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19 April 2024

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Abstract. The development of novel high-performance thermal insulating products represents a key-action within the deep energy retrofit strategies on the existing building stock. In the framework of the Horizon 2020 project Wall-ACE, a highly efficient thermal insulating plaster, based on silica aerogels, has been developed. In this paper, the development process aimed at achieving a thermal conductivity lower than 0.030 W/mK is presented. Moreover, the hygrothermal characterization process aimed at assessing the data for the dynamic heat and moisture transfer (HMT) simulations, according to EN 15026:2008, is described.

Keywords: Aerogel, Plaster, Render, Hygrothermal, Thermal Conductivity.

1 Introduction

Around 83% of the buildings in EU countries were built before 1991 when poor energy saving measures were adopted. Around half of them were built before the 1960s, of which, a large part is so far non-insulated.

The thermal insulation of the building envelopes represents, therefore, one of the most effective energy-saving strategies. However, historical and technological constraints, as well as the need to save internal space, can represent a limitation for an extensive application of thermal insulation. For this reason, the development of suitable envelope retrofit solutions that can overcome the present-day barrier for a deep energy retrofit represents an essential step toward the next targets of 40% reduction of the emissions by 2030 (Paris agreement).

In the framework of the EU Horizon 2020 project Wall-ACE, a highly efficient thermal insulating plaster based on silica aerogels has been developed for the internal wall insulation.

According to the national legislation, all the retrofit solutions must be verified as far as the absence of surface and interstitial condensation is concerned, this assessment can be performed by using a simplified steady-state method based on EN ISO

13788:2012 Standard (Glaser method). Nevertheless, several studies highlighted that the application of this simplified procedure often determines an overestimation of the condensation risk [1-3]. For this reason, especially when internal insulation is adopted, dynamic coupled heat and moisture transfer (HMT) simulations, accordingly to EN 15026 Standard, are always preferable. Moreover, HMT analyses can be a valid support for designers to:

- assess the actual advantages achievable by adopting advanced thermal insulating layers in term of energy saving as well as Indoor Environmental Quality (IEQ);
- optimize the application of the insulating materials with the aim of achieving the best performance.

To perform reliable HMT simulations, a number of material's hygrothermal properties is needed. These properties have to be determined through a series of laboratory tests following measurement protocols suggested by various national/international standards.

Unfortunately, most of the materials that are under development and sometimes, even those that are already marketed, are not accompanied by such data. Indeed, only a few studies report a complete and exhaustive hygrothermal characterization [4-7] while most of the studies are mainly focused on thermal characterization.

In this framework, the aims of this paper are:

- to present the development process of a high performance aerogel-based thermal insulating plaster with a thermal conductivity below 0.030 W/mK;
- to describe the experimental protocol and the methodologies to be adopted for the material hygrothermal characterization;
- to provide guidelines and raise the awareness of manufacturers and designers that extensive dataset of physical properties should be always provided; thus an example of datasheet containing the minimum information to fully simulate the hygrothermal behaviour of materials is also shown.

2 Development of a high performance insulating plaster

One of the goals of the H2020 project Wall-ACE is to develop an internal thermal insulating plaster characterised by a thermal conductivity λ lower than 30 mW/mK (project target value) adopting the Kwark® granular aerogel as aggregate. To achieve this goal, an optimization process was pursued, starting from a series of mixtures in which a different aerogel and other mineral lightweight aggregates ratio was analysed (samples A-B-C). Since the λ-value achieved by the first set of samples was not considered satisfactory (0.06-0.09 W/mK, **Fig. 1**), in a second phase a complete replacement of the mineral lightweight aggregate with a full aerogel aggregate formulation was done. Further formulations were then developed by increasing the aerogel content to 24-25 % in mass (D-E-F). The difference in the density as well as in the thermal conductivity of the three mixtures depends on the different aerogel particle

size and distribution. Although the use of 24-25 m-% of aerogel as lightweight aggregate determine an important reduction of the thermal conductivity (down to 0.042 W/mK, **Fig. 1**) the results achieved did not fulfil the project target value of 0.030 W/mK. For this reason, a new formulation (sample G) was developed, starting from the same particle size of sample F, which had shown a better performance if compared with the previous mixtures (D-E). Limiting the aerogel content to ~50% aerogel in mass (a good compromise considering the main aspects involved, as thermal properties, mechanical properties and cost) it was possible to reach the project target value, achieving the lowest thermal conductivity of ~0.024 W/mK (**Fig. 1**) and the lowest dry-bulk density (~139 kg/m^3).
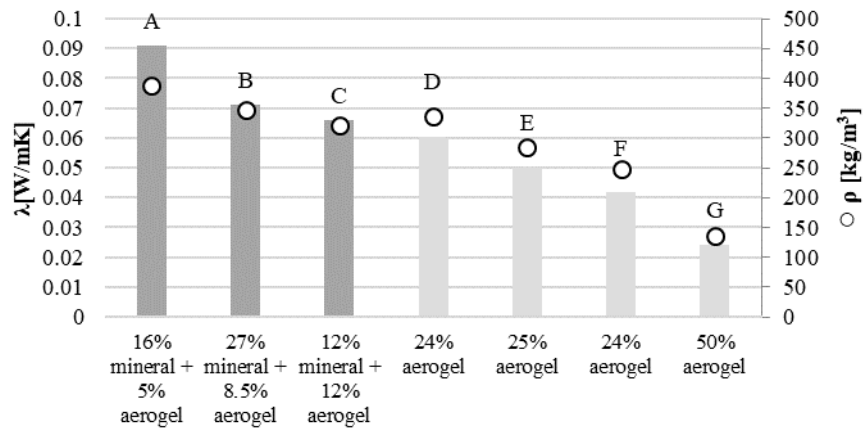
Fig. 1. Comparison of the thermal conductivity and density of the different thermal insulating plasters developed

## 3 Experimental characterization methods

Several standardized methods must be adopted to characterize the hygrothermal properties of insulating plasters; thus, in the following paragraphs, a summary of the methodologies is reported for each physical property to be measured.

The presented experimental tests allow to obtain the minimum required data to simulate the combined effect of the heat and the water vapour transmission. Nevertheless, to allow the effect of liquid water transport to be analyzed, an additional liquid water absorption test, according to EN 1015-18:2004 [11], should be performed.

3.1 Thermal conductivity

To be classified as thermal insulating plaster, the material must reach a thermal conductivity lower than 0.2 W/mK. The EN 12667:2001 standard [8] has to be followed to measure the thermal performance since the product has a medium-high thermal

resistance. The instruments that can be adopted for this purpose are the heat flux meter or the guarded hot plate apparatus. The heat flux meter (HFM, **Fig. 2a**) presents two plates that can be heated/cooled at different temperatures.
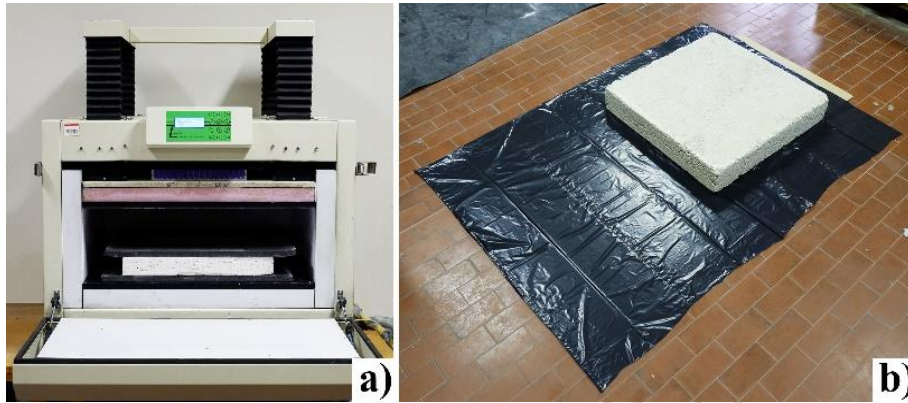
Fig. 2. a) HFM for thermal conductivity measurement; b) sealing operation of a plaster sample.

The sample size has to be in accordance with the size of the instrument adopted and its measurement area. Moreover, the standard reports the maximum and minimum thickness that the sample must have, according to the overall size and to the thermal conductivity value. In particular, the sample must have a thickness ≤ 8 times the side size (a prismatic sample of 40 x 40 x 5 cm has been adopted). In case of rigid products, as the thermal plasters, the samples must have smooth, plane and parallel surfaces. Furthermore, due to the irregular surfaces of the specimens and to reduce the contact resistance between the plates and the sample, contact sheets (e.g. rubber mats) must be placed at the interface. The surface temperature for the thermal conductivity calculation should be measured by means of external thermocouples placed between the rubber mats and the plaster specimen.

Temperature-dependent thermal conductivity.

To measure the thermal conductivity of the material, sufficient high heat flux through the samples must be generated. This can be achieved, imposing a temperature difference between the HFM plates in the range of 10 K to 50 K. In order to determine if the temperature can affect the thermal conductivity, the λ-measurements can be performed by using at least two different average temperatures between the plates (e.g. 10°C and 40°C). From the heat fluxes and the temperatures, measured by the HFM, the equivalent thermal conductivity of the specimens (eq. 1) can thus be determined:

$$\lambda = \left(\frac{s \cdot \phi}{\Delta \theta}\right) \tag{1}$$

λ: equivalent thermal conductivity [W/mK], s: sample thickness [m], φ: specific heat flux [W/m^2], Δθ temperature difference between the plates [°K].

Moisture-dependent thermal conductivity.

The moisture content can affect the thermal performance of the plasters. There-fore, the thermal conductivity measurement must be repeated starting from the dried specimens and then, step by step, moving to various cases in which the sample is tested at its equilibrium moisture content with increasing relative humidity.

For the λ measurement in dry conditions, the samples are previously dried at 105°C until the constant mass is reached. For the measurement performed on the moist material, the sample has to be conditioned in an environment at constant RH level.

Since the measurement can require several hours, before starting the test, in both dry and moist condition, the specimen must be sealed in a vapour-tight envelope to avoid any variation of the moisture content due to the migration of water vapour from the environment to the sample or vice-versa.

3.2 Specific heat capacity

The specific heat capacity measurements were performed according to the methodology reported in [9] through the adoption of a Heat Flux Meter (**Fig. 2a**).
The HFM measures the heat required to increase the temperature of the samples for a pre-set temperature difference. A step up of the temperature between an initial set point (T1, e.g. 15 °C) to the final set-point (T2, e.g. 25 °C) is applied to both the HFM plates. The HFM thus measures the heat fluxes exchanged with the lower and the upper plate during this transient phase (**Fig. 3**). The total thermal energy stored in the sample is finally obtained as the integral of the net heat fluxes:

$$H = \sum_{i=1}^{N}[\phi_{upper} + \phi_{lower}]\tau \left[\frac{J}{m^2}\right] \qquad (2)$$
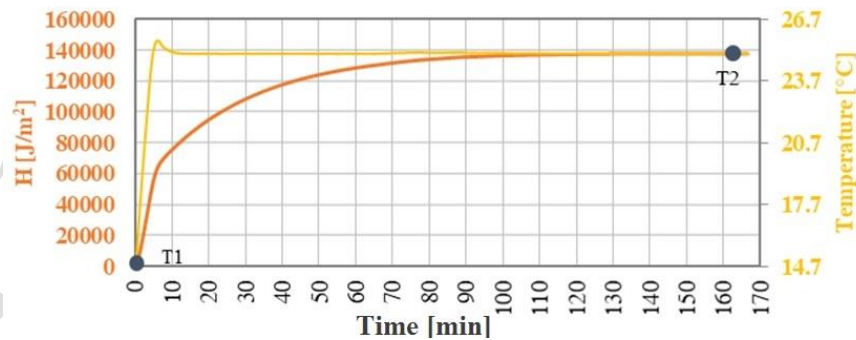

Fig. 3. The heat per unit of the area required by the sample to increase the temperature from T1 to T2;

From the H value, the specific heat, c_p [J/(kg·K)], can be calculated by means of the following equation (3):

$$c_p = \frac{H}{\Delta T \cdot s \cdot \rho} \quad \left[\frac{J}{kg \cdot K}\right]$$ (3)

Where: Δt is the difference between the first and the second set point temperature [°C]; s is the sample thickness [m]; ρ is the sample density [kg/m^3].

The specimens have to be preliminarily conditioned in an oven and then sealed in an envelope in the same way as for the λ measurement. Moreover, even for this test, the rubber mats should be adopted and placed at the interface between the instrument plates and the sample. In such a case, it is necessary a preliminary measurement of the specific heat capacity of the rubber sheets, that has to be subtracted from the value obtained for the assembly (sample + rubbers) results.

3.3 Water vapour diffusion resistance factor

The water vapour permeability test can be performed according to the wet cup test UNI EN ISO 12572:2016 [12] that allow characterizing buildings products and materials under isothermal conditions.

The samples (dimensions of 12 x 12 x 3 cm) were preliminarily stored in a controlled environment (23±1°C of temperature and 50 ± 5% of relative humidity) and has to be kept in such conditions for a time period long enough to have a variation of less than 5 % of weight over 5 consecutive weighing.

For the test, a saturated solution of water and potassium nitrate (KNO$_3$) was prepared. The aqueous solution allows reaching a relative humidity (RH) of ~94% inside the cup. The solution is then placed in the cup with a minimum depth of 15 mm. The sample must be sealed on the cup with an appropriate hydrophobic material (e.g. silicon, bee wax, aluminium tape), leaving an air gap between water and specimen of 15±5 mm. Then the specimen has to be stored in a controlled environment (23°C and 50% of relative humidity).

Fig. 4. The cup filled with the saturated solution, the tested specimen and the sealing operation;

To determine the water vapour permeability, it is necessary to calculate the average mass flow rate of the water vapour through the sample; this quantity is given by

the ratio between the mass change (Δm1,2) and the time interval during which the mass change occurred, that is:

$$\Delta\dot{m}_{1,2} = \frac{m_2 - m_1}{t_2 - t_1} \left[\frac{kg}{s}\right] \qquad (4)$$

The water vapour permeability δ of the material is calculated as:

$$\delta = d\frac{G}{A \cdot \Delta p} \left[\frac{kg}{m \cdot s \cdot Pa}\right] \qquad (5)$$

where G is the water vapour flow rate through the sample (the mean of five successive determination of $\Delta\dot{m}_{1,2}$), d is the sample thickness [m], A is the area of the specimen [m^2], and Δp is the water vapour pressure difference across specimen [Pa].

Finally, the water vapour diffusion resistance factor (μ) can be determined through eq. 6:

$$\mu = \frac{\delta_{air}}{\delta} \qquad (6)$$

where δ$_{air}$ is the water vapour permeability of air.

3.4 Hygroscopic sorption/desorption

To simulate the effect of the moisture storage in building components, the hygroscopic sorption properties must be determined according to UNI EN ISO 12571:2013 [10].

The samples must be exposed to an increasing or decreasing humidity level with a constant temperature of 23°C. When the equilibrium is reached at each humidity level, the moisture content (mass by mass, u) can be assessed. By repeatedly increasing or decreasing (step by step) the relative humidity, different values of u can be assessed and, finally, the adsorption or desorption isotherm curve can be obtained by plotting the equilibrium moisture content value against the relative humidity.

The specimen has to be representative of the material and, if the density is less than 300 kg/m^3, shall have an exposed area of almost 100 x 100 mm.

The adsorption curve measurement can be performed by using a climatic chamber. For this measurement, a preliminary drying phase of the specimens is required. The duration of this phase has to be prolonged until a constant mass is reached (according to ISO 12570). Then the specimen must be placed in the climatic chamber, and the humidity level shall be set at the lower level selected for the test. A minimum of four RH levels (varying between 30% and 95%) is required. The mass is considered constant when the change in mass between three consecutive weighings is less than 0.1% of the overall sample mass. When the constant mass is reached, it is thus possible to switch to the next and higher RH level. In the case of desorption curve, the first measurement point has to start with the higher relative humidity value (i.e. 95% or sample in free water saturation condition) and then switching to a

lower value until 35% of RH is reached. The equilibrium moisture content for every relative humidity level (u) is calculated according to equation (7):

$$u = \frac{m - m_0}{m_0} \left[\frac{kg}{kg}\right]$$ (7)

After the collection of at least 4 points at different RH value, the adsorption/desorption curve can be drawn plotting u value against RH, as shown in **Fig. 5b**.
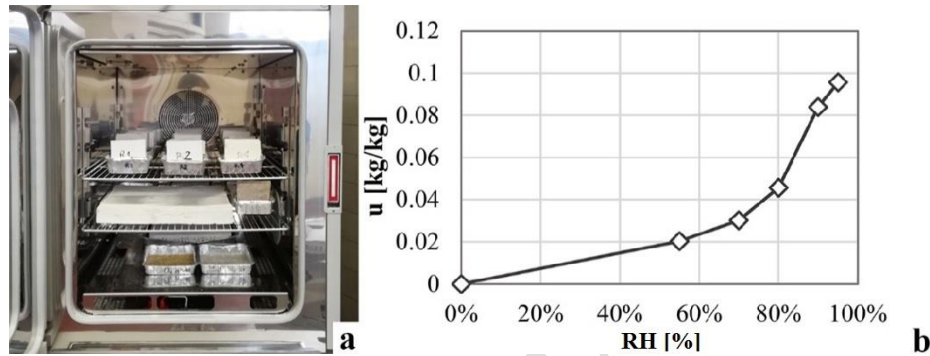
Fig. 5. a) Sample conditioned in the climatic chamber; b) an example of adsorption curve for the thermal plaster (F).

4   The hygrothermal properties datasheet

The previously reported standards allow obtaining an exhaustive overview of the hygrothermal properties of thermal insulating plaster. Generally, all the Heat and Moisture Transfer simulation software (e.g. WUFI® [13], Delphin [14]) requires the definition of all these properties for an accurate hygrothermal simulation. In **Fig. 6**, an example of technical datasheet is reported with the aim of simplifying the filling phase on the software schedule and also to have a summarized, but a complete overview, of the material properties.
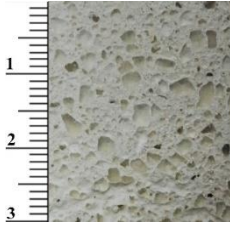
Plaster F				
General information				
Binders		Mineral, < 0.5% organic		
Aggregates		Aerogel		
Other compound		(e.g. fibres)		
Water ratio		1:1		
Application		Internal		
Other				

Technical data				
	Value		**u.m.**	
ρ_{dry}	337		kg/m^3	
$\lambda_{10,dry}$	0.060		W/mK	
cp	988		J/kgK	
μ	8		-	
C_m	n.a.		$kg/(m^2min^{0.5})$	
Thermal conductivity as function of temperature [a]				
$T_{average}$ [°C]	10	25	40	…
λ [W/mK]	0.060	0.061	0.062	..
Thermal conductivity as function of moisture content [a]				
m.c. $[kg/m^3]$	0	…	…	240
λ [W/mK]	0.060	…	…	0.176
Adsorption curve [b]				
R.H. [%]	55%	70%	80%	90%	95%
u [kg/kg]	0.0203	0.030	0.046	0.084	0.096
[a] At least 2 point are required;			[b] At least 4 point are required		

Fig. 6. Example of a datasheet for material hygrothermal simulation.

5   Conclusions

The study shows the development process of a new high-performance aerogel-based internal plaster and a synthesis of the tests to be performed to obtain a comprehensive hygrothermal characterization of the materials, that is useful to perform heat and moisture transfer simulations.

Increasing the aerogel content to a 50% (in mass) allows reaching the project target value ($\lambda \le 0.03$ W/mK), with thermal conductivity of ~ 0.024 W/mK.

Furthermore, moisture and temperature-dependent thermal conductivity, specific heat capacity, hygroscopic sorption and water vapour diffusion resistance factor were identified, as the required properties, to perform accurate heat and moisture transfer simulations. For each identified hygrothermal property, the associated test procedure that was adopted for the characterization of the newly developed products was described.

Finally, an example of datasheet containing the minimum required information to fully simulate the hygrothermal behaviour of building insulating materials is reported.

Acknowledgments

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