

## MONITORING OF RADIONUCLIDES IN CARBON STEEL BLOOMS PRODUCED BY EAF PROCESS

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### Abstract

*Because natural and artificial isotopes in steel might originate from steel scrap or from the residue of the material that was used in the technological process, thus monitoring especially artificial radionuclides  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{192}\text{Ir}$  deserve special attention.*

*The analysis by g-spectrometry has been applied to determine the presence of natural isotopes  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$  and  $^{238}\text{U}$  as well as of the artificial isotope  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{192}\text{Ir}$  and their activity in the produced steel round blooms in the Steel Mill of CMC Sisak d.o.o. At the same time the content of radionuclides in the other materials (ferroalloys, bauxite, fluorite, lime, coke, graphite electrodes, refractory blocks) used in the same steel making process was investigated.*

*The measured values regarding the presence of individual isotopes and their activity in steel were as follows:  $^{40}\text{K}$  all values were less than  $1.6 \text{ Bqkg}^{-1}$ ;  $^{232}\text{Th}$  all activities values were less than  $0.02 \text{ Bqkg}^{-1}$ ;  $^{226}\text{Ra}$  all activities values were less than  $0.01 \text{ Bqkg}^{-1}$ ;  $^{238}\text{U}$  all activities values were less than  $1.10 \text{ Bqkg}^{-1}$ ;  $^{60}\text{Co}$  all activities values were less than  $0.02 \text{ Bqkg}^{-1}$ ;  $^{192}\text{Ir}$  all activities values were less than  $0.02 \text{ Bqkg}^{-1}$  and  $^{137}\text{Cs}$  all activities values were less than  $0.30 \text{ Bqkg}^{-1}$ .*

*Key words: Radioactivity; Radionuclide; Steel; G-spectrometry.*

### 1. Introduction

In the last ten years (2000-2010), the level

of global crude steel production [1] was between 849 and 1413 million tones/y. If we take on the assumption that there is an

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average of 1135 kg of steel scrap used per 1000 kg of crude steel [2], we obtain the consumption of about 964 to 1604 million tones/y of scrap in the observed period. At the same period, the level of crude steel production [1] in the EU (27) was from 173 to 210 million tones/y and consumption of steel scrap was 196 to 238 million tones/y as well.

Considering the immense significance of steel scrap as raw material in the steel making process, it is vital to be fully acquainted with its chemical composition and its disadvantages. Alongside the well-known additives, which, during melting or refining, pass to either slag or dust, or remain fully melted, steel scrap can also contain admixtures from the group of radioactive metals. Radioactive substances can become associated with steel scrap in various ways and if not discovered they can be incorporated into steel through the melting process [3]. Many radioactive sources found by recoveries and recyclers originally come from nuclear installations (fission and activation products) as well as from industrial and research irradiator activities ( $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ), teletherapy ( $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ), industrial radiography ( $^{60}\text{Co}$ ,  $^{192}\text{Ir}$ ,  $^{170}\text{Tm}$ ,  $^{169}\text{Yb}$ ), medical brachytherapy ( $^{226}\text{Ra}$ ,  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{192}\text{Ir}$ ), humidity gauges ( $^{241}\text{Am}$  /Be,  $^{226}\text{Ra}$ /Be) and density gauges ( $^{137}\text{Cs}$ ), industrial gamma gauges ( $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ), and various beta gauging ( $^{90}\text{Sr}$ ), and well logging ( $^{241}\text{Am}$ /Be,  $^{137}\text{Cs}$ ,  $^{252}\text{Cf}$ ). One can also find pipes contaminated with uranium and thorium and from the potash [4] industries ( $^{40}\text{K}$ ).

Numerous incidents have occurred in the past involving the discovery of radioactive

substances in steel scrap and, in some cases, in steel or other metal from the melting process. Data available in published work [5-27] indicate a relatively significant number of recorded cases of the presence of radioactive elements in steel and by-products created in the steel production process, and their order, according to element abundance is as follows:  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$ ,  $^{226}\text{Ra}$ ,  $^{192}\text{Ir}$ ,  $^{241}\text{Am}$ ,  $^{232}\text{Th}$  and  $^{90}\text{Sr}$ . Although today great amount of attention is being paid to the procedures of procurement, usage and disposal of radioactive sources, the number of cases in which they appear in steel and other metal scrap is still too big. Only in the Netherlands in the period from 2003 to 2007, 210 to 388 scrap loads with increased radiation level have been recorded annually [20]. Higher levels of radiation in steel scrap and steel may stem from losses, accidents or the inadvertent disposal of radioactive material. This can cause health hazards to workers and to the public as well as environmental concerns and it can also have serious commercial implications. In order to minimize the risks, radioactivity needs to be measured systematically. The United Nations Economic Commission for Europe (UNECE) has released recommendations to monitor and reduce the risks involving radioactivity in scrap metal [22]. While these recommendations are not legally binding, they provide guidance based on existing best practice to all interested parties (scrap yards, metal smelters, customs, regulatory authorities and transporters, amongst others).

Although many of the steel producers have installed equipment for radionuclide monitoring in steel scrap, it is impossible to prevent all incidents, because this equipment

cannot provide absolute protection. If radioactive material is not removed before the melting process, it could contaminate the steel and finished product, equipment, and the by-product generated during the electric arc furnace (EAF) process as well. When radionuclides are present in produced steel and by-product and/or waste, generated by steel making process, this material can be dangerous for the whole environment. It is clear that in the case of high activity, such materials belong to the radioactive waste but it is not so clear for which purposes such materials can be used when content of radionuclides is low. From this point of view, although radioactivity testing increase the total cost of quality [28], radionuclide testing is steel bloom deserve special attention.

Since the steel blooms typically come with certificates that are issued according to the requirements of EN 10204:2004-3.1 which does not control the eventual presence of radionuclides in steel, there is a need to provide the customers with this very important information.

Just as with the abovementioned and based on results of earlier studies [21] when the presence of an artificial radionuclide  $^{60}\text{Co}$  was detected in steel, and with the purpose of advancing the existing system of monitoring radionuclides CMC Sisak d.o.o., Croatia, throughout 2010, studies of occurrence and distribution of present radionuclides in round steel blooms from the process of production of carbon steel by electric arc furnace (EAF) have been conducted. The study was done with intent to show to the potential buyers steel blooms, that the the systematic quality control of steel exists, according to the norm EN

10204:2004-3.1. Specifically, with respect to steel scrap which is used to produce steel round blooms and supplies on the market of Southeast Europe, which in 90s were affected by war activities, and also, since the steel scrap from demolitions of nuclear installations appears environment, long-lived artificial radionuclides in steel scrap and steel produced round blooms deserve special attention.

## 2. Experimental

The testing has been conducted on round blooms (diameter 210-410 mm, length 3.25-5.25 m) produced by EAF and processing other materials used in steel making process during the production of carbon steel in the Steel Mill of CMC Sisak d.o.o., Croatia. The analysis by g-spectrometry has been applied to determine the presence of radionuclides and their activity in the produced round blooms and materials used in steel making process (graphite electrode, fluoride, ferroalloys, non-metallic additions and refractories).

### 2.1. Sampling

For the purpose of continuous monitoring of radionuclide presence in round blooms liquid steel was sampled simultaneously by casting into a copper mould, cooled in the air and marked. From the 100 steel samples collected during six months in 2010, 8 samples were selected and measured. These samples (Fig.1) were originally provided by casting in specially constructed cooper mould at the same time as samples for quality assurance in the steel mills.



Fig. 1. The cylindrical steel samples for the laboratory analysis by g-spectrometry

Dimensions of casted steel samples were adjusted to requirements of used g-spectrometry analyzer. The cylindrical samples which were used for the laboratory analysis were 60mm in diameter and 45mm high and have a mass of about 945-990 g and commonly the samples were measured without preparation.

Samples of materials used in the electric arc furnace steel making process i.e. Fe-Si-Mn, Fe-Cr, bauxite, fluorite, lime, coke, carburite, graphite electrodes, refractory blocks (about 5 kg each) were crushed in a ring mill to the grain size below 1 mm, homogenised, 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

All of the samples were transferred to

standard counting vessels of 125 cm<sup>3</sup> and weighed. The loaded vessels were sealed and stored for at least 4 weeks to allow the in-growth of gaseous <sup>222</sup>Rn (3.8 day half-life) and its short-lived decay products to equilibrate with the long-lived <sup>226</sup>Ra precursor in the sample.

At the end of the in-growth period, the samples were counted with HPGe multi-channel g-spectrometer. The activities of <sup>40</sup>K, <sup>226</sup>Ra, <sup>232</sup>Th, <sup>238</sup>U and <sup>137</sup>Cs <sup>60</sup>Co <sup>192</sup>Ir were determined by g-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 the National Bureau of Standards (USA), Amersham International (UK) and Analytix (USA).

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 <sup>226</sup>Ra were calculated from the 609.4 keV-peak of its <sup>214</sup>Bi progeny. Activities of <sup>232</sup>Th were calculated via <sup>228</sup>Ra from the 911.1 keV-peak of its <sup>228</sup>Ac progeny. Activities of <sup>40</sup>K were calculated from the 1460.7 keV-peak, activities of <sup>137</sup>Cs were calculated from the 661.6 keV-peak, activities of <sup>192</sup>Ir were calculated from the 468.1 keV-peak and activities of <sup>60</sup>Co from the 1173 keV-peak.

Activities of <sup>238</sup>U were calculated from the <sup>235</sup>U activities assuming the <sup>235</sup>U / <sup>238</sup>U activity ratio<sup>23</sup> of 0.046. All calculated <sup>238</sup>U activities are additionally checked via its <sup>234</sup>Th progeny at 92.6 keV-peak. <sup>235</sup>U activities were calculated from the 186 keV-peak, after subtraction of the overlapping



gas production industry, is mainly transferred to slag and dust, less than 1% of these radionuclides remain in the melt [39].

When steel scrap is charged to an EAF, chemical agents (fluxes) and ferroalloys are added to adjust the chemical compositions of the molten steel. The interactions among the flux, the refractories which lines the furnace and/or ladle, and the molten steel affect the final composition of the steel and hence the distribution of radionuclides among the melt, the slag, and the dust and it is necessary to determine how that contaminant is distributed in the various media, the melt, the slag, and the dust, following the melting of the steel scrap. The partition mechanism and mode of removal radionuclide from scrap to steel, slag and dust is a complex process that can be influenced by numerous chemical and physical factors, including the composition, thermodynamic conditions, solubility of radionuclides in molten steel, melting temperature, and melting practices such as the furnace type and size, melting time and the method of a carbon adjustment, and the physical and chemical properties of radionuclides [35]. Because of partitioning of radionuclides and distribution of mass in the various medium (steel, slag and dust), the concentrations of some radionuclides can be much higher in one of medium than the original concentrations in the steel scrap and/or total furnace charge.

To perform an exposure assessment of a given radionuclide in the EAF charge, it is necessary to estimate how that radionuclide is distributed in the various media during the steel making process. According Anigstein et al [37] and Cheng et al [38] the radionuclide concentrations or

specific activity in medium (steel, slag, dust) can be derived as follows:

The concentration of radionuclide  $i$  in medium  $m$  is calculated as follows:

$$C_{im} = (C_{if} \times M_s \times P_{im}) / M_m \quad (1)$$

Where

$C_{im}$  = specific activity of radionuclide  $i$  in medium  $m$  ( $Bqg^{-1}$  or  $pCig^{-1}$ )

$C_{if}$  = specific activity of radionuclide  $i$  in the furnace charge ( $Bqg^{-1}$  or  $pCig^{-1}$ )

$M_s$  = mass of scrap in furnace charge

$P_{im}$  = partition ratio (or distribution factor) of radionuclide  $i$  in medium  $m$

$M_m$  = mass of medium  $m$  produced from that charge

The ratio  $M_s/M_m$  in Equation 1 can be replaced by the mass partitioning factor of the medium  $m$ , the mass of each medium as a fraction of the mass of the furnace charge.

Just as the abovementioned partition mechanism and mode of removal of radionuclides from steel scrap to melt, slag, and dust is a complex process that can be influenced by the thermodynamic conditions, solubility of radionuclides in molten steel, melting temperature, and melting practices, such as furnace type and size, melting time, the physical and chemical properties of radionuclides and other phenomena which appear during the steel making process.

According to the data presented in Table 1, it is to be expected for the melt to contain 98–100% of radioactive  $^{60}Co$ ,  $^{63}Ni$ , and 98–99% of  $^{192}Ir$ , while the dust contains the majority of all evaporating  $^{137}Cs$ . Slag normally contains 80–100% of  $^{90}Sr$ , 95–100% of  $^{232}Th$ , 95–99% of  $^{147}Pm$ ,  $^{226}Ra$ ,  $^{238}Pu$ ,  $^{241}Am$ , and  $^{244}Cm$ , as well as 1–5% of

$^{60}\text{Co}$ ,  $^{63}\text{Ni}$ , and  $^{192}\text{Ir}$  all of which are being captured in the slag in the form of metal granules. That the distribution of radionuclides during melting is a quite complex and complicated process is also demonstrated by the results of Neuschütz et al's [36].

In this manner, the presence of natural and particularly artificial isotope were determined in the specimens of different steel quality (according to API Specification 5L and 5CT: J-55, H-40, N-80, X-42, X-52, Gr. B), and other material used in steel making process i.e. ferroalloys, bauxite, fluorite, lime, coke, carburite, graphite electrodes, refractories, and obtained results are presented in this work.

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

For the purpose of determining the artificial isotope  $^{60}\text{Co}$  and  $^{192}\text{Ir}$  presence in steel, the analysis by g-spectrometry has been applied to determine their presence and

natural radionuclides, as well as their activity, Tables 2.

Although, nickel is chemically similar to cobalt and should remain in the melt during steelmaking, according to the type of radiation ( $\beta$  - emitter), in this study  $^{63}\text{Ni}$  has not been taken into account.

Table 2 shows results of radionuclides determination in steel round steel blooms samples. As it was expected regarding partitioning factors for steel in previous research [21, 25, 32, 35-42], the steel samples contain artificial radionuclides  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{192}\text{Ir}$  as well as natural isotopes  $^{40}\text{K}$ ,  $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ , and  $^{238}\text{U}$  in concentrations under lower level of determination.

The measurements of radionuclide activities in round steel blooms steel samples showed that all measured values regarding the presence of individual isotopes and their activity were less than their minimum detection limit (MDC) as follows:  $^{40}\text{K}$  all values were less than  $1.6 \text{ Bqkg}^{-1}$ ;  $^{232}\text{Th}$  all activities values were less than  $0.02 \text{ Bqkg}^{-1}$ ;  $^{226}\text{Ra}$  all activities values were less than  $0.01 \text{ Bqkg}^{-1}$ ;  $^{238}\text{U}$  all activities values were less

Table 2. Results of g-spectrometric analysis of the EAF steel from CMC Sisak steel mil

Sample	Measured activity concentration $\pm$ measurement uncertainty						
	(Bqkg <sup>-1</sup> )						
	$^{60}\text{Co}$	$^{192}\text{Ir}$	$^{137}\text{Cs}$	$^{40}\text{K}$	$^{232}\text{Th}$	$^{226}\text{Ra}$	$^{238}\text{U}$
6000062	<0.02	<0.02	<0.30	<2.0	<0.06	<0.03	<1.8
6000144	<0.02	<0.02	<0.30	<1.9	<0.06	<0.02	<1.7
6000152	<0.02	<0.02	<0.30	<2.0	<0.06	<0.03	<1.7
6000213	<0.02	<0.02	<0.30	<1.6	<0.02	<0.01	<1.1
6000214	<0.02	<0.02	<0.30	<1.9	<0.06	<0.02	<1.7
6000215	<0.02	<0.02	<0.30	<2.0	<0.06	<0.03	<1.8
6000223	<0.02	<0.02	<0.30	<1.7	<0.06	<0.03	<1.7
6000250	<0.02	<0.02	<0.30	<2.0	<0.06	<0.03	<1.7

than 1.1 Bqkg<sup>-1</sup>; <sup>60</sup>Co all activities values were less than 0.02 Bqkg<sup>-1</sup>; <sup>192</sup>Ir all activities values were less than 0.02 Bqkg<sup>-1</sup> and <sup>137</sup>Cs all activities values were less than 0.30 Bqkg<sup>-1</sup>.

Obtained results of radionuclides determination in steel round steel blooms agree with previous obtained results, Table 3.

Table 3. Results of g-spectrometric analysis of the steel from CMC Sisak steel mil [43]

Sample	Measured activity concentration ± measurement uncertainty (Bqkg <sup>-1</sup> )				
	<sup>40</sup> K	<sup>232</sup> Th	<sup>226</sup> Ra	<sup>238</sup> U	<sup>137</sup> Cs
	ST1	< 4.37	< 0.43	<0.50	< 2.18
ST 2	< 4.10	< 0.40	< 0.44	< 2.05	< 0.007
ST 3	< 4.21	< 0.41	< 0.45	< 2.10	< 0.007
ST 4	< 4.18	< 0.41	< 0.45	< 2.09	< 0.007
ST 5	< 5.49	< 0.54	< 0.59	< 2.74	< 0.009
ST 6	< 4.98	< 0.49	< 0.53	< 2.49	< 0.008
ST 7	< 4.60	< 0.45	< 0.49	< 2.29	< 0.008
ST 8	< 4.19	< 0.41	< 0.45	< 2.09	< 0.007
ST 9	< 4.38	< 0.43	< 0.47	< 2.18	< 0.007
ST 10	< 4.17	< 0.41	0.80 ± 0.34	< 2.08	< 0.007
ST 11	< 4.25	< 0.41	< 0.45	< 2.12	< 0.007
ST 12	< 4.10	< 0.40	0.83 ± 0.34	< 2.05	< 0.007

For the purpose of testing the possible origin of the identified natural radionuclides in carbon steel obtained by EAF process, 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 g-spectrometric analysis of materials used in the EAF process

Material	Measured activity concentration ± measurement uncertainty (Bqkg <sup>-1</sup> )		
	<sup>40</sup> K	<sup>232</sup> Th	<sup>226</sup> Ra
FeSiMn	<5.93	<0.76	0.89±0.5
FeCr-carbure	<6.57	<0.85	<0.9
Bauxite	34± 7	59±6	60±2
Fluorite	10±2	119±7	124±2
Lime	<22.9	<3.25	16±3
Coke	70±24	21±7	41±6
Graphite electrode	46± 8	2.5±0.6	2.5±0.5
Carburite	191±27	15±4	40±5
<b>Refractory material</b>	<8.71	<1.13	6±1
Magne Hearth820			
<b>Refractory material</b>	14±8	2.4±1	7±1
C-MAG C220			

In order to solve these important problem steelmakers, including in particular those from EU member states over the past 20 years have access to systematic monitoring of the presence of radionuclides in steel scrap and produced crude steel. With regard to standards and regulations to determine the threshold for the content of radionuclides in steel scrap, steel and steel products, there is still a gap among EU countries, although it is working intensively on the adoption of uniform legislation, and in the meantime to use guidelines and recommendations issued by the International Atomic Energy Agency, IAEA.

Therefore, a group of specialists from United Nations Economic Commission for

Europe, UN-ECE who is responsible for the problem of radioactive pollution of metallurgical waste suggests a voluntary acceptance so-called business level the activity up to  $100\text{Bq kg}^{-1}$ , although in most European countries, this value varies in the range between  $100\text{Bqkg}^{-1}$  and  $300\text{Bqkg}^{-1}$ .

In countries outside the EU steel producers, there is also inconsistency in the approach to this problem and its resolution, as well as defining the limits or maximum allowable value of radionuclide activity in steel and steel products (Japan  $500\text{Bqkg}^{-1}$ , Russia  $370\text{Bqkg}^{-1}$ ).

#### 4. Conclusion

With the purpose of advancing the existing system of monitoring radionuclides, as well as improving the management system of waste from the steel production processes, and on the basis of earlier investigations, we have conducted a systematic control of radionuclide in round steel blooms from the process of carbon steel production by electric arc furnace (EAF).

The measurements of radionuclide activities in round steel blooms samples showed that all measured values regarding the presence of individual isotopes and their activity were less than their minimum detection limits (MDC).

The measurements of radionuclide activities in round steel blooms steel samples showed that all measured values regarding the presence of individual isotopes and their activity were less than their minimum detection limits (MDC) as follows:  $^{40}\text{K}$  all values were less than  $2.0\text{Bqkg}^{-1}$ ;  $^{232}\text{Th}$  all activities values were less than  $0.02\text{Bqkg}^{-1}$ ;  $^{226}\text{Ra}$  all activities values were less than

$0.03\text{Bqkg}^{-1}$ ;  $^{238}\text{U}$  all activities values were less than  $1.8\text{Bqkg}^{-1}$ ;  $^{60}\text{Co}$  all activities values were less than  $0.02\text{Bqkg}^{-1}$ ;  $^{192}\text{Ir}$  all activities values were less than  $0.02\text{Bqkg}^{-1}$  and  $^{137}\text{Cs}$  all activities values were less than  $0.30\text{Bqkg}^{-1}$ .

Activity of natural isotopes in investigated samples can be considered normal, because they are found in ferroalloys, bauxite, fluorite, lime, coke, carburite, graphite electrodes, refractories used in steel making process, in minor quantities. In addition, presence of natural isotopes can be considered usual and they may appear in raw materials and accessory materials that are used in the steel making process.

Because natural and artificial isotopes in steel might originate from steel scrap or from the residue of the material that was used in the technological process, thus monitoring especially artificial radionuclides  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$  and  $^{192}\text{Ir}$  deserving special attention.

Although these measured radioactivity values of natural and artificial isotopes in steel do not represent significant values, it is important to know the levels of these activities and to try to maintain or even reduce them by improving the existing monitoring system.

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