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# **Environmental Impacts of Mixed Aggregates for use in Unbound Layers in Road Construction**

Okoljski vplivi mešanih agregatov za uporabo v nevezanih materialih v cestogradnji

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#### Abstract

During carbon steel manufacturing, large amounts of electric arc furnace (EAF) slag are generated. EAF slag, if properly treated and processed into aggregate, is an alternative source of high-quality material, which can substitute the use of natural aggregates in most demanding applications in the construction sector, mostly for wearing asphalt courses. In this screening process of high-quality aggregates, a side material with grain size 0/32 mm is also produced, which can be used as an aggregate for unbound layers in road construction. In this study, the environmental impacts of slag aggregate (fraction 0/32 mm) were evaluated in mixed natural/ slag aggregates. Different mixtures of natural/slag aggregates were prepared from aged (28 days) and fresh slag, and their environmental impacts were evaluated using leaching tests. It was shown that among the elements, chromium (Cr) was leached from some mixed aggregates in quantities that exceeded the criterion for inert waste. The data from the present investigation revealed that mixed aggregates, prepared from aged slag (fraction 0/32 mm) and natural stone in the ratio 10/90, are environmentally acceptable and can be safely used in unbound materials for road construction.

**Keywords:** electric arc furnace slag, natural aggregate, mixed aggregates, environmental impacts, unbound materials for road construction.

#### Povzetek

Med proizvodnjo ogljičnega jekla v elektroobločnih pečeh nastajajo velike količine žlindre. Pravilno obdelana in predelana žlindra je odličen material, ki lahko uspešno nadomesti naravne agregate. Pred uporabo se žlindra stara, drobi in separira v agregate različnih frakcij, ki se uporabljajo v gradbeništvu, predvsem kot visokokakovosten agregat v obrabnih asfaltnih plasteh. Med separiranjem nastaja tudi agregat zrnavosti 0/32 mm, ki ni primeren za uporabo v asfaltnih plasteh, temveč se uporablja za nevezane nosilne plasti v cestogradnji. V našem delu smo preučili okoljske vplive mešanega agregata (frakcija 0/32 mm) žlindre in naravnega agregata za njegovo uporabo v nevezanih materialih v cestogradnji. Pripravili smo različne mešanice iz naravnega agregata in agregata iz žlindre. Uporabili smo svežo in starano (28 dni) žlindro ter ocenili okoljske vplive pripravljenih mešanic z izlužitvenimi testi. Rezultati naše raziskave so pokazali, da se iz nekaterih pripravljenih mešanic lahko izlužuje element krom (Cr) v količinah, ki presegajo mejno vrednost za inertne odpadke. Kot nevezane materiale v cestogradnji varno lahko uporabimo agregate, ki smo jih pripravili iz agregata iz starane žlindre (frakcija 0-32 mm) in naravnega agregata v razmerju med 10/90.

**Ključne besede:** žlindra iz elektroobločne peči, naravni agregat, mešani agregati, okoljski vplivi, nevezani materiali v cestogradnji.

# Introduction

Industrial wastes or by-products are increasingly being used as alternative materials that successfully substitute natural raw materials. Recycling and use of these materials lead to the preservation of natural resources, significant reduction of landfill load and protection of the environment [1]. Recycling of waste materials is in line with the main objectives of green building, i.e. application of processes that are environmentally responsible and resource efficient. The latter goals are strongly supported by the Directive 2018/851/ES on waste [2] and Construction Products Regulation No. 305/2011 [3]. During steel manufacture, huge amounts of waste materials, such as electric arc furnace (EAF) slag, ladle slag and EAF filter dust, are produced. In the Store Steel plant, Slovenia, the annual steel production is about 150,000 tonnes. Approximately 20,000 tons of EAF slag, 2,000 tons of ladle slag and 2,500 tons of EAF dust are generated as waste per year [4].

During the production of steel in an EAF, scrap metal or metallised ore, or both, are melted along with lime in refractory lined vessels. Throughout the melting process, oxygen is injected into the molten steel, which oxidises a part of metallic Fe and alloying components (e.g. Mn, Ni, Cr, Mo, V), as well as the impurities in steel scrap (e.g. Al, Si, Mn, P and C). The EAF steel slag is poured from the furnace in a molten state at the end of the process. About 15-20% of EAF slag occurs per equivalent unit of steel. The principal components of EAF steel slag are calcium silicates and ferrites, together with oxides and compounds of iron, magnesium, manganese and alumina, which together make up 95% of the slag [5]. The main minerals in slag are wustite, dicalcium silicate, tricalcium silicate and brownmillerite; and the accessory minerals can be spinel, barite, CaO and MgO. Both of the latter components are unstable. In the presence of moisture, they transform into Mg(OH)<sub>2</sub> and/or Ca(OH)<sub>2</sub>, which occupy a larger volume than the primary components. The result is swelling of the composite into which the slag has been placed. The slag must therefore be "aged" long enough for the CaO and MgO to be transformed into stable forms. Slow, controlled cooling also enables this material to develop a microtexture, which ensures long-term toughness and roughness and is comparable with the porphyric texture of volcanic rocks.

EAF slag is highly durable and is, in terms of its physico-mechanical characteristics, comparable to high-quality natural rocks. Due to its excellent physical and mechanical properties, it can be used as an alternative material that successfully replaces natural aggregates. Hence, the greatest potential for recycling and use of steel slag is in the construction sector [6–9]. EAF slag aggregate is used in different types of concrete [9–13], in sub-base layer constructions [14, 15], and especially in asphalt mixes [6, 7, 16–18]. Other EAF slag applications include water and wastewater treatment [9], as well as usage in synthesis of alkali-activated materials [19].

The use of materials that contain industrial waste and/or by-products is possible when such materials possess appropriate technical characteristics [15, 20-23] and are environmentally acceptable [20-24]. For evaluation of the environmental impacts of raw materials and final products, the extent of leaching of contaminants is estimated by applying different leaching tests, e.g. the European test method for leaching of aggregates (Slovenian Institute for Standardisation [SIST] EN 1744-3) [25] and the compliance test for the leaching of granular waste materials and sludges (SIST EN 12457-4) [26]. In these leaching tests, shaking of the solid material with water (a liquid-to-solid ratio of 10 L/kg) is performed over 24 h.

The EAF slag, which is generated during the metallurgical process of carbon steel production in Štore Steel (waste classification no.: 10 02 02), is first subjected to stabilisation by ageing. For this purpose, a process of slow cooling, wetting with water and temporary storage for a period of 28 days (ageing) is carried out. Before final crushing and screening of the aggregate, iron is extracted by magnetic separation. The 32/300 mm fraction is mostly used for unbound layers, the 32/63 mm fraction particles for railway ballast and the 2/4 mm, 4/8 mm and 8/11 mm fractions for asphalt mixes. During these screenings, an additional fraction (0/32 mm) is also formed, which is the material for unbound layers in road construction. Therefore, the aim of the present study was to evaluate the potential use of the mixed aggregate (0/32 mm) from the EAF slag aggregate and natural aggregate as the unbound material for road construction from the environmental point of view. Different mixed aggregates were prepared and their environmental impacts were estimated by applying leaching tests.

# **Materials and Methods**

### Reagents

Merck (Darmstadt, Germany) suprapur acids and Milli-Q water (Direct-Q 5 Ultrapure water system; Millipore, Watertown, MA, USA) were used for the preparation of samples and standard solutions. Certipur inductively coupled plasma spectroscopy (ICP) multi-element standard solution IV (1,000  $\pm$  5 mg/L in 5% HNO<sub>3</sub>) and single stock standard solution of Hg (1,000  $\pm$  5 mg/L in 5% HCl), both purchased from Merck, were used for the preparation of calibration curves in ICP-mass spectroscopy (MS) determinations. To control the stability of ICP-MS, Merck Ge, Rh, Sc and In  $(1,000 \pm 2 \text{ mg/L in 5\% HNO})$  were used as internal standards. The reagents for spectrophotometry were obtained from Hach Lange GmbH (Düsseldorf, Germany). Cellulose nitrate membrane filters (0.45 mm; Sartorius, Gottingen, Germany) were used for filtration. The certified reference material CRM 320R (trace elements in river sediment; Community Bureau of Reference, Geel, Belgium), the standard reference material SPS-SW1 (reference material for measurement of elements in surface waters; obtained from Spectrapure Standards, Oslo, Norway) and reference material Anions - Whole Volume (purchased from Merck) were used to check the accuracy of the analytical procedures.

#### Apparatus

The concentrations of elements in the leachates of natural aggregate, EAF slag aggregate and mixed aggregates were determined by ICP–MS on an Agilent 7700' spectrometer (Agilent Technologies, Tokyo, Japan). The ICP–MS operating parameters are presented in Table 1.

**Table 1.** ICP–MS operating parameters for determination of element concentrations

Parameter	Type/value Helium mode		No gas mode
Sample introduction			
Nebuliser	Mira Mist		
Spray chamber	Scott		
Skimmer and sampler	Ni		
Plasma conditions			
Forward power	1,550 W		
Plasma gas flow	15.0 L/min		
Carrier gas flow		1.05 L/min	0.75 L/min
Dilution gas flow		0.10 L/min	0.45 L/min
He gas flow		4.5 mL/min	
Quadrupole (QP) bias		–15 V	-3.6 V
Octapole (Oct) bias		–18 V	-8.0 V
Cell entrance		-40 V	-40 V
Cell exit		-60 V	–50 V
Deflect		-2.2 V	13.4 V
Plate bias		-60 V	-40 V
Sample uptake rate	0.3 mL/min		
Data acquisition			
parameters			
Isotopes monitored		<sup>52</sup> Cr, <sup>60</sup> Ni, <sup>63</sup> Cu, <sup>66</sup> Zn, <sup>75</sup> As, <sup>78</sup> Se, <sup>95</sup> Mo	<sup>111</sup> Cd, <sup>121</sup> Sb, <sup>137</sup> Ba, <sup>201</sup> Hg, <sup>208</sup> Pb
Isotopes of internal standards		<sup>45</sup> Sc, <sup>72</sup> Ge, <sup>103</sup> Rh, <sup>115</sup> In	<sup>45</sup> Sc, <sup>72</sup> Ge, <sup>103</sup> Rh, <sup>115</sup> In

The contents of chlorides, sulphates and fluorides were determined on the DR 3900 portable spectrophotometer (Hach, Manchester, Great Britain). The MARS 6 Microwave System (CEM Corporation; Matthews, NC, USA) was used for digestion of the slag samples. The WTW 330 pH meter (WTW, Weilheim, Germany) was used to determine the pH. The Mettler AE 163 (Mettler Toledo, Zürich, Switzerland) analytical balance was used for weighing.

#### Samples

The EAF slag samples from different batches were obtained from Štore Steel Company, Štore, Slovenia. After ageing of the slag and production of aggregate fractions, the 0/32 mm fractions from fresh and aged slags (time of ageing: 28 days) were used in the present study. Natural aggregate (dolomite) was obtained from Andraž Quarry, Slovenia. Mixed aggregates of different natural-to-slag ratios were prepared from fresh and aged slags.

#### Sample preparation

To determine the total content of elements in the slag and aggregate samples, about 0.25 g of sample was subjected to microwave-assisted digestion, using a mixture of nitric, hydrochloric and hydrofluoric acids [27], and the concentrations of the elements in the digested samples were determined by ICP-MS.

The extent of leaching of selected elements and anions from bulk natural aggregates, bulk slag aggregates and bulk mixed aggregates of different ratios was evaluated by the preparation of aqueous leachates (a liquid-to-solid ratio of 10 L/kg), following the SIST EN 1744-3 [25] test method for leaching of aggregates and the SIST EN 12457-4 [26] compliance test for the leaching of granular waste materials and sludges. The concentrations of elements and anions in the aqueous leachates were determined by ICP-MS and spectrophotometry, respectively. The results are presented on a dry mass basis.

#### **Results and Discussion**

#### Quality control of the analytical data

The accuracy of determination of the total metal concentrations in the EAF slag aggregate was checked by analysis of CRM 320R, the total metal concentrations in the leachates were analysed by the SPS-SW1 quality control method and determination of anions was done by analysing the reference material Anions – Whole Volume. The results are presented in Tables 2–4.

**Table 2.** Concentrations of elements in certified reference

 material CRM 320R (Trace Elements in River Sediment)

 determined by ICP-MS after microwave-assisted digestion

Element	Certified (mg/kg)	Determined (mg/kg)	
Fe	25,700 ± 1,300	$24,350 \pm 700$	
Mn	910 ± 50	940 ± 30	
Cr	59 ± 4	61 ± 2	

Note: The results represent the mean concentration from two parallel samples.

**Table 3.** Concentrations of elements in standard reference material SPS-SW1 (reference material for measurements of elements in surface waters) determined by ICP-MS

Parameter	Certified concentration (mg/L)	Determined concentration (mg/L)
As	$10.0 \pm 0.1$	$10.4 \pm 0.3$
Ва	50 ± 1	52 ± 1
Cd	$0.50 \pm 0.01$	$0.48 \pm 0.01$
Total Cr	$2.00 \pm 0.02$	$1.97 \pm 0.02$
Cu	20 ± 1	19.6 ± 0.3
Мо	$10.0 \pm 0.1$	$9.8 \pm 0.2$
Ni	$10.0 \pm 0.1$	$10.2 \pm 0.3$
Pb	5.0 ± 0.1	4.91 ± 0.01
Se	$2.00 \pm 0.02$	$2.03 \pm 0.02$
Zn	20ª	$21.0 \pm 0.2$

<sup>a</sup> Informative value.

Notes: The results represent the mean concentration from two parallel samples.

**Table 4.** Concentrations of chlorides, fluorides and sulphates in reference material Anions – Whole Volume determined by spectrophotometry

Parameter	Certified concentration (mg/L)	Determined concentration (mg/L)
Chlorides	$95.0\pm9.50$	$92.5\pm5.0$
Fluorides	$1.17\pm0.117$	$1.14\pm0.06$
Sulphates	$44.3\pm4.43$	$42.5\pm2.0$

Note: The results represent the mean concentration from two parallel samples.

Data from Tables 2–4 show good agreement between the determined and certified values (the agreement between the results is better than  $\pm$ 5%), which confirmed the accuracy of the applied analytical procedures for the determination of (a) elements in the EAF slag aggregate and (b) elements and anions in aqueous leachate samples.

#### Chemical composition of the EAF slag

The chemical composition of the EAF slag aggregate may differ according to the additives used in steel production. The concentration ranges of the main components in the EAF slag analysed are presented in Table 5. During EAF slag processing, elemental oxides are formed. Therefore, concentrations of the measured elements in Table 5 are expressed in the form of their corresponding oxides. Results showed that the content of  $P_2O_5$  and  $V_2O_5$  was <1%, while oxides of other elements (Mo, Ba, Ni and Zn) were found in trace amounts.

**Table 5.** Concentration ranges of elemental oxides in EAF slag aggregate samples (0–32 mm) determined by ICP-MS after microwave-assisted digestion

Parameter	Concentration (%)	
FeO and Fe <sub>2</sub> O <sub>3</sub>	33-46	
CaO	20-35	
SiO <sub>2</sub>	10-20	
MgO	3-13	
Al <sub>2</sub> O <sub>3</sub>	3-6	
Mn <sub>2</sub> O <sub>5</sub>	3–5	
Cr <sub>2</sub> O <sub>3</sub>	1-3	

Notes: Measurement uncertainty is better than ±3%. Results represent concentration ranges obtained from five EAF slag samples.

As can be seen from the data of Table 5, the main components in the EAF slag are oxides of Fe, Ca, Si and Mg, whereas oxides of Al, Mn and Cr in general occur in concentrations <5%.

# *Environmental impacts of mixed aggregates from natural and EAF slag aggregates*

In order to evaluate the environmental impacts of the mixed aggregates prepared from natural aggregate and EAF slag aggregate, leachates of bulk aggregates were prepared according to relevant test methods (a liquid-to-solid ratio of 10 L/kg) [25, 26]. First, natural aggregate from the Andraž Quarry, aged EAF slag aggregate (0/32 mm) and mixed aggregate containing aged slag aggregate (natural aggregate-to-EAF slag aggregate ratio 30/70) were examined. Concentrations of elements and anions in leachates were determined by ICP-MS and spectrophotometry, respectively. The measured parameters were selected based on the legislative requirements for inert waste [28]. Aggregates used in the unbound materials for road construction and earthwork structures are generally compacted at optimum moisture content in order to ensure maximum density and bearing capacity of the final product. In order to evaluate the environmental impacts of such aggregates, the worst-case scenario was considered. Therefore, the leaching procedure was not performed on the compacted aggregate sample but rather on the bulk sample without any pre-treatment. The results of these experiments, along with the concentration limits for inert waste [28], are presented in Table 6.

As evident, the leaching of elements and anions from natural aggregate is below the instrumental limits of detection for all measured parameters. Due to the presence of soluble Ca(OH), which is formed after reaction of CaO with water, the pH values of EAF slag aggregate and mixed natural/EAF slag aggregate are high (pH 12.5 and 12, respectively). As a consequence of the high pH, most of the elements in the aqueous leachates are precipitated. Hence, their concentrations in the leachates are very low, in general, far below the concentration limits for inert waste (Table 6). Attention should be paid to Cr, the concentration of which exceeded the concentration limit of 0.5 mg/kg for inert waste in EAF slag aggregate and in mixed natural/ EAF slag aggregates. Cr in EAF slag aggregate is present in the highly insoluble chromite mineral in its trivalent oxidation state. However, at high pH, traces of trivalent Cr are solubilised as the  $Cr(OH)_{4}$  complex. Although only a negligible amount, about 0.01% of the total Cr content in the EAF slag aggregate is leached with water; the solubilised Cr(III) is, under the highly alkaline conditions, almost completely oxidised to hazardous Cr(VI) by the presence of dissolved oxygen. As an oxyanion, chromate  $(CrO_{4}^{2-})$ , under alkaline pH values, is a highly mobile and

Natural aggregate	EAF slag aggregate (0-30 mm)	Mixed aggregate (natural aggregate-to-slag aggregate ratio 30/70)	Concentration limits for inert waste*
< 0.001	< 0.001	<0.001	0.5
< 0.02	14	2.2	20
< 0.002	< 0.002	< 0.002	0.04
< 0.002	1.4	0.95	0.5
< 0.001	< 0.001	< 0.001	2
< 0.001	< 0.001	< 0.001	0.01
< 0.002	0.26	0.24	0.5
< 0.002	< 0.002	< 0.002	0.4
< 0.005	0.45	0.15	0.5
< 0.001	0.08	0.02	0.06
< 0.003	< 0.003	<0.003	0.1
< 0.005	1.95	0.2	4
<7	<7	<7	800
<1	4.59	<1	10
<10	19.5	<10	1000
8.7	12.5	12.0	/
	Natural aggregate           <0.001	Natural aggregate (0-30 mm)           <0.001	Natural aggregate (0-30 mm)Mixed aggregate (natural aggregate ratio 30/70)<0.001

\*Decree on waste (2015): Official Gazette of Republic Slovenia RS, No. 37/15 and No. 69/15.

Notes: The results represent the mean concentration obtained from two parallel analyses of leachate. Measurement uncertainty for ICP-MS is better than  $\pm 3\%$  and, for spectrophotometry, it is  $\pm 5\%$ .

highly stable species [23]. Similarly to Cr, Mo also forms the oxyanion molybdate  $(MoO_4^{2-})$ , and thus, its leaching from slag aggregates is favourable at high pH values. Leaching of chlorides, fluorides and sulphates from mixed aggregates does not represent any environmental burden.

In the following experiments, mixed aggregates from stone and cured EAF slag of the same batch were prepared in different stone-to-EAF slag ratios. Based on the data from Table 6, only Cr and Mo were measured in the aqueous leachates (Figure 1).

It can be seen that Cr and Mo concentrations in the leachates decrease with higher natural aggregate-to-slag aggregate ratio in the mixed aggregate. As expected, the pH also gradually decreased from pH 12 to pH 10. At natural aggregate-to-slag aggregate ratio 30/70 and 90/10, about 0.25 and 0.05 mg/kg of Mo, respectively is leached from the mixed aggre-



**Figure 1.** Concentrations of Cr and Mo in aqueous leachates (a liquid-to-solid ratio of 10 L/kg) of bulk mixed aggregates from aged EAF slag aggregate of the same batch, applying different natural aggregate-to-slag aggregate ratios. Notes: Bars represent mean Cr and Mo concentrations determined by ICP-MS, while the error bars indicate the minimum and maximum concentrations from two parallel determinations in the leachate by ICP-MS.

gate. These concentrations are lower than the legislative requirements for inert waste



**Figure 2.** Concentrations of Cr in aqueous leachates (a liquidto-solid ratio of 10 L/kg) of bulk mixed aggregates from aged EAF slag of different slag batches, applying natural aggregateto-slag aggregate ratio 90/10. Notes: Bars represent mean Cr concentrations determined by ICP-MS, while the error bars indicate the minimum and maximum concentrations from two parallel determinations in the leachate by ICP-MS.

(0.5 mg/kg Mo) [28]. Cr concentration in the leachate at natural aggregate-to-slag aggregate ratio 30/70 is 0.95 mg/kg and gradually decreases with decrease in the amount of slag aggregate in the mixed aggregate. A concentration that is below the limit for inert waste (0.5 mg/kg Cr) [28] is reached at ratio 85/15.

In order to evaluate the variability of Cr leaching from mixed aggregates, aged slag aggregate from different batches was used for the preparation of mixed aggregates, applying natural aggregate-to-slag aggregate ratio 90/10. These results are presented in Figure 2.

The pH values of the leachates in the samples from Figure 2 were around 10. As evident from the data of Figure 2, the leaching of Cr from 13 mixed aggregates ranged from 0.1 mg/kg up to 0.65 mg/kg. Only two samples exceeded the maximal legislative value set for inert waste. High variability in leached Cr concentrations in the aggregates prepared from aged EAF slag at the same natural aggregate-to-slag aggregate ratio (90/10), with constant pH (10), but from different slag aggregate batches, indicates the high variability of Cr content in the slag aggregate samples from different batches. Cr content in the EAF slag aggregate is related to different compositions of Cr in steel.

Finally, the influence of EAF slag ageing on leaching of Cr from mixed aggregates was estimated. For this purpose, mixed aggregates from four different batches were prepared from fresh and aged EAF slag aggregates, applying natural



**Figure 3.** Concentrations of Cr in aqueous leachates (a liquidto-solid ratio of 10 L/kg) of bulk mixed aggregates from fresh and aged EAF slags of different slag batches, applying natural aggregate-to-slag aggregate ratio 90/10. Notes: Bars represent mean Cr concentrations determined by ICP-MS, while the error bars indicate the minimum and maximum concentrations from two parallel determinations in the leachate by ICP-MS.

aggregate-to-slag aggregate ratio 90/10. The results of this experiment are shown in Figure 3.

The pH of the leachates investigated was around 10. The data in Figure 3 demonstrate that, as a consequence of variable Cr content in different slag batches, leached Cr concentrations varied significantly. The ageing of EAF slag, which stabilises its mineralogical phases, results in lower leachability of Cr. Leaching of Cr from mixed aggregates made of aged EAF slag aggregate is 30–60% lower than that of aggregates prepared from fresh slag aggregate from the same batch. It is further evident that Cr leaching from mixed aggregates made of aged EAF slag aggregate (natural aggregate-to-slag aggregate ratio 90/10) from different EAF slag aggregate batches did not exceed 0.3 mg/kg.

# Conclusions

The results from the present study revealed that in bulk mixed aggregates (0/32 mm), made of dolomite natural aggregate and EAF slag aggregate, only Cr, among all elements and anions investigated, may exceed the concentration limit for inert waste (0.5 mg/kg Cr). To find the conditions for safe use of mixed aggregates from natural aggregate and EAF slag aggregate, leaching of Cr from samples with different natural aggregate-to-EAF slag aggregate ratios was studied, applying fresh and aged (28 days) EAF

slag aggregate from different batches. High pH of mixed aggregates (pH 10-12) enabled the leaching of Cr in the form of oxyanion chromate. Due to variable Cr content in the EAF slag aggregate from different batches, which is related to the composition of steel (different amounts of Cr added in the process to obtain the steel of desired properties), the extent of Cr leaching from mixed aggregates varied significantly. Leaching of Cr from mixed aggregates made from aged EAF slag aggregate was 30-60% lower than that from fresh EAF slag aggregate. Data of the present investigation demonstrated that mixed aggregates prepared from natural aggregate and EAF slag aggregate may be safely used in unbound materials for road construction if aged slag is used for preparation of mixed aggregates at natural aggregate-to-EAF slag aggregate ratio 90/10. The safe reuse of EAF slag leads to the protection of the environment and preservation of natural resources.

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# References

- Banaitė, D., Tamošiūnienė, R. (2016): Sustainable development: the circular economy indicators' selection model. *Journal of Security and Sustainability Issues*, 6(2), http://dx.doi.org/10.9770/ jssi.2016.6.2(10).
- [2] Regulation (EU) No 305/2011 of the European parliament and of the council, of 9 March 2011, Official Journal of the European Union, 4.4.2011.
- [3] Official Journal of the European Union. (2011). Regulation (EU) No 305/2011 of the European parliament and of the council laying down harmonised conditions for the marketing of construction prod-

ucts and repealing Council Directive 89/106/EEC, 88, pp. 5–43.

- [4] ARSO annual report (2019): The type and quantity of waste generated in manufacturing and service activities and the ways of managing (2018), https:// www.arso.gov.si/varstvo%20okolja/odpadki/poro%c4%8dila%20in%20publikacije/. Last assessed march 2020.
- [5] Yildirim, I., Prezzi, M. (2011): Chemical, mineralogical, and morphological properties of steel slag. Advances in Civil Engineering, pp. 1–13, doi:10.1155/2011/463638.
- [6] Milačič, R., Zuliani, T., Oblak, T., Mladenovič, A., Ščančar, J. (2011): Environmental impacts of asphalt mixes with electric arc furnace steel slag. *Journal of Environmental Quality*, 40, pp. 115–1161, doi:10.2134/jeq2010.0516.
- [7] Oblak, T., Ščančar, J., Vahčič, M., Zuliani, T., Mladenovič, A., Milačič, R. (2011): Environmental impacts of asphalt and cement composites with addition of EAF dust. *RMZ - Materials and Geoenvironment*, 58(2), pp. 181–192.
- [8] National Slag Association, Steel slag: A premier construction aggregate, Assessed on March 2020, from http://www.acobrasil.org.br/siderurgiaemfoco/CCABrasil/NSA%20Steel%20Furnace%20Brochure.pdf. Last assessed March 2020.
- [9] Lim, J.W., Chew, L.H., Choong, T.S.Y., Tezara, C., Yazdi, M.H. (2016): Overview of Steel Slag Application and Utilization, MATEC Web of Conferences 74, 00026, pp. 1–5, DOI: 10.1051/matecconf/20167400026.
- [10] Arribas, I., Vegas, J., San-José, J., Manso, J.M. (2014): Durability studies on steelmaking slag concretes. *Materials & Design*, 63, pp. 168–176, https://doi. org/10.1016/j.matdes.2014.06.002.
- [11] Arribas, I., Santamaría, A., Ruiz, E., Ortega-López, Manso, J.M. (2015): Electric arc furnace slag and its use in hydraulic concrete. *Construction and Building Materials*, 15, pp. 68–79, https://doi.org/10.1016/j. conbuildmat.2015.05.003.
- [12] Roslan, N.H., Ismail, M., Abdul-Majid, Z., Ghoreishiamiri, S., Muhammad, B. (2016): Performance of steel slag and steel sludge in concrete. *Construction and Building Materials*, 104, pp. 1–24, https://doi. org/10.1016/j.conbuildmat.2015.12.008.
- [13] Penteado, C.S.G., Evangelista, B.L., dos Santos Ferreira, G.C., Borges, P.H.A., Lintz, R.C.C. (2019): Use of electric arc furnace slag for producing concrete paving blocks. *Ambiente Construído, Porto Alegre*, 19(2), pp. 21–32, http://dx.doi.org/10.1590/s1678-86212019000200305.

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- [14] Wang, G.C. (2016): 8 Unbound slag aggregate use in construction. In: *The Utilization of Slag in Civil Infrastructure Construction*, Woodhead Publishing Series in Civil and Structural Engineering: Number 68, Wang, G.C. (ed.). Elsevier Ltd., SPi Global: India, pp. 155–184, https://doi.org/10.1016/B978-0-08-100381-7.00008-2.
- [15] Gonawala, R.J., Kumar, R., Chauhan, K.A. (2019): Usage of Crushed EAF Slag in Granular Sub-base Layer Construction. In: *Proceedings of ICIIF 2018*, Springer Nature, Singapore, Gateway East Singapore, pp. 257–266.
- [16] Skaf, M., Manso, J.M., Aragón, Á., Fuente-Alonso, J.A., Ortega-López, V. (2017): EAF slag in asphalt mixes: A brief review of its possible re-use. *Resources, Conservation and Recycling*, 120, pp. 176–185. http:// dx.doi.org/10.1016/j.resconrec.2016.12.009.
- [17] Maharaj, C., White, D., Maharaj, R., Morin, C. (2017): Reuse of steel slag as an aggregate to asphaltic road pavement surface. *Cogent Engineering*, 4(1), pp. 1–12, doi.org/10.1080/23311916.2017.1416889.
- [18] Vaiana, R., Balzano, F., Iuele, T., Gallelli, V. (2019): Microtexture performance of EAF slags used as aggregate in asphalt mixes: A comparative study with surface properties of natural stones. *Applied Sciences*, 9(15), 3197, pp. 1–14, https://doi.org/10.3390/ app9153197.
- [19] Češnovar, M., Traven, K., Horvat, B., Ducman, V. (2019): The Potential of Ladle Slag and Electric Arc Furnace Slag use in Synthesizing Alkali Activated Materials; the Influence of Curing on Mechanical Properties. *Materials*, 12(7), 1173, pp. 1–18, doi:10.3390/ma12071173.
- [20] Vahčič, M., Milačič, R., Mladenovič, A., Murko, S., Zuliani, T., Zupančič, M., Ščančar, J. (2008): Leachability of Cr(VI) and other metals from asphalt composites with addition of filter dust. *Waste Management*, 28, pp. 2667–2674, doi:10.1016/j.wasman.2008.01.003.
- [21] Šturm, T., Milačič, R., Murko, S., Vahčič, M., Mladenovič, A., Strupi Šuput, J., Ščančar, J. (2009): The use of EAF dust in cement composites: Assessment of environmental impact. *Journal of Hazardous Materials*, 166, pp. 277–283, doi:10.1016/j. jhazmat.2008.11.015.
- [22] Zalar Serjun, V., Mladenovič, A., Mirtič, B., Meden, A., Ščančar, J., Milačič, R. (2015): Recycling of ladle slag in cement composites: environmental impacts. *Waste Management*, 43, pp. 376–385, http://dx.doi. org/10.1016/j.wasman.2015.05.006.

- [23] Milačič, R., Zuliani, T., Oblak, T., Mladenovič, A., Ščančar, J. (2011): Environmental impacts of asphalt mixes with electric arc furnace steel slag. *Journal of Environmental Quality*, 40, pp. 115–1161, doi:10.2134/jeq2010.0516.
- [24] Wang, G.C. (2016): 7 Environmental aspects of slag utilization. In: *The Utilization of Slag in Civil Infrastructure Construction*, Woodhead Publishing Series in Civil and Structural Engineering: Number 68, Wang, G.C. (ed.). Elsevier Ltd., SPi Global: India, pp. 131–153, https://doi.org/10.1016/B978-0-08-100381-7.00007-0.
- [25] SIST EN 1744-3 (2002). Tests for chemical properties of aggregates. Part 3, Preparation of eluents by leaching of aggregates.
- [26] SIST EN 12457-4 (2004). Characterisation of waste Leaching – Compliance test for leaching of granular waste materials and sludges. Part 4, One stage batch test at a liquid to solid ratio of 10 L/kg for materials with particle size below 10 mm (without or with size reduction).
- [27] Vidmar, J., Zuliani, T., Novak, P., Drinčić, A., Ščančar, J., Milačič, R. (2017): Elements in water, suspended particulate matter and sediments of the Sava River. *Journal of Soils and Sediments*, 17(7), pp. 1917–1927, doi 10.1007/s11368-016-1512-4.
- [28] Decree on waste (2015): Official Gazette of Republic Slovenia RS, No. 37/15 and No. 69/15.