Waste Management 79 (2018) 251–259

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Characterization of sinter flue dust to enhance alternative recycling and environmental impact at disposal

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ARTICLE INFO

Article history:
Received 10 May 2018
Revised 16 July 2018
Accepted 18 July 2018

Keywords:
Characterization
Dust abatement
Environmental impact
Heavy metals
Leaching
Sinter dusts

ABSTRACT

Dust emission is one of the main environmental pollution impacts associated with steelmaking. In this sense, electrostatic precipitators (ESP) are regarded as the best available technique for treating this type of emission, thus generating two differentiated fractions: coarse and fine. Thorough chemical and structural characterization of both materials was carried out to recycle these byproducts in either the sintering process or other steps of pig iron production. Both types of dusts are crystalline heterogeneous materials mainly composed of sepiolite ($Mg_8Si_{12}(OH)_2{\cdot}12H_2O$), hematite ($Fe_2O_3$) and calcite ($CaCO_3$), the coarse fraction containing low amounts of Na (0.38 ± 0.04%) and K (0.17 ± 0.02%), which adversely affect blast furnace operation. Hence, the coarse fraction is suitable for recycling, whereas the fine one presents higher concentrations of these alkali elements. Besides, textural characterization revealed that dust particulates are essentially macroporous materials, with specific surface area values of 21.6 m^2/g for the coarse fraction and 33.7 m^2/g for dust fines. In order to ensure inoffensive dumpsites, the environmental behavior associated with dust particles accumulated in disposal areas was also evaluated by performing leaching studies simulating different rainfall scenarios. It was found that the specific leaching rates of Ca, Mg, K and S varied between 0.072 ± 0.001 and 0.75 ± 0.01 $\mu g_{element}/(g_{dust}{\cdot}d)$, whereas slower leaching rates were obtained for heavy metals (Fe, Mn and Cu), the values ranging from $(1.20 \pm 0.1) \times 10^{-4}$ to $(1.8 \pm 0.1) \times 10^{-3}$ $\mu g_{element}/(g_{dust}{\cdot}d)$. These low rates indicate that the leaching of sinter dusts compounds has minimal environmental impact.

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1. Introduction

In the steel making industry, all the processing steps which involve handling, crushing, charging, screening or transporting the raw materials cause continuous dust emissions. Dust is thereby-one of the main environmental impacts of steelmaking and creates pollution both inside and outside the plant (Pelino et al., 2002). This source of pollution includes sinter emissions, such as sinter dust with particulate matter and various gases: CO, CO_2, SO_x and NO. The outcome is a potential increase in the concentration of suspended particles in the air, mainly inside the factory, if these emissions are not treated efficiently. Over the last decades, the use of new technology has significantly reduced the amount of dust generated by a typical steel plant (Rabah, 1999). The emissions are usually collected and encapsulated in housing with suction hoods that are directly connected to cleaning devices. Cleaning of sinter off-gases is mainly achieved by dust abatement (BREF, 2012). Electrostatic precipitators (ESPs) are the most widely used abatement systems for dust emissions. Other alternatives rely on the use of cyclones, web scrubbers or fabric/baghouses filters. Nevertheless, ESPs are often preferred due to their simple design, low operating costs, high collection efficiency and low pressure drop (Vehlow, 2015; Bohidar et al., 2015). Usually, they consist of dry devices with three or four fields installed in sinter plants located within the European Union (Remus et al., 2013), and can reduce dust emissions with efficiencies above 95% and, in some cases, can yield final dust concentrations in the range of 20–50 mg/Nm^3 during normal operation periods, excluding start-ups and shutdowns (Reyes and Elholm, 2011). Therefore, adequate design and sound guidelines for the correct maintenance of advanced ESPs are necessary to ensure good performance, since this technique is considered the best available one (BAT) to reduce daily dust particulates emission.

Electrostatic precipitators are originally designed for sinter plants basing on both the gas and dust properties, making it

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https://doi.org/10.1016/j.wasman.2018.07.039
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necessary to conduct an in-depth study on dust characteristics, such as mineral composition and chemical behavior. The coarse fraction of the dust, which is prone to efficient separation by ESPs, results from the first part of the strand, and originates from the sinter feed and the lower layer. Dust fines are formed in the sintering zone, after the evaporation of water from the mixture is accomplished.

Since the physicochemical properties of the dust particles formed in the sinter plant are key parameters for suitable performance and efficiency of the electrostatic precipitators, their design requires a detailed study of the dust particulates characteristics. This study is part of a project led by ArcelorMittal, whose aim is to gain in-depth knowledge of the physicochemical properties of sinter dusts in order to find alternative uses to recycle them and develop highly efficient ESPs. Due to the new legislation and its impact on the waste management policies in Europe, efforts are driven to reduce the environmental and health impacts of waste and disposal activities and are also focused on improving Europe's efficiency in terms of resource utilization. To achieve these goals, it is required much higher levels of recycling and minimization of the consumption of natural resources according to 7th Environment Action Program of European Commission.

In this work, the study of the chemical and mineralogical composition and evaluation of the textural and morphological properties of such byproducts were carried out using a variety of techniques: XRF, XRD, elemental analysis, FTIR spectroscopy, TGA-DTG analysis, N_2 adsorption-desorption, Hg intrusion porosimetry, SEM and TEM. To the best of our knowledge, sinter dusts were previously characterized in just a few works (Chang et al., 2015; Lanzerstorfer and Steiner, 2016; Sinha et al., 2010) but such characterization was cursory in comparison with the one here presented, in which a great number of techniques were employed. Besides, this work addressed, for the first time ever, the study of the environmental behavior of dust particles accumulated in disposal sites for ensuring environmentally-friendly sinter dust dumpsites. Thus, the changes in the chemical composition of dust particulates, which were previously treated under simulated environmental conditions, were thoroughly analyzed.

2. Materials and methods

2.1. Experimental section

The samples of sinter dusts employed in this work were provided by an ArcelorMittal sinter plant located in Asturias (north of Spain), namely ArcelorMittal Veriña, and consisted of two fractions collected from different fields of the ESPs module corresponding to the sinter dust fine fraction (sample FF) and the sinter dust coarse fraction (sample CF). The samples were collected by qualified plant personnel for several weeks of dust production in order for it to be representative. Prior to the characterization, it was necessary to mill and sieve the sample CF and sieve the FF one, except in the case of N_2 adsorption-desorption analysis, in which the samples were used without modification. In both cases, the fraction with sizes below 75 μm was used. In the environmental tests, the samples FF and CF were also employed without modification.

2.2. Characterization methods

Shimadzu EDX-720 energy dispersive X-ray fluorescence (XRF) spectrometer was used to determine the chemical composition of sinter dusts. The content of C, H, N and S was measured by elemental analysis employing an Elementar Vario EL analyzer. Powder X-ray diffraction (XRD) patterns were obtained at room temperature in order to determine the mineralogical composition of the samples. PANalyticalXPert Pro powder diffractometer using Cu Kα radiation ($\lambda_{K\alpha}$ = 1.5406 Å) and a graphite secondary monochromator was employed. The diffractograms were recorded for 2θ values between 5° and 80° by 0.02° step, with a scan step time of 1 s. Quantitative phase analysis was carried out using the Rietveld refinement method (Taylor, 2001). FTIR spectra of the samples were recorded in the 4000–400 cm^{-1} range, by means of a Perkin-Elmer PARAGON 1000 spectrometer. The resolution employed was 4 cm^{-1}. The samples were pressed into small discs using a spectroscopically pure KBr matrix. Thermal analysis was carried out using a Mettler Toledo TGA/SDTA851e thermogravimetric analyzer. 45–47 mg of each of the samples were heated in alumina crucibles under a nitrogen atmosphere at a heating rate of 10 °C/min from 20 °C to 1000 °C. Textural properties (specific surface area and porous structure) were studied by nitrogen adsorption-desorption at 77 K using a Micromeritics ASAP 2020 instrument. Prior to the analysis, it was necessary to degas the dusts (400–450 mg) at 120 °C for 10 h. The presence of macroporosity was determined through Hg intrusion porosimetry using a Micromeritics Autopore IV instrument. The analysis was carried out with a sample mass of 950 mg at low and high pressure (0.1–60,000 psi) employing a penetrometer bulb for powdered materials. The morphological characterization of the sinter dusts and energy dispersive X-ray analyses (EDX) was performed using a JEOL JMS-6610LV scanning electron microscope (SEM) operating at 0.3–30 kV. Previously to the SEM characterization and EDX analysis, the samples were sputtered coated with gold. Transmission electron microscopy (TEM) was performed on a MET JEOL-2000 EX-II microscope operating at 160 kV. Samples were prepared by sonicating the powdered sample in ethanol and then evaporating several drops of suspension on copper grids.

2.3. Environmental behavior

Leaching tests were performed at room temperature (20 °C) in a double-neck flat bottom glass flasks of 250 mL. Three different liquid (L) to solid (S) ratios were studied: 10, 2.5 and 1.25 mL/g. Such L/S ratios were selected in order to simulate intense, medium and low rainfall conditions, respectively. In all cases, 150 mL of distilled water (as rainfall water) were added to the proper amount of sinter sample to give the desired L/S ratio. The mixture of water and sinter sample was stirred very slowly (50 rpm) in order to guarantee the homogeneity of the suspension. The mixture was kept under constant stirring and samples were taken at 4 different times in order to simulate various storage times: (i) 1 week, (ii) 1 month, (iii) 3 months and (iv) 6 months. After such periods, the mixture was centrifuged at 10,000*g* for 10 min in order to separate the polluted water from the solid. The polluted water was filtered through a PVDF membrane (pore size of 0.45 μm) to remove any remaining solids before the analysis of the leached elements. The analysed elements were alkali metals (K), alkaline earth metals (Ca, Mg), heavy metals (Cu, Fe and Mn), metals (Al), halogens (Br) and non-metallic elements (S). These elements were determined by ICP mass spectrometry (ICP-MS), using an Agilent 7500ce Spectrometer. Rhodium (^{103}Rh) was used as the internal standard.

3. Results and discussion

3.1. Sample characterization

3.1.1. Chemical composition

Elemental analysis of samples FF and CF (see Table 1) showed that the presence of both nitrogen and sulfur compounds is negligible in both dusts. Carbon content is low in sample CF

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