

AN EFFICIENCY STUDY OF FOAMED POLYISOCYANURATE (PIR) MATERIALS AS BUILDING INSULATORS

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Polymeric foams are one of the most efficient thermal insulation materials because of the extra low thermal conductivity blowing agent gases trapped inside of the closed porous structures. Thermal conductivity is one of the most exclusive properties of foamed polyisocyanurate (PIR) materials. The blowing agent gases are selected based upon their characteristics of low thermal conductivity and slow diffusion rates through the foam polymers. The atmospheric gases have a greater thermal conductivity and are typically much smaller molecules with much faster diffusion rates through the foam. PIR gains much of its thermal resistance value from the blowing agents, often a pentane gas mixture, which is trapped in the foam cells. Pentane isomers are commonly used in Europe in manufacturing rigid insulating polyurethane foams. Since the thermal conductivities of the pentanes (between 0.010 and 0.014 W/(m·K)) fall significantly below that of air, polyurethane insulation panels may achieve thermal conductivity as low as 0.024 W/(m·K). This paper analyses results of the thermal conductivity study and measurements according to two different parameters: the initial value of thermal conductivity and the rate of aging, i.e., the rate of increase in thermal conductivity over time. The initial thermal conductivity value is influenced by the thermal conductivity of the gas inside the cell and the average diameter of the cell.

Keywords: *Blowing agents, insulation materials, polyisocyanurate (PIR) materials, PIR aging, thermal conductivity.*

1. INTRODUCTION

Energy efficient renovation of public and private buildings is an important part of the European Green Deal, and its key initiative is to significantly increase energy efficiency in real estate sector throughout the European Union (EU). As an element of the European Green Deal, the building renovation initiative aims at increasing energy performance and efficiency in up to 35 million buildings by 2030, at least doubling the annual rate of energy renovations in the EU [1].

Polyurethanes (PU), a diverse group of polymers with a wide range of applications as adhesives, coatings, elastomers, and foams [2], were invented in 1937 by the German scientists, who recognised that the polyaddition of liquid polyester or polyether diols with liquid diisocyanates yielded products that were superior to existing polyolefin plastics [3]. In 1954, the accidental introduction of water to the reaction mixture was shown to produce flexible foams. Later, this chemistry was modified to produce rigid foams and elastomers. Today, several types of “polyols” and “polyisocyanates” are used to produce polyurethane materials that yield varying properties.

In 1967, the class of materials known as urethane modified polyisocyanurate (PIR) foams was introduced. These compounds are essentially an improvement on PU insulations, offering improved thermal stability, flame resistance, chemical resistance, and dimensional stability [4]. During PIR production, the polyol and polyisocyanate reaction takes place at higher temperatures when compared to PU production. This allows for excess isocyanate

to react with itself in what is called trimerization, producing strong chains of isocyanurate crosslinks. These crosslinks are stronger than normal PU bonds. Therefore, they are more difficult to break, resulting in improved properties.

During the manufacture of PIR insulation, the ratio of polyisocyanate to polyol (index) helps determine the final properties of the foam, and, therefore, plays a key role in determining the foam application. The low-index foam insulations exhibit behaviour closely resembling urethanes, since urethanes are produced at close to a 1/1 polyisocyanate/polyol ratio. As this ratio is increased, the trimerization reaction occurs, and the improved properties associated with PIR insulations become evident [5]. Although all these properties are important for certain applications, the improvements in dimensional stability, flame resistance, and thermal stability provide the biggest fundamental differences between PU and PIR foams [6], [7].

Polyisocyanurate insulations are produced in bunstock form, either continuously or individually box poured. Because of better consistency, performance, and quality, PIR insulation produced via the continuous process is usually preferred. These large, rectangular buns are then fabricated into various shapes, including flat boards and pipe shells, which are typically 3 to 4 feet in length. These pipe shells are designed to fit directly over nominal pipe size (NPS) pipe and tubing. Complex shapes can be fabricated to fit tightly around valves, fittings, and other equipment.

2. MATERIALS AND METHODS

The understanding of the cellular microstructure of foams is important for their thermal and mechanical performance. In fact, the cell geometry and cell parameters can be used to study heat transfer and gas permeability through analytical models. The earliest analysis of foam cellular microstructure using microscopic techniques was performed in the 1980s to observe the cellular structure of polyurethanes. The purpose of the imaging was to develop an analytical model to predict the diffusion coefficients of polyure-

thane gasses during the foam aging [8].

The imaging of foams was also done to compare alternative blowing agents when CFCs were being phased out. Spray and laminated polyurethane foams prepared with various blowing agents were imaged using optical microscopy to perform their cellular characterisation. Samples were analysed to determine the distribution of cell areas, aspect ratio, cell wall thickness, and cell strut thickness.

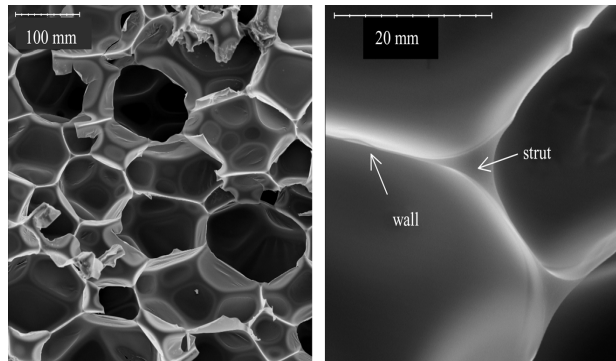


Fig. 1. Scanning electron microscope imaging of PIR foam cells [9].

PIR is a thermoset polymeric material that combines isocyanates and polyols. The manufacturing process creates a cross-linking chemical reaction that results in a robust, rigid material that does not

melt when heated. By comparison, a thermoplastic material, such as polystyrene or polyethylene, will soften and become liquid on heating.

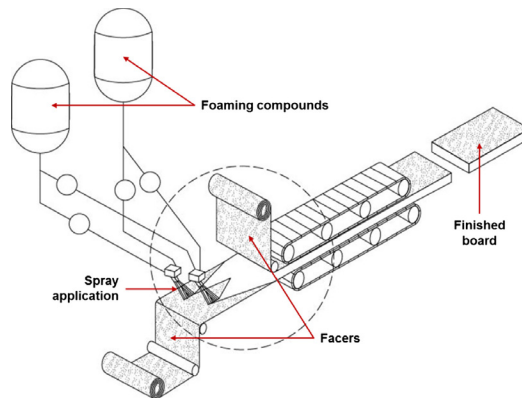


Fig. 3. PIR board production process [10].

PIR is mainly produced in the form of panels – coated panels are used in construction as thermal insulation of partitions; they can be used in refrigerators, fuel pipelines and elsewhere. PIR coatings are used to stabilise the thermal resistance of a product and to slow down the drift of thermal

conductivity. In the corners of the building where these products need to join, the coatings can be arranged in the direction of heat flow, and heat transfer can be increased by thermal bridges formed in the corner of the building wall, which should be discussed further.



(a)



(b)



(c)



(d)

Fig. 4. PIR facing types: (a) aluminium foil; (b) composite paper foil; (c) multilayer aluminized facing; (d) bitumen facing.

2.1. Analysis of Heat Transfer through Materials

In porous foams, such as PIR, heat is transferred through a solid material by conductivity, through cells by radiation, conductivity and convection, which can be described by the following formula:

$$\lambda = \lambda_p + \lambda_g + \lambda_c + \lambda_r, \quad (1)$$

where λ_p – thermal conductivity through solid;
 λ_g – thermal conductivity in gas;
 λ_c – convection in cells;
 λ_r – radiation through cell walls and cell itself.

When the cell size is small, heat transfer by convection through the cells is negligible, i.e., $\lambda_c = 0$.

In the production of PIR, many small, closed cells are formed, which means that the foaming agent evaporated during the foaming reaction fills these small cells as a gas. It is proposed that the total effective thermal conductivity of porous foam should have the following four inputs: thermal conductivity through solids, thermal conductivity through gas, thermal convection in cells, and thermal radiation through the cell walls and through the cells themselves. Thermal conductivity of PUR foam is tended to increase during the first 15 years after production, which is shown in Fig. 5.

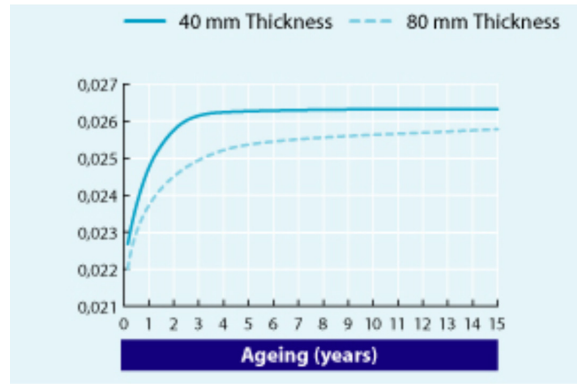


Fig. 5. Increase in thermal conductivity (W/m.K) of PUR foam during the first 15 years after production [11].

In addition to the diffusion, the blowing agent and the infusion of air, condensation affects the thermal conductivity of foams, as it creates the presence of a liquid phase and a reduction in the blowing pressure. Spray foams are prepared under high heat and high-pressure conditions.

The high vapour pressure of the blowing agent increases the total pressure inside the cell leading to expansion and initially prevents condensation risks. However, for closed cell foams, the following risks of condensation of the blowing agent result in an increase in the thermal conductivity. The impact of the condensation process on the thermal conductivity was described as a process that occurs in three regions: the first region is where the thermal conductivity decreases with the temperature up to the boiling point of the cell gas mixture; the second region is where the thermal conductivity increases as temperature decreases because the gaseous blowing agent condenses into its liquid phase; and the third region shows again a reducing thermal conductivity with the decrease of temperature, coherently with the kinetic gas theory.

Over time, when the blowing agents are absorbed into the solid polymer or escape from the material, the thermal conductivity of foams increases. The absorption of the

blowing agents into the polymer cell wall is dictated by the Fick's law, and depends on the effective diffusion constant, gas concentration and time. In particular, the diffusion constant is dictated by the ratio of the gas permeation rate and solubility of the gas on the polymer surface.

When placed in the building envelope of each specific building, thermal insulating materials are exposed to temperature and humidity levels, and their actual thermal performance differs from that predicted under standard laboratory conditions. Thermal conductivity is often reported to decrease at low temperatures, although this aspect is rarely considered. However, this is usually beneficial, as it increases heat savings. Although studies show that most fibrous insulation materials have a linear relationship between their thermal conductivity and operating temperature, for some foam materials this relationship is nonlinear and difficult to predict because they have shown increased thermal conductivity at low temperatures [12].

Over time, the thermal properties of polymeric materials deteriorate as atmospheric gases rapidly penetrate the cells and expel foaming gases from them, causing the so-called aging. Finally, the foaming gas is replaced by air and the thermal efficiency

of the foam reaches a steady state, so only aged thermal conductivity values should be used [13].

The aging process takes place in two stages. During the first stage, the gas composition in the pores changes very rapidly due to diffusion. This stage of aging is called primary, and when atmospheric gas diffusion stops, the thermal conductivity changes more slowly and this stage is called secondary.

The thermal conductivity dependence on temperature is close to linear for most

of insulation materials: it increases with increasing material temperature; however, the thermal conductivity of polyisocyanurate (PIR) thermal insulation material increases in the lower temperature range. The thermal conductivity of PIR also increases over the aging process, especially during the later service life, which is not covered by the standard aging procedures used to determine the declared thermal conductivity coefficient values for this material [14].

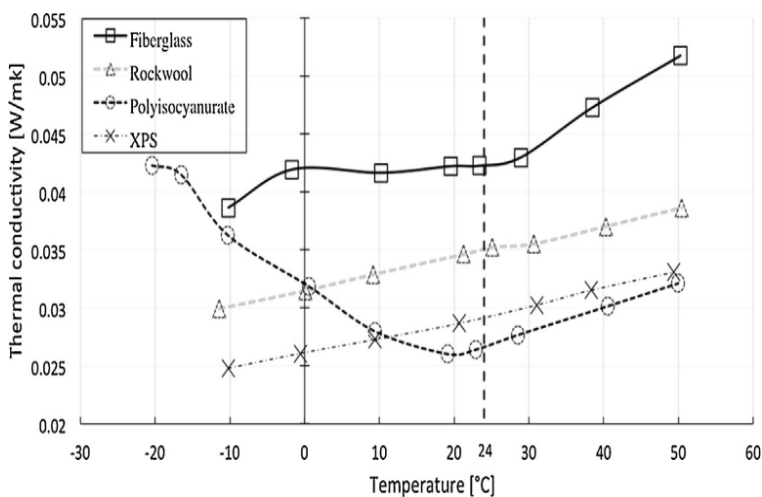


Fig. 6. Thermal conductivity dependence on temperature of thermal insulation materials [15].

2.2. Analysis of Standards for Determining Thermal Properties of Materials and Practical Measurements in the Laboratory

The main standard for rigid polyurethane foam (PUR/PIR) products is EN 13165:2012+A2:2016 [16], and the aged value can be determined in one of two ways according to this standard:

- the first is the thermal conductivity measured after 175 days of aging at 70 °C, with the addition of safety factor, which depends on the type of coating, the blowing agent of the foam and the thickness of the product. The safety factor may be adjusted based on a comparison

between the aged values at 23 °C and 70 °C;

- the second is the thermal conductivity measured after 21 days of exposure at 70 °C with addition of a fixed increment. Fixed increment depends mainly on the type of coating and is different for different blowing agents.

The results of thermal conductivity measurements are analysed according to two different parameters: the initial value of

thermal conductivity and the rate of aging, i.e., the rate of increase of thermal conductivity over time. The initial thermal conductivity value is influenced by the thermal conductivity of the gas inside the cell and the average diameter of the cell.

Aging procedures are performed differently depending on the standard used. According to the North American standard, accelerated aging is performed by cutting the product into thin slices, and according to the standard used in Europe, aging is performed by cutting a thin sample from the core of the product. There are some limitations in both standards: with increasing

temperature, the diffusion coefficients of all the different gases involved in the aging process do not change at the same rate and slicing does not consider the density of the PIR board, and the initial thermal conductivity is greatly influenced by cell filling gas composition and cell diameter.

When the temperature drops below the condensation point of the foaming gas, condensate forms in the cells and the value of the thermal conductivity coefficient increase due to the higher conductivity of the liquid phase compared to the gas phase of the foaming gas.

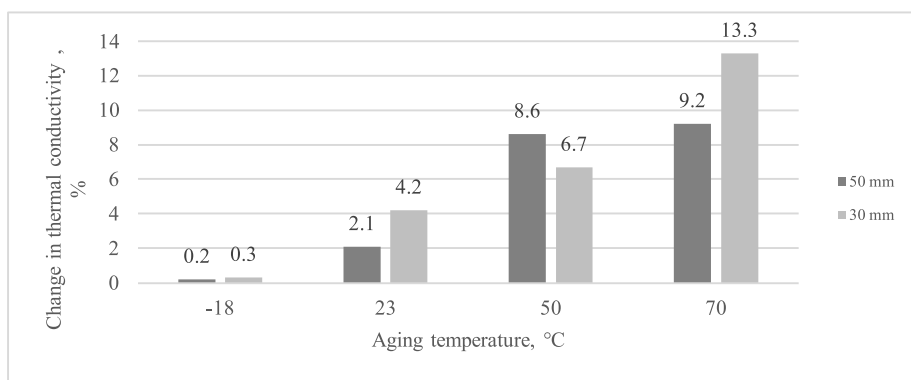


Fig. 7. Changes in thermal conductivity dependence on thickness and aging temperature.

The increase in thermal conductivity is mainly influenced by the samples stored at +70 °C. For 30 mm thick samples this change was on average 13 %, for 50 mm – about 9 %. Changes in thermal conductivity of samples stored at negative temperature were insignificant. This shows that at negative temperatures, the diffusion of the foaming gas takes place very slowly and does not change the composition of the gas to such an extent as to affect the changes in the value of thermal conductivity. For both thicknesses studied, an increase in the thermal conductivity of all types of specimens was recorded, especially in thinner specimens with removed facings. The thermal

conductivity of the thicker, covered from all sides specimens increased slightly compared to the specimens with the factory-made facing, and was lower than that of the specimens with the facings removed. This confirms the influence of the compacted surface layers on the reduced gas diffusion intensity. The differences in thermal conductivity obtained are likely to be more related to the structure of the samples or their facings than to the effects of diffusion of the foaming gas. In the case of negative temperatures, neither the facing nor the negative temperature has virtually any effect on the changes in thermal conductivity.

3. RESULTS

3.1. Analysis of the Use of Materials in Building Projects

A study was conducted to evaluate the extent to which facings can affect heat transfer through partitions, especially when the facing is made of aluminium foil. The construction of a ventilated wall used for numerical modelling is shown in Fig. 7. The thicknesses and thermal properties of the components making up the structure are given in Table 1. The heat transfer coefficient of the linear thermal bridge of the modelled wall of the building was compared with the joint of the same wall corner insulated with PIR without facings and adhesive between two boards in the corner joint. The increase in heat flow through a wall corner of 1 metre due to the influence of the facings was calculated using the given equation presented in the standard

EN ISO 10211:2017 [17]:

$$\Psi = L_{2D} - \sum_{j=1}^{N_j} U_j \cdot l_j, \text{ W/(m}\cdot\text{K)}, \quad (2)$$

where

L_{2D} – specific heat losses of the longitudinal thermal bridge, determined by calculating the two-dimensional temperature field for the component that separates two environments (inside and outside), W/(m·K);

U_j – the heat transfer coefficient calculated in the one-dimensional temperature field for component i, which separates the two environments, W/(m²·K);

l_j – the length of the two-dimensional geometric module for which the value of the heat transfer coefficient U_j is calculated, m; N_j – 1D number of components.

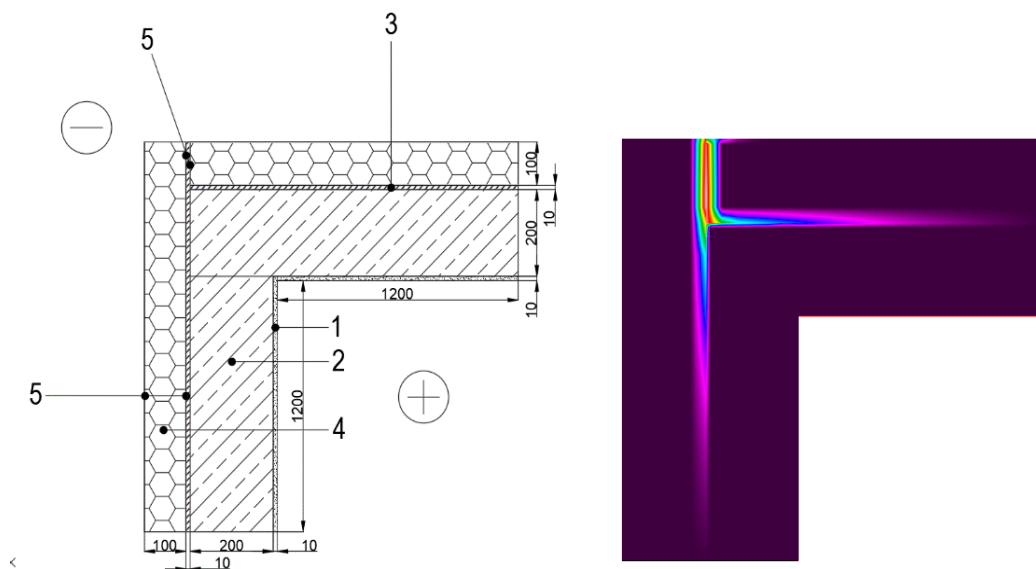


Fig. 8. Wall corner fragment (drawing and thermal imaging):

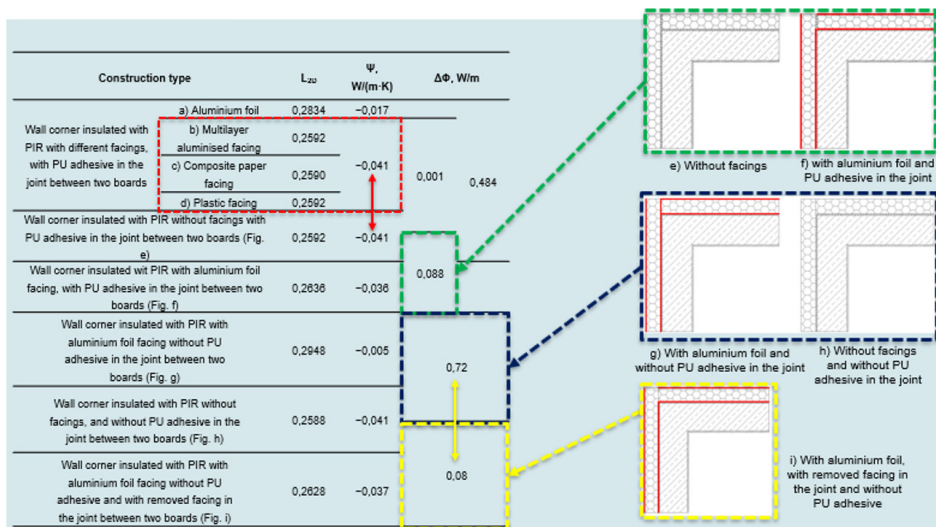
1 – plaster, 2 – aerated concrete block masonry, 3 – PU adhesive, 4 – PIR thermal insulation, 5 – PIR facing (see Table 1) (all dimensions are in millimetres).

Table 1. Materials of Wall Construction

No.	Material	Thickness, mm	Declared thermal conductivity λ , W/(m·K)
1	Plaster	10	0,9
2	Aerated concrete block masonry	200	0.13
3	PU adhesive	≤ 10	0.04
4	PIR board with facing:	100	0.022
	a) Aluminium foil	100 μm	211
	b) Aluminised multilayer facing	155 μm	0.125
	c) Composite paper facing	132 μm	0.066
	d) Plastic facing	103 μm	0.125

The value of the thermal bridge when using PIR with an aluminised multilayer, composite paper or plastic facing was the same. It was observed that wall corners with PIR without facings and with facings other than aluminium foil had the same values for thermal bridges, so it was not necessary to evaluate PIR facings if it was not aluminium foil. Another modelled wall corner structure was insulated with PIR with a removed aluminium foil facing in the joint between the boards and using a polyurethane adhesive in the joint. The difference in heat flow between these two structures can be of little significance. For comparison, another simulation was performed

when the PIR is unfaced and without adhesive in the joint. The difference in heat flow between the two options means that without the use of glue in the joint, the heat flow through the corner of the wall is increased. To reduce the heat flux to the outside that increases through the aluminium foil facing at the PIR connection, the facings on this connection must be removed. In this case, the difference in heat flow between the PIR with the aluminium foil facing removed in the joint and the adhesive in the joint, and PIR without the facing and without the adhesive in the joint is nine times smaller than the structures shown in Fig. 8.

**Fig. 9.** Heat flux in wall corners with different facings of PIR.

After determining the possible changes in the thermal conductivity of PIR in comparison with the standard value of thermal conductivity, the effect of these differences on the heat transfer of the selected wall was analysed. The selected conditions and the calculation results showed that the thermal conductivity of the PIR determined using the standard procedure met the conditions for the calculation of the seasonal heat loss and cooling energy loss of the building.

When calculating the heat loss of a building in the coldest months of the year, a lower thermal conductivity of 0.002 can be used, which would give about 8 % less heat loss through the specified enclosure. A higher value of 0.002 should be used to calculate energy losses in refrigerator buildings, which would reduce the estimated annual energy consumption for refrigeration but would not have a significant impact on actual energy consumption.

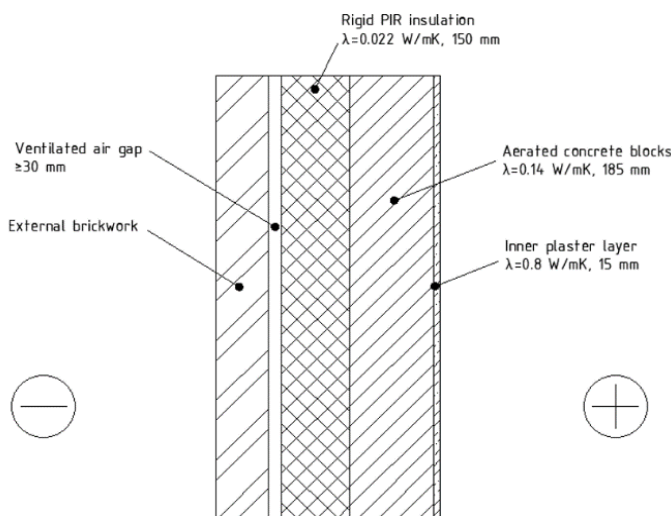


Fig. 10. A fragment of the wall of a nearly-zero energy building.

Table 2. Dependence of Wall Heat Transfer Changes on PIR Thermal Conductivity Changes

Exterior temp., °C	Interior temp., °C	Mean temp of PIR., °C	Purpose of calculation	Thermal conductivity		Heat transfer. U, W/(m ² ·K)
				Method of determination	λ, W/(m·K)	
0	+20	+10	Heating	Declared	0.022	0.120
0	+20	+10	Heating	Heat flow meter, + 10 °C	0.021	0.116
+30	+20	+25	Cooling	Heat flow meter, extrapolated	0.022	0.120
-10	+20	+5	Heating, the coldest month	“Hot box”	0.020	0.111
-5	-15	-10	Freezing	“Hot box”, extrapolated	0.024	0.129

3.2. The Use of PIR Panels to Increase the Tightness of the Building

When installing a pitched roof or a frame wall, mineral wool is often used, which is sandwiched between the rafters. In such a structure, it is necessary to install a vapour insulating layer from the inside. By

using a single layer of FF-PIR insulation from the inside, it is possible not only to form a tight and reliable vapour barrier, but also to additionally insulate the structure.

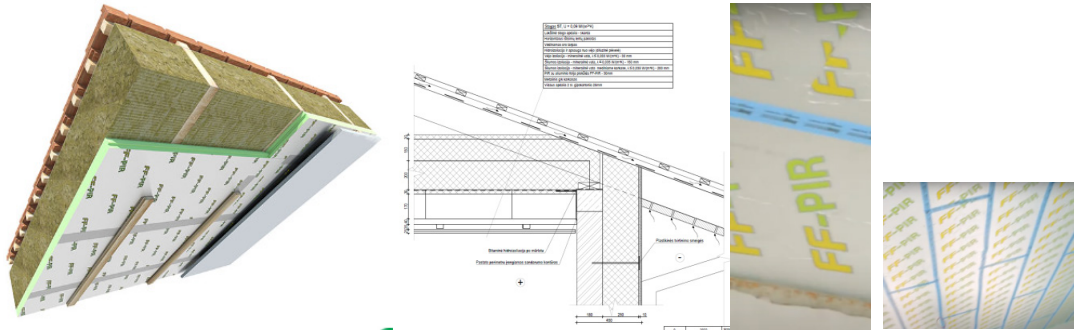


Fig. 11. Thermal insulation of ceiling and roof for improved air tightness [18].

Sandwich panels are manufactured in a wide range of thickness, with different thermal insulation cores, panel joints and profiling of the steel facings, all of which have a significant influence on the performance characteristics and the appearance of the structure.

Sandwich panels with polyisocyanurate foam (PIR) core are particularly suitable for the structures with very high thermal performance and fire safety requirements, as well as for the structures with the risk of water condensation in the thermal insulation [19].

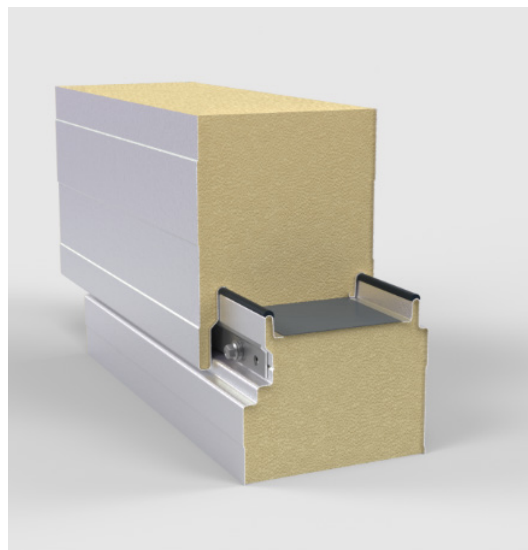


Fig. 12. Sandwich panels with polyisocyanurate foam (PIR) core [20].

4. CONCLUSIONS

1. The use of PIR products in negative temperature environments does not change their thermal conductivity measured under standard conditions, but specimens measured at low medium temperatures may increase the thermal conductivity by up to 20 % due to changes in blowing gas state: gas condenses, the distance between the molecules decreases; they form compounds which increase heat transfer between the surfaces of the material cells.
2. The largest increase in thermal conductivity of PIR products (~15 %) was found after their conditioning at +70 °C, when conditions are created for intensive diffusion of blowing gases. The thermal conductivity of the specimens with factory-made facings changed the least, and that of the thin specimens the most.
3. The standard procedure for determining the declared thermal conductivity of PIR when a specimen is cut from the core of a product does not meet the conditions for use of products with diffusion-proof facings and needs to be adjusted. The thermal conductivity of specimens aged in a high temperature environment after removal of their facings changes by more than 8 % compared to samples faced with factory-made facings. Subsequently, diffusion-proof films applied to the open sides of the specimen do not reduce the changes in thermal conductivity after aging, as they do not form such a barrier to the diffusion of blowing gases as the factory-made facings.
4. Numerical modelling and experimental measurements of wall corner joints insulated with PIR thermal insulation with contacting aluminium facing show that the increase of heat flow through the joint of walls with U value of $0.15 \text{ W}/(\text{m}^2 \cdot \text{K})$ compared to continuous PIR insulation is $0.72 \text{ W}/\text{m}$ wall height, which increases the heat loss of a medium-sized (up to 150 m^2) single-family home by 30–40 kWh per year.
5. The accuracy of the measurement of the thermal conductivity of PIR products by the heat flow meter method depends on the difference between the average ambient temperatures of the specimen and the measuring device. With a very low thermal conductivity of the material, a small heat flux is generated during the measurement, which can be distorted due to the lateral heat exchange between the environment, the specimen, and the measurement plates. When measuring the thermal conductivity of the specimens in a “hot box” with additional one or two heat flow sensors, the measurement accuracy can be increased by up to 5 % compared to the heat flow meter method.
6. The results of this research show that the changes in PIR are significantly smaller than those described in the results of previous studies. The decreasing effect of temperature on the thermal conductivity of PIR products can be explained by the continuous improvement of the material: by reducing the cell size, increasing the number of closed cells, the gas volume is further decomposed, reducing the effect of blowing gas condensation on heat transfer between cell surfaces and throughout the specimen.
7. When calculating the heat loss of a building in the coldest months of the year, it is recommended to use a revised value, i.e., $0.002 \text{ W}/(\text{m} \cdot \text{K})$ lower ther-

mal conductivity, which would give up to 8 % lower heat loss through the thermal insulation layer of the partition. A value higher than 0.002 W/(m·K) than the declared thermal conductivity of PIR products may be used to calculate energy losses in refrigerator buildings; this would reduce the estimated annual

energy consumption for refrigeration but would not have a significant impact on actual energy consumption, as heat flow through the enclosures of refrigerator buildings to the environment is significantly lower during periods of low outdoor temperatures.

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