



## Composite fillers and their influence on emissivity

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### ABSTRACT

The research work presented in this paper shows influence of filler composition in inorganic composite on its emissivity. Chromium Oxide is an active ingredient, which is present in the basic formula of BG HitCoat<sup>®</sup>. Also other active ingredients (Iron powder, Kaolin, Silicon Carbide, Boron Carbide, Boron Nitride and Aluminum Nitride) were built in the matrix of the composite. Thermal stability up to 900 °C was analyzed. Spectral dependence of emissivity for different temperatures was measured by FTIR spectrometer for each sample. These tests demonstrated that filler composition is able to influence emissive properties in the range of short IR wavelength (1.5–4.0 μm) within 0.7–1.5 times (against the reference).

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## 1. Introduction

The inorganic composite materials have been used for protection of materials during the last decade, which are pelted with heat, clinkering, abrasion and high temperature corrosion. If the materials also have a high emissivity, they can be used in various heat plants (industrial furnaces, combustion chambers, heat exchangers, etc.) to improve energy efficiency [1–5]. More research and development work in this area deals with new materials and technologies for their application [6], and the practical use of these coatings [7,8]. Development of new methods for measuring emissivity is required [9].

### 1.1. Emissivity

The emissivity of a material (usually written  $\varepsilon$  or  $e$ ) is the relative ability of its surface to emit energy by radiation. It is the ratio of energy radiated by a particular material to energy radiated by a black body at the same temperature. It is a measure of a material's ability to radiate absorbed energy. A true black body would have a value of  $\varepsilon = 1$  while any real object would have  $\varepsilon < 1$ . Emissivity is a dimensionless quantity with no units.

Thermally emitted radiance from any surface mainly depends on two factors: (1) the surface temperature, which is an indication of the equilibrium thermodynamic state resulting from the

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energy balance of the fluxes between the gray body surface and its surroundings; (2) the surface emissivity, which is the efficiency of the surface for transmitting the radiant energy generated on the surface into its surroundings. The latter depends on the temperature (but the relationship between emissivity and temperature is not definite, depending on surface parameters and wavelength), material composition, surface roughness, coating thickness, wavelength, and physical parameters of the surface [9]. This contribution is dealing right about material composition.

Spectral emissivity represents the value of emissivity at a wavelength of radiation and total emissivity characterizes the total power emitted over all wavelengths [10].

Spectral zone in near IR band (1.5–6.4 μm) is interesting in furnaces with operational temperature 700–1200 °C. 90% of the heat energy is transferred in these temperatures and wavelengths [7].

### 1.2. Objective

The aim of this paper was to determine the influence of using different fillers (or active ingredients) in the composite system on its emissivity.

Various substances with high emissivity and high thermal resistance were selected. It was an attempt to incorporate these substances into the composite matrix and analyze the thermal stability of fillers and measure the spectral emissivity.

Similar researches were carried also in other research teams, but they were based on another base material and examined different parameters. For example Babrekar et al. studied the effect of the size of fillers in composite system aluminum/polystyrene and found that emissivity decreased when using coarse grains or flakes of aluminum. While they used the aluminum nanoparticles (10–50 nm)

there was no change in emissivity compared to commonly used aluminum grain sizes (micrometer units) [20]. Yu et al. reached similar conclusion. They examined the size and shape of the particles of copper in the composite system copper/EPDM (ethylene-propylene-diene monomer). Spherical particles of copper in micrometer units ensured the highest emissivity, whereas emissivity rapidly declined with increasing size of copper (spherical shape, tens of micrometers) or change in shape (plate, ticket) [21]. Both of these studies were carried out under normal temperature.

Dan et al. [6] used emissive coatings for high temperature applications. These coatings were based on inorganic fillers, and they examined  $\text{Al}_2\text{O}_3$ ,  $\text{Cr}_2\text{O}_3$ ,  $\text{ZrO}_2$  and  $\text{SiC}$ . Also they reached total emissivity of 0.90.

## 2. Inorganic composite BG HitCoat<sup>®</sup>

Research is carried out on coatings BG HitCoat<sup>®</sup> (produced by BG SYS HT Ltd, Pardubice, Czech Republic), which are inorganic composite systems based on silicon. The coatings have been developed to protect heat transfer surfaces exposed to heat. They are water based and dry at normal temperature. The composition of the composite can be adapted to different types of substrates.

### 2.1. Binders

The binders are not true solutions. They are mixtures of soluble ionic and covalent silica-based cross linkers, colloidal silica both pure and alkali enhanced, and metal-oxide particles partially soluble in  $\text{ion}^{++}$  form. The binder provides the flexibility, the bonding properties, some of the thermal and chemical resistance and the integrity of the coating. The volume of the binder is between 30% and 55%. When the binder dries, it forms an amorphous layer, which binds the fillers and the active ingredients together and to the substrate. This functions by combining silicon dioxide with a cross-linking agent and an alkali-metal oxide. The resultant solution has an extremely high pH and functions by dissolving the initial atomic layers of the surface, breaking the atomic bonding, leaving a chemically active surface looking for oxygen and anions to bond to. The binder provides these thereby achieving chemical equilibrium and chemically bonding the coating to the surface simultaneously. Finally bonding strengths are extremely high (above 15 MPa).

### 2.2. Fillers

Fillers are added to the binder to improve the physical properties of the coatings. Because the filler constituents are usually larger than the binder constituents their surface chemistry is less important. The shape and size of the filler particles is, however, of critical importance to their performance. Spherically shaped particles allow for maximum solids loading but there is some trade-off in porosity and coating viscosity. Platelet and fiber shaped particles may be added to reinforce the coating's ability to handle both internal forces (such as drying stress) and externally applied forces. However, they can only be used at relatively low levels since they increase the coatings' viscosity and porosity. The particle size will affect the hardness and the uniformity of the coating. It must also be compatible with the desired coating thickness.

Some preferred fillers and their effects are as follows:  $\text{TiO}_2$  is used to improve the bonding of the coating to the substrate.  $\text{CeO}_2$  is used to alter the electrical properties of the coating, as well as to cross-link to the polymer chains in the wet binder stage. It provides extremely strong adhesive and cohesive bonds, which are chemically inert and heat resistant. Clay, flint, iron and

carbides are used to provide a hard coating, which is thermally resistant. To help in providing thermal shock resistance clay is used as a plasticizer to provide stress release of temperature reduction stresses. Some fibrous material provides increased tensile strength, which is critical during the curing stage, as well as in normal use.

### 2.3. Active ingredients

Active ingredients are special type of fillers. They are added to the composites to provide the particular physical properties. Their purpose is not to improve the mechanical or chemical properties, such as abrasion or corrosion resistance, but to affect the optical and/or heat transfer properties of the surface and bulk. For example emissivity, thermal conductivity/resistivity, and electrical conductivity/resistivity are all properties, which can be tailored. Subject of this paper is to investigate the influence of various active compounds on emissive properties. Substances with high emissivity at normal temperatures, which managed to incorporate into the matrix of this composite system, are used (such as chromium oxide, iron powder) [11–13] as well as mixtures of these substances (such as carbides, nitrides) [14,15].

### 2.4. Samples preparation

The same binder was used for all the samples. There was 45–55 vol% of binder. The rest contained fillers while the active ingredient formed 20–30 vol% in the filler. The substances used were Chromium Oxide ( $\text{Cr}_2\text{O}_3$ ) produced in BASF Belgium, Iron powder (Fe) produced in BASF Germany, kaolin ( $\text{Al}_4(\text{OH})_8\text{Si}_4\text{O}_{10}$ ) produced in Sedlecký kaolin Ltd., Czech Republic, Boron Nitride (BN) produced in Saint Gobain USA, Aluminum Nitride (AlN) produced at ANCeram Germany, Silicon Carbide (SiC) produced in Korund Benatky Ltd, Czech Republic and Boron Carbide ( $\text{B}_4\text{C}$ ) produced in Saint Gobain Ceramics USA. The list of samples is shown in Table 1. Sample S02 corresponds to the original formulation. The properties of the active ingredients are shown in Table 2. Thus prepared mixtures of fillers were incorporated into the aqueous solution of binder. Correct dispergation was assisted by ultrasound. The mixing container was equipped with a probe that transferred high frequency voltage to ultrasonic waves. With this method of mixing the fillers particles are equally dispersed in suspension. Formed suspension was air-sprayed on steel specimens (steel substrate EN X8Cr30 (diameter 25 mm, thickness 5 mm)). The coating dried at normal temperature. The film formed an amorphous structure after drying. The thickness of the dry layer was ca. 150  $\mu\text{m}$ .

### 2.5. Samples testing

Surface microstructures of all samples, which were heated up to 500 °C, are shown in Fig. 1. No damages or melting are seen in these pictures.

**Table 1**  
List of measured samples.

Sample	Active ingredient
S02	Chromium Oxide
S04	Iron Powder
S06	Chromium Oxide and Iron Powder
S15	Kaolin
S16	Carbides (SiC and $\text{B}_4\text{C}$ )
S17	Nitrides (BN and AlN)

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