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2 Review article

Influence of chemical composition and physical structure on normal radiant emittance characteristics of ash deposits

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ABSTRACT

Ash residues that arise during combustion of solid fuels form deposits on heat exchanger surfaces which hinder the heat transfer to the working fluid steam. The thermal conductivity and – especially within the combustion chamber – the optical properties of the deposits determine the transferred amount of heat. Thus, knowledge of the optical properties, here represented by the normal emittance, are crucial for the design and operation of a steam generator.

Since the emittance is dependent on both the physical structure of the surface as well as the mineralogical composition, both parameters are subject to the current investigation. In the work presented the spectral normal emittances of mineral samples as well as their dependence on both the chemical composition and physical structure were investigated experimentally. In the course of the research the degree of complexity was increased gradually. Starting with pure quartz sand the influence of the surface structure on emittance has been investigated. By mixing quartz and hematite powder the influence of chemical enrichment with this typical ash component has been investigated. Measurements with real coal ashes complete the experimental program. Additionally, the latter samples were undergone thermal treatment to investigate the effect of particle agglomeration and melting.

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61 **1. Introduction**

52 Slaggings and foulings form thermal insulations on heat 53 exchanger surfaces that hinder the heat transfer to the working

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http://dx.doi.org/10.1016/j.fuel.2014.05.047 0016-2361/© 2014 Published by Elsevier Ltd. fluid steam. Accordingly, these surfaces have to undergo regular cleaning procedures. The heat insulation characteristics of the coverings are on the one hand caused by the rather low thermal conductivities of ashes in the order of 1 W/mK. On the other hand, due to the high temperatures present in combustion chambers, radiant heat is the dominant heat transfer mechanism. So the optical properties of the ashes determine the amount of heat absorbed.

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Experience shows that even very thin coatings on heat exchanger surfaces can lead to a significant decrease in absorbed heat and thus to an increase of furnace exit temperature (see Gwodsz et al. [13]). Since the thickness of these coatings is in the order of just millimeters the decreased heat transfer by conduction cannot be the root cause for this effect. In contrast, the effect can be attributed to the fact that ash layers change the optical properties of the walls which in turn leads to an increased amount of radiant heat that gets reflected back into the combustion chamber.

Since the reflectance of a surface – and hence also the emissivity – is affected by both its physical structure and its chemical composition the question arises which is the dominant influence. The goal of the current article is to contribute to the clarification of this question. In the course of this article "emissivity" is used for theoretical values derived from electromagnetic wave theory or Mie scattering theory, whereas "emittance" is used for measured values of real surfaces.

88 In the current paper the normal emittance perpendicular to the 89 sample surface is measured and not the hemispherical or the angular distribution of emittance. However, because typical ash oxides 90 91 behave like dielectrics which show low angular dependency of 92 emittance the normal emittance is a good first estimate for the 93 hemispherical emittance (a typical correlation: hemispherical 94 emittance normal emittance 0.92, see for example Wall [9]). Hemi-95 spherical emittance is used in most CFD software packages as 96 boundary condition for radiation computation.

The importance of the exact knowledge of ash emittance for reliable design and CFD assisted lay-out of industrial boilers is highlighted for example in [15–19].

100 **2. Review on previous experience**

101 Mulcahy et al. [1] were among the first to carry out systematic experimental investigations of the total emittances of ashes and 102 slags taken from a pulverized coal fired boiler as well as synthetic 103 slags mixed from pure mineral powders and glasses. The experi-104 105 mental results of all samples showed similar characteristics: The 106 emittances decreased with increasing temperatures up to a certain 107 point from which on the emittances started to increase again. By 108 repeated measurements at lower temperatures these emittance 109 increases were found to be irreversible and were attributed to 110 structural changes of the sample surfaces caused by the onset of 111 sintering and fusion.

112 Boow and Goard [4] have extended the sample spectrum of [1] 113 to more than 30 ashes and slags in order to investigate the effect of particle size and chemical composition on emittance. Qualita-114 tively, the measured emittances showed the same characteristics 115 116 as those found in [1]. However, the authors measured vast differences for ashes with different thermal histories. The effect of the 117 thermal history on the emittances of the samples was mainly 118 119 attributed to differences in particle sizes resulting from the thermal 120 treatment. To further examine the influence of particle size and 121 chemical composition on emittance they examined synthetic slags. 122 Samples composed of SiO₂, Al₂O₃ and CaO, partly enriched with "coloring agents" Fe₂O₃ and carbon, respectively, were molten, 123 ground and sieved into different particle size fractions. Samples of 124 125 the same particle sizes showed increasing emittances with increasing Fe₂O₃ or carbon content. Similarly, the emittances of pure as 126 127 well as enriched samples were found to increase with increasing 128 mean particle diameter. For the non-enriched synthetic slag the 129 authors were even able to derive a numerical correlation between 130 particle diameter d and emittance at a temperature of 500 °C:

131 **133**

$$\varepsilon(d) = 0.25 \log_{10} d + 0.13$$

Since the emittances of the synthetic slags enriched with Fe₂O₃ were significantly increased at higher temperatures, it was assumed that the enrichment especially influences the absorption index at shorter wavelengths. This was experimentally confirmed by Goodwin and Mitchner [7] who were the first to directly measure both components of the complex refractive indices of synthetic and natural slags. Starting with a base mineral mixture similar to [4] consisting of SiO₂, Al₂O₃ and CaO they successively added Fe₂O₃ and TiO₂ and created thin slabs of slag by means of melting and surface polishing. For wavelengths up to about 4 µm their results showed a strong dependence of the absorption index on the amount of iron oxide added to a slag, whereas the real part of the refractive index was mostly unaffected by the enrichment. Their measured values have since then served as a base for many numerical simulations involving absorption and scattering of radiation by slag particles, e.g. Bhattacharva [10].

Further experimental results were provided by Markham et al. [6], who measured spectral emittances of several boiler ashes and slags. They compared the emittance of an original sample composed of sintered material with those of ground, fused and molten samples prepared from the same material. They found that the emittance of the sample grinded to powder was the lowest over the whole wavelength range examined, followed by the sintered and the fused sample, respectively; the molten sample in turn showed the highest emittance. These results were confirmed by measurements of a powdery fly ash sample, which also showed a significant emittance increase after fusion. The authors concluded that the surface morphology is the dominant influence on the emittance of samples of identical composition.

Similar experimental results were reported by Zygarlicke et al. [5], who measured spectral emittances of six different coal ashes which were each separated into five different particle size fractions by means of a staged cyclone. They found that the sample emittances were dominated by the particle sizes and that the different ash compositions only played a minor role on the optical properties. Thus, their measurements and those of Markham et al. [6] confirmed the results of Boow and Goard [4], who found the particle size to have a more significant influence on emittance than chemical composition.

In more recent research, Shimogori et al. [2] and Saljnikov et al. [3] measured spectral emittances of coal ashes. The former measured at a fixed wavelength of 1.6 μ m and found increasing emittances with increasing temperatures at that wavelength, whereas the latter measured in a wavelength range between 2.5 μ m and 25 μ m and found decreasing total emittances with increasing temperature. As a common result, repeated measurements at lower temperatures showed the emittance developments to be irreversible, which both authors attribute to structural surface changes caused by sintering and fusion. Both authors confirmed the hysteresis in the temperature-dependent development of emittances originally found by Mulcahy et al. [1].

Finally, Wall et al. [9] reviewed the experimental results of [1,4,6,7] and theoretical investigations regarding the emissivity of single particles based on the Mie scattering theory [8] and combined them into a correlation between particle size, refractive index and emissivity. They showed that the emissivity of chemically identical particles is dominated by their respective size. Furthermore, the influence of the refractive index of the particle material on its emissivity is dependent on particle size: The effect of the chemical composition on particle emissivity gets more significant with increasing particle diameter. This relationship is discussed in more detail in the following section.

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