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Influence of a Novel Carbon-Based Nano-Material on the Thermal Conductivity of Mortar

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Abstract: The paper presents the results of research work to assess the thermal conductivity of mortar incorporating a novel carbon-based nano-material (CBN). The data from the laboratory tests served as the starting point in training an artificial neural network (ANN) based on the Levenberg–Marquardt backpropagation algorithm that was used to predict the values of the thermal conductivity at later ages. The used CBNs were essential precursors of multi-walled carbon nano-tubes but different from their counterparts in the fact that they were capped at the ends. This configuration should result in lower surface tension and should prevent the bundling even without the use of surfactants and sonication. The obtained results show that the mortar mixes with CBN exhibit higher values for the thermal coefficient at early ages compared to the reference mix, even at very low percentages of CBN by weight of cement. The ANN is able to accurately predict the experimental results both at 28 days and at later ages. The obtained results should serve as the starting point for further investigations into the microstructure of cement-based materials enhanced with CBNs.

Keywords: mortar; carbon-based nano-materials; thermal coefficient; ANN; backpropagation

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1. Introduction

The continuous demand for construction materials with improved properties has sparked the interest of many researchers, and a lot of effort has been invested into creating new, better materials [1], which are still convenient enough from the point of view of their manufacturing process, design guidelines and practical applications.

Nanomaterials quickly gained an important momentum in terms of their application to construction materials due to the added value in terms of improved material properties and behavior [2–4]. The general consensus is that their use improves the physical and mechanical properties [5], and an optimal percentage of 0.075% was proposed as being optimal for yielding lowest sorptivity, lowest porosity and improved mechanical strength of cementitious matrices [6]. Percentages between 0.01–0.07% of carbon nano-tubes (CNTs) in conjunction with nano-silica were reported to improve the microstructure of cement mortar in a synergistic effect and resulted in better corrosion resistance and improved durability. Nano-silica enhanced the pozzolanic activity, whereas the CNT acted as fiber reinforcement between the aggregates and the paste, bridging the micro-cracks and arresting their development, thus leading to improved mechanical properties [7].

There are two distinct types of CNTs that are currently being investigated in terms of their beneficial effects in cementitious materials. Single-walled carbon nanotubes (SWCNTs) are basically tubes of graphite, sometimes capped at the ends, with their wall made of a single layer of molecules. Multi-walled carbon nanotubes (MWCNTs) consist of a series of concentric graphite tubes [8]. The advantage of SWCNTs is their greater flexibility in terms of being twisted, flattened or bent around sharp edges (of aggregates) compared to

MWCNTs. On the other hand, taking into account the large quantities of CNTs that would be necessary in large scale concrete elements, the use of MWCNTs seems the better choice, although their structure is less well understood and they are prone to more defects than their SWCNT counterparts [9].

Although the influence of CNTs on the mechanical properties of cementitious materials is intensely studied, their influence on the thermal conductivity does not seem to attract so much interest [10]. Studies were carried out on characterization of the CNTs themselves, and it was reported that they have excellent electrical and thermal conductivity. The electrical conductivity was in the range of 10^6 S/m for SWCNTs and $>10^5$ S/m for MWCNTs, whereas the thermal conductivity was determined to be 6000 W/mK for an individual SWCNT and >3000 W/mK for an individual MWCNT [11,12]. In their study, Lee et al. [13] concluded that 0.125%wt of cement resulted in the best thermal transfer capabilities of mortar, whereas 0.250%wt resulted in a decrease in the thermal transfer due to agglomerations of MWCNTs during the mixing process. Similar results were reported for steel fiber reinforced concrete, with the main difference being in the fact that the increase in CNT percentage did not result in a decrease in the thermal characteristics of concrete [14]. This might be because the agglomeration effect of CNTs that impacted the thermal properties of mortar was counteracted or diminished by the presence of steel fibers.

One of the most challenging goals in the construction industry is the reduction of carbon footprints [15]. This may be achieved by either developing new materials with enhanced mechanical and durability properties that require less maintenance and can resist higher loads, or by substituting a part of the raw materials by recycled components and by-products of other industries. Both such approaches require years of research and laboratory tests that result in large quantities of waste worldwide. However, due to the recent advances in computer technology and computer science, an alternative has been developed—computer simulation tools based on mathematical models that try to replicate, as accurately as possible, the chemical processes that take place inside the materials, their behavior under loads as well as the evolution of their long-term properties.

One such useful tool is the artificial neural network (ANN) that, if properly designed and trained, can help in the prediction process of long-term material properties. This would result in less waste generated during research works and significant time savings for determining those characteristics that require lengthy laboratory tests. Compressive strength is one of the most investigated characteristics of cement-based materials. It is the defining parameter of classifying materials for their structural or non-structural application. Although concrete can incorporate a large variety of components, some of them by-products or other industries, e.g., with treated palm oil fuel ash, the use of the ANN in predicting the compressive strength has proven to yield accurate results [16]. Other studies have shown the suitability of using the ANN when it comes to rubberized concrete [17] and concrete using fly-ash as a partial substitute for Portland cement [18]. It should be pointed out that even slight changes in concrete composition may result in different values for the mechanical properties. This is why the ANN is considered to be a very powerful tool that can be used to obtain accurate results if properly trained [19,20].

There is very little information in the scientific literature on the thermal conductivity of cement-based materials, e.g., mortar or concrete, using nano-materials, although this can be a key parameter in terms of either the comfort of living for the inhabitants or, more importantly, a characteristic of the material that shows its ability to distribute or inhibit the distribution of heat during a fire. Hence, the paper presents the results obtained from laboratory tests on the thermal conductivity of a cement mortar containing a novel carbon-based nano-material. Three different ratios, by mass of cement, were considered for the addition of nano-material. The thermal conductivity was measured on a standardized sample at different ages, up to 140 days. The developed ANN was used to predict the results at the age of 28 days and all subsequent ages, based on training datasets that were randomly selected. Its accuracy was validated against experimental results.

2. Materials and Methods

2.1. Materials

A CEM II B-M (S-LL) 42.5R, rapid hardening cement complying with standard specifications [21], commercially available, was used. It is a composite cement consisting of 65% ÷ 79% cement clinker and 21% ÷ 35% a mixture of ground-granulated blast-furnace slag (GGBS) and limestone. Natural sand, readily available on the market, with particle diameters ranging from 0 ÷ 4 mm, was considered. Tap water was used in the mortar mix.

The novel carbon-based nano-material (CBN) was produced in the laboratory based on the chloride mediated chemical vapor deposition (CVD) method [22]. The length of the CBNs was about 300 μm, the outer diameter was around 30 nm and the inner diameter was 6 nm. The CBNs are essentially precursors of MWCNTs with the difference that they are capped at their ends, as seen from Figure 1. One end is capped by a Fe particle (the white end in Figure 1), from the $FeCl_2$ powder used as a catalyst, whereas the opposite end is closed by a C particle coming from the C_2H_2 that was used as a carbon source in the CVD method. This would imply a lower surface energy compared to "traditional" MWCNTs.

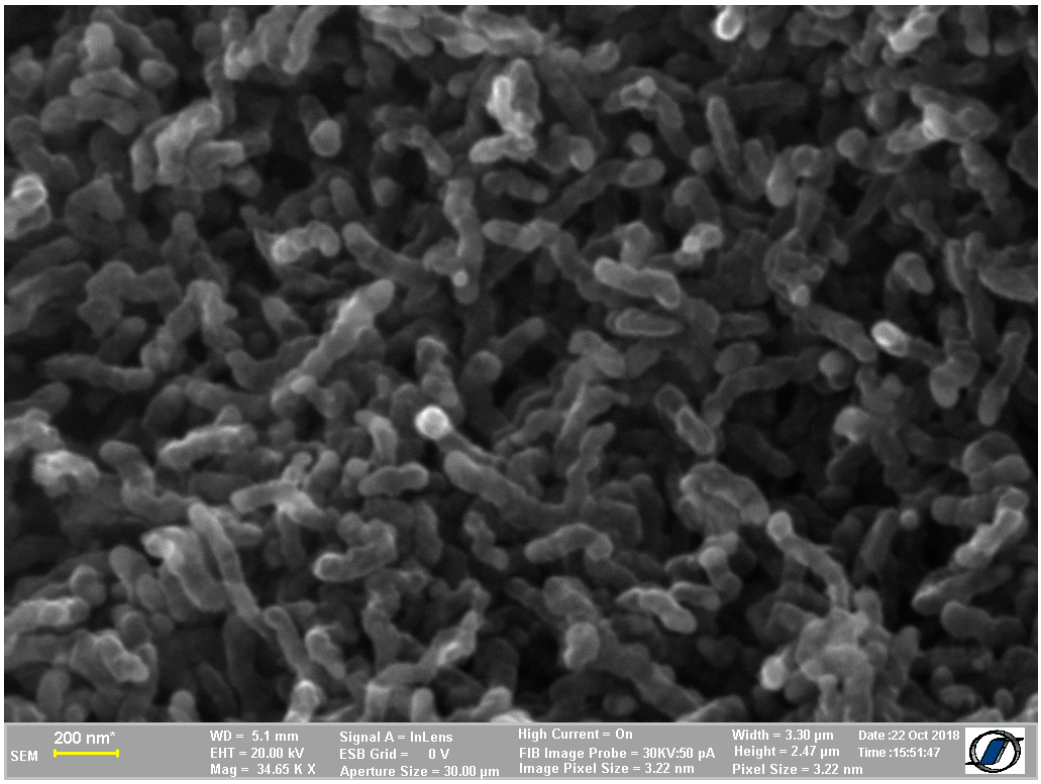


Figure 1. SEM image of carbon-based nano-material.

One of the main drawbacks of using MWCNTs is their high tendency to bundle together. This is mainly due to their high surface tension, which for the novel CBN is much lower. There are currently two widely used approaches to overcome this issue: the mechanical and the chemical approach. The latter involves the use of surfactants to lower the surface tension [23], while the former implies the use of sonication. Both approaches have their limitations: the use of surfactants creates a large volume of voids inside the cement matrix, while improper use of sonication may lead to breaking of MWCNTs [24]. The novel CBN aims at addressing the issue of using either surfactants or sonication to ensure their uniform distribution and relies only on the mechanical process during mixing.

2.2. Methods

The mix proportions used in this research are shown in Table 1. Three different percentages of CBNs, by mass of cement, were used. While 0.05% and 0.1% of MWCNTs are frequently reported in the scientific literature, the lower percentage, 0.025%, was seldom used. However, in the study performed by Xu et al. [25], a 6.25% increase in the value of the flexural strength was obtained for 0.025% MWCNTs for the cement paste. Morsy et al. [26] concluded that a 0.02% MWCNT by mass of cement yielded the highest improvements in the mechanical properties of mortar mixed with nano-clay.

Table 1. Mix proportions.

Mix Designation	Cement [g]	Sand [g]	Water/Cement	CBN/Cement [%]
REF	500	2050	0.6	-
CBN-025				0.025
CBN-05				0.05
CBN-1				0.1

Three standard cylindrical samples [27], 30 mm × 60 mm (h × d), were cast for each mix shown in Table 1. The samples were stored in water, after demolding, until the age of 28 days. From 28 days to 140 days the samples were stored at room temperature, 22 ± 1 °C and air humidity 50~60%.

Heat transfer (or thermal transmittance) occurs, to different degrees, in any structure. Heat flow can be measured, and subsequently expressed, as U-value (or thermal transmittance coefficient), being the heat flow through one square meter of a structure when the temperature on either side of the structure differs by 1 °C. U-values are commonly used to describe the thermal performance of building elements and, subsequently, the overall energy performance of a building. Therefore, the U-value is dependent on the thermal conductivities (λ) of the building materials and their respective thicknesses. Thermal conductivity (or conduction coefficient) is a physical property of the material through which heat transfer occurs and it defines its ability to conduct heat.

The measurements were carried out using the ISOMET model 2114, as shown in Figure 2, which is a hand-held instrument for direct measurement of heat transfer properties of a wide range of isotropic materials, plastics, glasses and minerals according to ASTM-5334-14 [27]. The operation principle is based on applying heat flow impulses on the material sample and analyzing the temperature response. Three measurements were conducted on each of the three samples cast for each mix proportion, resulting in nine measurements/mixes/curing ages.

Artificial neural networks (ANN) are incredibly adaptive and powerful tools that can be used to solve complex engineering problems. They have a wide range of applications, from municipal solid waste management and rainfall prediction to the properties of concrete by considering various parameters [28,29]. The ANN configuration used in the present research consisted of two neurons in the input layer, five neurons in the hidden layer and one neuron in the output layer. The ANN was based on the backpropagation (BP) algorithm and the Levenberg-Marquardt BP algorithm. The BP neural network has many advantages, such as being very fast and simple, but it may sometimes suffer from slow convergence and oscillations during the training process. One of the solutions proposed to overcome these disadvantages is the Levenberg-Marquardt algorithm, which is based on the combination of the gradient descent method and the quasi Newton method in order to ensure fast convergence and overall better performance [30].

The main parameters of the research were the CBN content and the curing age of mortar. The considered ages were: 7, 14, 21, 28 and 35 days (for the five weeks from the day of casting). Some of the experimentally obtained data was used to train the ANN, whereas other sets of data were used to check the accuracy of the predicted results. The ANN was then used to predict the values of the thermal conductivity (λ) for all four mixes at the ages of 100 days and 140 days.

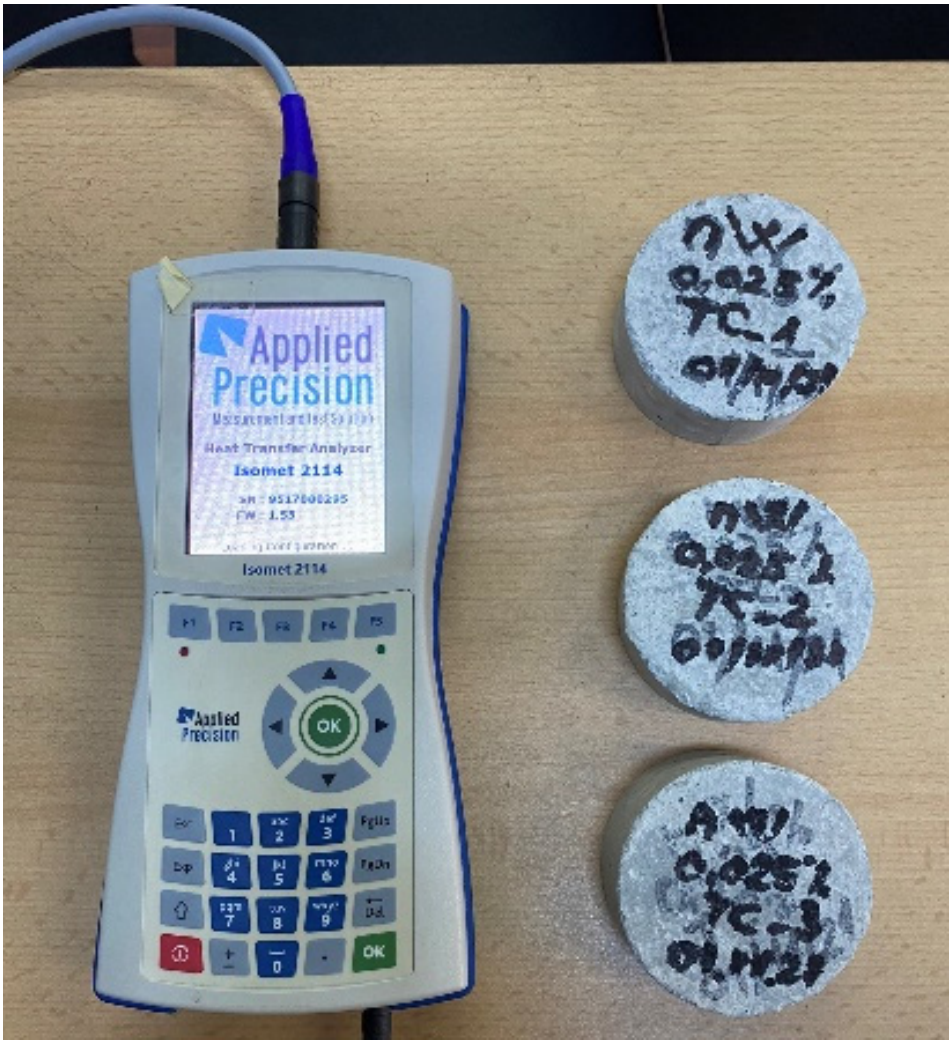


Figure 2. Measuring equipment and samples.

3. Results

3.1. Density

One of the key parameters influencing the values of the thermal conductivity is the density. Liu et al. [31] concluded that the increase in density results in an increase in the values of the thermal conductivity of iron ore sand cement mortar. Subsequent studies [32,33] have confirmed earlier findings. The density at the ages of 28 days and 140 days was assessed, taking into account the two different storage conditions. The variation of the mortar density with curing age and mix proportion is shown in Figure 3.

It can be observed that the mixes containing CBNs exhibit higher values for the density at both 28 and 140 days. The changing of curing conditions from fully immersed in water to air curing resulted in a decrease in the values of the density for all mixes.

The change, however, did not exceed 2% of the values obtained at 28 days. At the same time, the influence of adding CBNs to the mortar mix resulted in a maximum 3% increase in the values of the density. This maximum increase was also observed at the age of 140 days. It can, therefore, be concluded that the curing conditions influence all mortar mixes equally. At the same time, the very small variations in the values of density, compared to the reference mix, leads to the conclusion that, in this case, the influence of density on the values of thermal conductivity could be neglected.

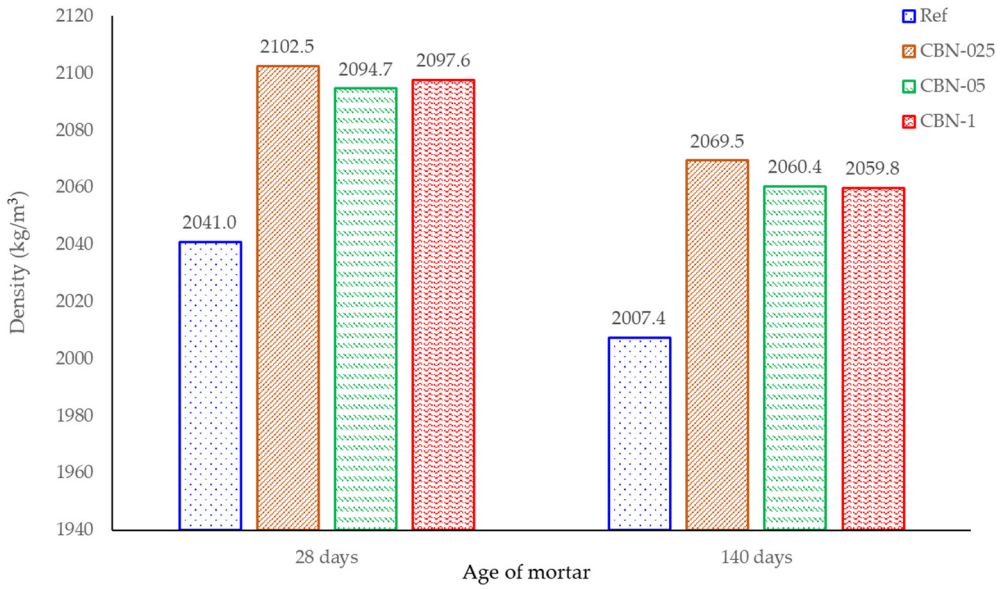


Figure 3. Variation of mortar density with curing age and mix proportion.

3.2. Thermal Conductivity (λ)

The change in the values, average values, of the thermal conductivity for all mix proportions is shown in Figure 4. It can be observed that for all considered curing ages, the mortars with CBN exhibit higher values for thermal conductivity. This is in line with the trends reported earlier in the scientific literature [13].

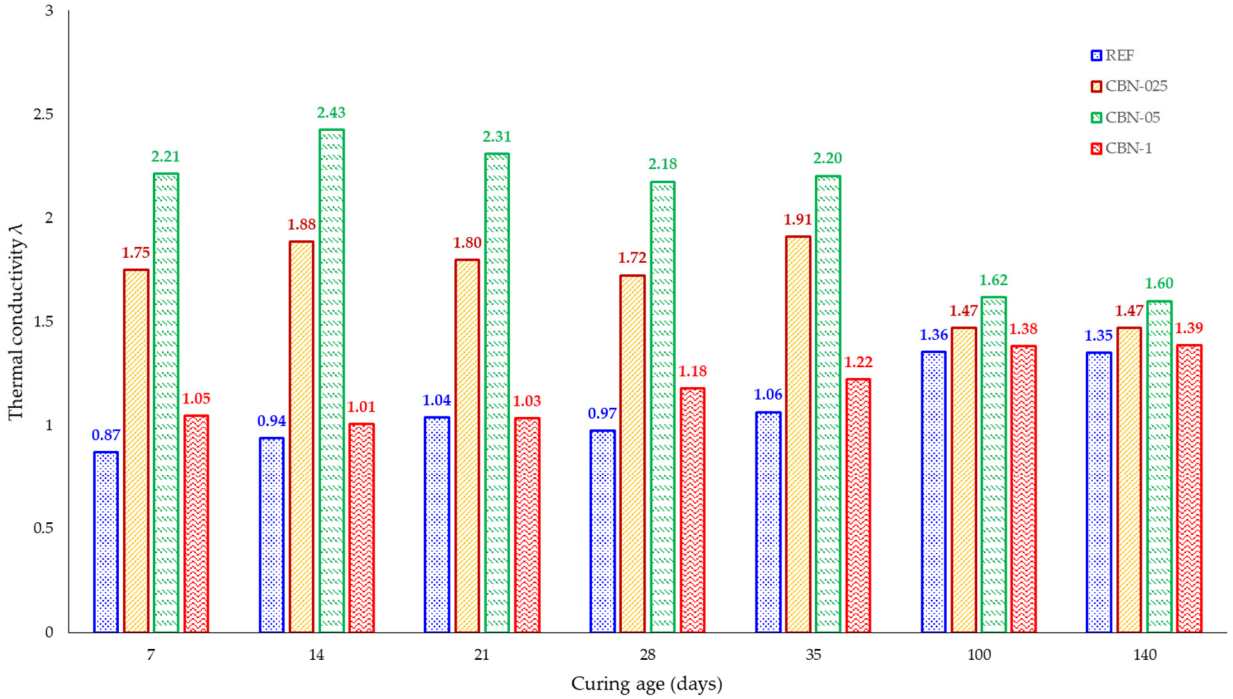


Figure 4. Variation of the thermal conductivity with curing age and composition of mortars.

Considering the mix proportion for the reference sample, the obtained values are within the limits of the interval 0.6~1.4 W/mK specified in construction practice. The values show fluctuations of 19% during the first 5 weeks, between 0.87 W/mK and 1.06 W/mK. However, they tend to stabilize from 100 days onwards.

The mixes containing CBN showed two distinct evolution patterns. CBN-025 and CBN-05 exhibited very high initial values of thermal conductivity only to drop later on by 23% and 27%, respectively, and remained constant afterwards. The CBN-1 mix, however, showed a similar trend to the reference mix, continuously increasing from 7 to 140 days.

The statistical data is presented in Table 2 in terms of standard deviation. It can be observed that, even though the CBN was directly mixed with the water and then added into the mixer, the obtained results are not scattered, meaning that a uniform mixture of the CBN was achieved. It should be pointed out that no surfactants or sonication procedures were applied.

Table 2. Thermal conductivity.

Mix Designation	Curing Age (Days)						
	7	**14**	**21**	**28**	**35**	**100**	**140**
REF	0.87 ± 0.23	0.94 ± 0.16	1.04 ± 0.23	0.97 ± 0.19	1.06 ± 0.26	1.36 ± 0.02	1.35 ± 0.01
CBN-025	1.75 ± 0.21	1.88 ± 0.22	1.80 ± 0.27	1.72 ± 0.29	1.91 ± 0.24	1.47 ± 0.06	1.47 ± 0.05
CBN-05	2.21 ± 0.1	2.43 ± 0.22	2.31 ± 0.24	2.18 ± 0.22	2.20 ± 0.1	1.62 ± 0.09	1.60 ± 0.07
CBN-1	1.05 ± 0.16	1.01 ± 0.22	1.03 ± 0.23	1.18 ± 0.16	1.22 ± 0.31	1.38 ± 0.02	1.39 ± 0.02

3.3. Thermal Conductivity (λ) Using Artificial Neural Network

At the age of 28 days, the ANN was trained using 101 randomly chosen sets of data (70% of the total data sets), 21 sets were used for the validation (15% of the total data sets) and another 21 sets were used to test the accuracy of the ANN (15% of the total data sets). The data sets were obtained by measuring the thermal conductivity of all samples at the ages of 7, 14, 21 and 28 days. The obtained results are shown in Figure 5.

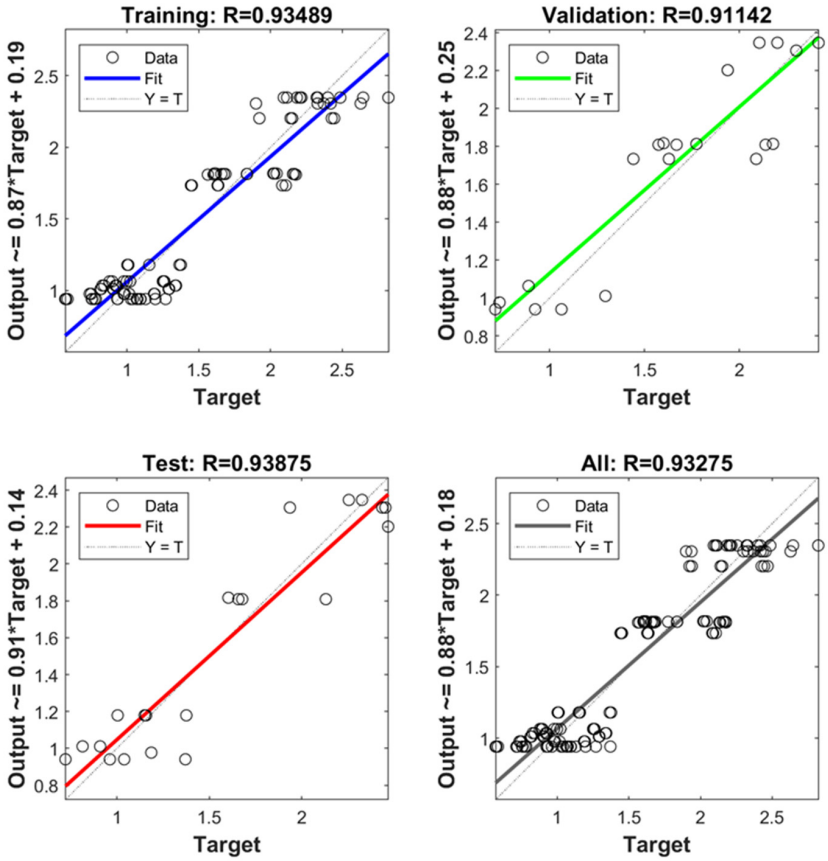


Figure 5. Performance of ANN at the age of 28 days (143 data-sets).

Different configurations of the ANN were also investigated but the currently chosen configuration yielded the best predictions in terms of the values for the thermal conductivity of mortar mixes, quantified by means of the R^2 coefficient and mean square error (MSE).

Figure 6 presents the ANN performance at the age of 35 days. As it can be seen, the performance is similar to the one obtained at 28 days. Considering the values of the thermal conductivity, presented in Figure 4, the obtained results were expected even without further training of the ANN. The somehow lower performance compared to the age of 28 days could be attributed to the fact that the curing conditions changed from being stored in water to being stored in air. The ANN was retrained using the experimentally obtained data from all previous measurements, with a total number of 180 data sets, which were divided using the same percentages as with 28 day predictions (70% for training, 15% for validation and 15% for testing). The retraining of the ANN, from anew, was considered necessary in order to avoid the over-training phenomenon [34].

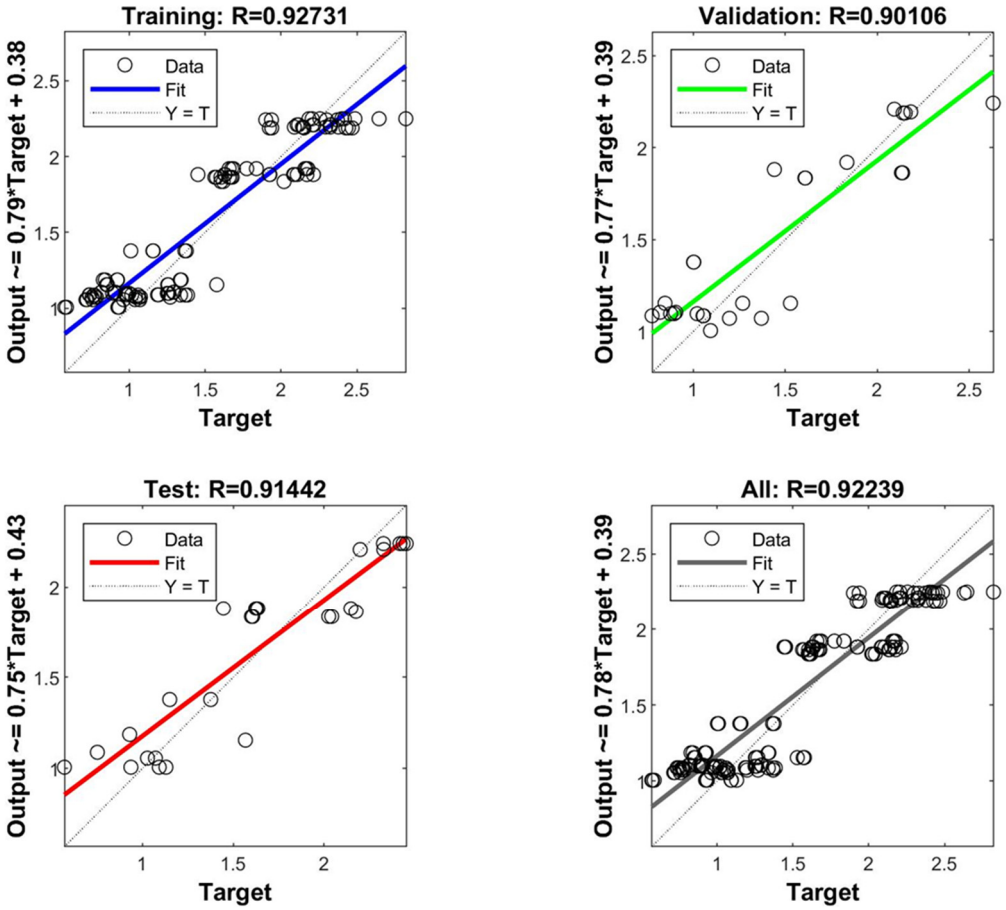


Figure 6. Performance of ANN at the age of 35 days (179 data-sets).

Considering the performance of the ANN and knowing that the properties of cement-based materials do not vary significantly in time after the age of 28 days, we decided to conduct the next set of measurements at the age of 100 days.

The ANN was further trained with an additional 36 sets of data and predictions were made for the values of the thermal conductivity at the age of 100 days. However, given the large time interval between the two curing ages, coupled with a change in storage conditions, the predicted results were significantly different from the measured data. The predicted values differed from the measured ones, from −0.29 W/mK to 0.5 W/mK, which means that the standard error varied between −20% to 30%.

Because of the significant differences between the predicted and measured values of the thermal conductivity, more values were experimentally determined at the ages of

100 and 140 days. The number of determinations doubled, which resulted in 18 data sets/mixes/curing ages. The newly determined values were used to re-train the ANN in the same percentages: 70% training, 15% validation and 15% for testing. The number of data sets increased to 251. The performance of the ANN improved significantly, as can be seen in Figure 7.

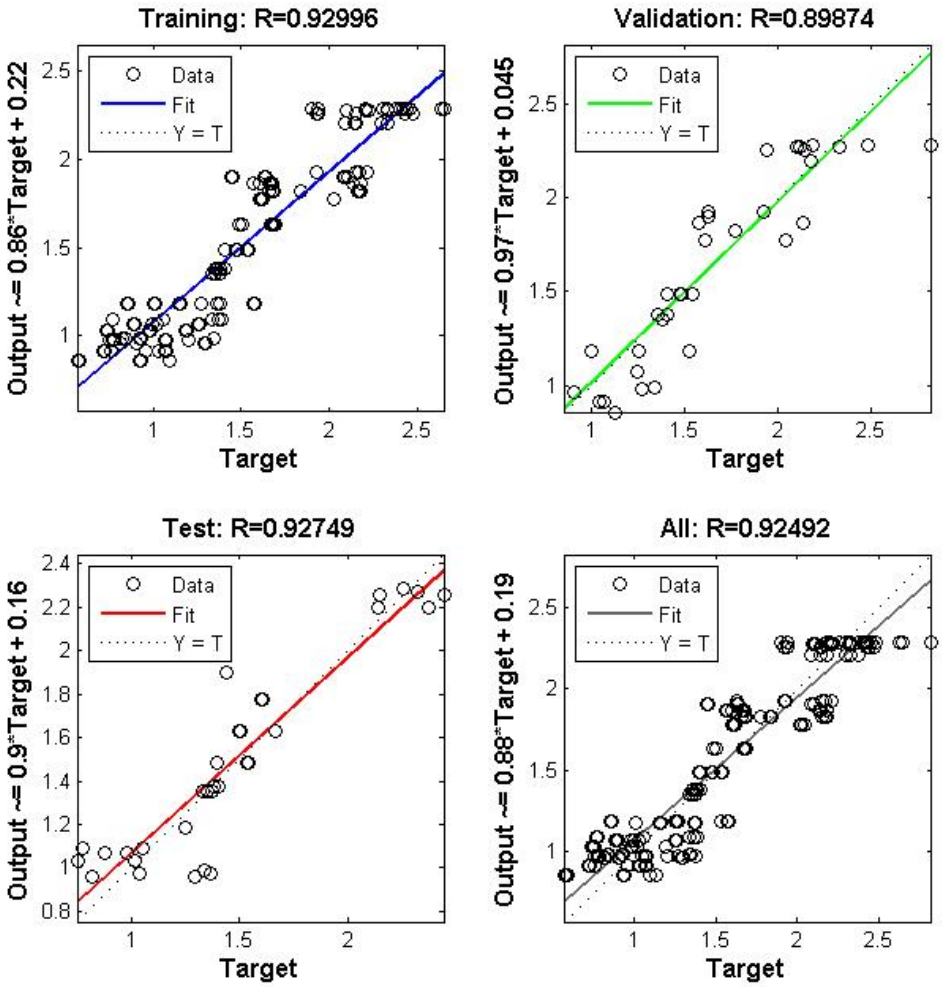


Figure 7. Performance of ANN at the age of 100 days (251 data-sets).

Similar performance of the ANN was obtained for the age of 140 days after using the new data for training

4. Discussion

The fluctuations in the measured data during the first 28 days could be attributed to the fact that the samples were stored in water. They were taken out of the water 12 h before the measurements were conducted in order to ensure a dry surface. However, the water was still present inside the specimens and was coupled with the ongoing chemical reactions due to the hydration of cement, which may have influenced the results. This could also be seen in Table 2, where quite significant values of the standard deviation were obtained compared to the mean value. The same trend in the scattering of the results was observed at 5 weeks (35 days of curing), as shown in Figure 8.

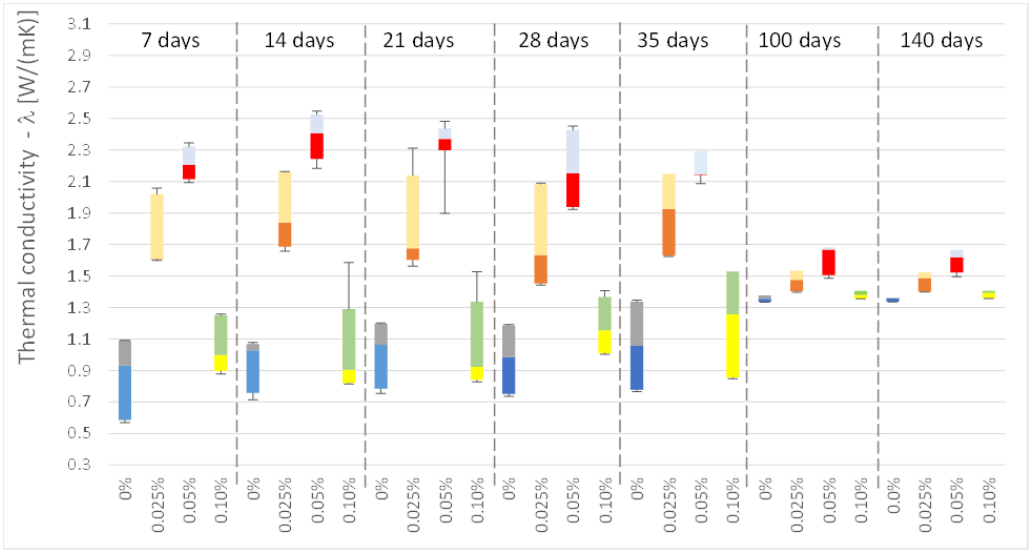


Figure 8. Boxplot for measured data.

Once the samples were taken out of the water and stored at room temperature and a relative air humidity of 50–60%, the scattering of the results was significantly reduced. The slowing down of the chemical reactions inside the specimens after 28 days could also be an influencing factor. This can be seen from the values of the R^2 coefficient at 35 days compared to the values from 28 days.

The main parameters of the research were the amount of carbon nano-material and the curing age and their influence on the values of the thermal conductivity. Other influencing parameters such as density, porosity, temperature and humidity were not considered at this stage of the research.

As seen from Figure 3, the variation of density between mortar mixes was very small and, therefore, its influence on the thermal conductivity would be negligible. Temperature is another influencing factor for thermal conductivity as highlighted in [31]. However, all specimens used in the present study were kept in the same conditions, temperature and humidity in order to even out their influence on the results among the various mix proportions.

Porosity is another influencing factor on the values of thermal conductivity. Higher porosity results in lower values of thermal conductivity [33,35]. The use of MWCNTs, even in very small percentages by mass of cement, resulted in a denser structure of the mortar and, consequently, decreased porosity [36,37] compared to reference mixes without MWCNTs. Hence, CBN mixes would exhibit higher values for thermal conductivity compared to the reference mix. In a recent study by Francioso et al. [35], it was proven that the values of the thermal conductivity change with the change in moisture content both for recycled aggregate mortar and natural aggregate mortar. Earlier research works reported an increase in the value of the thermal conductivity with the moisture content by up to 200% [38].

Based on the data presented in the scientific literature, the experimental program was devised in such a way so that to reduce, as much as possible, the influence of temperature, density, porosity and humidity on the thermal conductivity of investigated mortar mixes. This would help assess the influence of using the novel carbon-based nano-material on the thermal conductivity as well as the curing age.

The presence of CBNs clearly influences the values of the thermal conductivity of mortar, especially at low percentages of 0.025% and 0.05%wt of cement. Owing to their excellent thermal conductivity characteristics, coupled with uniform distribution within the mortar samples, they have resulted in higher values of the thermal conductivity of mortars. A 0.1%wt of cement of CBN does not seem to yield impressive results, although the obtained values are slightly higher compared to the reference mix. The fact that no surfactants or sonication procedures were used during the mixing process may have favored

the coalescence of CBNs. Further investigations by means of SEM should be conducted to confirm this assumption.

The scattering of the results at 100 and 140 days significantly decreases, with values of the coefficient of variation between 1.5% and 5.3%. The free water inside the samples may have evaporated and only the chemically bound water remained [39]. Therefore, it can be assumed that at later ages, 100 and 140 days, the thermal coefficient in the dry state was measured. Although the configuration of the adopted ANN may have seemed simplistic at first glance, the obtained values for the thermal coefficient at the ages of 28 and 35 days prove the suitability of the chosen configuration as well as the employed algorithms. The two neurons on the input layer accounted for the two main parameters of the research: the curing age and the CBN content of the specimens. There was some concern about the small number of datasets used to train the ANN, especially at early ages, which would lead to erroneous predictions. Therefore, the accuracy of the predicted results was assessed by means of the R^2 coefficient and the mean square error (MSE). The values are summarized in Table 3.

Table 3. Performance of trained ANN.

Curing Age (Days)	R^2	MSE
28	0.93275	0.0434
35	0.92239	0.0464
100	0.92492	0.0434

The change in storage conditions resulted in a sharp decrease of the thermal coefficient, irrespective of the mortar mix. The predicted results by the ANN underestimated the experimental results by 20% and, in some cases, overestimated the experimental results by 30%. Since humidity plays an important role in the thermal conductivity of a material, the experimental data followed the theoretical predictions. Retraining the ANN with the new data, in addition to previous data sets, improved the performance, and the relative error between the predicted and measured values of the thermal coefficient at the age of 140 days was 5%. The error histogram at 100 days is presented in Figure 9. It can be observed that the envelope curve is very close to a classical Gaussian distribution.

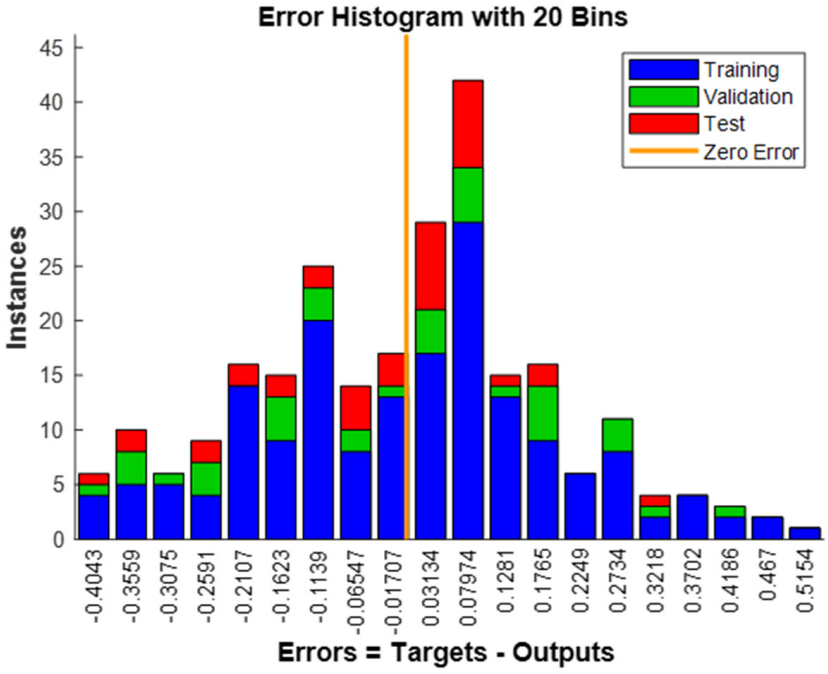


Figure 9. Error histogram using 251 datasets at 100 days.

5. Conclusions

The paper presents the results obtained from laboratory tests on the thermal conductivity of mortars made with different percentages of novel carbon-based nano-materials. A backpropagation artificial neural network with the Levenberg-Marquardt BP algorithm was used to predict the values of the thermal conductivity at different ages. Based on the obtained results, the following conclusions could be drawn:

1. The use of a novel carbon-based nano-material leads to higher initial values of thermal conductivity compared to the reference mix, even at very low percentages of 0.025%wt of cement. The carbon-based nano-material was added to the mortar mix without the use of surfactants or sonication procedure. The low scattering of the experimental results between the samples of the same mix is a good indicator of the uniform spreading of the CBN inside the mortar, especially for lower percentages. The highest benefit in terms of the values for thermal conductivity is obtained for the mix with a CBN content of 0.05%wt of cement. A higher percentage leads to a significant drop in terms of thermal conductivity. This may be attributed to the bundling effect, which should be investigated by means of SEM during the next stage of the research.
2. The change in storage conditions for the mortar samples leads to a drop in the values of thermal conductivity. The slowing down of the chemical reaction due to cement hydration coupled with evaporation of water from capillaries results in a 23–27% drop in the values of thermal conductivity for mixes with 0.025% and 0.05% CBNs. However, the thermal conductivity of the reference mix and of the mix with 0.1% CBN continues to increase.
3. A backpropagation artificial neural network with a Levenberg-Marquardt BP algorithm has been developed and is used to predict the values of the thermal conductivity. Very good predictions are made for 28 and 35 days. The change in storage conditions impacts the accuracy of the ANN and initial predictions differ by 20–30%, in some cases being underestimated and in other cases being overestimated. Retraining the ANN improves the accuracy, and only a 5% relative error is obtained for the curing ages of 100 and 140 days, irrespective of mortar mix, owing also to a significantly larger number of data sets. The ANN proves to be an efficient tool for running long term predictions on the evolution of material characteristics as long as accurate data is used for the training stage. However, in view of the future developments of the ANN to include other input parameters, additional methods will be used for cross-checking the accuracy of the predicted results, such as the Hashin–Schtrikman bounds and the Eshelby model.
4. Further research is deemed necessary in the direction of developing reliable predictions in terms of the thermal conductivity of cement-based construction materials incorporating either "traditional" nano-materials or newly developed nano-materials. The underlying phenomena that lead to the trends observed during the experimental program, e.g., the mix with a higher percentage of CBN exhibiting lower values of thermal conductivity, should be better understood and explained by using SEM and XRD analyses.
5. The use of CBNs may prove useful, especially at early ages, because the obtained mortar exhibits high values of thermal conductivity. This would ensure a more rapid dissipation of the hydration heat and may represent an alternative method to mitigate early cracking due to thermal stresses. Even at later ages, such as 140 days, the use of CBN in 0.05%wt of cement still manages to yield a 18.4% higher value of thermal conductivity compared to the reference mix. These higher values of thermal conductivity may prove advantageous in case of fire hazards as it would help dissipate the heat over a much wider area and, therefore, decrease the local temperature and prevent degradations in terms of load bearing capacity of the elements. In normal, day-to-day applications, the benefit is, however, in significant increases in the values of the mechanical properties, as reported in the scientific literature for MWCNTs. The main advantage of the new CBN relies on the fact that it does not need surfactants to prevent

coalescence, and neither does it need sonication to ensure uniform dispersion prior to mixing. Further in-depth investigations will be conducted to provide additional information in this direction.

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