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RECENT TRENDS IN THE APPLICATION OF GREEN TECHNOLOGIES AND THEIR MANAGEMENT

Steel slag quality control for road construction aggregates and its environmental impact: case study of Vietnamese steel industry—leaching of heavy metals from steel-making slag

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Abstract
Steel slag is an industrial by product of steel manufacturing processes and has been widely utilized within civil and construction materials for road materials and environmental remediation in countries like Japan, USA, and European Union nations. However, the current utilization of steel slag in Vietnam is very low mainly because of lack of quality control of slag treatment and chances for reuse of treated steel slag. This paper presents the up to date steel slag production status in Vietnam through the extensive survey and sampling at seven large steel factories. The paper also highlights the environmental and quality control issues of these steel slags to use as road construction aggregates by assessing the heavy metals concentration in the leachate. The basic oxygen furnace (BOF) and electric arc furnace (EAF) slag samples were collected to evaluate leaching properties of metals leached from the slags. The two standardized batch leaching tests of steel slag roadbed material in Japan (JIS K 0058-1) and toxicity characteristics leaching procedure (TCLP—EPA method 1311) were performed to the evaluated the hazardous metals. The results of the leaching test show that almost all of the concentration of the metals in the leached solution does not exceed the National Standard for Industrial Wastewater Discharge (QCVN 40-2011). The pH and parameters such as total chromium, nickel, copper, lead, arsenic, and manganese differ from the two test methods. The acidic conditions employed in the EPA 1311 were not representative of condition excepted during slag reuse in road constructions because in the operation condition of the road, acidic liquid is absent. The leaching test results confirmed that JIS test which uses deionized water with gentle mixing prevents the slag sample from size degradation is suitable for the environmental assessment of steel slag use for roadbed material. This research suggests that the adjustment of pH value prior to disposal or reuse as base materials and official guideline should be promulgate by the authorities to ensure the leachate meet the surface water quality standard.

Keywords Steel-making slag · Leaching test · Industry by-product · Environmental impact · Quality control · JIS K 0058-1 · EPA method 1311

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Abbreviations

BOF Basic oxygen furnace
EAF Electric arc furnace
IF Induction furnace

Introduction

Steel slag has been widely utilized as construction materials for road construction and environmental remediation because of its similarity in physical properties as natural materials such as gravel (Chen et al. 2020a, b; Hainin et al. 2015; Horii et al. 2013; Liyun et al. 2017; Mombelli et al. 2016; Piatak et al. 2015). However, in Vietnam, steel slag is currently facing challenges of disposing and reuse.

According to the Steel Industry Report (2019), the steel-making industry in Vietnam is the fastest growing industry with the growth rate of 17% per year from 2014 to 2018 and forecast a 9% growth from 2019 to 2023. Together with the increase of steel products, the amount of slag generated in 2018 is about 4.2 million tons. While there is increasing demand for utilizing steel slag for road construction in Vietnam, due to the lack of national regulations and guidelines, most of the slag remains unutilized and kept in the storage yard in the factory area. On the other hand, there is the concern that bulk utilization of steel slag may pose a significant impact to the environment due to leaching of hazardous substances from steel slag matrix, which may become mobile in ground water and soil environment (Chaurand et al. 2007; Riley and Mayes 2015; Spanka et al. 2018). Because slag is widely reused as construction materials and environmental application globally, the assessment of environmental aspects of slag would contribute to encourage the utilization of steel slag in Vietnam.

Vietnam Steel Association (VSA) reported that there are more than 70 factories producing crude and finished steel. In 2020, crude steel production was 17.2 million tons. Vietnam steel market will become one of the fastest growing markets in the world from 2020 to 2024. According to VSA, steel slag generation from steel making will increase to 10 million tons in 2025 and 15 million tons in 2030.

Steel slag is generated from the steel production process. Depending on the type of the furnace, the steel slag is characterized into two types: basic oxygen furnace slag—BOF slag and electric arc furnace slag—EAF slag.

Basic oxygen furnace slag is generated during refining of hot metal produced at blast furnaces, into steel in blast oxygen furnace. While carbon in iron is eliminated as gaseous carbon monoxide by injected oxygen, the impurities like silicon, manganese phosphorus, and iron are also oxidized to form liquid state slag with lime and dolomite; the mixture of these oxides phase are called slag. Due to the difference in density with molten steel, liquid slag will float to the top and be removed from the furnace to form the steel slag after cooling. BOF slag is slowly cooled by natural air, or sprays water at the cooling yard. The yield of BOF slag is about 100–150 kg per ton of molten steel.

Electric arc furnaces use graphite electrodes to increase the temperature to melt by arcing and refining raw materials (scrap steel, lump cast iron). Some other metals like iron, alloys are added to balance the required chemical compositions of steel, and oxygen is also used for refining of molten steel. Oxidizing slag and reducing slag (reduction process) are generated from each stage of the steel-making process. The yield of oxidizing slag and reducing slag is 100 and 50 kg per ton of molten steel, respectively. The manufacturing of steel slag was illustrated in Fig. 1. Most of the steel slag produced is then dumped in the steel plant areas while hot and cooled in the slag storage yards. In some factories, slag was processed by sieving and magnetic separation for metal recovery. Because lacking national standards, technical regulations and guidelines for the safe use of steel slag as construction material, the excess steel slag is stockpiled in steel plants without being reused.

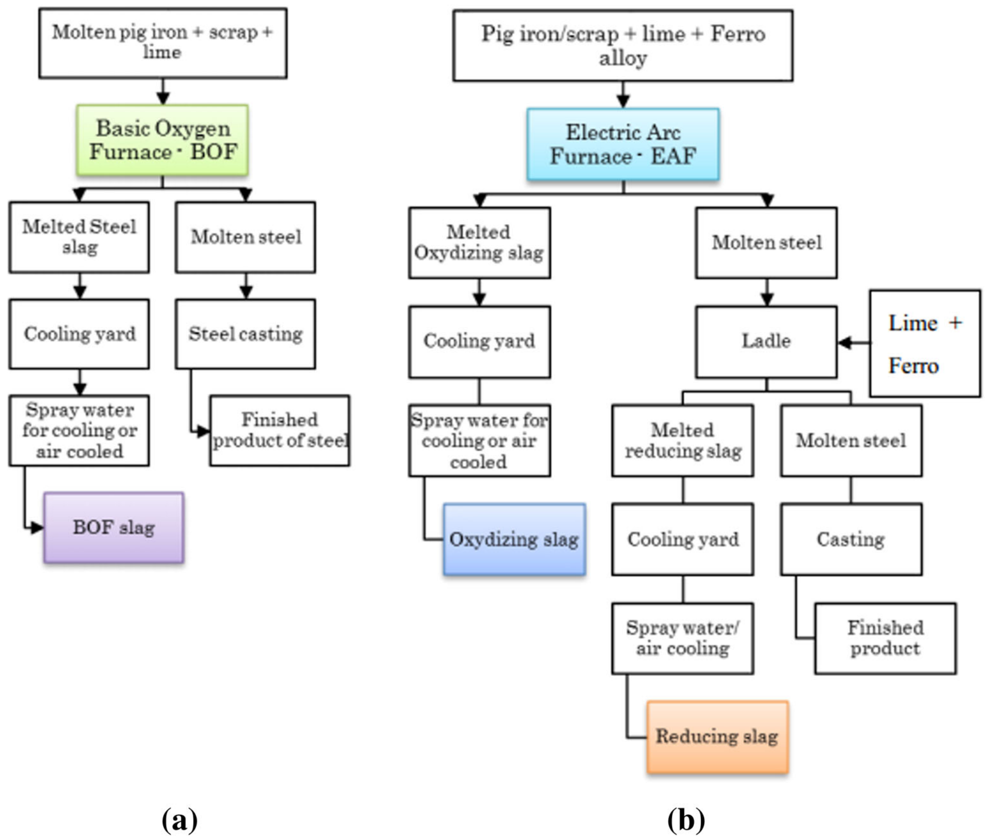
This paper discusses the potential of Vietnam steel slags to use as road construction aggregates in consideration of environmental and quality control issues. The BOF, EAF, and IF slag samples were collected from the seven steel-making factories in Vietnam with relatively high crude steel production to evaluate leaching amount of metals from the slags. The leaching test methods were compared using the leaching test of steel slag roadbed material in Japan (JIS K 0058-1) and EPA method 1311 widely adopted in Vietnam.

Materials and methods

Steel slag collection and pre-treatment

The BOF and EAF slag samples were collected from seven dumping sites of the steel plants during May 2019. The total number of slag samples was 35 samples (5 samples from each steel plants). The slags were stored in plastic containers and transported back to the laboratory for analysis. At the laboratory, the coarse particles were crushed and sieved to meet TCVN 8857:2011—natural aggregate for road pavement layers—specification for material, construction, and acceptance type C − D_{max} = 25mm (Table 1 and Fig. 2). Although the type C consists of fine and coarse particles, then the particles of 10 mm or more are eliminated for the leaching EPA 1311 leaching test and 2mm or more for the JIS K 0058-1 leaching test; use of large particles means large vessel and equipment, and large sample volume, even though the contribution to

Fig. 1 Schematic process of BOF steel slag (**a**) and EAF steel slag (**b**)



leaching test is relatively low due to low specific surface area. In order to match the actual environment as close as possible (carbonation treatment) of steel slag, 100 g of slag sample was put into the container at room temperature and add 7 g of water by spraying (Sano et al. 2015). After 1 week, 7g of water was added and the leaching procedure was performed (Fig. 3).

Experimental procedure

To investigate the mobility of trace metals in steel slags, the two types of single batch test were performed according to the US-EPA 1311 which is an evaluation method of waste and JIS K 0058-1 which is a standard for evaluating the environmental safety quality of steel-making slag road base material in Japan (Fig. 4). The EPA 1311 standard procedure uses dilute acidic leachant solutions (acetic acid or acetate buffer) with a contact time of 18 h and continuous end-over-end rotation of 30 rpm, while the JIS K 0058-1 uses deionized water with a contact time of 6 h and constant stirring frequency of 200 rpm. Leachate from JIS K 0058-1 test was analyzed follow the JIS K 0102—2016 testing methods for industrial wastewater standard by Japanese Industrial Standard. Leachate from EPA 1311 test was analyzed follow TCVN 6665:2011 (ISO 11885:2007—water quality—determination of selected elements by inductively coupled plasma optical emission spectrometry (ICP-OES)) and pH measurement method follow the TCVN 6492: 2011 (ISO 10523: 2008—water quality—determination of pH). The detection limit of ICP-OES for metals is shown in Table 2.

Table 1 Particle range before crushing for leaching test (%)

Sieve (mm)	25	9.5	4.75	2	0.425	0.075
Sample (%)						
Separate	32.5	17.5	12.5	15	12.5	10
Accumulate	100	67.5	50.0	37.5	22.5	10
Type C (%) (Requirement)	*100*	*50–85*	*35–65*	*25–50*	*15–30*	*5–15*

Results and discussion

The leachate concentration results for both standardized tests are presented in Table 3 and Fig. 5. We can see that pH and alkalinity of the leachate from JIS K 0058-1 test were notably higher than those from EPA 1311 test. The reason is that JIS standard uses deionized water while EPA test employs acetic acid for leachant. The leached pH in most samples were generally alkaline is higher than the National Technical

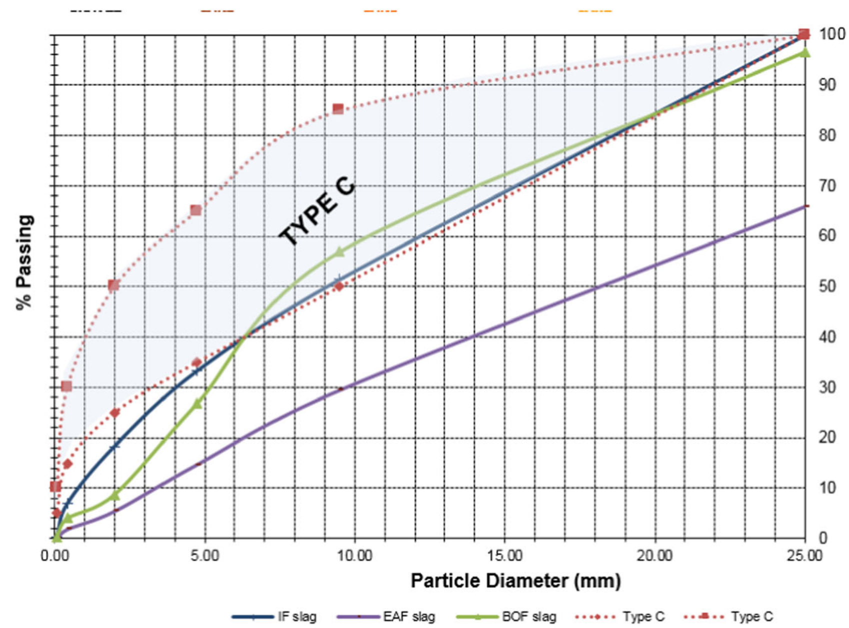
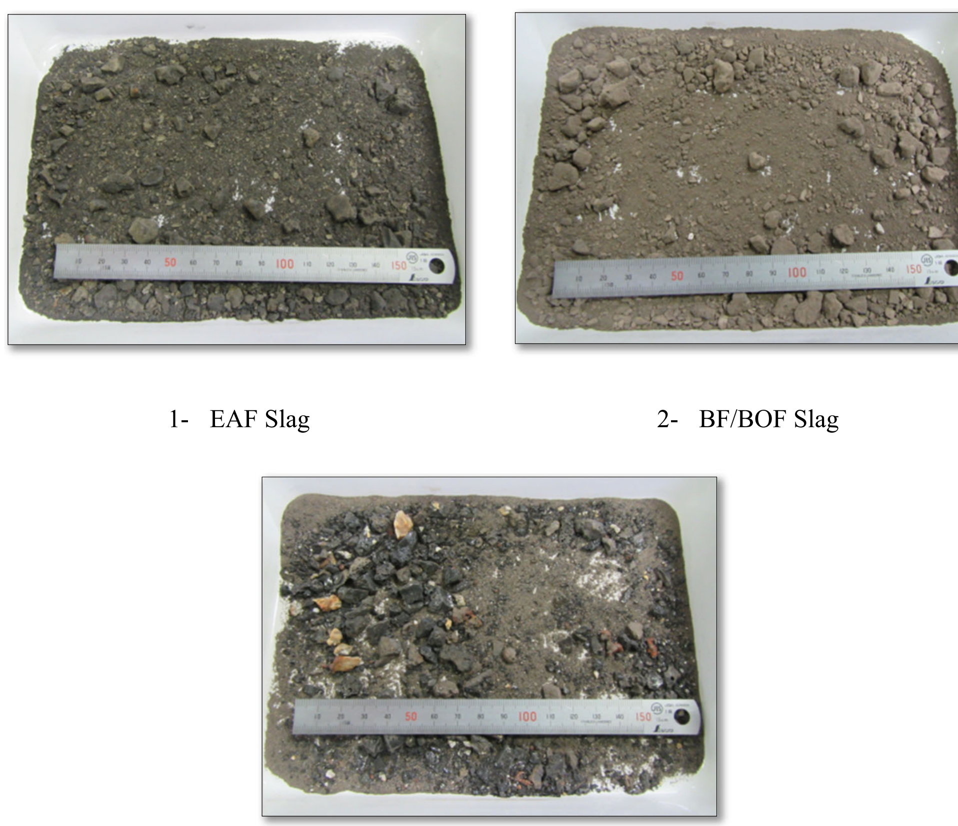


Fig. 2 Slag samples size distribution test

Regulation on Industrial Wastewater (QCVN 40:2011/BTNMT). The high alkaline level was also found in other studies, because of the dissolution of Ca silicates, oxides, and/or carbonates to form Ca^{2+} ion and hydroxyl OH^{-}(Chand et al. 2019; Piatak et al. 2015; De Windt et al. 2011; Gomes et al. 2018). The dissolution of these metals would result in the increasing of pH value to 1012.4, much higher than the typical range of natural water (Gomes et al. 2018; Mayes et al. 2008).



1- EAF Slag

2- BF/BOF Slag

3 – IF slag

Fig. 3 Slag samples after aging and crushing

Table 2 Detection limit for ICP-OES

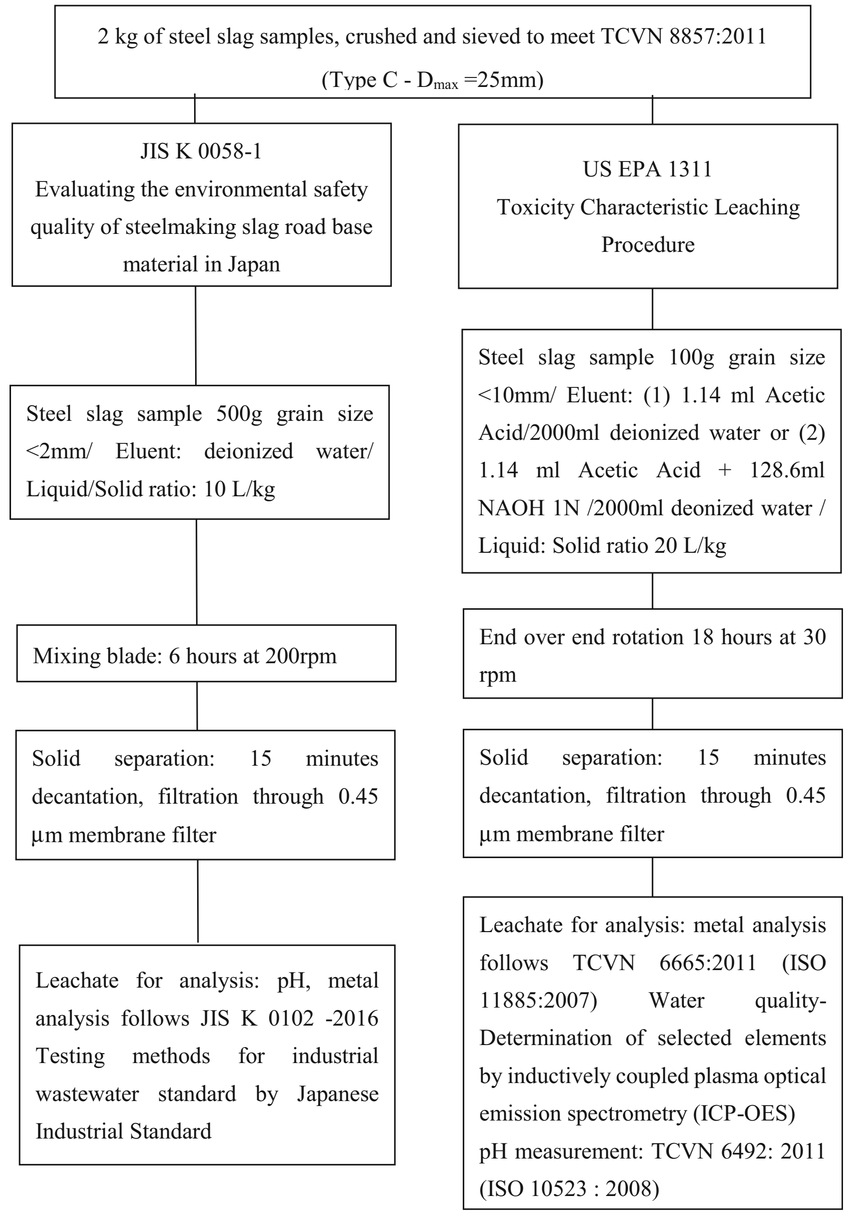
Metal	Pb	Hg	Se	As	Cd	T-Cr	Cr^{6+}
Detection limit (mg/L)	0.005	0.005	0.002	0.005	0.001	0.01	0.02
Metal	**F**	**B**	**Mn**	**Ni**	**Zn**	**Cu**	
Detection limit (mg/L)	0.08	0.1	0.01	0.1	0.1	0.1	

The pH is a major parameter controlling the leaching of many elements in slags and shown clearly from the results of the two different leaching procedures employed. The standard leaching test EPA results show that some hazardous elements were leached from the samples with the amount may be harmful to the environment such as F, Pb, As, Hg, Cd, Mn, Ni, T-Cr, and B. The release of trace metals was likely because of lower pH of the buffer solution and enhanced extraction by acetic acid (Ettler et al. 2009; Proctor et al. 2000). The concentration of the leached element from EPA 1311 procedure was significantly higher than that from JIS K 0058-1 procedure, especially for Mn, T-Cr, and F (Table 3, Fig. 5). The high concentration of Mn in the leachate suggests that most of the concentration of toxic elements in leachate results from JIS K 0058-1 method were lower than detection limit and far below the standard for industrial wastewater discharge

Table 3 Chemical characteristics and min/max element leachate concentration in steel slag samples (bold numbers indicate over Vietnamese standard for industrial wastewater discharge—QCVN 40-2011)

Parameters			pH	Pb	Mn	Hg	Ni	Se	Zn	As	Cd	T.Cr	Cr(+6)	Cu	F	B
QCVN 40-2011			6-9	0.1	0.5	0.005	0.2	0.01	3	0.05	0.05	0.2	0.05	2	5	
Sample ID				mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L
1-E-AF	EPA 1311	Mean	6.240	-	**110.00**	-	0.102	-	1.430	-	-	0.016	-	-	0.711	0.680
		SD	0.126	0.003	11.52	0.002	0.020	-	0.200	0.003	-	0.017	-	-	0.143	0.035
	JIS K 0058-1	Mean	9.930	-	-	-	-	-	-	-	-	-	-	-	0.865	-
		SD	0.109	-	-	-	-	-	-	-	-	-	-	-	0.097	-
2-BOF	EPA 1311	Mean	12.310	-	0.100	0.002	0.101	-	1.430	-	-	0.020	-	-	0.432	0.258
		SD	0.116	0.001	-	0.000	-	-	0.082	0.002	-	0.002	-	-	0.012	0.049
	JIS K 0058-1	Mean	12.380	-	-	-	-	-	-	-	-	0.015			0.159	-
		SD	0.049	-	-	-	-	-	-	-	-	0.001	-	-	0.016	-
3-IF	EPA 1311	Mean	3.730	0.070	**18.500**	-	0.167	-	**3.010**	-	0.004	**0.246**	-	0.260	1.040	1.620
		SD	0.033	0.010	1.362	-	0.012	-	0.387	-	0.000	0.047	-	0.084	0.095	0.145
	JIS K 0058-1	Mean	8.590	-	-	-	-	-	-	-	-	0.017	-	-	0.142	0.431
		SD	0.129	-	-	-	-	-	-	-	-	-	-		0.016	0.065
4-BOF	EPA 1311	Mean	7.540	-	**3.220**	-	0.014	-	0.262	-	-	-	-	-	6.790	0.868
		SD	0.164	-	1.741	-	0.003	-	0.076	-	-	-	-	-	1.913	0.046
	JIS K 0058-1	Mean	9.710	-	-	-	-	-	-	-	-	0.017	-	-	1.600	0.314
		SD	0.066	-	-	-	-	-	-	-	-		-	-	0.169	0.023
5-EAF	EPA 1311	Mean	7.140	-	**2.560**	-	-	-	-	-	-	-	-	-	0.092	0.276
		SD	0.040	0.003	0.051	-	-	-	-	-	-	-	-	-	0.009	0.003
	JIS K 0058-1	Mean	**10.970**	-	-	-	-	-	-	-	-	-	-	-	-	-
		SD	**0.100**	-	-	-	-	-	-	-	-	-	-	-	-	-
6-EAF	EPA 1311	Mean	7.730	-	**5.830**	-	0.041	-	-	-	-	-	-	-	**8.000**	4.000
		SD	0.360	-	2.121	-	0.013	-	0.054	0.003	0.001	-	-	-	0.699	0.517
	JIS K 0058-1	Mean	9.860	-	-	-	-	0.003	-	-	-	-	-	-	2.670	4.510
		SD	0.332	-	-	-	-	0.000	-	-	-	-	-	-	0.787	1.394
7-EAF	EPA 1311	Mean	4.330	0.109	**120.00**	-	0.268	-	0.935	0.006	-	0.066	-	0.512	1.320	0.832
		SD	0.048	0.078	17.516	-	0.016	-	0.245	0.000	0.002	0.004	-	0.136	0.132	0.102
	JIS K 0058-1	Mean	9.740	-	-	-	-	-	-	-	-	-	-	-	0.112	-
		SD	0.265	-	-	-	-	-	-	-	-	-	-	-	0.054	-

Fig. 4 Standardized batch leaching procedure



(QCVN 40-2011/BTNMT). These results are identical to those of the studies by Liyun et al. (2017) and Oh et al. (2012). Hexavalent chromium was not detected by either method because it was not formed during steel-making process (BOF slag) or in very low concentration in EAF slag (Proctor, 2000). In steel slags, the leachability of trace elements depends on the leachability of iron. The leachate from JIS K 0058-1 test which uses deionized water as buffer solution accelerates the precipitation of iron oxides and hydroxides due to the alkaline condition (Chand et al. 2017) which prevent the leaching of other heavy metals into the environment. However, high alkaline condition would associate with high concentrations of some metals and metalloids; notably, those that form oxyanions mobile under alkaline conditions such as Cr, Mo, V (Hobson et al. 2017; Matern et al. 2013).

The particle size of slag and liquid to solid ratio also affect the leaching of metals during leaching tests (Chand et al. 2019; Liyun et al. 2017; Mizutani et al. 2006; Piatak et al. 2015; Riboldi et al. 2020; Han et al. 2019). During the EPA 1311 test, the end-over-end rotation produces finer particles then compared to the agitation in the JIS K 0058-1 test which accelerate the release of toxic elements to the leachant. The increasing of water or leachant volume may increase the amount of dissolved slag and increase the leached concentration of trace metals; in this case, the liquid over solid ratio of EPA 1311 test was 2 times higher than JIS K 0058-1 test(Fig. 5).

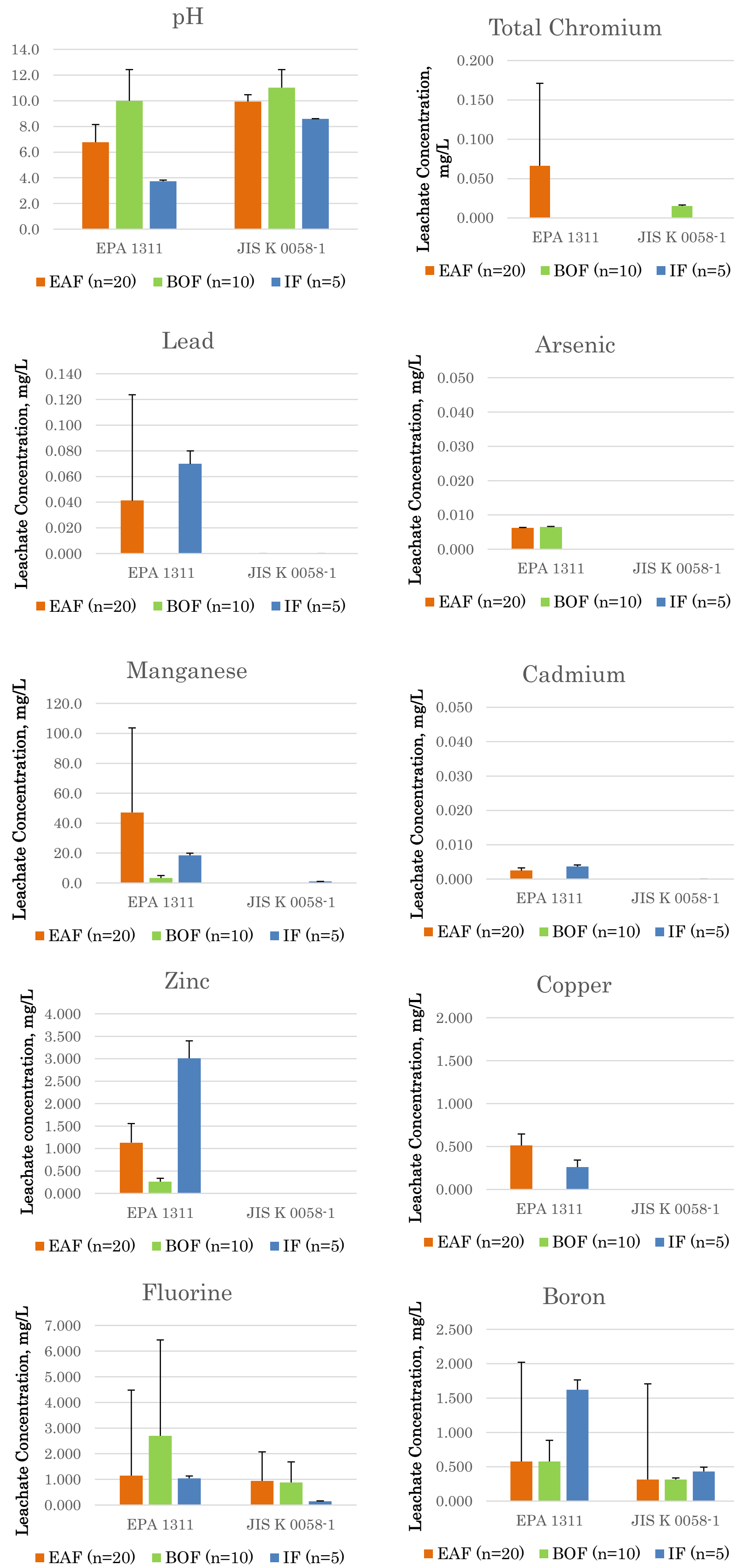


Fig. 5 Chemistry of slag leachate from EPA 1311 and JIS 0058-1 (Mean value with standard deviation)

The adjustment of pH value prior to disposal or reuse as base materials (e.g., back filling, road aggregates) is required by the authorities to ensure the leachate meet the surface water quality standard. In the surveyed factories, slag was stored in an open area for natural weathering which takes several months to reduce lime content. Other conditioning methods to prevent the highly alkaline leachate include aeration, acid dosing, or accelerated carbonation (Chen et al. 2020a, b). According to Mombelli et al. (2016), the high basicity slag, in terms of CaO/SiO2 ratio, could enhance Cr leaching; high CaO and MgO content had better retaining behavior against V release. Because of the correlation between chemical composition and leaching test results, Mombelli et al. (2016) suggested that to control a correct balance of chemical composition during the production of steel to generate stable and safe slag especially for EAF slags.

At the surveyed steel factories, the total amount of slag generated was 1677 ton per year. Depending on the steel-making process, steel slag generation rate ranges from 70 to 400kg slag per ton of steel. Some applications of steel slags from these factories are ground granulated blast-furnace slag for concrete and mortar, aggregate for road base and backfilling. According Decree 40/2019/ND-CP dated May 15, 2019, of the government (Clause 26, Article 3), steel slag when categorized as nonhazardous substance shall be reused as materials for construction to promote sustainable reuse of waste, in compliance with the Law on Environmental Protection. Some of Vietnamese steel-making slag that meets the requirement of leached heavy metals is safely applicable to road aggregate for the conservation of natural resources. The criteria need to be promulgate by the Government such as the official guideline or standard to promote recursive use of by-product as long as it is safe in construction and in service; if not, those shall be disposed complying with the regulations of Vietnam.

Conclusions

For a better understanding of the status quo of Vietnamese steel-making slag when applied to road building material, several steel-making slags were collected from seven steel-making factories in the region of Ha Noi, Da Nang, and Ho Chi Minh City. The steel slags were crushed and sieved to meet the size distribution for road aggregate. Two types of single batch leaching test were performed according to the US-EPA 1311 and JIS K 0058-1 to analyze the possible environmental impact of the leached elements from slag when applying as roadbed aggregates. Some remarks can be obtained from the results of leaching tests:

- The pH value is a major parameter controlling the leaching of many elements in slags and shown clearly from the results of the two different leaching procedures employed. The EPA 1311 method is more suitable for assessing the leachable amount of metals when disposing slag as waste in the landfill environment, while the JIS K 0058-1 method is more suitable to assess the mobility of heavy metals in slag under neutral condition like in the road construction settings;
- The concentration of the leached element from EPA 1311 procedure was significantly higher than that from JIS K 0058-1 procedure, especially for Mn, T-Cr, and F. Most of the concentrations of hazardous element in leachate result from JIS K 0058-1method were lower than detection limit and far below the standard for industrial wastewater discharge (QCVN 40-2011/BTNMT). The notable high pH value beside the concentration of heavy metals in the leachant would suggest the possibility of safe utilizing slag for roadbed construction when pH adjustment of the slag to neutral condition is performed prior to reuse (e.g., natural weathering or accelerated carbonation);
- The particle size of slag and liquid to solid ratio are among the fundamental factors influencing the leaching of metals during leaching tests. It is suggested that a nonacidic, neutral or possibly alkali water that imitates groundwater or rainwater, and a gentle agitation without reduction in particle sizes, are suitable for the assessment of slag aggregates when used for road building such as JIS K 0058-1 procedure.
- Official guideline or standard is needed to be promulgated by the Vietnamese government to promote recursive use of industrial by-product especially steel slags with consideration of standardized environmental assessment methods.

Author contribution LHN, YT, and TLN analyzed and interpreted the data regarding the leaching test. TDN, LHN, TPN, DLN, YT, TI, and NTN performed field study and sampling. LHN was a major contributor in writing the manuscript. YT, TI, TVNT, THN, TPN, RS, HST, NTN, TLN, and HGN contributed in review and editing the manuscript. All authors read and approved the final manuscript.

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Data availability The data presented in this study are available on request from the corresponding author. The data are not publicly available due to the information security conditions of the project.

Declarations

Ethics approval and consent to participate Not applicable

Consent for publication Not applicable

Competing interests The authors declare no competing interests.

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