

T. SOFILIC*, D. BARIŠIĆ**, U. SOFILIC***

NATURAL RADIOACTIVITY IN STEEL SLAG AGGREGATE

NATURALNA RADIOAKTYWNOŚĆ W KRUSZYWIE Z ŻUŻŁA STALOWNICZEGO

Present day steelmaking slags are being successfully used as a high quality mineral aggregate for the building industry. With this, it is of vital importance to be familiar with the technical significance of the secondary application of steel slag, because some steel slag might contain increased concentration of substances harmful to human health. In terms of steel slag impact on the environment, radionuclides are the least researched of all pollutants emitted from the metallurgical processes.

This paper presents the results of radiochemical testing of steel slag and steel slag aggregates for the purpose of its use in the production of construction material.

Obtained results of measurements show that ^{40}K , ^{226}Ra and ^{232}Th in all examined steel slag samples have the activity concentration from 45.3 to 62.9Bqkg^{-1} , 15.2 to 21.4Bqkg^{-1} and 12.9 to 15.4Bqkg^{-1} , respectively. Results of measurements of radionuclide activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in slag aggregates show similar values for all radionuclides ranges as follows: ^{40}K from 14.1 to 23.3Bqkg^{-1} ; ^{232}Th from 8.6 to 14.4Bqkg^{-1} and ^{226}Ra from 14.8 to 26.8Bqkg^{-1} . Activities index (I1, I2, I3) of ^{226}Ra , ^{232}Th and ^{40}K were compared with values recommended by Croatian legislation. Radium equivalent concentrations (Ra_{eq}) of ^{226}Ra , ^{232}Th and ^{40}K for examined steel slag and steel slag aggregates are harmonious with the results presented by other authors for the same by-product.

The testing has been conducted on steel slag created during the production of carbon steel by electric arc furnace in Steel Mill of CMC Sisak, Croatia.

Keywords: radioactivity, radionuclide, slag, aggregate, industrial by-product

Obecnie żużle stalownicze są z powodzeniem używane do wytwarzania wysokiej jakości kruszywa mineralnego dla budownictwa. Z tego powodu, istotnego znaczenia nabiera znajomość wtórnego zastosowania żużli, ponieważ niektóre żużle mogą zawierać zwiększone stężenie substancji szkodliwych dla zdrowia ludzkiego. Pod względem wpływu żużli na środowisko, radionuklidy są najmniej zbadane ze wszystkich zanieczyszczeń emitowanych z procesów metalurgicznych.

W pracy przedstawiono wyniki badań radiologicznych żużla stalowniczego i kruszyw przeznaczonych do produkcji materiałów budowlanych.

Otrzymane wyniki badań próbek żużla wskazują, że aktywność promieniotwórcza: ^{40}K , ^{226}Ra , ^{232}Th wynosi odpowiednio: 45.3 do 62.9Bqkg^{-1} ; 15.2 do 21.4Bqkg^{-1} ; i 12.9 do 15.4Bqkg^{-1} . W przypadku kruszywa otrzymano podobne wyniki dla wszystkich radionuklidów tj.: ^{40}K od 14.1 do 23.3Bqkg^{-1} ; ^{232}Th od 8.6 do 14.4Bqkg^{-1} ; i ^{226}Ra od 14.8 do 26.8Bqkg^{-1} . Indeks aktywności (I1, I2, I3) dla ^{226}Ra , ^{232}Th i ^{40}K został porównany z wartościami rekomendowanymi przez chorwackie prawo. Równoważne stężenia radu (Ra_{eq}) dla ^{226}Ra , ^{232}Th i ^{40}K dla badanych kruszyw są zgodne z wynikami przedstawionymi przez innych autorów dla tego samego materiału.

Badania przeprowadzone zostały na żużlu stalowniczym otrzymanym w trakcie produkcji stali w łukowym piecu elektrycznym w hucie stali CMC Sisak w Chorwacji.

1. Introduction

Because large quantities of industrial by-products and/or waste are generated in different industrial processes, waste management has become one of the major environmental concerns globally. With the increase of

environmental awareness, devastation of land-fill areas and due to its ever increasing cost, industrial by-products and/or waste utilization have become an attractive alternative to disposal. High levels of consumption of raw materials from natural sources, high amounts of industrially generated by-products and/or wastes and environ-

* CMC SISAK D.O.O., BRAČE KAVURIĆ 12, 44010 SISAK, CROATIA

** RUDJER BOŠKOVIĆ INSTITUTE, BIJENIČKA CESTA 54, 10000 ZAGREB, CROATIA

*** TINA UJEVIĆA 25, 44010 SISAK, CROATIA

mental impact require new solutions for a sustainable development. Recent studies [1-3] show that applications of nontraditional materials, also called industrial by-products and/or non-hazardous waste, have been considered in the construction industry. Application of industrial by-products and/or waste for incorporation in building material can be seen as a factor of preservation of natural non-renewable sources of mineral aggregates as well as simultaneous undesirable effects on the environment of all activities connected with the exploitation and transport from the origin point to the installation point. It is these factors exactly that impress upon us the need to better understand the environmental and economic aspects of using natural mineral raw materials in relation to alternative sources – industrial waste materials – which can in many ways contribute to the improvement of sustainable development in the construction industry.

Metallurgical industry influences the environment directly with its by-products, i.e. various hazardous and non-hazardous technological wastes (slag, refractories, sludge, dust, mill scale, etc.) which are most commonly disposed of at their inadequate landfills. The most common technological waste inadequately disposed of in the said manner is unprocessed steel slag. Through awareness of environmental considerations and more recently, the concept of sustainable development, extensive research and development has removed slag from industrial waste into modern industrial product which is effectively and profitably used for many industrial purposes, especially as raw material in the numerous building applications [4-6].

Present day steelmaking slags are being successfully used as a high quality mineral aggregate for the construction industry and from this reason slags are promoted as “sustainable” building materials, mainly on the basis that slags substitute the natural raw materials directly or indirectly [7,8]. The steel slags are in many features similar to building materials like crushed stone, sand, cement etc. that are extracted from natural raw materials but sometimes slag building materials even exceed them regarding their physical-mechanical properties. Having this in mind, it is of vital importance to be familiar with the technical significance of the secondary application of steel slag, but also with its possible environmental effects. Even though its application as building material reduces the quantity of landfills, which is why it can be considered as partial or total steel slag management, some steel slag might contain increased concentrations of substances harmful to human health and/or the environment. In terms of steel slag impact on the environment, radionuclides, highly hazardous elements, are

the least researched of all pollutants emitted from the metallurgical processes.

Taking into consideration that in Croatia we expect a significant increase in steel production by electric arc furnace (EAF), it is vital to pay more attention to the issue of steel slag disposal as most highly represented waste. Therefore, it is indispensable to consider the EAF steel slag as a by-product and not classify it as metallurgical waste, but to examine it in detail and, in accordance to final results, apply it as a valuable raw material in building material industry (road construction, paving, cement industry, concrete production, etc.). This paper presents the results of radiochemical testing of steel slag and steel slag aggregates with the purpose of its characterization as the type of by-product of EAF processes. Special attention has been directed at investigating the possibilities of it being used in building material production.

2. Experimental

The testing has been conducted on steel slag created during the production of EAF carbon steel in Steel Mill of CMC Sisak, Croatia. The content of radionuclides in the electric arc furnace slag and slag aggregates was determined, as well as in other materials (ferro-alloys, bauxite, fluor-spar, lime, coke, carburite, graphite electrodes, and refractories) used in the same steel making process.

2.1. Sampling

For the purpose of determination of radionuclide presence in electric arc furnace, steel slag was, after being poured out of the electric furnace, cooled with water, after which it was subjected to the following procedures: grinding, magnetic separation in order to remove left-over particles of the cooled steel melt and sieving. Slag samples and samples of materials used in the electric arc furnace steel making process i.e. ferro-alloys, bauxite, fluor-spar, lime, coke, graphite electrodes, refractory blocks (about 5 kg each), were crushed in a ring mill to the grain size below 1 mm, homogenized, and quartered to the quantity of 1.00 kg. They were dried at 378 K for 24 hours, transferred to glass bottles with ground cap, and marked.

2.2. Quantitative determination

The prepared samples were transferred to standard counting vessels of 125 cm³ and weighed. The loaded vessels were sealed and stored for at least 4 weeks to allow the in-growth of gaseous ²²²Rn (3.8 day half-life) and its short-lived decay products to equilibrate with the long-lived ²²⁶Ra precursor in the sample.

At the end of the in-growth period, the samples were counted with HPGe multi-channel γ -spectrometer. The activities of ^{40}K , ^{226}Ra , and ^{232}Th , were determined by γ -ray spectrometry, using a low background hyper pure germanium semiconductor detector system coupled to 8192-channel CANBERRA analyzer. Detector system was calibrated using standards supplied by both the National Bureau of Standards (USA) and Amersham International (UK).

Depending on sample activity, spectra were recorded for times ranging 100,000-200,000 seconds, and analyzed using the GENIE 2000 CANBERRA software.

Activities of ^{226}Ra were calculated from the 609.4 keV-peak of its ^{214}Bi progeny. Activities of ^{232}Th were calculated via ^{228}Ra from the 911.1 keV-peak of its ^{228}Ac progeny. Activities of ^{40}K were calculated from the 1460.7 keV-peak.

Efficiency of the system was checked during International Atomic Energy Agency inter-comparison runs. Precision and accuracy of the system were checked additionally by simultaneous measurement of IAEA Reference Materials (International Atomic Energy Agency). It should be mentioned that efficiency was calculated as function of energy and geometry at the base of experimental data.

Limit of detection (LD) was determined according to Currie [9] relation for aired observation and zero blank. From LD, a lower limit of detection (LLD) was estimated at the base of known efficiency, counting time, energy intensity and sample mass.

3. Results and discussion

Most building materials of natural origin contain small amounts of naturally occurring radioactive material (NORM), mainly radionuclides from the ^{238}U and ^{232}Th decay chains and the radioactive isotope of potassium, ^{40}K . The activity concentration of the radionuclides in building material varies considerably, depending on both the nature and the origin of the raw material compounds. Generally, natural building materials reflect the geology of their site of origin. The average activity concentrations of ^{226}Ra , ^{232}Th and ^{40}K in the Earth's crust are about 40, 40 and 400 Bqkg^{-1} respectively [10]. Available literature shows typical and maximum activity concentrations in common building materials and industrial by-products used for building materials in the EU, Table 1. The radioactivity concentrations found in certain kinds of industrial by-products (fly ash, phosphogypsum, etc.) can often be significantly higher in comparison with most common building materials [10].

Table 1 shows contents of some natural radionuclides in most common building materials and most common industrial by-products used in building materials. As was expected, the raw materials commonly used in the construction industry may contain NORM at different activity concentrations, according to their place of origin. The present ranges of activity concentration of the natural radionuclides in industrial by-products depend on the production process and the origin of the used raw materials.

TABLE 1
Typical and maximum activity concentrations in common building materials and industrial by-products used for building materials in the EU [10]

Material	Typical activity concentration (Bqkg^{-1})			Maximum activity concentration (Bqkg^{-1})		
	^{226}Ra	^{232}Th	^{40}K	^{226}Ra	^{232}Th	^{40}K
Most common building materials (may include by-products)						
Concrete	40	30	400	240	190	1600
Aerated and light-weight concrete	60	40	430	2600	190	1600
Clay (red) bricks	50	50	670	200	200	2000
Sand-lime bricks	10	10	330	25	30	700
Natural building stones	60	60	640	500	310	4000
Natural gypsum	10	10	80	70	100	200
Most common industrial by-products used in building materials						
Phosphogypsum	390	20	60	1100	160	300
Blast furnace slag	270	70	240	2100	340	1000
Coal fly ash	180	100	650	1100	300	1500

During the steel making process, with large volume streams, naturally and artificial occurring radionuclides may be concentrated to a radiologically relevant level. Electric arc furnace slag may also contain radionuclides originating from steel scrap and other material used in the steel making process. Very often steel scrap used in EAF process can become contaminated with radionuclides arising from different origins. Some of the main origins are the demolition or decommissioning of industrial facilities which used to process raw materials and still contain naturally occurring radionuclides, the decommissioning of nuclear installations and other facilities, loss of sealed radioactive sources, demolition of equipment in which radioactive sources have been used, etc. If such a slag is used as raw material for the production of building material, the final product will also contain this radionuclide (natural and artificial), depending on both the physical-chemical properties of the radionuclide and thermodynamic conditions in EAF during the melting process.

Data from previous publications [11,12] indicate the appearance of radionuclides in the products and by-products from steel production processes, and the most common radionuclides are the following: ^{137}Cs , ^{60}Co , ^{226}Ra , ^{192}Ir , ^{241}Am , ^{232}Th , ^{90}Sr , ^{40}K , and ^{226}Ra which are distributed among the melt, slag and electric furnace dust during the technological process of steel production, depending on their chemical and physical properties. The results of earlier studies point to the distribution of radionuclide during the process of production of steel by EAF procedure [12,13], during which ^{60}Co , ^{63}Ni and ^{192}Ir remain melted, ^{90}Sr , ^{147}Pm , ^{226}Ra , ^{232}Th , ^{238}Pu , ^{241}Am and ^{244}Cm pass to slag, and the evaporating ^{137}Cs accumulates in the dust.

In line with the said, in order for the electric furnace steel slag aggregates to be used as supplement in the production of building material, it is essential to be familiar with the composition and amount of radionuclides in such a material, which is exactly why it was exposed to a γ -spectrometric analysis. In this manner, the presence of natural isotopes ^{40}K , ^{232}Th and ^{226}Ra was determined in the specimens of steel slag and steel slag aggregates. The obtained results are shown as presented in Tables 2 and 3.

It should be mentioned that every single sample was counted three times and results in all tables presents the average activity value with standard deviation computed from these values and single counting error.

Table 2 shows results of measured activity concentration with measurement uncertainty of radionuclides in steel slag (without sieving and granulometric separation). In line with expectations, the electric arc furnace slag samples contain natural isotopes ^{40}K ,

^{226}Ra , and ^{232}Th . The measured values regarding presence of individual isotopes and their activity were as follows: ^{40}K from $45.3 \pm 10.3\text{Bqkg}^{-1}$ (sample 10) to $62.9 \pm 12.1\text{Bqkg}^{-1}$ (sample 1); ^{232}Th from $12.9 \pm 2.75\text{Bqkg}^{-1}$ (sample 9) to $15.4 \pm 2.95\text{Bqkg}^{-1}$ (sample 3) and ^{226}Ra from $15.2 \pm 2.05\text{Bqkg}^{-1}$ (sample 7) to $21.4 \pm 2.80\text{Bqkg}^{-1}$ (sample 1).

TABLE 2
Results of γ -spectrometric analysis of the average steel slag samples (without sieving and granulometric separation)

EAF slag (bulk)	Activity concentration (Bqkg ⁻¹)		
	⁴⁰ K	²³² Th	²²⁶ Ra
1	62.9 ± 12.1	15.1 ± 2.8	21.4 ± 2.8
2	57.0 ± 11.3	13.3 ± 2.6	19.5 ± 2.5
3	54.6 ± 11.3	15.4 ± 2.9	20.8 ± 2.7
4	50.3 ± 10.5	14.5 ± 2.8	18.0 ± 2.4
5	58.5 ± 11.5	14.5 ± 2.9	20.4 ± 2.6
6	52.5 ± 10.8	13.5 ± 2.6	19.1 ± 2.5
7	49.1 ± 9.33	13.2 ± 2.4	15.2 ± 2.0
8	55.9 ± 11.2	13.0 ± 2.6	19.0 ± 2.5
9	57.9 ± 11.5	12.9 ± 2.7	19.7 ± 2.5
10	45.3 ± 10.3	14.9 ± 2.9	19.5 ± 2.5

TABLE 3
Results of γ -spectrometric analysis of the steel slag aggregates

EAF slag aggregate	Activity concentration (Bqkg ⁻¹)		
	⁴⁰ K	²³² Th	²²⁶ Ra
0-4mm	22.0 ± 2.8	14.4 ± 0.9	24.0 ± 0.8
4-8mm	19.3 ± 1.7	10.4 ± 1.2	26.8 ± 3.0
8-16mm	14.2 ± 6.2	9.7 ± 2.1	16.9 ± 2.2
16-32mm	14.1 ± 6.8	10.2 ± 2.1	14.8 ± 2.0
0-32mm	23.3 ± 1.4	8.6 ± 0.9	22.0 ± 1.2

Table 3 presents results of radionuclides determination in slag aggregates. The measured values regarding presence of individual isotopes and their activity are as follows: ^{40}K from $14.1 \pm 6.8\text{Bqkg}^{-1}$ (aggregate 16-32mm) to $23.3 \pm 1.4\text{Bqkg}^{-1}$ (aggregate 0-32mm); ^{232}Th from $8.6 \pm 0.9\text{Bqkg}^{-1}$ (aggregate 0-32mm) to $14.4 \pm 0.9\text{Bqkg}^{-1}$ (aggregate 0-4mm) and ^{226}Ra from $14.8 \pm 2.0\text{Bqkg}^{-1}$ (aggregate 16-32mm) to $24.0 \pm 0.8\text{Bqkg}^{-1}$ (aggregate 0-4mm).

For the purpose of testing the possible origin of the identified natural radionuclides in electric arc furnace slag, the testing has been conducted for determination of the composition of the radionuclides in the materials added into the electric arc furnace as non-metal additives, graphite electrodes, as well as in ferroalloys and

other materials used in the process itself, as presented in Table 4.

TABLE 4
Results of γ -spectrometric analysis of materials used in the EAF process

Material	Activity concentration (Bqkg ⁻¹)		
	⁴⁰ K	²³² Th	²²⁶ Ra
FeSiMn	< 5.93	< 0.767	0.89±0.5
FeCr-carbure	< 6.57	< 0.85	< 0.9
Bauxite	34.2± 7.2	58.9±6.3	59.6±1.9
Fluorite	10±2.0	118.5±7.5	123.8±2.3
Lime	< 22.9	< 3.25	16.4±3.4
Coke	70.2±24.7	20.8±7.29	40.5±6.4
Graphite electrode	46.4± 8.7	2.5±0.6	2.5±0.5
Carburite	190.5±27.8	14.5±4.30	39.7±5.1
Refractory material Magne Hearth820	< 8.71	< 1.13	5.59±1.2
Refractory material C-MAG C220	13.7±8.6	2.37±1.4	7.20±1.6

In the last ten years (1999-2009), the level of global crude steel production was between 789 and 1329 million tones/y. If we take on the assumption that there is an average of 162 kg of steel slag generated per 1000 kg of crude steel [14], we obtain the production of about 128 to 215 million tones/y of slag in the observed period. In the same period, the level of crude steel production in the EU (27) was from 182 to 210 million tones/y and generation of steel slag was 29 to 34 million tones/y as well. Steel slag is used for many industrial purposes, especially as raw material in cement production, landfill cover material, and as crushed aggregate in the numerous building applications.

When steel slag is added into mixture that will be used for house building material, it is essential to fulfill the very often prescribed values of maximum limit radioactivity of building material [15], which should not exceed the following concentration of activities: 300 Bqkg⁻¹ for ²²⁶Ra, 200 Bqkg⁻¹ for ²³²Th and 3000 Bqkg⁻¹ for ⁴⁰K. From the radiological point of view, the maximum value of the activity index (I1) for house building material must be as follows:

$$I1 = (C_{Ra}/300) + (C_{Th}/200) + (C_K/3000); I1 \leq 1 \quad (1)$$

where C_{Ra} , C_{Th} and C_K are the concentrations of appropriate radionuclides in Bqkg⁻¹. If the activity index I1 is 1 or less than 1, the steel slag can be used as building material, as far as the radioactivity is concerned, without restriction.

When steel slag is added into material that will be used for road construction, according to the Finnish Radiation and Nuclear Safety Authority (STUK) Guide ST

12.2 [16], the activity index for materials used in road, street and related building (I2) must be as follows:

$$I2 = (C_{Ra}/700) + (C_{Th}/500) + (C_K/8000); I2 \leq 1 \quad (2)$$

where: C_{Ra} , C_{Th} and C_K are the concentrations of appropriate radionuclides in Bqkg⁻¹. If the activity index I2 is 1 or less than 1, the steel slag can be used as building material in road construction, as far as the radioactivity is concerned, without restriction.

Through awareness of environmental considerations and, more recently, the concept of sustainable development, extensive research and development has removed steel slag from industrial waste into modern industrial product which is effectively and profitably used for many industrial purposes, especially as landfill cover material. According to the same Finnish model [16], the activity index for materials used in landfill and landscaping is (I3) must be as follows:

$$I3 = (C_{Ra}/2000) + (C_{Th}/1500) + (C_K/20000); I3 \leq 1 \quad (3)$$

where: C_{Ra} , C_{Th} and C_K are the concentrations of appropriate radionuclides in Bqkg⁻¹. If the activity index I3 is 1 or less than 1, the steel slag can be used as landfill cover material, as far as the radioactivity is concerned, without restriction.

Table 5 presents the calculated values of activities concentrations (I1, I2, I3) in the specimens of the analyzed electric arc furnace steel slag aggregates.

From the data listed in Table 5 we reach the conclusion that the analyzed steel slag and steel slag aggregate, from the radiation point of view, can be used as raw material in the numerous building applications, especially for house building, road construction, landfill covering and landscaping.

In order to compare the specific activities of materials containing different amounts of ²²⁶Ra, ²³²Th and ⁴⁰K, numerous authors [17,18] have applied an index called the radium equivalent concentration Ra_{eq} and defined on the basis of previous investigations, that 370 Bqkg⁻¹ of ²²⁶Ra, 259 Bqkg⁻¹ of ²³²Th and 4810 Bqkg⁻¹ of ⁴⁰K produce the same gamma dose rate.

Therefore, the Ra_{eq} of building material can be written as:

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_K \quad (4)$$

where C_{Ra} , C_{Th} and C_K are the activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K, respectively, in Bqkg⁻¹.

From the measured activity concentrations values of ²²⁶Ra, ²³²Th and ⁴⁰K of investigated EAF slag and steel slag aggregates, the radium equivalent concentrations were computed and the results are presented in Table 6.

Index values of activities concentrations in the EAF slag and slag aggregates

EAF Slag/ Slag aggregate	Activity concentration (Bqkg ⁻¹)			Activity Indeks (Bqkg ⁻¹)		
	⁴⁰ K	²³² Th	²²⁶ Ra	I1	I2	I3
1	62.9 ± 12.1	15.1 ± 2.8	21.4 ± 2.8	0.17	0.07	0.02
2	57.0 ± 11.3	13.3 ± 2.6	19.5 ± 2.5	0.15	0.06	0.02
3	54.6 ± 11.3	15.4 ± 2.9	20.8 ± 2.7	0.16	0.07	0.02
4	50.3 ± 10.5	14.5 ± 2.8	18.0 ± 2.4	0.15	0.06	0.02
5	58.5 ± 11.5	14.5 ± 2.9	20.4 ± 2.6	0.16	0.07	0.02
6	52.5 ± 10.8	13.5 ± 2.6	19.1 ± 2.5	0.15	0.06	0.02
7	49.1 ± 9.3	13.2 ± 2.4	15.2 ± 2.0	0.13	0.05	0.02
8	55.9 ± 11.2	13.0 ± 2.6	19.0 ± 2.5	0.15	0.06	0.02
9	57.9 ± 11.5	12.9 ± 2.7	19.7 ± 2.5	0.15	0.06	0.02
10	45.3 ± 10.3	14.9 ± 2.9	19.5 ± 2.5	0.15	0.06	0.02
0-4 mm	22.0 ± 2.8	14.4 ± 0.9	24.0 ± 0.8	0.16	0.07	0.02
4-8 mm	19.3 ± 1.7	10.4 ± 1.2	26.8 ± 3.0	0.15	0.06	0.02
8-16 mm	14.2 ± 6.2	9.7 ± 2.1	16.9 ± 2.2	0.11	0.05	0.02
16-32 mm	14.1 ± 6.8	10.2 ± 2.1	14.8 ± 2.0	0.11	0.04	0.02
0-32 mm	23.3 ± 1.4	8.6 ± 0.9	22.0 ± 1.2	0.12	0.05	0.02
Allowed value (House building)	3000	200	300	≤ 1.00	–	–
Allowed value (Road construction)	8000	500	700	–	≤ 1.00	–
Allowed value (Landfill & Landscaping)	20000	1500	2000	–	–	≤ 1.00

TABLE 6

The radium equivalent concentration of ²²⁶Ra, ²³²Th, ⁴⁰K in EAF steel slag

EAF slag aggregate	Activity concentration (Bqkg ⁻¹)			<i>Ra_{eq}</i> (Bqkg ⁻¹)
	⁴⁰ K	²³² Th	²²⁶ Ra	
1	62.9 ± 12.1	15.1 ± 2.8	21.4 ± 2.8	47.84
2	57.0 ± 11.3	13.3 ± 2.6	19.5 ± 2.5	42.91
3	54.6 ± 11.3	15.4 ± 2.9	20.8 ± 2.7	47.03
4	50.3 ± 10.5	14.5 ± 2.8	18.0 ± 2.4	42.61
5	58.5 ± 11.5	14.5 ± 2.9	20.4 ± 2.6	45.64
6	52.5 ± 10.8	13.5 ± 2.6	19.1 ± 2.5	42.45
7	49.1 ± 9.3	13.2 ± 2.4	15.2 ± 2.0	37.86
8	55.9 ± 11.2	13.0 ± 2.6	19.0 ± 2.5	41.89
9	57.9 ± 11.5	12.9 ± 2.7	19.7 ± 2.5	42.61
10	45.3 ± 10.3	14.9 ± 2.9	19.5 ± 2.5	44.30
0-4 mm	22.0 ± 2.8	14.4 ± 0.9	24.0 ± 0.8	46.29
4-8 mm	19.3 ± 1.7	10.4 ± 1.2	26.8 ± 3.0	43.16
8-16 mm	14.2 ± 6.2	9.7 ± 2.1	16.9 ± 2.2	31.86
16-32 mm	14.1 ± 6.8	10.2 ± 2.1	14.8 ± 2.0	30.47
0-32 mm	23.3 ± 1.4	8.6 ± 0.9	22.0 ± 1.2	36.09

Table 6 presents results of radium equivalent concentrations in steel slag and steel slag aggregates. The obtained calculated values were from 37.86Bq kg⁻¹ (sample 7) to 47.84Bq kg⁻¹ (sample 1). The calculated values of R_{aeq} were from 30.5Bq kg⁻¹ in aggregate 16-32mm to 46.2Bq kg⁻¹ in aggregate 0-4mm. The obtained results of R_{aeq} for investigated steel slag and steel slag aggregates are lower than the results presented by other authors for the same by-product [18].

4. Conclusion

This study determined the natural radionuclide activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K, activities index (I1, I2, I3) and radium equivalent concentrations (R_{aeq}) from some steel slag and steel slag aggregates.

Results of measurements show that ⁴⁰K has the largest activity concentration in all examined steel slag bulk samples and its activity concentration ranges from 45.3 to 62.9Bqkg⁻¹. The obtained measured values of ²²⁶Ra and ²³²Th activity concentrations were from 15.2 to 21.4Bqkg⁻¹ and 12.9 to 15.4Bqkg⁻¹, respectively.

Results of measurements of radionuclide activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K in steel slag aggregates show similar values for all radionuclides ranges as follows: ⁴⁰K from 14.1 to 23.3Bqkg⁻¹; ²³²Th from 8.6 to 14.4Bqkg⁻¹ and ²²⁶Ra from 14.8 to 26.8Bqkg⁻¹.

Activities index (I1, I2, I3) calculated on the basis of measured radionuclide activity concentrations of ²²⁶Ra, ²³²Th and ⁴⁰K were less than 1, i.e. it shows that neither of the samples exceeds values recommended by the Nuclear Safety Authority (STUK) Guide ST 12.2 and Croatian Ordinance.

Results of calculated Radium equivalent concentrations (R_{aeq}) of ²²⁶Ra, ²³²Th and ⁴⁰K for investigated steel slag and steel slag aggregates are in line with the results presented by other authors for the same by-product.

The measured and calculated values all of considered parameters of naturally occurring radionuclides in examined electric arc furnace steel slags and steel slag aggregates are significantly lower than the maximum values recommended by the Nuclear Safety Authority (STUK) Guide ST 12.2 as well as Croatian Ordinance allowed limit, so that steel slags, in terms of the radionuclides present, may be applied as aggregates in the production of various building materials as concrete aggregate and as a cement ingredients, aggregates in road construction, roofing material, landfill cover material, landscape aggregate, etc.

- [1] L. Chateau, Environmental acceptability of beneficial use of waste as construction material-State of knowledge, current practices and future developments in Europe and in France. *J Hazard Mater* **139**, 3, 556-562 (2007).
- [2] Y. Huang, R.N. Bird, O. Heidrich, A review of the use of recycled solid waste materials in asphalt pavements. *Resour Conserv Recycl* **52**, 1, 58-73 (2007).
- [3] V. Ducman, B. Mirtič, The applicability of different waste materials for the production of lightweight aggregates. *Waste Manage* **29**, 8, 2361-2368 (2009).
- [4] A.G. Svyazhin, E.Kh. Shakhpazov, D.A. Romanovich, Recycling the slags of iron and steel industry. *Metallurg* **42**, 4, 25-27 (1998).
- [5] D.M. Proctor, K.A. Fehling, E.C. Shay, J.L. Wittenborn, J.J. Green, C. Avent, R.D. Bigham, M. Connolly, B. Lee, T.O. Shepker, M.A. Zak, Physical and Chemical Characteristics of Blast Furnace, Basic Oxygen Furnace, and Electric Arc Furnace Steel Industry Slags, *Environ Sci Technol* **34**, 8, 1576-1582 (2000).
- [6] J.M. Manso, M. Losañez, J.A. Polanco, J.J. Gonzalez, Ladle furnace slag in construction. *J Mat In Civ Engrg*.
- [7] M. Dziarmagowski, Possibilities of converter slag utilization. *Arch. Metall Mater* **50**, 3, 769-782 (2005).
- [8] K.H. Lee, K. You, J.W. Ahn, B.C. Ban, Use of Non-Hazardous Components as Substitutes for Traditionally Hazardous Components as well as the Treatment of Waste Materials – A study of alternative construction materials for slag derived from steel making industry. *Mater Sci Forum* **544** 503-506 (2007).
- [9] L.A. Currie, Limits for Qualitative Detection and Quantitative Determination. *Anal Chem* **40**, 586-593 (1968).
- [10] Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials, Radiation Protection 112, European Commission, Directorate-General, Environment, Nuclear Safety and Civil Protection, 11 (1999).
- [11] I. Malatova, S. Foltanova, P. Rulik, Contamination of Steel Produced in the Czech Republic by ⁶⁰Co, Proc. of the Workshop on Radioactive Contaminated Metallurgical Scrap, Eds.: UNITED NATIONS, EC, Prague, Czech Republic, 26-28 May, 43-56 (1999).
- [12] D. Neuschütz, D. Spirin, U. Quade, J. Meier-Kortwig, L. Holappa, M. Hamalainen, M.A. Heredia Lozano, M.J. Guio Bonany, Inadvertent Melting of Radioactive Sources in BOF or EAF: Distribution of Nuclides, Monitoring, Prevention, *ISIJ International* **45**, 2, 288-295 (2005).
- [13] R. Bartley, V. Ciani, E. Morere Molinero, B. Deckers, D. Dellatre, B. Dodd, D. Harvey, N. Holmberg, A. Ishikawa, M.

Isakov, M. Mattia, K. Terashima, G. Van der Reijden, Report on the Improvement of the Management of Radiation Protection Aspects in the Recycling of Metal Scrap, UN New York and Geneva, EC for Europe, IAEA, 2002, Chapter VII, pp. 63-67.

- [14] Integrated Pollution Prevention and Control. BAT for the Production of Iron and Steel. 2008. EC Directorate General JRC Joint Research Centre. European IPPC Bureau. 379p.
- [15] Ordinance on the conditions, methods and terms as well, for systematically research and monitoring of types and activities of radioactive substances in air, soil, sea, rivers, lakes, underground waters, solid and liquid rain-falls, drinking water, food and stuff of commonly usage and housing and business rooms as well. Official Gazette No. 60/2008 (in Croatian).
- [16] Radiation and Nuclear Safety Authority (STUK) – GUIDE ST 12.2 The radioactivity of building materials and ash, Helsinki 2005, ISSN 0789-4554 Available from Internet: <<http://www.finlex.fi/pdf/normit/23857-ST12-2e.pdf>
- [17] J. Beretka, P.J. Mathew, Natural Radioactivity of Austrian Building Materials, Industrial Waste and By-Products, Health Phys **48**, 87-95 (1985).
- [18] T. Mondal, D. Sengupta, A. Mandal, Natural radioactivity of ash and coal in major thermal power plants of West Bengal, India, Current Science **91**, 10, 1387-1393 (2006). Available from Internet: <<http://www.ias.ac.in/currsci/nov252006/1387.pdf>

Received: 10 February 2011.