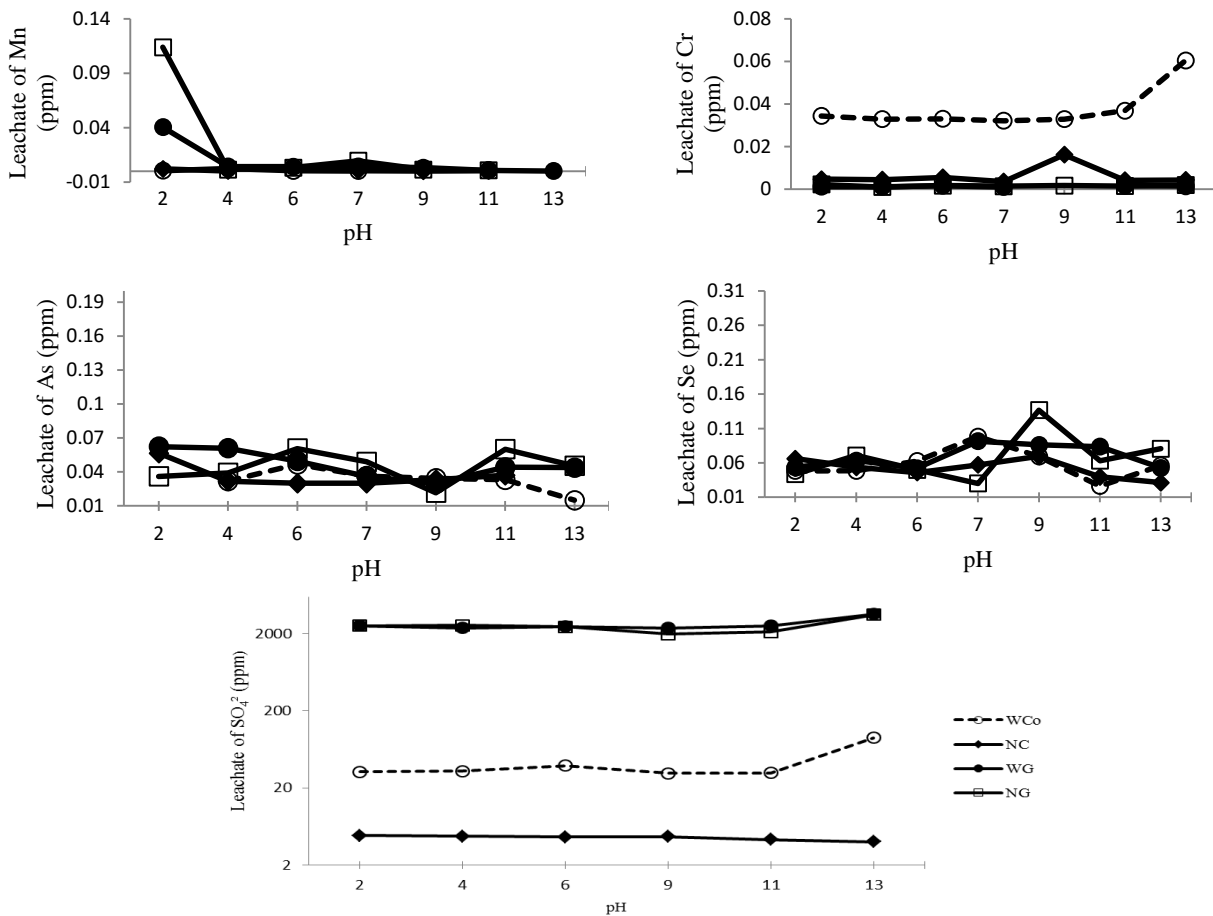


Figure 3. pH dependent leaching behavior of inorganic elements

## CONCLUSIONS

The common disposal practice of CDW requires proper management since such waste can be utilized as recycled concrete for building and infrastructure construction purposes. Feasibility study on CDW is necessary to produce environmental friendly products, which comply with the standard requirements of concrete. Characterization of inorganic elements via XRF demonstrated both waste consist of the elements of Ca, Mg, Fe, Zn, Mn, Pb, Cu, Cd, As, Cr, Se, Ni, Cl and  $SO_4^{2-}$ , while, XRD analysis proved that WCo was dominated by quartz, calcite, ettringite, cordierite, diopside but WG contained high amount of gypsum. However, the highest concentration and variety of inorganic element were found in WG were  $SO_4^{2-}$  > Ca > Cl > Mg > Zn > Cu > Fe. The leaching behavior of WG demonstrated pH dependent particularly for the elements of Ca, Mg, Fe, Zn, Cu and Mn, but only the elements of Ca and Cr for WCo were shown to be pH dependent. In term of environmental concern, the element of  $SO_4^{2-}$  from WG strongly requires regulation and control before it can be utilized as part of the raw

materials in recycled building materials, since,  $SO_4^{2-}$  displayed higher reading than WC without influence of pH and this result had exceeded the standard limit of EU Landfill Directive [19].

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**Persian Abstract**

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**چکیده**

بازیافت پسماند ساختمانی (CDW)، (اعم از ساخت یا تخریب) با هدف به حداقل رساندن تولید ضایعات و کاهش وابستگی به منابع طبیعی انجام می‌شود. از جمله اهداف این تحقیق، شناسایی عناصر غیرآلی و تعیین رفتار آبشویی زائادات ساختمانی (بتن و گچ) توسط آزمایشات لیچینگ بوده است. نتایج بدست آمده با استانداردهای دفع زبالات در دستورالعمل اتحادیه اروپا مقایسه گردید. در هر کدام از این زائادات حاوی عناصری همچون کلسیم، منیزیم، آهن، روی، منگنز، سرب، مس، کادمیم، کروم، سلنیوم، نیکل، کلر و سولفات یافت شد. بالاترین غلظت و تنوع عناصر غیرآلی در زائادات گچی به صورت SO<sub>4</sub><sup>2-</sup> >Ca>Cl>Mg>Zn>Cu>Fe وجود داشت. آنالیز پراش اشعه ایکس (XRD) اثبات نمود که عناصر معدنی غالب در زائادات بتنی شامل کوارتز، کلسیت، اترینگایت، کوردیریت، دیوپسید بوده، در حالی که تنها عنصر معدنی غالب در زائادات گچی، گچ بوده است. رفتار لیچینگ زائادات گچی نشان‌دهنده وابستگی فوق‌العاده pH به عناصر کلسیم، منیزیم، آهن، روی، مس و منگنز بوده، اما در زائادات بتنی این وابستگی تنها نسبت به عناصر کلسیم و کروم نشان داده شد. عنصر سولفات از زائادات گچی عدد بالاتری را نسبت به زائادات بتنی بدون تاثیر pH نشان داد و به طور برجسته‌ای ضرورت تنظیم غلظت سولفات زائادات گچی و کنترل آن پیش از مصرف، به عنوان بخشی از مواد خام در تولید مواد بازیافت ساختمانی دستدار محیط زیست، را نشان می‌دهد.

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