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# Application of a non-stationary method in determination of the thermal properties of radiation shielding concrete with heavy and hydrous aggregate

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ABSTRACT

Results of measurements of the specific heat and the thermal conductivity of concrete with blended special aggregate for neutron and gamma radiation shielding are presented. Experimental tests were performed on concrete with heavyweight aggregate (magnetite, barite), hydrogen-bearing aggregate (serpentine) and amphibolite aggregate. The thermal properties of concrete were determined using a nonstationary method. The highest specific heat was found for concrete with serpentine aggregate. Simple models for predicting the specific heat and the thermal conductivity on the basis of concrete mix design were evaluated to include the blends of heavyweight and hydrogen-bearing aggregates. The thermal conductivity of concrete was found to be linearly dependent on the concrete density in the range from 2200 to 3500 kg/m$^3$. Its increase due to water saturation of concrete was not dependent on the open porosity of concrete. It was found that the specific heat can be fairly well predicted using the rule of mixtures formula. The thermal conductivity of concrete can be approximately predicted using a parallel model in the case of water-saturated concrete. The thermal conductivity prediction for dry concrete is also discussed.

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## 1. Introduction

The design of radiation shielding concrete usually requires the use of special mineral aggregates, recommended for the attenuation of a particular type of radiation. Gamma and neutron radiation are the most difficult to shield [1]. The essential thickness of the shield in a specific radiation field is influenced by the elemental composition of the constituents of concrete, especially the content of heavy elements (of high atomic mass), light elements (mainly hydrogen) and privileged elements (e.g. boron) [2]. According to [3], preferable rocks consist of minerals such as goethite, ilmenite, barite, magnetite or hematite or also minerals containing a large number of hydrogen atoms in their elemental composition (usually in the form of water bound in a crystal structure), such as serpentine. These aggregates, apart from their features that determine their suitability for shielding applications, are usually characterized by specific thermal parameters that are different from those used in conventional structural concrete. Because the volume of aggregate is usually 70–77% of the volume of concrete, its thermal parameters determine the thermal parameters of the material as a whole.

Thermal parameters of concrete such as coefficient of thermal conductivity and specific heat are known as important parameters of thermo-hydro-mechanical description of the functionality of biological shields around PWR and BWR nuclear reactors. Their importance is even greater in the case of high temperature reactors in nuclear power plants (NPP) [2].

As for ordinary concrete, its thermal properties have been the subject of many studies and their values are quite well defined and known from the literature e.g. [4]. In the case of shielding concrete, with heavy and hydrous aggregate, research devoted to its thermal properties is much less, and the parameters available in the literature are very diverse [2,5]. The differences between them are at least twice.

The thermal properties of concrete depend to a large extent on the type and content of the aggregate. It is true that in the literature one can find the values of thermal conductivity coefficient and specific heat of minerals and rocks, but they are determined for solid rocks excavated in specific locations, not crushed like aggregates.

Determining the thermal parameters of aggregates used for heavy concrete in order to determine the properties of the concrete

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mix made with their application encounter at least two difficulties. The first is that aggregates for concrete (both ordinary and shielding) are generally polymineralic, and the content of individual components differs much depending on excavation location. It can be seen very clearly when analysing the data collected in cross-sectional publications [6]. An example may be barite aggregate, which may contain both more than 90% $BaSO_4$ as well as about 65% $BaSO_4$.

The second difficulty, which mainly concerns the rocks used to produce heavy aggregates, is the insufficient amount of data on their thermal properties. It results from a much smaller demand for this type of rocks and their less frequent occurrence in natural deposits. In the case of magnetite and barite, only limited data on the thermal properties of pure minerals are available. The situation is much better in the case of serpentine, although the problem here is a significant variation of data available in the literature.

Biological shielding structures of reactors are subject to a number of additional requirements: apart from the appropriate ability to attenuate the radiation flux, appropriate load-bearing capacity, impermeability for liquids and gases, as well as durability during its life cycle are required [7,8]. The tightness of the structure in the normal operation of a reactor must be ensured in conditions in which one side of the biological shield operates at a temperature of about 65 °C (even up to about 90 °C locally), and the other at ambient temperature. As a result, a temperature gradient arises in the material, resulting in coupled mass (moisture) and heat transport [9]. The parameters of this transport significantly determine the thermal properties of the material.

In addition to the mineral composition of aggregates, the thermal properties of concrete may also be influenced by the degree of binder reaction in the cement matrix, which is related to the conversion of free water to the bound water in hydration products. Unbound water may undergo diffusion in the concrete capillary pore system and due to the change of RH (and even evaporation) affect the thermophysical properties of concrete. That is why it is an important issue to determine these parameters with varying degrees of concrete saturation with water (vapour) or at least in the limit states - full saturation and dry state.

It should be taken into account that the literature data on thermo-physical properties of shielding concrete are to a large extent historical since they come from 40 to 70 years ago, when the majority of currently operating nuclear power plants were built. Modern cements are generally finer and contain components not used at that time, therefore their hydration rate is different and the degree of conversion in a relatively short time is high, which affects the content of bound and unbound water mentioned above.

Heavy and hydrous aggregates often come from soft rocks and therefore crumble into irregular grains, and when they are crushed, a lot of dust is formed. It is true that the thermophysical properties of concrete do not depend on the grain size and shape of the aggregate grains, but physical and mechanical properties already do. Such aggregate characteristics may cause ambivalent effects. It may preferably shape the thermal parameters, but adversely affect the strength or permeability of the concrete. Therefore, when assessing thermal properties, it is necessary to simultaneously evaluate the mechanical and durability properties of the shielding concrete in order to optimize the composition of the concrete mix.

The expected reliability of passive safety systems in NPP depends on the accuracy of prediction of thermophysical properties (as well as strength and durability) of concrete. There are known models for determining the coefficient of thermal conductivity and specific heat of multiphase materials. One can use for this purpose the rule of mixtures, the parallel model, the serial model, or other from quite numerous two-phase models and fewer three-phase models. However, their suitability for concrete depends largely on the knowledge of the thermophysical properties of the ingredients, including aggregate and hardened cement paste. Such data are either unavailable or very diverse in relation to heavy and hydrous aggregates. In addition, the use of two-phase models for concrete, which is a material consisting of greater number of phases, is fraught with the risk of making a big mistake. However, neither the magnitude of this error has been estimated yet, nor is it known whether to expect the overestimation or underestimation of the thermal parameters of concrete for specific models or their combinations. This publication is at least partly to fill this gap.

Despite the wealth of two-phase material models, there are no guidelines on the applicability of specific formulas for determining the coefficient of thermal conductivity and specific heat of concrete. A special case here is blended aggregate concrete, especially in the case of mixtures of heavy/hydrous aggregates with aggregate used typically in construction concrete. Due to the considerable diversity of properties of both types of aggregates in a blend, each of them should be treated as a separate phase, which greatly limits the number of available models of computational thermal properties and requires the use of non-standard calculation procedures (e.g. iterative calculations, computational aggregation of components, etc.). The results of such calculations have not yet been verified by the results of experimental studies and this knowledge gap is also to be partially completed by this publication.

The purpose of the research presented in this work was to determine the basic thermal properties of concrete made with the use of heavy aggregates: magnetite and barite, as well as hydrogen-bearing aggregate - serpentine. The thermal conductivity coefficient $\lambda$ and the volumetric heat capacity $c_V$ were measured and the specific heat $c_p$ was calculated on the basis of the density of the concrete. In the case of thermal conductivity and specific heat, the obtained values were analysed using selected models that were used for this type of analysis in literature [10,11]. The analysis included both the effect of composition and humidity on the thermal parameters of shielding concrete. A similar analysis of the composition impact, based on literature data in relation to conventional concrete used in pavements, can be found in Panchmatia [12]. The influence of humidity on thermal parameters was analysed, among others, in [13,14].

## 2. Materials and methods

### 2.1. Materials and specimens

The concrete specimens were manufactured using a variety of mineral aggregates and only one cement type CEM I 42.5 N SR5/LH/NA (a special low heat low alkali, high sulphate resistant cement described in [15]) and a constant water to cement ratio w/c = 0.48. The composition of the concrete mixes is shown in Table 1. For fine aggregate, local quartz sand was used in an amount of 20% of the volume of the combined aggregate, together with fine fractions of crushed aggregates. An exception is the C-A concrete series, in which the proportion of sand in the whole aggregate volume was 30% due to workability issues. The following types of crushed coarse aggregates were used: amphibolite, barite, magnetite and serpentine. Both single coarse aggregates were used as well as blends of serpentine aggregate and one of the heavy aggregates (barite or magnetite) in volume proportions of 2:1 and 1:2. The density according to manufacturers' data and thermal properties of the rocks with a similar mineralogical composition are given in Table 2. The set of aggregates and cement type was selected to fit the local availability for planned construction of nuclear power plant in Poland.

The use of chemical admixtures (WR and HRWR) was justified to maintain the proper rheological properties of the mixtures.

**Table 1**
Mix design of concrete.

| Content [kg/m$^3$] | Concrete mixture | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | C-A | C-B | C-BB | C-BS | C-M | C-MS | C-S | C-SB | C-SM |
| Cement CEM I 42.5 N SR5/LH/NA | 350 (in all the mixtures) | | | | | | | | |
| Water | 168 (in all the mixtures) | | | | | | | | |
| Quartz sand 0–2 mm | 556 | 371 | – | 371 | 371 | 371 | 371 | 371 | 371 |
| Crushed amphibolite aggregate 2–8 mm | 912 | – | – | – | – | – | – | – | – |
| Crushed amphibolite aggregate 8–16 mm | 507 | – | – | – | – | – | – | – | – |
| Crushed barite aggregate 0–16 mm | – | 2349 | 2936 | 1566 | – | – | – | 783 | – |
| Crushed serpentine aggregate 0–2 mm | – | – | – | – | – | – | 273 | – | – |
| Crushed serpentine aggregate 2–8 mm | – | – | – | 485 | – | 485 | 909 | 788 | 485 |
| Crushed serpentine aggregate 8–16 mm | – | – | – | – | – | – | 273 | 182 | 485 |
| Crushed magnetite aggregate 0–5 mm | – | – | – | – | 839 | 772 | – | – | 895 |
| Crushed magnetite aggregate 0–16 mm | – | – | – | – | 1846 | 1018 | – | – | – |
| Optima 100 (WR) [% cem. mass] | 0.39 | 0.74 | 0.74 | 0.74 | 0.67 | 1.51 | 1.97 | 1.03 | 0.97 |
| Fluid Optima 185 (HRWR) [% cem. mass] | – | – | – | – | – | – | 1.13 | – | 0.31 |

WR – water reducing admixture, HRWR – high-range water reducing admixture.

**Table 2**
The properties of mineral aggregates (based on manufacturers data and literature studies).

| Type of aggregate | Density [kg/dm$^3$] | Thermal conductivity [W/m·K] | Specific heat [J/kg·K] |
|---|---|---|---|
| Crushed amphibolite | 2.90 | 1.00–4.00 | 670–1260 |
| Crushed serpentine | 2.60 | 1.40–2.90 | 880–1130 |
| Crushed magnetite | 4.80 | 3.58–9.70* | ~600* |
| Crushed barite | 4.20 | 1.33–2.99* | ~450* |
| Quartz sand | 2.65 | 1.36–7.69* | 698–700* |

* No data available for rocks, parameters for minerals.

Otherwise the workability of mixes containing barite or serpentine aggregate would be not acceptable for ready-mix concrete technology.

A laboratory mixer was used to produce the concrete mixes. The PN-EN 206 standard 150 mm cube specimens were cast for the determination of the compressive strength, the open porosity and the thermal properties of concrete. The workability of mixtures was determined using the slump test according to PN-EN 12350-2: the target range of slump of 40–100 mm was maintained. The specimens were wet cured at a constant temperature of 20–22 °C for at least 56 days to ensure the proper development of hydration of slow-hardening cement used.

### 2.2. Test methods

European standard test methods were used for the determination of the compressive strength and the bulk density of concrete. The open porosity accessible to water was determined using the French standard method NF P18-459.

The measurement of thermal properties was carried out using an ISOMET 2114 device, which was used to measure the thermal properties of the materials using the non-stationary method. A surface probe IPS 1105 was used with three measurement ranges, which gave a total ability to determine the thermal conductivity coefficient in a range of 0.04–6.0 W/m·K, and the volumetric heat capacity in a range of 0.04–3.0 MJ/m$^3$·K. The measurement accuracy declared by the manufacturer is 10% for the thermal conductivity coefficient and 15% + 1 kJ/m$^3$·K for volumetric heat capacity. Determining the thermal properties of the ISOMET device is based on the analysis of changes in the surface temperature of the specimen in two stages, first when it is heated with constant power, and then during its cooling. On the basis of the solution of the inverse problem of non-stationary heat flow, two parameters were determined: thermal conductivity of the tested material $\lambda$ and its volumetric heat capacity $C_V$. Additionally, if the material bulk density is known, its specific heat $C_p$ can be calculated.

For each of the concrete series, 10 specimens were tested. The plate specimens with dimensions of $150 \times 135 \times 25$ mm were cut out of 150 mm cube specimens after curing period. The plate specimens were further stored in water for 6 to 8 weeks and then weighed. Apart from determining their mass, the hydrostatic weighing of specimens was also performed in order to calculate the bulk density of the material.

The measurements of the thermal properties were first carried out on concrete specimens in the state of their maximum water saturation, available in a natural way under the atmospheric pressure. In order to prevent the specimens from drying out during the measurements, they were wrapped in aluminium foil, in which a small opening for a probe was left, after being removed from the water. After being tested in their saturated state, the specimens were placed in a laboratory drier where they were dried to a constant mass at 65 °C, avoiding any damage that might occur during drying at a higher temperature. Achieving a constant mass was confirmed if weighing two subsequent specimens at least 24 h apart showed no further weight loss. The specimens were weighed on electronic scales with an elementary weight unit of 0.1 g, which in the most unfavourable case meant an inaccuracy of 0.01% in determining the change in the specimens' weight The specimens that were dried to a constant weight at 65 °C were tested under laboratory conditions after cooling to 22 ± 2 °C.

### 2.3. Method of statistical data analysis

The obtained results of the thermal properties measurements were subjected to statistical processing using 30 results obtained for each series of concrete (three measurements on 10 specimens). These results were first subjected to the procedure of identifying outliers. The criterion defining outliers was 1.5 times the interquartile range (IQR). After the rejection of outliers, the average values of the thermal parameters were calculated and the measurement uncertainty was determined using the Student's t-distribution.

After statistical processing, the results obtained for each series were compared with each other, and also plotted in the figures illustrating the obtained relationships between the compositions of individual concrete mixtures and their thermal properties. Then, using the models available in literature, the thermal properties of the used aggregates were determined. In these calculations, only those concrete mixes were used in which only one type of special aggregate was included, i.e. the C-B, C-M and C-S concrete mixtures, and also the reference mixture C-A. The values of the thermal properties of the aggregates determined in this way were used to predict the thermal properties of the concretes in which mixes of two different special aggregates were used, i.e. concrete designated as C-BS, C-MS, C-SB and C-SM. In the predictive calculations concrete mixtures C-A and C-BB were omitted, because of their specific composition that made them useless in the calculations. For the prediction, the same models were used that were used to determine the thermal properties of the aggregates. The values calculated in the prediction procedure were then compared to the values obtained from experimental studies for the same concrete mixtures.

## 3. Results of measurements and calculations

### 3.1. Strength and open porosity of concrete

The compressive strength of concrete falls in the range between 45 and 67 MPa. That range is roughly representative for C35/45 concrete strength class. The apparent density of concrete is within the range from 2220 $kg/m^3$ and 3520 $kg/m^3$ as a result of the changing content of the heavyweight aggregate and the normal-weight aggregate. The open porosity of concrete specimens is presented in Table 3. The aggregate changes in the mix resulted in an increase of the open porosity up to 2.3%. The porosity accessible to water from 11.0 to 13.5% is representative for normal quality of concrete [7,16]. The scatter of results of the open porosity and apparent density determination was low: up to 5.9% and up to 2.6%, respectively.

### 3.2. Thermal conductivity

The values of the thermal conductivity obtained from measurements are presented in Table 4.

Comparison of the obtained results in the case of the concrete containing one type of special aggregate tested in the state of water saturation leads to the conclusion that in comparison with the reference concrete, the mix with magnetite aggregate was markedly more conductive (the difference was around 40%), and the mixture with barite aggregate had a significantly lower conductivity (by about 22%). The value of the thermal conductivity coefficient of the concrete with serpentine aggregate turned out to be very close to the value obtained in the case of the reference concrete (a difference of about 3%). The same relations of results were obtained for the dry specimens, except that the differences were 38%, 22% and 2% in relation to the reference concrete with magnetite, barite and serpentine aggregate, respectively.

In the case of concrete with mixes of two special aggregates, it can be noticed that the obtained values of the thermal conductivity coefficient increase with the increasing volume content of aggregate with higher conductivity, which was to be expected. The concrete with a mix of barite and serpentine aggregates has thermal conductivity values correspondingly greater than that of the concrete with coarse barite aggregate alone, and smaller than that of the concrete with only the serpentine aggregate. The situation is similar in the case of the concrete containing a mix of serpentine and magnetite aggregates, in which an increase in the content of the second aggregate causes an increase in the value of the thermal conductivity coefficient. The recorded increases and decreases are proportional to increase of density of concrete. This is illustrated well in Fig. 1, where the coefficients of the thermal conductivity of the concrete with special aggregates are plotted as a function of density.

Analysis of the obtained relationship clearly suggests that the value of the thermal conductivity coefficient of the concrete is almost linearly dependent on the density. Increasing density results from increasing content of special aggregate. In case of barite aggregate, which has lower conductivity than serpentine aggregate, the thermal conductivity coefficient decreases with the increasing density. In case of concrete with magnetite aggregate the trend is reverse, which results from the higher conductivity of magnetite aggregate compared to serpentine aggregate. The relationship described above is almost identical for both the saturated and dry specimens. The value of the thermal conductivity coefficient of concrete with amphibolite aggregate is close to the value obtained for concrete with serpentine aggregate.

### 3.3. Volumetric heat capacity and specific heat

The measured values of the volumetric thermal capacity $C_V$ are summarized in Table 5. The calculated concrete bulk density values

**Table 3**
Concrete porosity accessible to water and apparent density (the average values of 3 specimens).

| Concrete property | Concrete series | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | C-A | C-B | C-BB | C-BS | C-M | C-MS | C-S | C-SB | C-SM |
| Apparent density of fresh mixture [$kg/m^3$] | 2450 | 3100 | 3330 | 2870 | 3520 | 3050 | 2220 | 2480 | 2630 |
| Porosity accessible to water [%] | 11.0 | 12.2 | 13.2 | 12.5 | 12.3 | 12.2 | 12.9 | 13.3 | 12.4 |

**Table 4**
Measured values of the thermal conductivity coefficient.

| Parameter | Concrete mixture | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | C-A | C-B | C-BB | C-BS | C-M | C-MS | C-S | C-SB | C-SM |
| | *Specimens saturated with water* | | | | | | | | |
| Average thermal conductivity $\lambda$ [W/m·K] | 2.23 ± 0.03 | 1.87 ± 0.03 | 1.52 ± 0.02 | 1.90 ± 0.01 | 3.13 ± 0.05 | 2.65 ± 0.07 | 2.29 ± 0.03 | 1.97 ± 0.03 | 2.52 ± 0.02 |
| | *Specimens oven-dried in 65 °C* | | | | | | | | |
| Average thermal conductivity $\lambda$ [W/m·K] | 1.82 ± 0.01 | 1.42 ± 0.01 | 1.21 ± 0.01 | 1.49 ± 0.01 | 2.51 ± 0.02 | 2.08 ± 0.04 | 1.78 ± 0.01 | 1.53 ± 0.02 | 1.99 ± 0.01 |

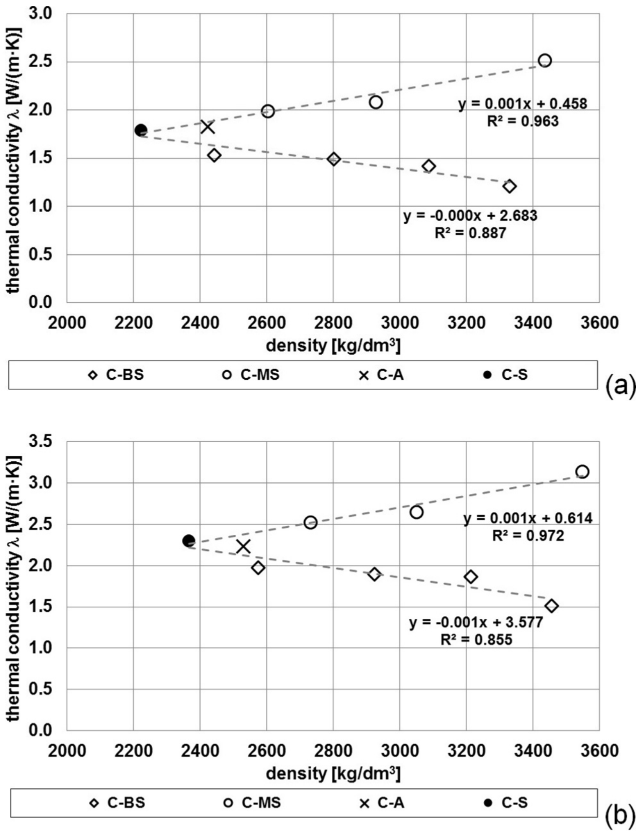


**Fig. 1.** Average values of the thermal conductivity of dry (a) and wet (b) concrete specimens.

$\rho$, which were used to calculate the specific heat of the concrete, are listed in Table 6, and the specific heat values $C_p$ in Table 7.

Comparison of the results of the thermal volumetric capacity test leads to several observations. First of all, it can be noted that when compared to the reference concrete, only concrete containing magnetite aggregate alone has a higher average value of volumetric thermal capacity. Admittedly, the average results calculated for the saturated specimens indicate that the concrete made of mixes of magnetite and serpentine aggregates could be included in this group, however, when taking into account the measurement uncertainties, this conclusion should not be considered.

The small variability of the results also attracts attention. The difference between the maximum and minimum values obtained in the tests is about 11%, both for the results obtained for the dry and saturated specimens. The differences between the dry and water saturated specimens made of the same concrete range from about 7% (for C-A concrete) to about 15% (for C-MS and C-SM concrete).

Analysis of the calculated results of the specific heat of the concrete made using only one type of special aggregate allows the concrete to be divided into two groups. In the first group, concrete with a specific heat value above 750 J/kg·K is included. It contains the C-A reference concrete and the C-S serpentine aggregate concrete. The second group contains concrete mixtures with a specific heat value below 600 J/kg·K. It includes the concrete with barite aggregate and magnetite aggregate. The given limit values are independent of the saturation state of the tested specimens, although they do have a clear influence on the obtained values of specific heat. The results for the dry specimens are lower, and the differences for the concrete with special aggregates range from less than 6% for the C-B concrete to over 11% for the C-M concrete. For the C-A reference concrete, the difference is even smaller and is about 2%.

Analysis of the all calculated values of the specific heat leads to the conclusion that these results are almost proportional to the concrete density for both wet as well as dry concrete. The $R^2$ parameter values for both linear functions linking the concrete density with its specific heat are over 0.90. What is interesting, the slopes of both of them are practically the same. This can be clearly seen in Fig. 2.

## 4. Thermal parameters prediction results

### 4.1. Specific heat

Specific heat is an additive parameter and can be determined in accordance with the rule of mixtures. Therefore, this rule was used to analyse its values. Due to the lack of information on the specific heat values of the aggregates used, the analysis was divided into

**Table 5**
Measured values of the volumetric heat capacity.

| Parameter | Concrete mixture | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | C-A | C-B | C-BB | C-BS | C-M | C-MS | C-S | C-SB | C-SM |
| | *Specimens saturated with water* | | | | | | | | |
| Average volumetric heat capacity $C_V$ [MJ/m$^3$·K] | 1.94 ± 0.04 | 1.89 ± 0.04 | 1.83 ± 0.03 | 1.93 ± 0.05 | 2.09 ± 0.06 | 2.01 ± 0.07 | 1.91 ± 0.05 | 1.90 ± 0.04 | 2.01 ± 0.07 |
| | *Specimens oven-dried in 65 °C* | | | | | | | | |
| Average volumetric heat capacity $C_V$ [MJ/m$^3$·K] | 1.81 ± 0.01 | 1.72 ± 0.02 | 1.70 ± 0.02 | 1.73 ± 0.01 | 1.89 ± 0.03 | 1.75 ± 0.03 | 1.70 ± 0.02 | 1.70 ± 0.03 | 1.75 ± 0.03 |

**Table 6**
Bulk density of concrete mixtures.

| Parameter | Concrete mixture | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | C-A | C-B | C-BB | C-BS | C-M | C-MS | C-S | C-SB | C-SM |
| | *Specimens saturated with water* | | | | | | | | |
| Average bulk density ρ [kg/m$^3$] | 2529 ± 3 | 3212 ± 8 | 3455 ± 9 | 2924 ± 28 | 3549 ± 26 | 3050 ± 27 | 2366 ± 3 | 2574 ± 19 | 2731 ± 10 |
| | *Specimens oven-dried in 65 °C* | | | | | | | | |
| Average bulk density ρ [kg/m$^3$] | 2422 ± 4 | 3086 ± 9 | 3329 ± 10 | 2801 ± 29 | 3436 ± 23 | 2928 ± 29 | 2223 ± 4 | 2442 ± 19 | 2603 ± 10 |

**Table 7**
Calculated values of specific heat of concrete mixtures.

| Parameter | Concrete mixture | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | C-A | C-B | C-BB | C-BS | C-M | C-MS | C-S | C-SB | C-SM |
| *Specimens saturated with water* | | | | | | | | | |
| Average specific heat $C_p$ [J/kg·K] | 765 ±17 | 589 ±12 | 531 ±8 | 659 ±15 | 589 ±17 | 659 ±22 | 809 ±21 | 736 ±17 | 737 ±24 |
| *Specimens oven-dried in 65 °C* | | | | | | | | | |
| Average specific heat $C_p$ [J/kg·K] | 751 ±5 | 557 ±7 | 511 ±5 | 613 ±6 | 530 ±8 | 602 ±7 | 762 ±7 | 694 ±14 | 672 ±10 |

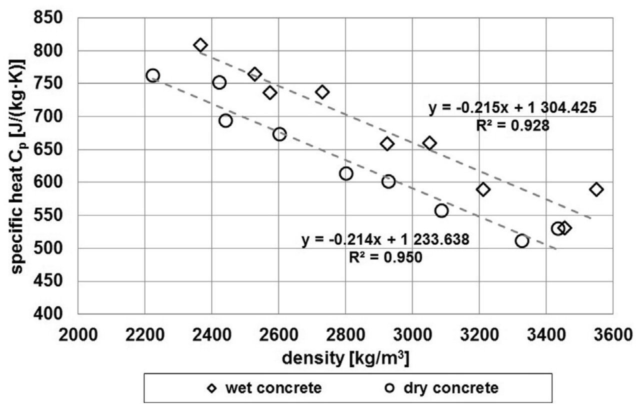


**Fig. 2.** Average values of the calculated specific heat of concrete specimens.

two stages. In the first stage, the values of the specific heat of the special aggregates, based on the specific heat values of the C-A, C-B, C-M and C-S concrete, were estimated. The values of the specific heat of the remaining components of the concrete were taken from the literature or tables. In the case of water, the specific heat was equal to 4200 J/kg·K, for cement paste to 703 J/kg·K [17], and for quartz sand (quartz) to 698 J/kg·K [6].

According to the rule of mixtures, one of the formulas (1) or (2) can be used to calculate the specific heat of concrete,

$$c_e = \frac{m_p c_p + m_s c_s + m_w c_w + m_a c_a}{m_p + m_s + m_w + m_a} \tag{1}$$

$$c_e = \frac{m_p c_p + m_s c_s + m_a c_a}{m_p + m_s + m_a} \tag{2}$$

in which $m$ and $c$ denote, respectively, mass and specific heat, and the indices $p$, $s$, $w$ and $a$ refer respectively to paste, sand, water and special aggregate. In the case of saturated concrete, it will be formula (1), and in the case of dry concrete, formula (2), in which the influence of air due to its negligible mass share in concrete is not taken into account.

The calculations were carried out assuming that the weight of the coarse aggregate and sand is as assumed in the concrete recipe. The mass of the cement paste in the hardened concrete was assumed as 1.23 times the mass of the cement in the recipe, assuming, according to [18], that it consists of cement and non-evaporable water in an amount of 23% of the cement mass. The mass of vaporizable water in the saturated concrete was calculated from the difference in the density of the saturated and dry concretes. On the basis of the above assumptions, the specific heat of the aggregates was calculated for the dry and saturated concretes separately. The results are presented in Table 8. In literature [6], the following values of the specific heat of the rocks and minerals that make up the test aggregate can be found: amphibolite (rock) 670–1260 J/kg·K, barite (mineral) 450 J/kg·K, magnetite (mineral) 600 J/kg·K, serpentine (rock) 880–1130 J/kg·K. Comparison of the values obtained from the measurements and subsequent calculations leads to the conclusion that they are similar, in the case of the results obtained for the dry concrete, to the quoted values measured with the use of the geological material. In the case of amphibolite aggregate, the obtained value is within the given range, which is quite wide and in the case of serpentine and barite aggregates, the differences amount to approx. 11% (for serpentine the distance from lower limit of the range was calculated). Larger differences were obtained in the case of magnetite aggregate because the value from the calculations was lower by approx. 20%. However, the results obtained for the saturated concrete deviate very much from the values given in literature.

**Table 8**
Calculated values of the specific heat of the special aggregate.

| Aggregate type | Specific heat $c_p$ [J/kg·K] | |
|---|---|---|
| | Saturated concrete | Dry concrete |
| Amphibolite aggregate | 532 | 774 |
| Barite aggregate | 358 | 503 |
| Magnetite aggregate | 386 | 475 |
| Serpentine aggregate | 560 | 789 |

On the basis of the calculated values of the specific heat of the special aggregates, the specific heat of the concrete containing mixtures of the two aggregates were calculated. Due to the significant discrepancies in the specific heat values of the aggregates, depending on the state of concrete saturation, both calculated values of this parameter were used depending on whether the specific heat of the concrete in the saturated or dry state was calculated. The values of the specific heat calculated in this way are shown in Table 9, where they were compared with the results obtained from the measurements.

The comparison of the results indicates a very good agreement between the results obtained from the tests and the results of the predictive calculations based on the simplified dependencies resulting from the rule of mixtures. This is also the case for the predictive calculations carried out with the use of the values of the specific heat determined from the measurements of the saturated

**Table 9**
Values of the specific heat concrete from predictive calculations and tests.

| Parameter | Concrete mixture | | | |
|---|---|---|---|---|
| | C-BS | C-MS | C-SB | C-SM |
| *Specimens saturated with water* | | | | |
| Predicted specific heat value [J/kg·K] | 630 | 640 | 714 | 717 |
| Specific heat value from tests [J/kg·K] | 659 | 659 | 736 | 737 |
| Absolute difference [%] | 4.4 | 2.9 | 3.0 | 2.9 |
| *Specimens oven-dried in 65 °C* | | | | |
| Predicted specific heat value [J/kg·K] | 611 | 587 | 678 | 661 |
| Specific heat value from tests [J/kg·K] | 613 | 602 | 694 | 672 |
| Absolute difference [%] | 0.4 | 2.5 | 2.4 | 1.7 |

concrete specimens. This compliance indicates that the rule of mixtures can be used to correctly estimate the specific heat values of concrete made with the use of more than one special aggregate, provided that the values of the specific heat of individual components are known. The rule of mixtures can also be used to determine the unknown value of the specific heat of concretes that are made with the use of special aggregate if the values of the specific heat of each component are known by solving a kind of the reverse problem.

### 4.2. Thermal conductivity

Analysis of the obtained values of the thermal conductivity coefficient faces two types of difficulties. First of all, in literature the results of conductivity tests for rocks and minerals can be found, but their scattering is usually very large. For example, in the case of barite, the range of conductivity coefficient values (including uncertainty of measurement) in [19] is 1.66–2.99 W/m·K, while in [6], a value of 1.33 W/m·K can be found. In the case of magnetite, the range of values in [19] is admittedly quite small - 4.19–5.10 W/m·K, however, in the same work the value of 9.70 W/m·K is also provided. In addition, the values given are usually assigned to rocks from specific locations, or refer to pure minerals that form rocks. Differences in the value between the same rocks from different locations are significant.

The second difficulty is the modelling of the thermal conductivity of multiphase materials, due to the variety of models and their variants found in literature. Two basic models of the thermal conductivity of such materials that limit the effective value from the bottom and from the top are the serial model (bottom limit) [20]:

$$\lambda_e = \sum_i \lambda_i \phi_i \tag{3}$$

and the parallel model (top limit) [20]:

$$\lambda_e = \frac{1}{\sum_i \frac{\phi_i}{\lambda_i}} \tag{4}$$

These two models can be called open models because they can include any number of phases that build the material. Other models found in literature are usually limited to two phases: the dissolving phase and the dissolved phase. Such models are far from sufficient when describing shielding concrete, in which, in a simplified case, at least four phases with a differentiated thermal conductivity coefficient can be distinguished: cement paste, fine aggregate, coarse aggregate and pores. Another limitation in the use of two-phase material models is the fact that many of them contain empirically determined factors. This makes them useless in the case of materials for which the values of these coefficients have not been determined.

In literature, there are also models of three-phase materials, including those based only on measurable parameters. These include the Maxwell model modified by Brailsfold [20]:

$$\lambda_e = \frac{\lambda_0\phi_0 + \lambda_1\phi_1\left(\frac{3\lambda_0}{2\lambda_0+\lambda_1}\right) + \lambda_2\phi_2\left(\frac{3\lambda_0}{2\lambda_0+\lambda_2}\right)}{\phi_0 + \phi_1\left(\frac{3\lambda_0}{2\lambda_0+\lambda_1}\right) + \phi_2\left(\frac{3\lambda_0}{2\lambda_0+\lambda_2}\right)} \tag{5}$$

or the Lichtnecker model (adopted from [21]):

$$\lambda_e = (\lambda_0)^{\phi_0}(\lambda_1)^{\phi_1}(\lambda_2)^{\phi_2} \tag{6}$$

They can be used in the case of concrete provided certain simplifications are made. If the cement paste and sand are treated as one phase, the special aggregate is homogeneous, and the saturation state of the concrete allows the assumption that there is only one phase in the pores (liquid or gas); the concrete can then be treated as a three-phase material. In addition, in the case of the Lichtnecker model, due to its formulation, it can be developed with factors that include successive phases. It is also possible to recursively use two- and three-phase models by aggregating two or three phases into one phase in each iteration. In this case, however, an increasing estimation error should be expected.

Due to the above limitations, the analysis of the thermal conductivity coefficient values in this paper was carried out in two stages using the four above presented models and by making some simplifications. In the first stage, the values of the thermal conductivity coefficient of the special aggregates were estimated using the values obtained from the conductivity tests of the C-B, C-M and C-S concrete. The models used were: the parallel and serial models and the Lichtnecker model extended with the fourth factor. It was assumed that concrete is a four-phase material whose three phases have known values of the thermal conductivity coefficient. The unknown value, determining the conductivity of the special aggregate, was calculated based on the selected model and the thermal conductivity coefficient of the concrete obtained from the tests. In the second stage, the coefficients of the thermal conductivity of the concrete with special aggregate mixtures, i.e. C-BM, C-BS, C-SM and C-SB, were calculated. The special aggregate thermal conductivity values estimated in the first stage were taken as the basis for the calculations. First, they were used in the same models from which they were earlier obtained. This time, however, assuming that they are five-phase materials (the Lichtnecker model has been extended by a fifth factor). Subsequent calculations were made using the Maxwell model. First, using the model, the coefficient of the thermal conductivity of the matrix was calculated, which was treated as a three-phase material composed of cement paste, sand, and water or air. The calculations were made on the basis of the data of the C-B, C-M and C-S concretes, and the results were averaged separately for the results obtained using the dry and saturated specimens. The Maxwell model was then used for the second time, but this time in the case of the C-BM, C-BS, C-SM and C-SB concretes. It was assumed that the three phases that make up each concrete are the matrix and two types of special aggregate. The values of the thermal conductivity coefficient of the individual aggregates were taken from the calculations from the first stage.

The following values of the thermal conductivity coefficient were used in the calculations: cement paste - 0.52 W/m·K [17], sand - 2.30 W/m·K [6], water - 0.61 W/m·K and air - 0.0257 W/m·K. Depending on the assumed saturation of the concrete, it was assumed that the entire pore space is occupied by either air or water. The values of the special aggregate thermal conductivity coefficient were calculated separately in both states, and the results are given in Table 10.

In the case of the serial model, the results obtained are the closest to those found in literature. According to data collected in [6], the thermal conduction coefficients of serpentine from the Swiss Alps are in the range from 1.72 to 2.51 W/m·K, and in the case of the same rock from the Kola Peninsula, it even reaches 2.99 W/m·K. The amphibolite from the Bohemian Massif, according to the same study, has a thermal conductivity in the range of 2.8–3.5 W/m·K. In the case of the other two aggregates, only data for the main minerals that build them are available in literature. And so the thermal conductivity of barite according to [6] is 1.33 W/m·K, and according to [20] is 1.66–2.99 W/m·K. In the case of magnetite, the values are as follows: 5.10 W/m·K [6] and 4.19–5.10 W/m·K [20]. The results obtained using the parallel model do not have any physical sense, as the thermal conductivity cannot take negative values. This model w. The values obtained from the calculations with the use of the Lichtnecker model are the highest, especially in the case of dry concrete, which is all the more surprising that the parallel and the serial model should theoretically limit the range of achievable results. Despite this, a decision was taken to use all the values obtained for further calculations.

**Table 10**
Calculated values of the thermal conductivity coefficient of aggregate.

| Aggregate type | Thermal conductivity coefficient λ [W/m·K] | | | | | |
|---|---|---|---|---|---|---|
| | Saturated concrete | | | Dry concrete | | |
| | Model (3) | Model (4) | Model (6) | Model (3) | Model (4) | Model (6) |
| Amphibolite aggregate | 3.23 | −2.59 | 5.19 | 3.36 | −0.11 | 10.56 |
| Barite aggregate | 2.46 | −8.24 | 3.39 | 2.59 | −0.12 | 6.69 |
| Magnetite aggregate | 4.72 | −1.97 | 8.55 | 4.84 | −0.12 | 16.86 |
| Serpentine aggregate | 3.22 | −3.40 | 4.88 | 3.35 | −0.11 | 10.19 |

The results of the second stage of calculations, determining the thermal conductivity coefficient of concrete with a mixture of two special aggregates (i.e. C-BS, C-MS, C-SB and C-SM), are shown in Table 11. The same table also shows the absolute percentage difference between the calculated and measured values.

In the case of the Maxwell model, the thermal conductivity coefficient of the mortar was first calculated. Depending on the saturation, it was assumed that the pores are filled entirely with air or water. The values of the thermal conductivity coefficient from the predictive calculations were 0.874 W/m·K in the case of the fully saturated mortar and 0.584 W/m·K for the dry mortar. Thus, the calculated values, together with the values given in Table 11, were used to calculate the thermal conductivity coefficient of the C-BS, C-MS, C-SB and C-SM concrete in the second stage. The obtained results of the second stage of the calculations are presented in Table 12, together with the absolute values of the differences between the values obtained from the calculations and measurements. The calculation results obtained using model (4) were omitted, as they were either negative (in the case of the saturated concrete), or an order of magnitude lower than the results obtained from the measurements (in the case of the dry concrete).

The values of the thermal conductivity coefficient of the concrete made with the mixtures of special aggregates obtained as a result of the calculations are presented in Fig. 3 (water saturated concrete) and Fig. 4 (dry concrete). In the case of calculations using the Maxwell model, the method for determining the thermal conductivity coefficient of the aggregates is given in the legend after the stroke. In both figures, the densities of the tested concretes are given under the designation of each series.

The analysis of the diagrams leads to the conclusion that in the case of saturated specimens, the thermal conductivity coefficient values calculated with the use of different models differ only slightly from the measurement results. The exceptions are calculations with the use of the Maxwell model that were carried out using the thermal conductivity coefficients of the aggregates obtained by means of the serial and parallel models. They lead in the first case to underestimating the calculated coefficient, and in the second to obtaining values that do not have physical sense (are negative). In the case of the dry specimens, the results obtained with the use of different models show significantly greater deviations from the measurement results and also greater variation among themselves. At one pole there was a parallel model, which in the case of the three concrete mixtures, overestimated the thermal conductivity coefficient the most. In turn, in the case of the fourth concrete - C-SM, this model allowed the value closest to the measured one to be determined. At the other extreme was the Maxwell model with the coefficients of the thermal conductivity of aggregates determined using a parallel model. In this case, the obtained values were very much underestimated. Values closest to those obtained from the measurements for three concrete mixtures were obtained using the Maxwell model and data obtained using the serial model. In the case of the concrete

**Table 11**
Values of the thermal conductivity coefficient from the predictive calculations and measurements.

| Parameter | Concrete mixture | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | C-BS | | | C-MS | | | C-SB | | | C-SM | | |
| Formula used to calculate the thermal conductivity coefficient of aggregates and concrete | Eq. (3) | Eq. (4) | Eq. (6) | Eq. (3) | Eq. (4) | Eq. (6) | Eq. (3) | Eq. (4) | Eq. (6) | Eq. (3) | Eq. (4) | Eq. (6) |
| *Specimens saturated with water* | | | | | | | | | | | | |
| Calculated thermal conductivity coeff. λ [W/m·K] | 2.01 | 1.97 | 1.99 | 2.85 | 2.78 | 2.82 | 2.15 | 2.12 | 2.14 | 2.57 | 2.52 | 2.54 |
| Absolute difference between the calculated and the measured value [%] | 5.7 | 4.0 | 5.0 | 7.5 | 5.0 | 6.4 | 8.8 | 7.4 | 8.2 | 2.0 | 0.1 | 0.9 |
| *Specimens oven-dried in 65 °C* | | | | | | | | | | | | |
| Calculated thermal conductivity coeff. λ [W/m·K] | 2.01 | 108 | 2.08 | 2.85 | 4.26 | 2.85 | 2.15 | 4.48 | 2.18 | 2.57 | 1.92 | 2.52 |
| Absolute difference between the calculated and the measured value [%] | 35 | >100 | 40 | 37 | >100 | 37 | 41 | >100 | 43 | 29 | 3.4 | 27 |

**Table 12**
Values of the thermal conductivity coefficient from the predictive calculations (using the Maxwell model) and measurements.

| Parameter | Concrete mixture | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | C-BS | | C-MS | | C-SB | | C-SM | |
| Formula used to calculate the thermal conductivity coefficient of aggregates and concrete | Eq. (3) | Eq. (6) | Eq. (3) | Eq. (6) | Eq. (3) | Eq. (6) | Eq. (3) | Eq. (6) |
| *Specimens saturated with water* | | | | | | | | |
| Calculated thermal conductivity coeff. λ [W/m·K] | 1.65 | 1.98 | 2.05 | 2.56 | 1.73 | 2.09 | 1.93 | 2.38 |
| Absolute difference between the calculated and the measured value [%] | 13 | 4.0 | 23 | 3.5 | 12 | 5.9 | 23 | 5.6 |
| *Specimens oven-dried in 65 °C* | | | | | | | | |
| Calculated thermal conductivity coeff. λ [W/m·K] | 1.38 | 2.01 | 1.65 | 2.30 | 1.44 | 2.08 | 1.57 | 2.23 |
| Absolute difference between the calculated and the measured value [%] | 7.0 | 35 | 21 | 11 | 5.8 | 36 | 21 | 12 |

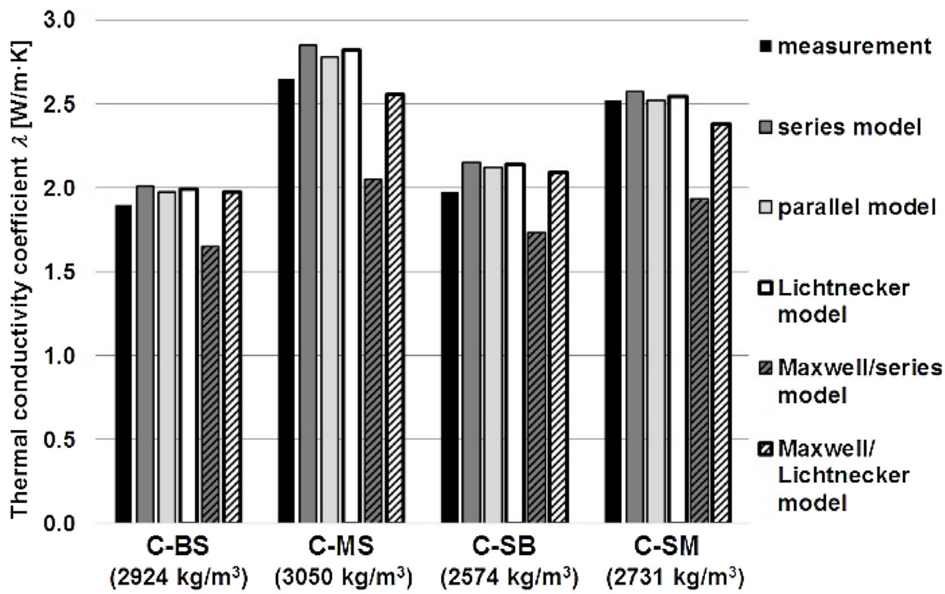


**Fig. 3.** Thermal conductivity coefficient from the measurements and predictive calculations (saturated concrete).

with a mix of serpentine and barite aggregates, this agreement is clearly better than in the case of the other two concrete mixtures.

### 4.3. Discussion

The non-stationary methods arouse some controversy [22] i.a. due to the indirect way of determining the thermal conductivity coefficient based on direct measurements of the thermal diffusivity and volumetric heat capacity. Despite these reservations, these methods are used quite commonly both in the case of cement paste [10], concrete [13], and concrete with recycled aggregate [23–25] with the addition of SCM materials [25,26], or also with the addition of waste materials [25,27]. The main advantage of non-stationary methods is the test time, which in the case of one measurement can range from several to over a dozen minutes when compared to several hours of testing in stationary apparatus [22]. The research and calculations presented above seem to indicate that reservations about non-stationary methods are not justified and that, after making appropriate assumptions, reliable results can still be obtained using them.

The obtained results confirmed that the rule of mixtures is the right model for calculating the specific heat of multiphase materials, including concrete. This formula can also be used to calculate the specific heat of an unknown material component, assuming that the heat values of the other components and the material as a whole are known. From the measurements and calculations presented above, it results, however, that in the case of porous material, when measurements are made at a transient heat flow to obtain reliable results, the material should be tested in a dry state. The specific heat values of the aggregates calculated using the rule

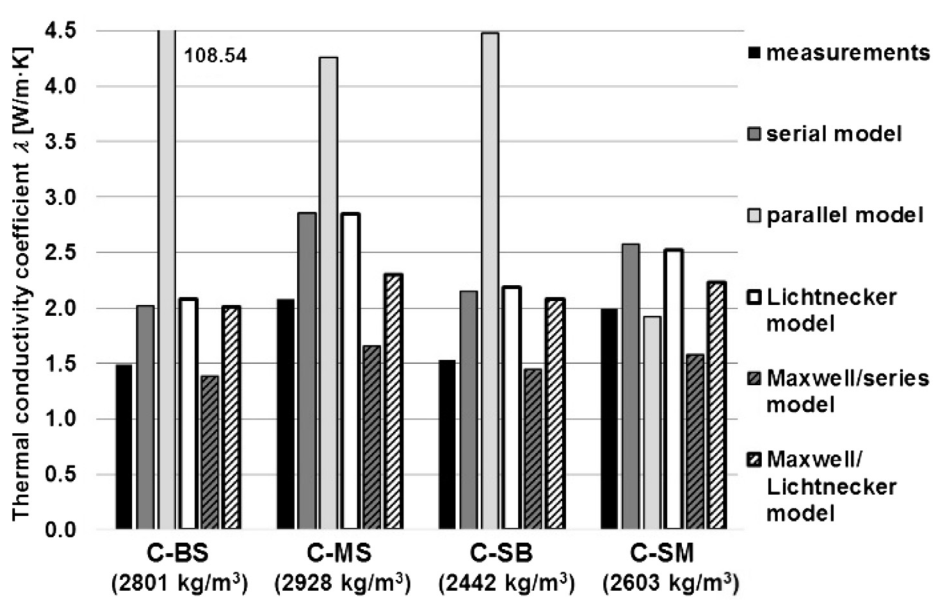


**Fig. 4.** Thermal conductivity coefficient from the measurements and predictive calculations (dry concrete).

of mixtures in the case of the measurements on the dry specimens are close to the values which can be found in literature referring to rocks or minerals that form the aggregates used. However, the values calculated from the measurements on the saturated specimens turned out to be much lower than the values known from literature and cannot be considered as correct.

Additional analyses have shown that to obtain results on saturated specimens similar to those obtained on dry ones, it should be assumed that the pore volume filled with water is 8–10 times smaller than the actual one. The probable cause of these discrepancies is the relatively short test time and the high value of the specific heat of water. During the test with the ISOMET device, the specimen is heated for about 200 s with a power that does not exceed 1.5 W. In such a short time and at such low power, only a small part of the water present in the pores had a chance of reaching a temperature like the concrete matrix, and only this part was included in the measurements. In the case of the dry specimens, this was not the case due mainly to the mass fraction of air in the specimen being negligibly small.

Regarding the obtained values of the specific heat of the tested concrete, the boundaries of the obtained range of values were outlined by the results of testing the concrete made with one type of special aggregate. From the bottom, this interval is limited by the concrete with barite aggregate and the concrete with magnetite aggregate. The upper limit is the value of the specific heat of the concrete with serpentine aggregate. The value of the specific heat of the reference concrete with amphibolite aggregate was close to the upper limit of the interval. Inside this interval, the values of the specific heat of the concrete were found, in which serpentine aggregate mixed with barite or magnetite aggregate was used. The values of specific heat obtained in these cases were proportional to the share of each aggregate in the aggregate mix.

In the case of the thermal conductivity coefficient, the obtained results indicate that it is possible to predict the value of this coefficient in the case of concrete with a mixture of aggregates, even if the thermal conductivity of these aggregates is unknown. However, it is necessary to measure the thermal conductivity coefficient of concrete with each of the aggregates used separately. With such values, it is possible, using the models available in literature, to calculate the thermal conductivity coefficient values of the aggregates used and to make predictions on this basis. The analyses carried out indicate that in order to obtain calculation results close to the measured values (within the measurement or calculation uncertainties), measurements of the thermal conductivity coefficient should be made on specimens of concrete completely saturated with water. In the case of the dry specimens, the prediction errors were much greater. The probable cause of these discrepancies is a few orders of magnitude lower thermal conductivity of air, which is significant in the case of materials with high porosity. Due to the fact that water has a thermal conductivity coefficient value similar to that of concrete, its presence in the material does not significantly distort the results.

As in the case of specific heat, the thermal conduction coefficient values obtained as a result of testing concrete made with one type of special aggregate determined the limits of the variability interval of the tested property. This time, however, at its one end there was concrete with barite aggregate with the lowest value of thermal conductivity coefficient, and at the opposite end concrete with magnetite aggregate. Between these values, although clearly closer to the minimum, there were thermal conductivity coefficient values obtained for concrete made with serpentine aggregate and the reference concrete. Between the values obtained for the concrete with barite aggregate and the concrete with serpentine aggregate, the values of the thermal conductivity coefficient of concrete with mixtures of these aggregates were found. However, between the obtained results and the composition of aggregate mixtures, there is no linear dependence. The situation is similar for concrete with magnetite aggregate and serpentine aggregate, and mixtures thereof. In both cases, the replacement of 1/3 of the aggregate that had lower conductivity with the aggregate that had greater conductivity results in a significant increase in the whole concrete conductivity, but the replacement of the next 1/3 causes a much smaller increase in conductivity and only the replacement of the whole aggregate causes a significant increase in the thermal conductivity coefficient value.

## 5. Conclusions

The performed test and analysis resulted in the following conclusions.

1. The thermal conductivity of concrete is linearly dependent on the bulk density of concrete in the range from 2200 to 3450 kg/m$^3$. It found to increase up to 47% when increasing the content of magnetite aggregate and to decrease by a maximum 41% for increasing content of barite aggregate.
2. The thermal conductivity of concrete is found to increase due to the saturation of concrete with water by an average of 21% (±3%) and the increase is not related to the open porosity of concrete.
3. The specific heat is linearly decreasing with increasing the bulk density of concrete by a maximum of 34% in the range of densities from 2200 to 3450 kg/m$^3$. The same relationship holds for all mixtures of heavyweight and hydrous aggregates.
4. The increment of specific heat due to water saturation of concrete is between 4 and 10% and it does not depend on the open porosity of concrete.
5. The use of heavyweight and hydrous aggregates in concrete resulted in an open porosity of concrete from 12.2 to 13.3%, meaning an increase of the open porosity by 1.2–2.3% in comparison with concrete containing amphibolite aggregate.
6. The simplified rule of mixtures was found effective to predict the specific heat of concrete containing both heavyweight aggregate and hydrogen-bearing aggregate. The difference between the predicted and measured values were within the margin up to 2.5% and 4% for dry and saturated specimens.
7. The prediction of the thermal conductivity of concrete with blended special aggregates calculated with the use of multiphase material models was found effective for the water saturated specimens: the differences between the predicted and measured values lied within the 7% margin. The best fit was obtained using the parallel model and the Maxwell model with the thermal conductivity coefficients of aggregates calculated using the Lichtnecker model.
8. For the dry concrete specimens with blended aggregates the prediction of thermal conductivity was much less accurate, probably due to the considerable porosity of concrete up to 13% and potential, significant influence of the thermal conductivity of air.

The obtained results permit to recommend the specific heat testing of radiation shielding concrete using a non-stationary method to be performed on dry specimens, while the thermal conductivity tests should rather be performed on water saturated specimens. For calculating values of thermal properties in other saturation states, the authors propose the use of appropriate calculation models.

## Conflict of interest

None declared.

## Ethical statement

Authors state that the research was conducted according to ethical standards.

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