

Technologically enhanced ^{210}Pb and ^{210}Po in iron and steel industry

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ABSTRACT

Iron and steel manufacture has been ranked as the largest industrial source of environmental contamination in the USA; the wastes generated in their production processes contain heavy elements that can be a source of contamination, and natural radionuclides that can produce an occupational and/or public radiological impact. In this work the potential occupational effective dose rate ($\mu\text{Sv/y}$) due to inhalation in four integrated steel-making factories from Egypt has been evaluated, by assuming a well defined scenario and with basis in the ^{210}Pb and ^{210}Po activity concentrations determined in ore and wastes collected in the aforementioned factories. Activity concentrations, in Bq/kg, of ^{210}Pb and ^{210}Po , and leachable Pb and Fe were measured using gamma-ray spectrometry based on HPGe detector, alpha particle spectrometry based on PIPS detector, and inductively coupled plasma-mass spectrometry (ICP-MS). Levels of ^{210}Pb and ^{210}Po in the range of $<\text{DL}-4238$ and $1-5660$ Bq/kg, respectively, were found. According to the assumed scenarios, the occupational exposures by inhalation are much lower than the recommended annual effective dose limit, although the environmental impact due to waste storage and/or use should be considered based on case by case evaluation.

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1. Introduction

The US Environmental Protection Agency has ranked the iron and steel industry as the largest industrial source of environmental contamination. About 2–4 tonnes of various solid wastes (slag, sludge, dusts and scales) are generated per ton of steel production. These wastes contain notable concentrations of heavy elements and radionuclides that could be a source of environmental contamination and occupational exposure (Clemens, 2001; IAEA, 2003; Hofmann et al., 2000).

As a consequence of thermal processes during steel production, ^{210}Pb and ^{210}Po activity concentrations are enhanced in some wastes while other natural occurring radionuclides (NOR) are not affected (Ryan et al., 2005). This study aims at measuring ^{210}Pb and ^{210}Po activity concentrations in the iron and steel industry in Egypt and estimating the occupational exposure due to the potential resultant dust inhalation.

1.1. Iron and steel production processes

There are two main process routes for iron and steel production;

- Reduction of iron ore, mainly via the blast furnace (BF) – basic oxygen furnace (BOF) converter process (Process A), and
- Melting of steel scrap in an electric arc furnace (EAF, Process B).

The flowchart of both iron and steel production processes is shown in Fig. 1.

In the Process A, pig (primary) iron (hot metal) can be manufactured from sintered, pelletized, or lump iron ores. Iron ore sinters are produced from mixtures of ore, dolomite, cokes and recycled dust. The sinters are fired with gas until the temperature reaches up to 1400°C in the combustion zone, and a multi-step dry cleaning procedure is applied where a large fraction of the dust particles is recycled into the sinter process. Only the dust from the last cleaning step is taken out as waste. For wet pellet production, ore mixes are dried and ground, sieved and mixed with water and bentonite. The formed wet pellets are fired into hard pellets and cooled with air.

The sintered, pelletized, or lump iron ores can be directly treated using coke and limestone in BF (Process A1). It produces iron and slag, both fluid, and BF gas carrying dust. The hot gas from the BF is cooled and cleaned in a multi-step process. The coarse-grained fraction is recycled into the sinter process. The fine-grained fraction undergoes a wet cleaning process using water and is then converted into sludge. As an alternative, the raw material (sintered, pelletized or lump iron ores) can be treated by the so called smelting reduction process which operates over two stages. First,

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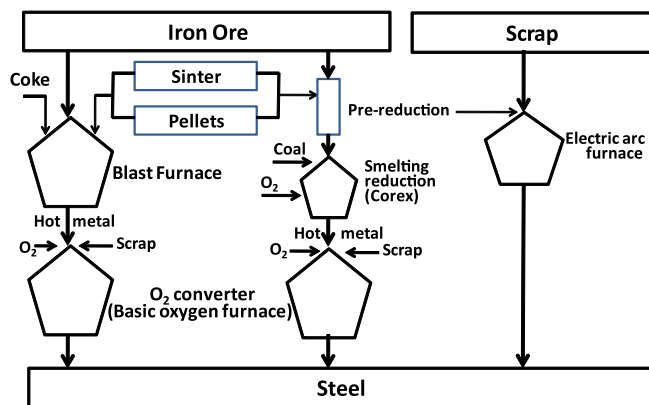


Fig. 1. Flowchart of the integrated manufacturing process for iron and steel.

Table 1

List of iron and steel industry samples.

Code	Type	Process	Factory
O1	Ore	–	A
D1	Dust	Sinter plant	
D2	Dust	Blast furnace	
S1	Slag	Sinter plant	
S2	Slag	Blast furnace	
S3	Slag	Transformer	
R1	Washing residue	Ore	
R2	Washing residue	Blast furnace	
D3	Dust	Electric arc furnace	B
S4	Slag	Electric arc furnace	
D4	Dust	Electric arc furnace	C
S4	Slag	Water treatment plant	
SC	Scale	Electric arc furnace	
O2	Ore	–	D
SL	Sludge	Direct reduction	
P	Product	Direct reduction	

the ores are reduced to sponge iron and then transferred into hot metal, similar to that from BF (Process A2). Direct reduction of the raw material (sintered, pelletized on lump iron ores) does not produce any molten hot metal. Only the oxygen is extracted from the ores, and the gangue constituents remain in the sponge iron product (DRI = Direct Reduction Iron).

There is another process which utilizes mainly scrap as an iron source, mixed with some DRI (Process B). In this process, the mixture of scrap and DRI is melted in an electric arc furnace (EAF) to produce molten steel (Ryan et al., 2005; Doring et al., 2008; Morito and Emi, 2003).

2. Materials and methods

2.1. Sampling and samples preparation

A total of sixteen industrial (ore, dust, slag, washing residue, sludge and scale) samples were collected from four factories that represent the steel industry in Egypt (Table 1). All the samples were oven dried, pulverized, homogenized and sieved through a 2 mm-mesh sieve. Aliquots of all the samples were sent to ALS-Chemex laboratory, Vancouver, Canada, where they were treated with aqua regia to extract the leachable portion of Pb and Fe.

2.2. Analytical techniques

2.2.1. Gamma spectrometric analysis

The dried-homogenized samples were transferred into polyethylene containers of 100 cm³ capacity. Lead-210 activity concentration was measured via its 46.4 keV gamma-ray energy line using a well calibrated gamma spectrometry system based on a hyper-pure germanium (HPGe) detector. The HPGe detector had a relative efficiency of 50% and full width at half maximum (FWHM) of 1.95 keV for ⁶⁰Co gamma energy line at 1332 keV. A standard solution of ²¹⁰Pb was used for efficiency calibration, and matrix self attenuation corrections were performed to all the determinations. Self attenuation corrections were carried out according to the method proposed by Cutshall et al. (1983). Further details of the self attenuation correction procedure are described in Khater and Ebaid (2008).

For analytical quality control, reference soil and sediment samples, IAEA-384, IAEA-326 and IAEA-327, from International Atomic Energy Agency (IAEA) were analyzed.

2.2.2. Polonium-210 analysis

Three gram portions of the dried samples were spiked, for chemical recovery determination, with ²⁰⁹Po ($E_{\alpha} = 4.9$ MeV) and

dissolved using mineral acids (HNO₃, HF and HCl). The sample residual was dissolved in 35 mL of 0.5 M HCl and heated up to 80 °C, while 100 mg ascorbic acid was added in order to reduce Fe(III). Finally, polonium was self-plated from the sample solution at 80 °C onto a stainless steel disk fixed in a rotating Teflon holder (Hamilton and Smith, 1986). The plated disks were counted with an alpha spectrometer (CANBERRA, Model 7401 VR) based on passivated implanted planar silicon (PIPS) detectors of 450 mm² surface area, coupled to low noise preamplifiers, amplifiers, and a multi-channel analyzer operating with Genie 2000 Software (CANBERRA). The samples were counted for 1000 min, obtaining a lower limit of detection of 1 mBq (Currie, 1968). The average chemical recovery was 75%, the individual values ranging from 50 to 100%.

Different reference materials (such as the IAEA-326 soil sample) were used for analytical quality control and quality assurance for ²¹⁰Po analyses.

2.2.3. Inductively coupled plasma-mass spectrometry (ICP-MS) analysis

Sample aliquots were treated with aqua regia to extract the leachable portion of Pb and Fe with other elements. The Pb and Fe were analyzed in adjusted leaching solutions using a Perkin–Elmer Elan 9000 ICP-MS system at ALS-Chemex laboratory - Canada. These results will give an indication about the distribution pattern of Pb and Fe in the samples.

3. Results and discussion

3.1. Activity concentrations

Activity concentrations of ²¹⁰Pb and ²¹⁰Po (in Bq/kg), the corresponding ²¹⁰Pb/²¹⁰Po activity ratios, and the amount of leachable Pb (mg/kg) and Fe (%) in all the samples analyzed are given in Table 2. The levels of ²¹⁰Pb and ²¹⁰Po in the two iron ore samples analyzed are relatively low and quite different: both ²¹⁰Pb and ²¹⁰Po in ore sample O1 were 3- to 4- fold higher than that in O2. According to unpublished data, the activity concentrations of other radionuclides from the uranium-238 series have the same trend in these samples. This variation is because hematite, the most important iron mineral, occurs in many different types of host-rocks. It occurs associated with vein deposits (igneous, metamorphic, and sedimentary rocks) and as product of the weathering of magnetite. Therefore, naturally occurring radionuclides (NOR: ²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²¹⁰Po, ²²⁸Ra and ⁴⁰K) concentrations vary in ores according to their geochemical properties and their concentrations in source rock materials (IAEA, 2003). For example, NOR activity concentrations in hematite

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