# EXPERIMENTAL AND THEORETICAL APPROACH TO DETERMINATION OF HEAT EVOLUTION IN ELECTRICALLY CONDUCTIVE ALUMINOSILICATES

by

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*Design of progressive building materials with increased utility value is the key issue for the development of reliable modern building structures. Compared to the conventional materials, progressive building materials are supposed to exhibit not just adequate mechanical, and thermal properties, but they are also supposed to be applicable in sophisticated solutions, such as in self-sensing, self-heating or magnetic-shielding systems. In terms of electric properties, the most of building materials are electric insulators which is the main limiting factor for their applicability in such sophisticated solutions. However, this deficiency can be solved by the addition of a proper amount of electrically conductive admixtures. Within the paper, electrically conductive alkali-activated aluminosilicate with 8.89 mass.% of carbon black admixture was designed and its materials properties necessary for calculations of heat evolution by the action of an electric source were experimentally determined. The electrical conductivity of such material equal to $5.57 \times 10^{-2}$ S m$^{-1}$ was sufficiently high to ensure self-heating ability. It was observed good agreement of experimentally determined data with those modeled by means of heat equation on sample with dimensions $40 \times 40 \times 10$ mm. Finally, one- and two-layered large-scaled heating elements based on materials with experimentally determined properties were designed and calculations were conducted to determine the voltage level necessary for one-hour heating from 268.15 K and 273.15 K to 278.15 K in the middle-top point of the construction.*

Key words*: alkali-activated aluminosilicates, finite element method,
carbon black, electrical conductivity, heat evolution*

## Introduction

Cement-based materials belong to the most frequently used in the world. They have been utilized for building structures in the past and remained dominant until present due to their good mechanical properties and durability. In 2016, the total production of cement in the world was estimated to be 4.2 Gt [1]. It was observed that China has been the leading cement producer in the world since 1985 and produced 2.35 Gt in 2015. The cement industry is responsible for nearly 6-8% of total carbon emission, consumes about 12-15% of the total industrial energy and is the second largest $CO_2$ contributor after power industry [2]. Therefore, the rising demand for alternative materials with less negative impact to the environment is obvious.

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Alkali-activated aluminosilicates (AAA) can be considered as a good alternative to the cement-based materials. The AAA involve binders obtained by reaction between a source of alkaline salt (solid or dissolved) and solid silicate powder [3]. This solid component can be calcium silicate or more aluminosilicate-rich material, such as metallurgical or blast-furnace slag (AAS), natural pozzolan, and fly- or bottom ash. Alkaline salts used for the activation of aluminosilicates are represented by soluble substances which raise pH of the reaction mixture, namely alkali hydroxides, silicates, oxides, sulfates, carbonates, or aluminates. The AAA are characterized by low hydration heat [4], good mechanical performance from very early ages [5], good resistance to sulfate attack [6], resistance to reinforcement corrosion [7], resistance to freeze-thaw cycles [8], and good performance at high temperatures [9, 10].

Both cement-based materials and AAA can be enhanced by addition of electrically conductive admixtures in order to attain sufficiently high electrical conductivity. In fact, admixtures in a proper amount build up electrically conductive net within the solid matrix which leads to the increase of electrical conductivity of the whole composite. Materials enhanced in such a way can be then utilized in smart applications, such as in self-sensing systems able to detect structural defects of the material by itself [11], in systems used for counting passing vehicles [12], in self-heating systems suitable for indoor floor heating [13], or in deicing systems embedded to roads, bridges or pavements in winter seasons [14]. The decisive presumption of correct functionality of a material in a smart system is the correct amount of electrically conductive admixture. Such topic was discussed, *e. g.*, in [15]. In the present, there are available various fibrous or particle-shaped electrically conductive admixtures, such as carbon fibers (CF), steel fibers (SF), steel slag (SS), carbon nanotubes (CNT), carbon black (CB), nickel powder (NP), or graphite powder (GP) [16].

This paper is focused on the experimental determination of material properties of AAS with 8.89 mass.% carbon black admixture important for calculation of the Joule's heat evolution. The ability of the analyzed materials to generate heat is proved experimentally by means of thermography and self-heating experiment. Subsequently, finite element calculations are conducted with experimentally determined input variables for two types of heating elements and different initial temperatures.

*Experimental part*

Alkali activation was conducted on AAS, subset of AAA, represented by SMŠ 380 produced by Kotouč Štramberk, Ltd. The fineness of the slag is of 380 $m^2$ $kg^{-1}$ and fulfilling the requirements of ČSN EN 197-1 [17]. Water glass Britesil $C_20_5$ with $SiO_2/Na_2O$ molar ratio equal to 2.07 was us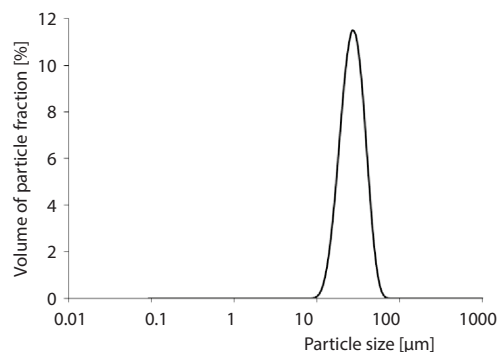ed as an alkali activator. Filler complying with ČSN EN 196-1 was produced by Filtrační pisky, Ltd., affiliate Chlum u Doks, Czech Republic. It is represented by three normalized CEN fractions of silica sand, PG1 (0/0.5 mm), PG2 (0.5/1 mm), PG3 (1/2 mm) in the weight ratio 1:1:1. The CB VULCAN 7H is electrically conductive filler produced by company CS CABOT. It is mainly used for the production of treads of premium passenger car tires, truck tires, solid tires, and conveyor belts. It is a promising material that offers a significant increase in the electrical



**Figure 1. Particle size distribution of CB VULCAN 7**

Fiala, L., *et al.*: Experimental and Theoretical Approach to Determination of ...
THERMAL SCIENCE: Year 2020, Vol. 24, No. 2A, pp. 787-794

789

conductivity of the AAS composite when used as an admixture due to the dominant particle size laying in the range of 10 – 100 mm, fig. 1, and of high surface area of the particles (up to 120 m$^2$/g).

The AAS samples were prepared CB suspension of 20 mass.% was prepared from a given amount of solid CB and an appropriate amount of water. Non-ionic surfactant Triton X in amount of 2 g was added and homogenizer IKA T-18 was used at 20.000 rpm for 15 minutes in order gain well-dispersed suspension. Besides, water glass in amount of 90 g was mixed with 105 g of water. Water glass solution was then mixed with 200 g of 20 mass.% CB suspension. Other components were mixed in the following sequence: 450 g of slag, 450 g of fine sand fraction (PG1 – 0/0.5 mm), 450 g of middle-coarse sand fraction (PG2 – 0.5/1 mm), and 450 g of coarse sand fraction (PG3 – 1/2 mm). The total amount of water (265 g) involving water present in CB suspension (160 g), and the additional water (105 g) was sufficiently high to ensure good workability of the mix. Water/slag and CB/slag ratio were equal to 0.59 and 0.0889, respectively. Composition of the studied AAS is given in tab. 1.

**Table 1. Composition of AAS with 8.89 mass.% CB**

| Component | Amount [g] [%] |
|---|---|
| Granulated blast-furnace slag | 450 |
| Waterglass | 90 |
| Sand PG1 (0/0.5 mm) | 450 |
| Sand PG2 (0.5/1 mm) | 450 |
| Sand PG3 (1/2 mm) | 450 |
| CB suspension | 20* |
| Amount of CB suspension | 200 |
| Additional water | 105 |

The mixture was put into 40 mm × 40 mm × 160 mm and 70 mm × 70 mm × 70 mm molds and vibrated. The samples were then demolded after 24 hours and cured in water for 28 days. The samples were finally dried in an oven in order to remove a majority of the physically bound water and placed into desiccator with silica gel to remove residual water during cooling to the ambient temperature.

The bulk density $\rho_v$ [kgm$^{-3}$] was determined using the gravimetric method by simple measurements of dimensions and weight. The thermal conductivity $\lambda$ [Wm$^{-1}$K$^{-1}$], and the specific heat capacity $c$ [Jkg$^{-1}$K$^{-1}$] was measured by the transient thermal-pulse method by an ISOMET 2104 device (Applied Precision, Ltd.) on samples with dimensions 70 mm × 70 mm × 70 mm [18]. The measurements were based on analysis of the temperature response to generated heat flow pulses. Heat flow was induced by a resistor heater placed in the probe having a direct thermal contact with the surface of the sample. The accuracy guaranteed by manufacturer is 5% of reading + 0.001 W/mK in the range of 0.015-0.7 W/mK and 10% of reading in the range of 0.7-6 W/mK. The measurements were carried out on samples dried in a dessicator with silica gel in order to keep them in dry state and conduct the experiment in environment without air-flow.
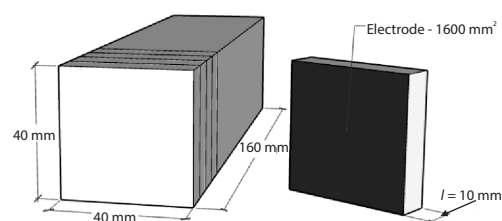


**Figure 2. Samples used for measurement of electric properties**

Electrical properties represented by the electrical conductivity were determined on samples with dimensions of 40 mm × 40 mm × 10 mm that were cut from demolded samples with dimensions 40 mm × 40 mm × 160 mm, fig. 2.

Samples were dried in an oven and put into a dessicator for several days in order to achieve the dry state without any residual water. In order to achieve good contact of the test-

ed samples with the measuring apparatus, two opposite lateral sides (40 mm × 40 mm) were painted by a conductive carbon paint (SPI Supplies) and additionaly pasted by an adhesive copper tape. The adhesive copper tape was added with respect to the previous observations which revealed the fact that carbon paint electrode is not sufficiently effective in terms of good distribution of the electric charge on the electrodes' surface and insufficiently tight connection between the carbon paint and the wire connecting the electrode with the power source.



**Figure 3. The electrical conductivity measurement**

The electrical resistance was determined in 2-electrode configuration by Fluke 8846A multimeter with 6-1/2 digit precision, fig. 3.

The electrical conductivity $\sigma$ [Sm$^{-1}$] was calculated with respect to the determined electrical resistance and the shape ratio of the samples according to:

$$\sigma = \frac{1}{R}\frac{l}{S} \tag{1}$$

where $R$ [W] is the resistance of the sample, $l$ [m] – the distance between electrodes, and $S$ [m$^2$] – the surface area of electrodes.
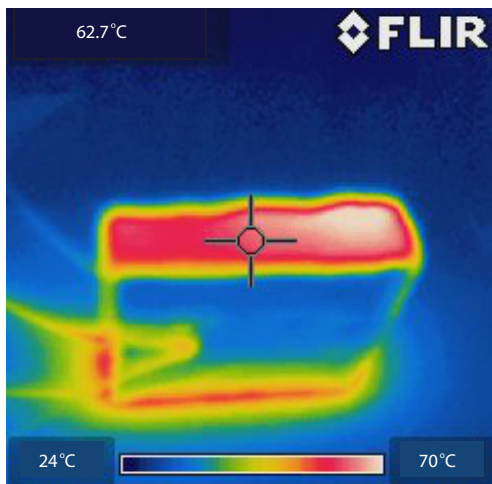


**Figure 4. Thermography measurement**

Self-heating ability of the studied AAS with 8.89 mass.% CB admixture was additionally proved by means of thermography measurements. The self-heating experiment was conducted on 40 × 40 × 10 mm sample under 40 V DC voltage load for 10 minutes. The temperature increase from 293.15 K up to 343.15 K proved the fact that such material is due to a sufficiently high electrical conductivity ($\sigma = 5.57 \cdot 10^{-2}$ S/m) able to evolve the Joule's heat, fig. 4.

**Theoretical part**

The Joule's heat evolution in electrically conductive materials is described by the heat equation with incorporated heat sources representing the heat evolved by-passing of the electric charge through the material [19]. Taking into account constant thermal conductivity, equation is expressed:

$$\rho_v c \frac{\partial T}{\partial t} = \lambda \Delta T + \sigma E^2 \tag{2}$$

where $\rho_v$ [kgm$^{-3}$] is the bulk density, $c$ [J kg$^{-1}$K$^{-1}$] – the specific heat capacity, $T$ [K] – the thermodynamic temperature, $t$ [s] – the time, $\lambda$ [Wm$^{-1}$K$^{-1}$] – the thermal conductivity, $\sigma$ [Sm$^{-1}$] is the electrical conductivity, and $E$ [Vm$^{-1}$] – the electric field intensity.

The following assumptions were taken into account within the model: material is homogeneous, isotropic and sufficiently electrically conductive in order to evolve the Joule's

heat, the contact resistances at electrode-material interface are negligible, it is considered a homogeneous electric field and constant electric field intensity. Input materials parameters used in finite element method calculations were determined experimentally.

In order to validate the model, self-heating experiment was conducted on $40 \times 40 \times 10$ mm sample by application of 20 V DC voltage. The temperature evolution in the middle of the sample's lateral side with dimensions 40 mm × 10 mm, (fig. 5 – experimental data), and ambient temperature, (fig. 5 – environment – probe 1 and 2), were monitored by *K*-type thermocouples and logged by the Comet MS55D data logger. The heating and subsequent cooling of the sample lasted 300 seconds and 2700 seconds, respectively, fig. 5. The heat transfer coefficient $\alpha = 25$ W/m²K was defined according to the ČSN 730540-3 standard [20], initial and boundary conditions corresponded to the temperature of the sample and ambient at the beginning of the experiment. The input data for the calculations are given in tab. 2.

Subsequently, 3-D models of two types of heating elements were designed. The first element, fig. 6, was formed just by the electrically conductive material CB (AAS with 8.89 mass.% CB), whereas the second element was a two-layered system made of CB and the covering non-conductive protective layer represented by a common oven-dried concrete (CC), fig. 7. The input model data represented by materials properties of involved materials, distance of the electrodes and initial and boundary condition are given in tab. 3. The 3-D models of heating elements were simplified into 2-D and calculations were carried out by means of the finite element method.

The main objective of the calculations was to identify the voltage level that would be necessary to increase the temperature from initial 268.15 K and 273.15 K up to 278.15 K in one hour of CB and CB + CC elements heating.
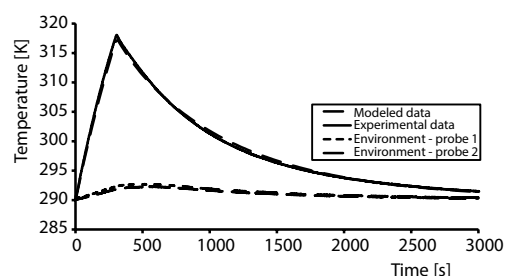


**Figure 5. Temperature evolution in the 40 mm × 40 mm × 10 mm AAS sample with 8.89 mass% CB**

**Table 2. Input parameters for model validation**

| Parameter | AAS-CB 8.89 [mass.%] |
|---|---|
| Applied voltage, $U$ [V] | 20 |
| Electrical conductivity, $\sigma$ [Sm⁻¹] | $5.57 \times 10^{-2}$ |
| Bulk density, $\rho$ [kgm⁻³] | 2022 |
| Specific heat capacity, $c$ [Jkg⁻¹K⁻¹] | 996 |
| Thermal conductivity, $\lambda$ [Wm⁻¹K⁻¹] | 1.16 |
| Distance between electrodes, $d$ [m] | 0.01 |
| Initial condition [K] | 290 |
| Boundary condition [K] | 290 |

**Table 3. Input parameters for calculations of heat evolution in CB and CB + CC heating element**

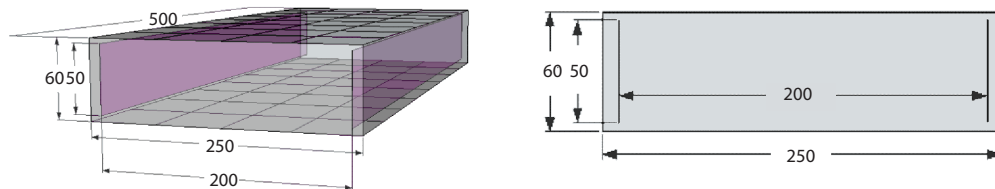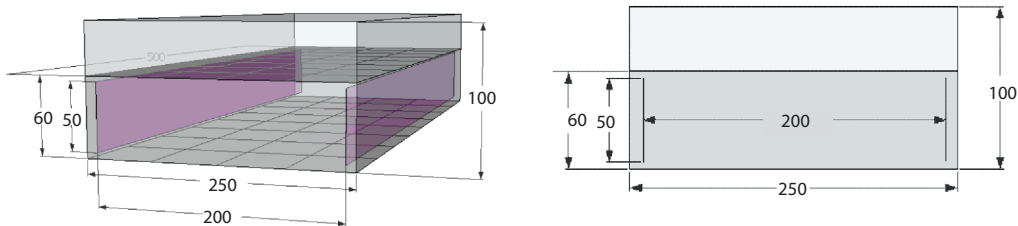| Parameter | AAS-CB 8.89 [mass.%] | CC |
|---|---|---|
| Electrical conductivity, $\sigma$ [S⁻¹] | $5.57 \times 10^{-2}$ | $1 \times 10^{-9}$ |
| Bulk density, $\rho$ [kgm⁻³] | 2022 | 2240 |
| Specific heat capacity, $c$ [Jkg⁻¹K⁻¹] | 996 | 737 |
| Thermal conductivity, $\lambda$ [Wm⁻¹K⁻¹] | 1.16 | 1.807 |
| Distance between electrodes, $d$ [m] | 0.2 | – |
| Initial condition [K] | 26815273.15 | 26815273.15 |
| Boundary condition [K] | 26815273.15 | 26815273.15 |

**Figure 6. The CB heating element [mm]**



**Figure 7. Two-layered (CB + CC) heating element [mm]**

## Results and discussion

In fig. 4, the initial experiment used for the verification of material's ability to evolve the Joule's heat and validation of the model is presented. It was proved that the material is sufficiently electrically conductive and therefore, able to generate the Joule's heat and that the modeled data are in good agreement with those experimentally determined.

The results of calculations performed for two types of heating elements described in section *Theoretical part* revealed the fact that in case of the heating element based on CB material, 110 V DC was needed to heat up the system from 268.15-278.15 K in one hour, to heat up such system in the same time from 273.15-278.15 K, just 76 V DC was necessary. In case of two-layered CB + CC system where the bottom layer was used as a heater and top layer as a protective cover that just conducted heat generated in the bottom the voltage needed to be increased. In case of the initial temperature of 268.15 K, it was necessary to use 165 V DC to heat up the element; when the initial temperature was 273.15 K, 114 DC V was needed.
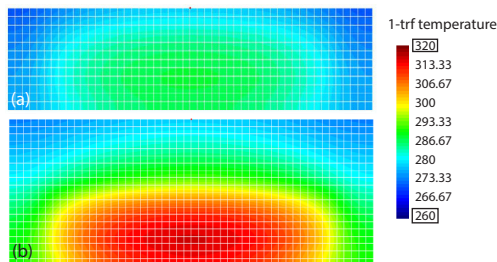
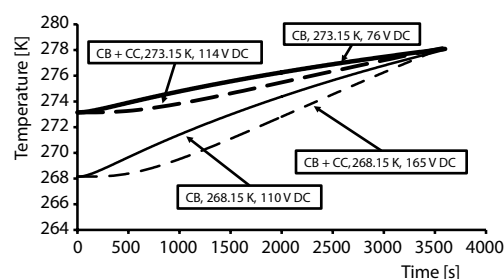In fig. 8, temperature maps of CB and CB + CC heating elements after 1 hour of acting of the given voltages are presented. Apparently, higher voltage was required for the two-layered CB + CC system compared to the one-layered CB system. The obvious reason is that the CC layer is not able to generate heat, it only spread out the heat generated in the CB layer by means of conduction.



**Figure 8. Temperature fields in (a) CB and (b) CB + CC element after 1 hour of heating**



**Figure 9. Temperature evolution in CB and CB + CC elements (top-middle point)**

In fig. 9, the heat evolution in CB and CB + CC elements in the top-middle point is presented. There was a difference in the depen-

dence of temperature on time for CB and CB + CC. While for CB the $T(t)$ curve was concave, for CB + CC it was convex due to the delay of heat transfer from the heat producing CB to the just thermally conducting CC.

## Conclusions

Both, experimental and theoretical approach was applied for the characterization of the Joule's heat evolution by acting of a DC power source in AAA represented by AAS with carbon black admixture.

The experiments showed that the analyzed material is able to evolve the Joule's heat which was proved by acting of 20 V power source. It is due to a conductive net of CB bulit in the solid AAS matrix. The effective electrical conductivity of the composite was determined by 2-probe method and was of 0.0557 S/m. On the basis of experimentally determined basic, thermal, and electric properties, finite element method calculations of heat evolution in two types of designed heating elements were conducted. The first element was composed of electrically conductive CB material. Such element was embedded by electrodes. The second element was composed of CB material with embedded electrodes and the protective CC layer. The CC is electrically non-conductive material with relatively high thermal conductivity that helps to effectively conduct heat. Finally, finite element method was used for identification of the voltage necessary for the temperature increase from 268.15-278.15 K and 273.15-278.15 K.

With respect to the calculations, the temperature increase of CB heating element in one hour from 268.15-278.15 K and 273.15-278.15 K can be provided by 110 V DC and 76 V DC power source, respectively. In case of two-layered CB + CC, the necessary voltage for the same temperature increase is 165 V DC and 114 DC V, respectively.

Heating elements based on electrically conductive AAS can find utilization in practice in a future, *e. g.*, in self-heating pavements or roads. Future work will be devoted to design and preparation of AAS mixtures with different electrically conductive admixtures and determination of mechanical properties that can be deteriorated with an increasing amount of electrically conductive admixtures.

## Acknowledgment

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