ISSN 1392–1320 MATERIALS SCIENCE (MEDŽIAGOTYRA). Vol. 21, No. 3. 2015

Moisture Dependent Thermal Properties of Hydrophilic Mineral Wool: Application of the Effective Media Theory

Iñigo ANTEPARA, Lukáš FIALA, Zbyšek PAVLÍK [*], Robert ČERNÝ

Czech Technical University in Prague, Faculty of Civil Engineering, Department of Materials Engineering and Chemistry, Thákurova 7, 166 29 Prague 6, Czech Republic

http://dx.doi.org/10.5755/j01.ms.21.3.7334

Received 15 June 2014; accepted 19 September 2014

Thermal properties of mineral wool based materials appear to be of high importance because the most widespread practice is using them as thermal insulation boards. Their thermal conductivity is easily found, i.e. data sheets from producers, often including specific heat capacity, but usually only characteristic values for dry state. Exposure to outside climate or any other environment containing moisture can negatively affect the thermal insulation properties of the mineral wool. This is why the presence of water inside the mineral wool is undesirable for the majority of applications; so, they are often provided with hydrophobic substances, whereas hydrophilic additives are seldom used. However, the later combination has good potential for some applications (i.e. desalination of masonries and green roofs). In those cases, mineral wool will work with a certain moisture content, which will change the thermal properties it had in the dry state. On this account, moisture dependent thermal properties (thermal conductivity and specific heat capacity) of hydrophilic mineral wool (HMW) are studied in a wide range of moisture content using impulse technique. The experimentally determined thermal conductivity data are analysed using a several homogenization formulas based on the effective media theory. In terms of homogenization, a porous material is considered as a mixture of two or three phases: solid (basalt fibres) and gaseous (air) phase for the dry state, adding the liquid phase (water) when moisten. The homogenization techniques are first applied to calculate the thermal conductivity of the solid matrix. Then, it dependence on moisture content is evaluated using some mixing formulas. To verify the obtained results, Wiener's and Hashin-Shtrikman's bounds are used. As a summary, the application of homogenization techniques can successfully estimate measured data for a highly inhomogeneous fibrous material (i.e. mineral wool), even consuming less time.

Keywords: hydrophilic mineral wool, moisture content, thermal conductivity, specific heat capacity, homogenization techniques.

1. INTRODUCTION

Mineral wool is widely used as a thermal insulation material on buildings in the Central Europe. It is due to its very good insulating properties, such as high vapour permeability that avoids condensation problems, excellent thermal properties and easy application on the building structures [1]. Moreover, this material is gentle on environment without any known effects on human health.

Despite the fact that mineral wools are mostly used as an insulating material for building industry, further applications are being tested. Mineral wools with hydrophilic admixtures accelerate the transport of liquid water [2], which can help with desalination process in masonries or can be applied as a water reservoir in green roofs [3]. It can find use also in interior thermal insulation systems [4, 5].

Thermophysical properties of HMW in the dry state are well described by producers, but not so high attention is paid to the of thermophysical parameters of this material in moistened state. Therefore, the material properties need to be experimentally determined and presented in the full range of moisture content. The first, most time consuming possibility in thermal properties' identification is to experimentally evaluate an empirical relationship of the HMW's thermal conductivity in dependence on the moisture content. Example is described in [2]. However, it is desirable to identify the thermal properties by a less time consuming method. Homogenization techniques present a good candidate for this task.

This article deals with the measurement of basic physical and thermophysical properties of mineral wool with hydrophilic admixture and subsequently with determination of the effective thermal conductivity by means of homogenization techniques.

The thermal conductivity is an important heat transport material parameter that is in case of dry hydrophilic mineral wool very low, typically about 0.04 W/mK. The thermal conductivity of water is more than one order of magnitude higher, so the material loses its insulating properties with increasing moisture content.

Heat accumulation is represented by the specific heat capacity which can be easily calculated for the moistened HMW using a simple mixing formula. As the specific heat capacity of water is much higher than that of air, the specific heat capacity of moistened HMW increases with increasing moisture content.

The anisotropy of heat transfer was not reported yet for this type of materials, at least it was not analysed in details. Measurements differentiating between hard and soft parts of the HMW could help with better description of commercially produced boards' anisotropy.

[*] Corresponding author. Tel.: +420 224 355436; fax: +420 224 357071.
E-mail address: *zbysek.pavlik@fsv.cvut.cz* (Z. Pavlik)

2. EXPERIMENTAL

The studied HMW was manufactured by Rockwool CZ. Hydrophilic admixtures in mineral wool accelerate liquid moisture transport, and thus make new application possibilities for mineral wool products.

2.1. Basic physical properties

The matrix density ρ_m (kg/m^3) was measured by the pycnometric method, the bulk density ρ_b (kg/m^3) was determined by the gravimetric method, and the porosity ψ (-) was calculated using Eq. (1):

$$\rho_{mat} = \frac{m_0}{V(1-\psi)}, \tag{1}$$

where m_0 (kg) is the mass of a dry sample, and V (m^3) is sample volume.

The measurements of HMW basic parameters were carried out in air-conditioned laboratory at the temperature 22 ± 1 °C and 25 – 30 % of relative humidity. Each result represents an average from four or five measured values.

2.2. Thermal conductivity and specific heat capacity

The thermal conductivity and specific heat capacity measurements were performed by the heat transfer analyser Isomet model 2104 (Applied Precision). At first, the commercial HMW samples consisting of the hard and soft layers were measured as sold (see Fig. 1 for HMW board arrangement).

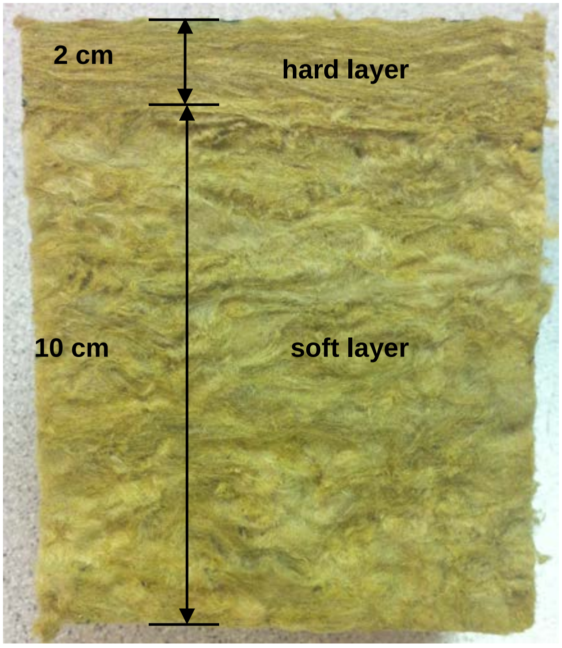

Fig. 1. Studied HMW board

Here, the measurements were done from the soft as well as from the hard side of the board. The thickness of soft layer in original HMW board is 100 mm and the thickness of hard layer is 20 mm.

Then, the samples were cut and the hard and the soft parts were measured separately respecting the orientation of mineral fibres. The hard part was measured in the dry state only.

All the measurements were carried out by using the needle probes (see Fig. 2). The probes with the most suitable thermal conductivity range were used, nominally probes with measuring range 0.015 – 0.05 W/mK for the dry state, and probes with range 0.035 – 0.2 W/mK or 0.2 – 1.0 W/mK for the moistened states. The relative standard uncertainty of the thermal conductivity measurement was 5 %, and for the specific heat capacity it was 10 %. This data is provided by Isomet producer.

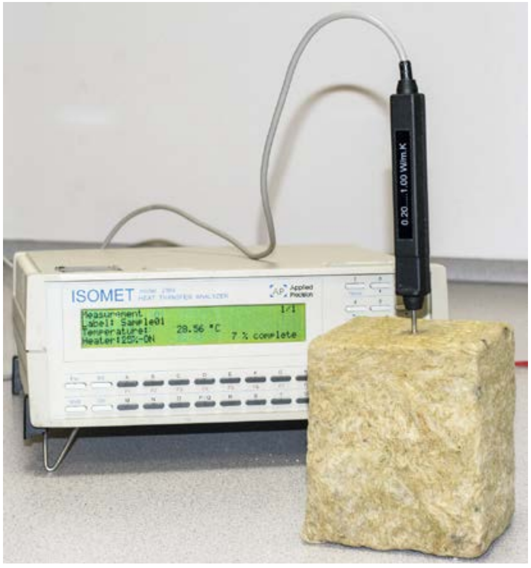

Fig. 2. Isomet 2104 and needle probe during test of a HMW sample

The thermal conductivity measurement of the HMW in dependence on moisture content was performed in the whole moisture range up to full saturation. Within the measurement, specific volume of distilled water was forced into the particular samples. Then, the samples were put into a plastic bag and kept several days until the moisture was uniformly distributed.

Due to the gravity, water was accumulating in the bottom part of the samples. Thus, before every measurement the samples were kept in the opposite position to the desired one for more than 30 minutes, and put up side down just a few seconds before the measurement.

Six different measurements were carried out in parallel as well as perpendicularly to the orientation of fibres layers, whereas three different samples were analysed and two measurements per sample were done.

2.3. Homogenization techniques: theory and models

Homogenization theory is an important science discipline that enables to determine the effective properties of heterogeneous materials. The theory is based on mixing of homogeneous phases of known properties. Each phase is described by its volume fraction and the studied property. Mostly studied physical quantities in materials engineering are the effective relative permittivity [6] as well as the effective thermal conductivity. These quantities are significantly affected by the amount of absorbed water. Therefore, the effective values are not calculated just for a dry state, but also for the different levels of water saturation.

Heterogeneous building materials in the dry state consist of two phases, solid matrix and gaseous air, that is represented by pores. When a part of pores' volume is filled by water, the third liquid water phase needs to be added to the mixture and in the case of more precise effective property identification also the fourth phase, bound water, is considered [7]. While free water phase fills the most of pores volume, bound water phase is formed by a few layers of water molecules bound to the pore walls by van der Waals forces. Thermal and dielectric properties of bound water are usually considered similar to the properties of ice.

In the practice, just a few simple models for a rough estimation of the effective properties are used. Typically, Wiener's serial model is used to predict the maximal value of effective property and Wiener's parallel model for the estimation of the minimal effective property [8]. Although the actual effective value lies between these two bounds, it is not precisely identified by these bounds. Further refinement can be achieved by an application of Hashin-Shtrikman bounds that are narrower [9]. However, the results are still not precise enough.

Therefore more precise models were developed [10]. Lichtenecker's power-law formula [11], one of the commonly used models that generalizes Wiener's bounds, can be expressed as

$$\lambda_{eff}^{k} = \sum_{j=1}^{3} f_j \lambda_j^{k}, \qquad 2)$$

where f_j (-) is the volumetric fraction of the particular phase - matrix, air, water ($f_1 + f_2 + f_3 = 1$), and λ_j (W/m K) is the thermal conductivity of a given phase. The parameter k varies within the closed interval [-1, 1], whereas $k = 0$ is excluded. The bounds of the interval correspond to the Wiener's parallel and serial model where k may be considered as an anisotropy transition from $k = -1$ (parallel) to anisotropy $k = 1$ (serial) that describes a different spatial arrangements of matrix, air and water layers.

For $k = 0$, Lichtenecker's formula is expressed as

$$\ln \lambda_{eff} = \sum_{j=1}^{3} f_j \ln \lambda_j. \qquad (3)$$

Further qualitative improvement of Lichtenecker's power-law formula can be achieved by adding the term representing the bound water. Direct generalization then leads to the 4-phase Dobson's formulation [12] that can be expressed as

$$\lambda_{eff}^{\ \beta} = w_{fw}\left(\lambda_{fw}^{\ \beta} - \lambda_a^{\ \beta}\right) + w_{bw}\left(\lambda_{bw}^{\ \beta} - \lambda_{fw}^{\ \beta}\right) + (1-\psi)\lambda_s^{\ \beta} + \psi\lambda_a^{\ \beta}, \qquad (4)$$

where w_{fw} (m^3/m^3) is the amount of free water, w_{bw} (m^3/m^3) the amount of water bonded on pore walls, λ_{bw} (W/mK) the thermal conductivity of bound water (the bound water is assumed to have the same thermal conductivity as ice, so near -20 °C it is equal to 2.4 W/mK), λ_{fw} (0.56 W/mK) the thermal conductivity of free water, λ_a (0.025 W/mK) the thermal conductivity of air, ψ (-) the total open porosity, and β (-) is an empirical parameter corresponding with the Lichtenecker's k parameter.

The 4-phase Dobson's model is appropriate to use in case of hygroscopic materials, where a significant amount of water can be adsorbed on the pore walls from the environment of high relative humidity.

3. RESULTS AND DISCUSSION

3.1. Basic physical properties of HMW

The results of basic physical parameters measurement are presented in Table 1.

Table 1. Basic physical parameters of HMW

Material	Bulk density, kg/m^3	Porosity, vol. %	Matrix density, kg/m^3 (SD %)
Commercial HMW (as sold)	95.0	-	-
Soft part of commercial HMW	79.5	96.9	2 557 (0.46 %)
Hard part of commercial HMW	172.4	93.4	2 602 (0.57 %)

The porosity matches typical values for this type of material. From the quantitative point of view, the obtained high values of porosity give studied material good thermal insulation properties and positively affect also the possible water vapour transfer through the HMW board. The matrix density corresponds with the values typical for basalt.

3.2. Thermal conductivity of basalt

For an application of homogenization techniques described in section 2.3, there was necessary to get information on the thermal conductivity of basalt fibres. The thermal conductivity of the natural basalt given in the literature is ranging from 1.5 to 4.0 W/mK, depending on material porosity. The thermal conductivity of air-dry Icelandic basalt gives an average value of 1.7 W/mK [13].

This is in agreement with 12 measurements carried out in our laboratory with the Isomet device and a surface probe on basalt paving plates. Here, we obtained the mean value of basalt thermal conductivity equal to 1.57 W/m K.

According to ISO TC 163/SC 2 [14], the thermal conductivity value of basalt having matrix density in the range from 2 700 to 3 000 kg/m^3 is considered as 3.5 W/mK, and the specific heat capacity is equal to 1 kJ/kgK. Here, the values for fully densified basalt are given what explains the higher thermal conductivity compared with above given data. In this paper, ISO value of the thermal conductivity was used for further calculations within the homogenization process.

3.3. Thermal conductivity of HMW

Table 2 collects the data from the manufacturer as well as the measured data obtained in perpendicular direction to fibres layers. The measurements of the HMW thermal conductivity in a dry state do not differ in any probe position. Table 2 also presents the average results for the measurement of the difference between HMW hard and soft parts.

Table 2. Thermal conductivity of the studied HMW

Material	Thermal Conductivity (W/mK)	
Rockwool data sheet	0.038	
Measured for original HMW board	From soft side	From hard side
	0.042	0.044
Measured for particular layers	Soft part	Hard part
	0.042	0.050

The measured data on the moisture dependent thermal conductivity of the HMW are given in Table 3. Here, the effect of fiber orientation is also presented. We can see an influence of moisture on the acceleration of heat transfer in researched materials. In this case, the materials anisotropy was not such significant, because of the dominant moisture effect on the thermal conductivity values.

Table 3. Thermal conductivity of researched HMW

HMW	MC, m^3/m^3	Direction	Thermal Conductivity (W/mK)
Soft part	Dry	Perpendicular	0.042
		Parallel	0.040
Hard part	Dry	Perpendicular	0.050
		Parallel	0.048
Soft part	0.05	Perpendicular	0.192
		Parallel	0.200
Soft part	0.25	Perpendicular	0.199
		Parallel	0.207
Soft part	0.35	Perpendicular	0.233
		Parallel	0.225
Soft part	0.5	Perpendicular	0.249
		Parallel	0.244
Soft part	0.75	Perpendicular	0.398
		Parallel	0.402
Soft part	Fully saturated (0.93)	Perpendicular	0.546
		Parallel	0.545

Looking at Fig. 3, the thermal conductivity does not follow a linear relationship until MC reaches value of 0.5 m^3/m^3. As the anisotropy effect on the measured thermal conductivity is negligible, an average of the data measured in two directions is used in the graph. The obtained values of thermal conductivity were higher than expected, whereas for higher MC a linear dependence of thermal conductivity on moisture was observed.

The differences in particular measurements were quite high and were higher as MC increased, except for the fully saturated samples. Within the measurement of thermal parameters that took usually 30 minutes, gravity is carrying some of the water to the sample bottom. This can affect negatively the reproducibility of measurement due to the changes in moisture distribution. It is also a problem for calculation of the real MC in the area of sensor placing, as the water is not uniformly distributed.

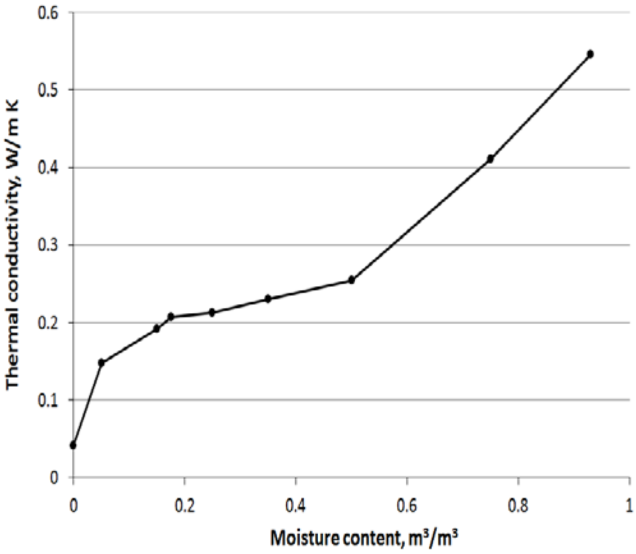

Fig. 3. Thermal Conductivity of HMW as a function of MC

Due to the production system of the HMW boards, two samples can be significantly different, even if cut from the same board. This problem we partially overcame by using three different samples for each of the measurements. However, this is another significant source of data variation.

3.4. Volumetric and specific heat capacity

The data on specific and volumetric heat capacity measured for the dried material samples is given in Table 4. In case of reference data from producer data sheet, the volumetric heat capacity was calculated from given specific heat capacity and known bulk density.

Table 4. HMW specific and volumetric heat capacity for the dry state

Material	Specific, J/kg K	Volumetric, $kJ/m^3 K$	
		Measured	Calculated
Rockwool data sheet	840	-	80
Commercial HMW (dry)	965	92	-
HMW soft (dry)	1 163	94	-
HMW hard (dry)	990	171	-

If moisture content MC (m^3/m^3) is known, specific heat capacity C_{wet} (J/m^3K) can be calculated using Eq. (5):

$$C_{wet} = \frac{\rho_{dry} \cdot C_{dry} + \rho_{water} \cdot C_{water} \cdot MC}{\rho_{dry} + \rho_{water} \cdot MC}, \tag{5}$$

where ρ_{dry} and C_{dry} are the values measured for the dry HMW, ρ_{water} and C_{water} are the values for water taken from chemical tables.

The moisture dependent volumetric heat capacity calculated using Eq. (5) is given in Table 5. Here, only the data for soft part of the original board are given. One can observe a high dependence of HMW heat capacity on moisture content. The increase of moisture content enhances the heat storage capacity of HMW due to the high volumetric heat capacity of water.

Table 5. HMW volumetric heat capacity in dependence on MC

MC, m^3/m^3	Volumetric heat capacity, kJ/m^3K
0.05	276
0.25	694
0.35	1 112
0.5	2 157
0.75	3 203
Fully saturated (0.93)	3 955

3.5. Homogenization

For determination of the HMW effective thermal conductivity dependence on moisture content, Lichtenecker's model was applied. At first, the thermal conductivities of particular material phases were identified as listed in Table 6.

Table 6. Thermal conductivity of HMW components

Phase	Thermal Conductivity (W/mK)
Solid matrix – basalt	3.5
Gaseous air	0.025
Liquid water	0.58
Bound water	2.5

The amount of bound water was determined from the measured sorption isotherm. Here, the hygroscopic moisture content of 0.000262 m^3/m^3 that corresponds to the 97 % of the environment relative humidity, was assumed to be equal to the amount of bound water.

Due to the negligible amount of bound water it is quite obvious that the Dobson's model is not suitable for the HMW thermal conductivity calculations. On this account, it was not applied in calculations of the effective thermal conductivity as function of moisture content.

Within the homogenization process, thermal conductivity function presented in Fig. 3 was divided in two subcurves. The first part of the curve consisted of the first four points, the second part of the rest of points. This approach was chosen due to the different shape of the thermal conductivity function for low and high amount of moisture content.

The results calculated by means of Lichtenecker's formula are presented in Fig. 4 and Fig. 5. The coefficient k and the thermal conductivity of matrix λ_m were identified within the optimization process that was carried out in order to find the best agreement between measured and calculated data. The optimization was performed by an application of genetic algorithms for the coefficient k in interval [-1; 1] and with respect to the value of basalt matrix thermal conductivity that was assumed equal to 3.5 W/mK. Here, λ_m was assumed in interval [3.4; 3.6] W/mK.

The best agreement of modelled data with the measured data in the first part of the curve was reached by optimized Lichtenecker's model with $k = 0.47$ and $\lambda_m = 3.4$ W/mK. Here, Root Mean Square Error (RMSE) was 0.06. Applied model overestimates the value of the thermal conductivity in the dry state. The rest of modelled points are underestimated as compared to the measured data.

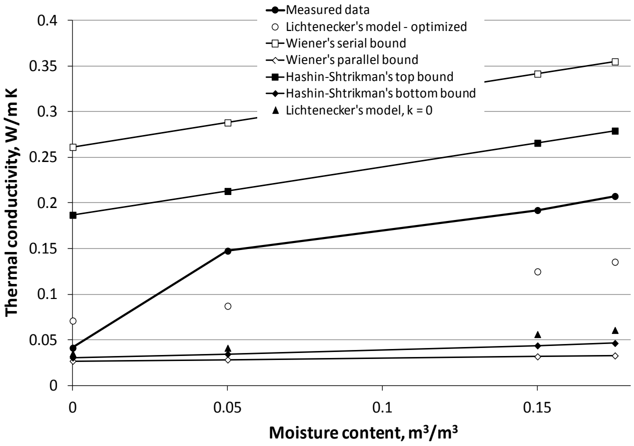

Fig. 4. The thermal conductivity dependence on MC (1st part of the curve)

The best agreement in the second part of the curve was reached by application of Lichtenecker's model with $k = 0.54$, $\lambda_m = 3.4$ W/mK, RMSE = 0.085. Optimized Lichtenecker's model overestimates values of the thermal conductivity from the volumetric moisture content greater or equal to 0.5 m^3/m^3.

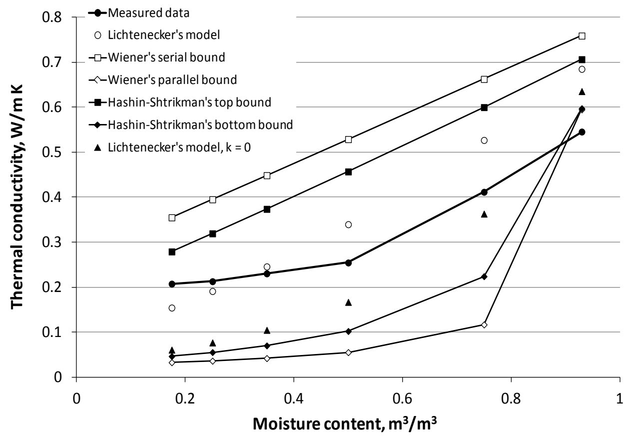

Fig. 5. The thermal conductivity dependence on MC (2nd part of the curve)

On the other hand, for lower moisture content the agreement between measured and modelled data can be considered as good in general. The thermal conductivity for the volumetric moisture content $w = 0.93$ is located out of Wiener's parallel and Hashin-Shtrikman bottom bounds, whereas the measured value is lower than expected. This finding we assign to the effect of measurement error that must be always considered in comparison of measured and calculated data.

4. CONCLUSIONS

Experimental work presented in this paper confirmed that hydrophilic mineral wool possesses good prerequisites for utilization in the building industry.

The measurement of thermal conductivity of moistened HMW gave information on the high influence of moisture on the increase of thermal conductivity values. The effect of material anisotropy on the thermal conductivity of HMW was not found statistically important.

Concerning the homogenization techniques, several models were applied to predict the thermal conductivity as a function of MC, namely Wiener's and Hashin-Shtrikman bounds, Lichtenecker's model with $k = 0$ and Lichtenecker's model with optimized parameters k and λ_m. The obtained data can be used for a rough estimation of the HMW moisture dependent thermal conductivity; the accuracy is higher than in the case of the often used Wiener's and Hashin-Shtrikman bounds. The results also indicate that more sophisticated models should be used for accurate HMW thermal conductivity prediction.

Acknowledgments

This research was supported by the European social fund within the framework of project "Support of inter-sectorial mobility and quality enhancement of research teams at Czech Technical University in Prague", project No. CZ.1.07/2.3.00/30.0034.

REFERENCES

1. **Michálek, P., Tydlitát V., Jerman M., Černý, R.** Desalination of Historical Masonry Using Hydrophilic Mineral Wool Boards *WIT Transactions on Modelling and Simulation* 46 2007: pp. 377 – 385.
2. **Jiřičková, M., Černý, R.** Effect of Hydrophilic Admixtures on Moisture and Heat Transport and Storage Parameters of Mineral Wool *Construction and Building Materials* 2006: pp. 425 – 434.
3. **Jim, C. ., Tsang, S. W.** Modeling the Heat Diffusion Process in the Abiotic Layers of Green Roofs *Energy and Buildings* 43 (6) 2011: pp. 1341 – 1350.
4. **Pavlík, Z., Černý, R.** Hygrothermal Performance Study of an Innovative Interior Thermal Insulation System *Applied Thermal Engineering* 29 (10) 2009: pp. 1941 – 1946.
5. **Pavlík, Z., Černý, R.** Experimental Assessment of Hygrothermal Performance of an Interior Thermal Insulation System using a Laboratory Technique Simulating on-site Conditions *Energy and Buildings* 40 (5) 2008: pp. 673 – 678.
6. **Sihvola, A.,** Electromagnetic Mixing Formulas and Applications. The Institution of Electrical Engineers, London, 1999. http://dx.doi.org/10.1049/PBEW047E
7. **Mnahoncakova, E., Jirickova, M., Pavlik, Z., Fiala, L., Rovnanikova, P., Bayer, P., Cerny, R.** Effect of Moisture on the Thermal Conductivity of Cementitious Composite *International Journal of Thermophysics* 27 (4) 2006: pp. 1228 – 1240.
8. **Wiener, O.** Die Theorie des Mischkorpers fur das Feld der stationaren Stromung, Abhandlungen der Mathematischen-Physischen Klasse der Königlichen Sächsischen Gesellschaft der Wissenschaften 32 1912: pp. 509 – 604 (in German).
9. **Hashin, Z., Shtrikman, S.** A Variational Approach to the Theory of the Effective Magnetic Permeability of Mul-tiphase Materials *Journal of Applied Physics* 33 1962: pp. 3125 – 3131.
10. **Pavlík, Z., Fiala, L., Vejmelková, E., Černý, R.** Application of Effective Media Theory for Determination of Thermal Properties of Hollow Bricks as Function of Moisture Content *International Journal of Thermophysics* 34 2013: pp. 894 – 908.
11. **Lichtenecker, K.** Die dielektrizitatskonstante naturlicher und kunstlicher mischkorper *Physikalische Zeitschrift* 27 1926: pp. 115 – 158 (in German).
12. **Dobson, M. C., Ulaby, F. T., Hallikainen, M. T., El-Rayes, M. A.** Microwave Dielectric Behaviour of Wet Soil. Part II: Dielectric Mixing Models *IEEE Transaction on Geoscience and Remote Sensing* 23 1985: pp. 35 – 46.
13. **Flóvenz, Ó. G., Saemundsson, K.** Heat Flow and Geothermal Processes in Iceland *Tectonophysics* 225 1993: pp. 123 – 138.
14. ISO TC 163/SC 2 Building Materials and Products – Hygrothermal properties – Tabulated design values and procedures for determining declared and design thermal values.