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Geochemistry and Leaching of Heavy Elements of Magnetite and Hematite Iron Ore Tailings in Un-Stabilized and Stabilized State

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Abstract

It is proven that the financial benefits of using the proposed new tailings materials in terms of environmental sustainability are much higher when compared to normal materials. The research has been conducted to investigate the possible environmental problems of using iron ore mine tailings in civil engineering projects (road construction materials). Therefore, two main iron ore mine tailings of Gol-e-Gohar complex produced in magnetite and hematite plants were evaluated in terms of leaching heavy elements after application as road materials. To address the potential application of tailings as road materials, the main characteristics of tailings such as elemental analysis, pollution indicators, and engineering properties after stabilization by Portland cement were measured. The leaching of dangerous elements in a natural and stabilized state of compacted tailings at different times and pHs were assessed. The total amount of elements by ICP analysis showed that the Mn, Fe, Zn, Cd, Ni, and Cu are more abundant in the two tailings and their leaching must be tested. The general results showed that leaching with different leakage times and solutions at different pH have no effect on the release of elements through tailings, so their efficiency as road materials from an environmental point of view is acceptable providing meet the engineering standards as materials. In addition to leaching experiments, contamination, enrichment, and geo-accumulation indices also confirmed leaching results.

Keywords: Environmental sustainability, Elements Releasing, Engineering properties, Chemical properties

1. Introduction

Mining is one of the most energy-intensive industries worldwide. It also provides a critical source of raw materials for the manufacturing, transportation, construction, and energy sectors [1]. Iron mining is an extremely important economic activity in many countries [2]. Iron ore production is expected to rise steadily to reach 2.7Bt by 2025, according to Global data's estimates. Limited by science and technology, the overall utilization rate of tailings is not high at present; nearly 78% of the tailings are piled on the surface, resulting in a considerable waste of resources [3]. Increased mining activity of iron ore has led to the generation of voluminous wastes of various nature, especially during the different stages of its extraction and production. One such waste which is generated during the beneficiation process of iron ore is waste iron ore tailings, which is also termed as Iron Ore Tailings (IOT) [4]. On the other hand, the large volume of tailings generated, IOTs, is a huge problem for the mining companies, governments, and main populations around the mines due to storage in dams and the risk of accidents [2]. Mine tailings are a potential source of environmental pollution because they typically contain potentially toxic elements (PTEs) and the residue of chemical compounds used during extraction processes [5].

Mines provide essential materials for life and human progress; however, on the other hand, the possibility of life and having healthy environment is adversely affected by increasing pollution. Mine waste (tailing) is one of the pollutants generated by mines that should be properly managed. Nowadays, strategic management by using appropriate tools and technology is essential for the proper management of waste everywhere, including the urban, rural, medical, industrial, and mining environments [6]. The problem of new researches to solve is how to convert these tailings into value added products for civil engineering applications for building and construction projects. This will reduce or eliminate the problems that mine tailings bring to the environment and human health. It will also ensure economic and environmental sustainability of the mining industries and provide cheaper alternative materials for building and construction projects. Comprehensive utilization of mine wastes, especially in building and construction applications are seen to have the greatest potential in finding a sustainable solution to these problems. This is because; it will ensure high volume utilization of mine wastes to significantly reduce the quantity of mine wastes in the environment or possibly to eliminate them completely [7].The improper disposal of IOT on open ground will adversely affect the natural resources such as water, air and soil, and utilizing IOT in construction industry can alleviate the environmental problems associated with disposal of such wastes [4]. Leaching is a hydrometallurgical process where metals are extracted by chemical dissolution, leaving behind a residue of inert minerals originally present as well as insoluble decomposition products of the reacted mineral [8]. The leaching from mine tailings results in contamination of nearby ground waters and rivers by hazardous metals like copper (Cu), zinc (Zn), and iron (Fe) [9]. The main IOT applications are in civil construction (aggregates for concrete, mortar, and Portland cement additives), ceramic industry, geo-polymer, synthesis of new materials such as zeolites, meso-porous silica, carbon nanotubes, adsorbents, and catalysts for different reactions, in batteries and in fuel cells [2]. The most promising technical achievement of stabilization is that the cement-waste mixture can be used in the base and sub-base layers of roads and highways around Gol-e Gohar mine (Sirjan, Iran), and the development of infrastructure around the mine can be done at a lower cost [10]. Gol-e Gohar mine zone is one of the oldest area of iron ore extraction with several thousand years of mining history. By possessing half billion tons of iron ore reserves, currently is one of the most active mining and industrial zones in the Middle East. This paper aims to explore the environmental issues related to magnetite and hematite iron ore tailings when these wastes use as road materials. In fact, it is necessary to evaluate the leaching of pollutants through raw and cement-stabilized tailing and if there are hazardous elements more than the permissible levels for the environment, its use should be prohibited or a solution should be provided.

2. Materials & Methods

2.1 Study Area

Gol-e Gohar Mining Zone With its rich iron ore mines as one of the most prominent active mining poles in the Middle East. These mineral deposits are located in Kerman province, 50 km southwest of Sirjan city. Gol-e Gohar iron ore deposit is located in six separate anomalies with a reserve of about 1200 million tons in a range of approximately 10 km in length and approximately 4 km in width. Products extracted from this mine are: iron ore pellets, iron ore concentrate and granulated ore.

2.2 Study Materials

Iron ore tailings (IOTs) are a form of solid waste produced during the beneficiation process of iron ore concentrate [11]. In this study, two iron ore tailings including magnetite and hematite tailings which has been collected from hematite and magnetite plants of Gol-e Gohar strict, where considered. Due to changes in feed conditions and operational problems, 75 tons per hour of hematite with dimensions less than 80 microns and 38% iron grade is transferred to the tailing dams [12].The other materials used in this study include Portland cement type II as stabilizer, sulfuric acid (98%) and hydroxide sodium for adjustment of pH values.

2.3 Physico-chemical properties of tailings

Soil classification [13, 14], Atterberg limits [15], specific gravity [16], compaction [17, 18], unconfined compressive strength (UCS) [19] and permeability [20] tests were performed according to ASTM standard for raw and cement-stabilized tailings. Tailings were stabilized using 5% of Portland cement by weight of dry tailings and sample were compacted with standard energy cured for 28 days in plastic bags at room temperature. This percentage of Portland cement satisfied the minimum allowable unconfined compressive strength (2.1 MPa) for base materials. The total elements of tailings was measured by ICP Mass model ELAN 6100 DRC-e made by Perkin Elmer analysis. The composition of the elements as oxides was measured using an X-ray fluorescence spectroscope (XRF) model PW1410 made by PHILIPS Company, Netherlands.

2.4 Leaching and concentration of elements

The column (Figure. 1) was oriented vertically and slowly saturated from the top with deionized water. Then, the soil column was allowed to stabilize for 24 h. Experiments were conducted in triplicate at each pH (4.7, 7 and 9.5) and time (1 hour, 24 hours and one week) treatments. The water head was constant until the end of experiment. The leachate from the soil column was collected to determine the heavy metal concentrations by AAS apparatus model Analyst 800 made by Perkin Elmer.

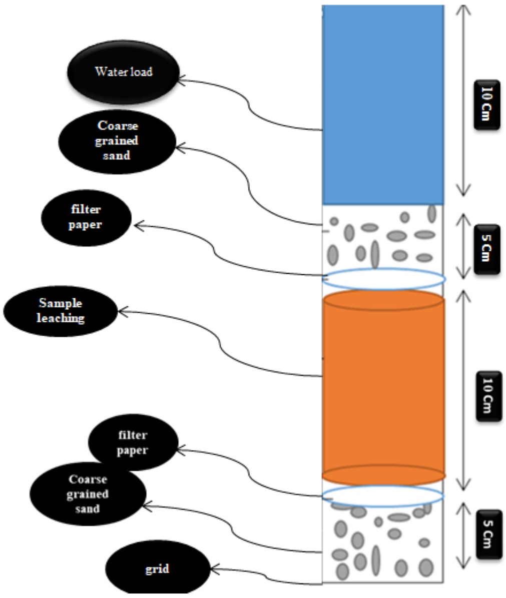

Figure 1. Soil column for leaching experiments

2.5 Contamination indices

Enrichment factor (EF): the enrichment factor was calculated based on Equation 1.

$$EF = \left[\frac{\left(\frac{Cm}{Cref}\right)sample}{\left(\frac{Cm}{Cref}\right)background}\right] \quad (1)$$

The concentration of the base metal in the same base medium (AL) [21].

Contamination Factor: the pollution factor was calculated based on Equation 2.

$$CF = \frac{(C)sample}{(C)background} \quad (2)$$

Pollution load index: the pollution load index was calculated based on Equation 3.

$$PLI = \sqrt[n]{CF1 \times CF2 \times \ldots \times CFn} \quad (3)$$

Where, CF is the cause of pollution of each metal element and n is the number of measured heavy metals [22].

Geo-accumulation Index: the geo-accumulation Index was calculated based on Equation 4.

$$Igeo = \log_2\left(\frac{Cm}{1.5 \times Bm}\right) \quad (4)$$

Where Bm is reference concentration and number 1.5 in the formula is due to the influence of terrestrial factors to eliminate the influence of different lithology in the region [23].

3. Results & Discussion

3.1 Characteristics of iron ore tailings

The physical properties of magnetite and hematite tailings are show in Table 1. According to Table 1, the magnetite iron ore tailing was classified as SM and A-2-4 according to the USCS and AASHTO systems, respectively, while the hematite iron ore tailing was classified as ML and A-4 according to the USCS and ASSHTO systems, respectively. The characteristic of the tailings depends on the type of ore mined. Sieve analysis of Itakpe (Kogi State, Nigeria) iron ore tailings showed that tailings were fine to medium sand which D50 of tailing particles was 0.2 [24]. The iron ore tailings were alkaline according to the results in Table 1.

Table 1. Properties of Magnetite and hematite iron ore mine tailings.

properties	Standard	Magnetite iron ore tailings	Hematite iron ore tailings
AASHTO	ASTM D3282	A-2-4	A-4
USCS	ASTM D2487	SM	ML
Specific Gravity	ASTM D854	3.0	3.16
Liquid limit	ASTM D4318	NP	19.7
Plastic limit	ASTM D4318	NP	15.9
Plasticity index	ASTM D4318	NP	3.8
pH	ASTM D4318	7.58	7.49

In brazil, the identified minerals of iron ore tailings by XRD, optical microscopy and thermal analyses were quartz, hematite (martitic, lamellar, granular, sinuous, specularitic), goethite (alveolar, amphibolitic, botryoidal), magnetite, kaolinite (5.3 wt%) and apatite (0.7 wt%) [25]. The major components of Itakpe iron ore mine (Nigeria) tailings were SiO_2 (71%) and Al_2O3 (2.62 %). The chemical analysis indicates that the higher the silica (quartz) and hematite content in the tailings the finer the particles [24]**.** The results of elemental analysis of the iron ore tailings by XRF are summarized in Table 2 .The results show that Si, Mg, Al, Ca, Na, K are the most abundant oxides in iron ore tailings, respectively.

Table 2. Chemical analyze of Magnetite and hematite iron ore tailings.

Element	Magnetite iron ore tailings (%)	Hematite iron ore tailings (%)
SiO_2	34.05	26.31
MgO	14.15	11.07
Al_2O_3	7.44	4.45
CaO	5.82	6.19
Na_2O	1.27	0.99
K_2O	1.25	0.44
TiO_2	0.54	0.36
MnO	0.06	0.05
BaO	0.02	0.01
Cr_2O_3	0.01	<0.01
LOI*	10.50	9.88

Table 3 shows the important characteristics of magnetite and hematite iron ore tailings. Particle size of hematite iron ore tailing is finer of magnetite iron ore tailing. Therefore percentage of optimum moisture and the UCS value for hematite tailing is higher than that for magnetite tailing. Addition of Portland cement to iron ore tailings leads to an increase in the optimum moisture content and a decrease in the maximum dry density [10]. According to the data in table 3, the optimum moisture content of hematite iron ore tailings increases with the addition of Portland cement. However, for magnetite iron ore tailings, the optimum moisture content does not change by adding Portland cement .The increase in maximum dry density can be due to the relatively higher specific gravity of cement, with a specific gravity of 3.17, has been added to the waste particles of iron ore, with a specific gravity of 3. Similar results have been reported by Yohanna et al. (2020)[26]. In general, the addition of Portland cement to iron ore tailings leads to an increase in UCS, and in addition, after curing the stabilized specimens, their unconfined compressive strength (UCS) was greatly increased [10]. Increasing the percentage of Portland cement increases the strength parameters such as UCS (unconfined compressive strength), E_0 (initial young's modulus) and ITS (indirect tensile strength) [27]. Optimum moisture content, maximum dry density, UCS, and permeability of raw and stabilized tailings are reported in Table (3). It is evident that the UCS of stabilized samples significantly increases after stabilization. Permeability of the tailings greatly depends on the percentage of fine particles. Permeability of the Itakpe iron ore tailings was 6.24×10^{-3} cm/sec [24]. Adding Portland cement as a stabilizer results in reducing the permeability coefficient of tailings.

Table 3. Important properties of Hematite and Magnetite tailings.

Iron ore tailings	K (Cm/s)	UCS (MPa)	OMC* %	MDD# (Cm^{-3})
Magnetite	7.04×10^{-7}	0.349	11.2	2.1
Hematite	1.68×10^{-7}	1.06	13	2.22
Magnetite + 5% Cement	2.64×10^{-8}	1.750	10.3	2.15
Hematite+ 5% Cement	5.745×10^{-7}	3.160	15.4	`2.11

3.2 Concentration of elements

The concentrations of two tailings elements, magnetite and hematite, were measured by ICP Mass and the results are shown in Table 6. The results show that the concentration of iron in both tailings is more than 10%.

Table 4. The result of ICP Mass analysis of Magnetite and Hematite iron ore tailings.

Element	Concentration in Magnetite (ppm)	Concentration in Hematite (ppm)	Element	Concentration in Magnetite (ppm)	Concentration in Hematite (ppm)
Ag	0.1	0.2	P	2561	2412
Be	0.6	0.4	Pb	48	71
Bi	0.5	0.5	Pr	5.6	4.93
Ca	38717	39483	Rb	34	13
Cd	0.3	0.4	S	>30000	27653
Ce	43	36	Sb	0.5	<0.5
Co	247	121	Sc	9.7	4.1
Cr	92	50	0.5	<0.5	<0.5
Cs	0.9	0.7	Sm	2.19	1.38
Cu	207	471	Sn	9.5	15.1
Dy	3.39	2.75	Sr	94.3	186
Er	1.86	1.51	Ta	0.51	0.39
Eu	0.83	0.37	Tb	0.6	0.49
Fe	>100000	>100000	Te	<0.1	0.12
Gd	3.67	3.16	Th	5.48	6.41
Hf	1.21	1.09	Ti	3006	1823
In	<0.5	<0.5	Tl	<0.1	<0.1
K	10055	4002	Tm	0.24	0.19
La	28	23	U	3.9	4.1
Li	12	12	V	109	135
Lu	0.28	0.21	W	<1	<1
Nb	8.5	7.9	Yb	18	14.6
Nd	19.7	17.2	Zn	60	87
Ni	94	123	Zr	17	20
Mg	>20000	>20000	Na	8657	5936
Mn	481	353	Mo	<0.1	<0.1

The leachate concentrations of Fe, Mn, Cu, Ni, Zn and Cd were measured as most abundant elements by atomic absorption spectrometry (Table 5). According to results shown in Table 5, the concentration of these elements in leachates is very low at different times and pH, so iron ore tailings can be used as materials in civil engineering applications without environmental concerns.The leaching behaviors of Zn, Cu, Fe and Mn in mine tailings from Dexing copper mine ore were investigated by a series of laboratory batch experiments indicated that the maximum leaching ratios of Zn, Cu, Fe and Mn at pH 2.0 were 5.4%, 5.8%, 11.1% and 34.1%, respectively. The dissolubility of all metals examined was positively correlated to the temperatures. The particle size would not change dissolution tendency of those heavy metals, but decrease the concentrations of leached heavy metal [28].The average concentration of some elements in Gol-e-gohar iron ore mine according to ICP-Mass analysis is show in Table 6. On average, the abundance of iron, aluminum, manganese, vanadium, nickel and cobalt in iron ore was higher than other analyzed elements [29].

Table 5. Heavy metal concentration of magnetite and hematite tailings in un-stabilized and stabilized state in leachate

Iron ore tailings	pH	Cu	Fe	Mn	Zn	Cd	Ni
Magnetite (un-stabilized)	4.70	0.04	0.42	0.55	0.02	0.02	0.11
	7.00	0.03	0.36	0.41	0.02	0.03	0.03
	9.50	0.06	0.33	0.38	0.01	0.03	0.00
Hematite (un-stabilized)	4.70	0.05	0.27	0.47	0.01	0.01	0.50
	7.00	0.05	0.27	0.15	0.01	0.03	0.05
	9.50	0.05	0.35	0.35	0.01	0.04	0.06
Magnetite + 5%Cement	4.70	0.02	0.25	0.05	0.00	0.02	0.05
	7.00	0.05	0.37	0.03	0.01	0.02	0.03
	9.50	0.39	0.31	0.00	0.00	0.01	0.00
Hematite +5% Cement	4.70	0.02	0.28	0.00	0.01	0.01	0.01
	7.00	0.01	0.22	0.00	0.00	0.01	0.02
	9.50	0.02	0.02	0.03	0.01	0.01	0.02

Table 6. Average concentration of some heavy metals- Gol-e-gohar iron ore [29].

Element	Concentration (ppm)	Element	Concentration (ppm)
Fe	85219.96	Cr	42.44
Al	18017.54	As	17.72
Mn	502.35	Pb	10.50
Ni	98.85	Mo	3.95
Co	81.61	Sb	0.17
V	70.09	Cd	0.14
Zn	63.04		

3.3 Analysis of Rare elements

The rare earth elements (REE) form the largest chemically coherent group in the periodic table.. Though generally unfamiliar, the REE are essential for many hundreds of applications (occupy 41% of global consumption). The versatility and specificity of the REE has given them a level of technological, environmental, and economic importance considerably greater than might be expected from their relative obscurity. Values were obtained from solutions with concentrations in the range of ppm of 14 elements corresponding to REEs: La, Ce, Pr, Nd, Sm, Sc, Y, Tb, Dy, Ho, Er, Tm, Yb, and Lu [30]. Table 7 shows the amount of trace elements in the Earth's crust [30].

Table 7. Rare elements in the earth's crust [30].

Element	Concentration (ppm)	Element	Concentration (ppm)
Sc	-	Tb	0.7
Y	-	Dy	3.8
La	30	Ho	0.8
Ce	60	Er	½
Pr	6.7	Tm	0.3
Nd	27	Yb	2
Sm	5.3	Lu	0.7
Eu	1.3	Gd	4

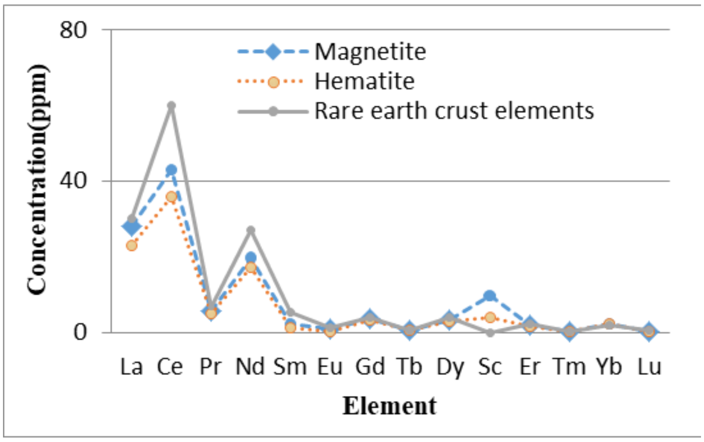

Figure 2. Rare trace elements of magnetite, hematite and earth crust

Figure 2 shows a comparison among amount of rare earth's crust elements, magnetite and hematite iron ore tailings. The rare elements levels in magnetite and hematite iron ore tailings are close to the earth's crust (Figure 2). The concentration of rare elements Ce and Nd in the earth's crust is higher than in iron ore tailings; but the amount of Sc element is less than that of iron ore tailings; however, the differences are not significant.

3.4 pollution assessment (EF, PLI, Igeo)An effective method for assessing the concentration of metal contaminants in soil is the enrichment coefficient (EF) [31]. By calculating the enrichment factor and comparing it with the standards, it can be seen that magnetite iron tailings have the least enrichment values for the study elements. To assess the general state of contamination of iron ore tailings with all contaminants measured simultaneously, the pollution load index (PLI) of heavy metal was calculated. This index indicates the number of times that the concentration of heavy metals in iron ore tailings has increased compared to the field concentration [23]. Contamination of arsenic, cadmium, cobalt, chromium, manganese, nickel, lead, vanadium and zinc in iron ore tailings of magnetite and hematite were low. The pollution load index is less than one for both tailings; indicates the class of “not contaminated with heavy metals”. Geo-accumulation index (Igeo) was first expressed by Müller (1996) for metal concentrations in particles smaller than 2 microns, which can determine the degree of sediment pollution and is calculated using the equation 3 [23]. Accumulation index values on arsenic, cadmium, cobalt, chromium, manganese, nickel, lead, vanadium and zinc tailings of magnetite and hematite showed that both tailings were not contaminated.

4. Conclusion

Silicon, Mg, Al, Ca, Na, K are the most frequent elements in magnetite and hematite iron ore tailings, respectively. The most abundant rare elements in the magnetite iron ore tailing are Ce (40 ppm), La (28ppm), Nd (19.7 ppm) and in the hematite iron ore tailings are Ce (36 ppm), La (23 ppm) and Nd (17.2 ppm) which are in accordance with the earth’s crust levels.

The amount of optimum moisture content (OMC) of magnetite iron ore tailing (11%) is lower than that for hematite iron ore tailing (13%) due to coarser particles. UCS of tailings followed the same trend, as well. The 5% of Portland cement (optimum amount) was added for stabilization of tailings and increasing the bearing capacity of these materials as road materials. After stabilization with Portland cement, the UCS of magnetite and hematite tailings increased from 0.349 MPa to 1.750 MPa and from 1.06 MPa to 3.160 MPa, respectively. The permeability coefficient of hematite tailings was 1.68×10^{-7} cm/s and magnetite tailings was 7.04×10^{-7}cm/s. The permeability coefficients of magnetite and hematite tailings after stabilization with Portland cement decreases to 2.64×10^{-8} and 5.745×10^{-7} cm /s, respectively. Therefore, it can be said that Portland cement results in reducing the permeability coefficient of tailings, because, coarse pores of tailings are filled with cement particles.Leaching test showed that the release of elements was negligible, at different pH solutions and leaching times. In fact, the use of these tailings as materials in road construction may not cause environmental concern in regard with elemental leaching; therefore, they will be suggested as materials providing pass the technical and engineering standards.

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