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Résumé

Une des techniques utilisées pour déterminer le vieillissement des plastiques à alvéoles remplies de gaz (PARG) consiste à établir une corrélation entre la résistance thermique de couches minces de ce matériau et celle de panneaux pleine épaisseur. On peut utiliser à cette fin les modèles de vieillissement ou la méthode des coefficients de changement d'échelle. L'auteur édudie ici cette dernière en comparant le vieillissement de couches minces et celui de couches épaisses de PARG, et il souligne ses limites.

Scaling Factors in Aging of Gas-Filled Cellular Plastics

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ABSTRACT: One of the techniques used to determine aging of gas-filled cellular plastics (GFCP) is to correlate the thermal resistance of thin material layers with that of full thickness boards. Either models of aging or scaling factors may be used for this purpose. This paper discusses the concept of scaling factors relating aging of thin and thick layers of GFCP, and points out their limitations.

1. INTRODUCTION

EVER SINCE MEASUREMENTS of thermal resistance following exposure to elevated temperature (28 days at 100°C) were introduced to the Canadian standard for polyurethane thermal insulation, their validity has been questioned. There are two main objections: 1) elevated temperature may cause rupture of some cells beyond that actually occurring in service, and 2) the correlation of thermal resistance determined in this test and in-situ has not been adequately established [1]. Recently, the Institute for Research in Construction, National Research Council Canada, undertook a review of procedures for evaluating long-term thermal resistance of GFCP. The discriminatory character of 100°C exposure was once more shown [2]. More importantly, it became evident that for some materials use of elevated temperature may not accelerate aging at all.

To demonstrate the limitations of elevated temperature as a means of determining long-term thermal resistance, several polyisocyanurate specimens cut from two production batches were exposed to elevated temperature for 3, 7, and 12 months and stored in room conditions for 24 months. Table 1

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Table 1. Conditions of exposure of PIR specimens.

Specimen Code/Batch		Initial Exposure Time, months	Initial Exposure Temp. °C	Laboratory Exposure Time, months	Laboratory Exposure Temp. °C
A	1	3	100	21	22
B	2	7	100	17	22
C	1	12	100	12	22
D	1	3	60	21	22
E	2	3	60	17	22
F	1	12	60	12	22
G	1		N/A	24	22

provides details of exposure. Thermal resistivity of the specimens was measured periodically (Figure 1).

The initial value of thermal resistivity is, for most of the specimens, between 51 and 52 m·K/W, i.e., within 2%. While initially small, the differences in thermal resistivity of various specimens has been increased with time, and after one year is as large as 4% (44.5 to 46.5 m·K/W). These differences appear to overshadow other effects, e.g., that of elevated temperature. Except for differences in aging curves of various specimens, one can-

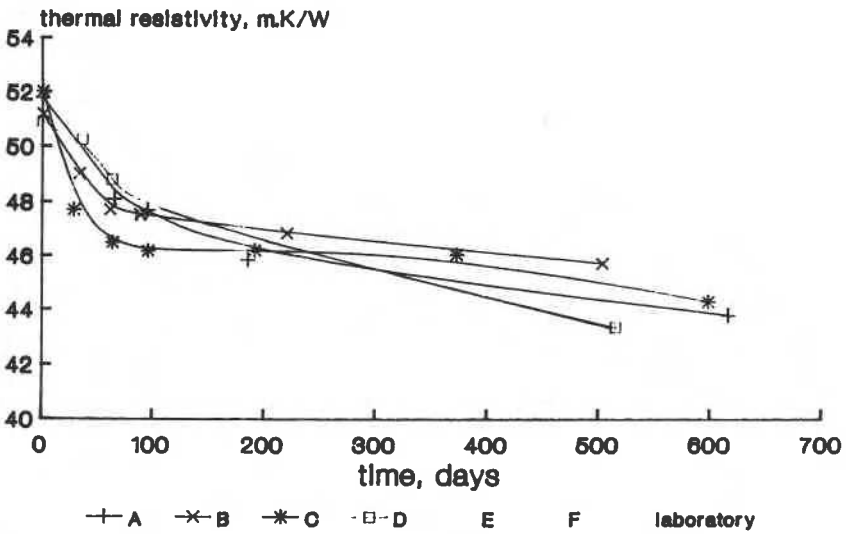

FIGURE 1. Aging of polyisocyanurate board aged for 3, 7 and 12 months at elevated temperature and later stored in the laboratory. Specimen codes are explained in Table 1.

not see in Figure 1 any trend that would reflect the accelerating effect of elevated temperature. Ironically, the highest thermal resistivities after two years are shown by specimens B and C, which were exposed to 100°C. Next highest thermal resistivity is shown by the specimens not exposed to elevated temperature but stored in the laboratory. The lowest thermal resistance values shown in Figure 1 are those of specimens E, D and F, which were exposed to 60°C. Figure 1 demonstrates that for some GFCP products the use of elevated temperature is not effective as a means of accelerating aging.

The only practical method of reducing duration of experiment is to measure aging of thin layers. Results must, however, be correlated with those of thick layers. Models of aging [3] or scaling factors [4] can be used. The latter approach has already been applied [5], but the limitations of the concept have not been discussed. To analyse the concept of scaling factors and the limitations of their use one may draw an analogy with heat and mass transfer.

2. ANALOGY BETWEEN HEAT AND GAS TRANSFER

The ingress of air into the GFCP slab or egress of blowing agent from the material may be described by the same equations as for heating or cooling of a material slab. If a slab with a uniform initial temperature T_1 has its surface temperatures lowered to level T_2 at time $t = 0$, the mean slab temperature, T_m, is as follows:

$$\frac{T_m - T_2}{T_1 - T_2} = \frac{8}{\pi^2} \sum_{n=0} \frac{1}{(2n + 1)^2} \exp\left(-Fo \cdot (2n + 1)^2 \cdot \pi^2\right) \qquad (1)$$

where

$$Fo = \frac{a \cdot t}{L^2} \text{ and } a = \frac{\lambda}{c \cdot \varrho}$$

L is slab thickness, t is time, Fo is Fourier number, a is thermal diffusion coefficient, λ is thermal conductivity coefficient, c is specific heat and ϱ is the density of the material [6].

For larger values of Fo (achieved at either longer time or a small thickness of the slab), the series in Equation (1) converges rapidly, permitting use of the first term only. Thus, Equation (1) may be simplified:

$$\frac{T_m - T_2}{T_1 - T_2} = \frac{8}{\pi^2} \exp\left(-Fo \cdot \pi^2\right) \qquad (2)$$

Equation (2) implies that at a certain value of time (for a given material thickness, L, and thermal diffusion coefficient, a, the process of cooling, or heating, of the slab becomes strictly exponential. This stage of the cooling process was designated by Kondratiev [7] "the regular regime" stage. We call it "an advanced stage" of the diffusion process, bearing in mind that the process of slab cooling constitutes a specific case of transfer phenomena governed by the theory of diffusion.

Gas transfer through closed-cell cellular plastic may also be treated as a process of diffusion [8]. In comparing heat and gas transfer phenomena, it is evident that temperature corresponds to gas pressure; thermal diffusion coefficient, a; to effective gas diffusion coefficient, D; and rate of temperature changes to rate of pressure changes.

One may analyze gas transfer phenomena with the help of Equations (1) and (2). Equation (2) indicates that logarithm of relative pressure of the gas (dimensionless) is linearly dependent on Fourier number. While the linear dependence is valid for larger values of Fourier number, Equation (1) states that the logarithm of relative gas pressure is always proportional to the Fourier number. So, to the extent in which Equation (1) describes the process of gas transfer, it may be used for "scaling" the transfer process itself, i.e., for determining the rate of diffusion on a thin specimen and relating it to that of a thick specimen. To ensure similitude of these processes, Fo must be constant. This means that to compare the diffusion rate determined for the slab with any set of properties (thickness, L, and effective diffusion coefficient, D) and the reference slab, the measured aging time must be multiplied by the scaling factor, S_f:

$$t^{\star} = t \cdot S_f \tag{3}$$

where

$$S_f = \frac{L_o^2 \cdot D}{L^2 \cdot D_o} \tag{4}$$

where L_o and D_o are the thickness and effective diffusion coefficients of the reference material layer. We restrict further discussion to a material for which one may assume that the effective diffusion coefficient remains unchanged ($D = D_o$). The scaling factor is therefore expressed as the ratio of the second power of thickness, S_L:

$$S_L = \frac{L_o^2}{L^2} \tag{5}$$

This scaling factor is used in the present paper.

3. THERMAL RESISTIVITY AS AN EXPONENTIAL FUNCTION OF TIME

If pressure changes in the advanced stage of diffusion follow an exponential curve, would thermal resistivity also follow the same type of dependence? Traditionally, laboratory measurements of thermal conductivity were presented against the logarithm of time [9–12]. This relation has never, however, been examined in a rigorous fashion. Now it can be done with the model of aging [3].

Figure 2 shows calculations of thermal resistivity performed for a 6 mm thick layer of GFCP placed in a Heat Flow Meter with a hot plate surface of 27°C, cold plate surface of 23°C, and the following effective diffusion coefficients: 0 for oxygen—0.2 × 10^{-11} m²/s for nitrogen and 0 for CFC-11. Changes in the cell-gas composition are related to only one diffusion process, nitrogen diffusion. Further, the calculations are performed with thermal conductivity of the cell-gas calculated in two ways: 1) assuming a linear dependence on molar concentration, 2) using the same equations as for the aging model [3]. The use of different approximations of thermal conduction of the gas mixture significantly affects the values of thermal resistivity, but the character of the aging curve remains the same.

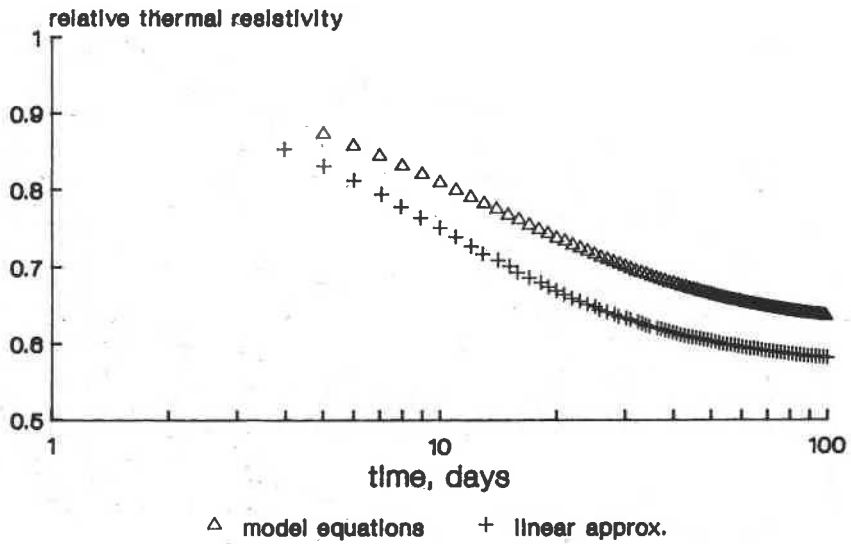

FIGURE 2. Thermal resistivity of 6-mm thick layer as affected by nitrogen diffusion only. Gas conductivity calculated in two ways: (1) as linear function of molar concentrations, (2) as in the model [3].

Figure 2 indicates that under idealized conditions, when only one gas is diffusing into a thin material layer and only the advanced stage of diffusion is considered, thermal resistivity becomes a strictly exponential function of time. Thus, although thermal resistivity will deviate from the exponential character in the initial and final stages of aging, it is on the whole beneficial to use the semi-logarithmic presentation of the experimental results.

4. USE OF SCALING FACTORS

4.1. Ideal Material

A number of curves were generated to represent aging of an ideal material [3]. The following material properties were used in the calculations: initial fraction of CFC-11 was 0.39, effective diffusion coefficients were 0.12×10^{-10} m²/s for oxygen, 0.2×10^{-11} m²/s for nitrogen and 0.1×10^{-13} m²/s for CFC-11. The density of the solid phase (polymer) was 1220 kg/m³, and the thermal conductivity of the solid matrix was 0.27 W/m·K. The density of the foam was 32 kg/m³; it had 85% of mass in the struts and only 15% in the cell-membranes. Calculated thermal resistivity values are shown in Figure 3 for three layers with thicknesses of 6, 24 and 72 mm.

To limit errors in the scaling process, a minimum Fourier number of 0.005 was selected (cf. curve II in Figure 12, page 102 of Carslaw & Jaeger [6]).

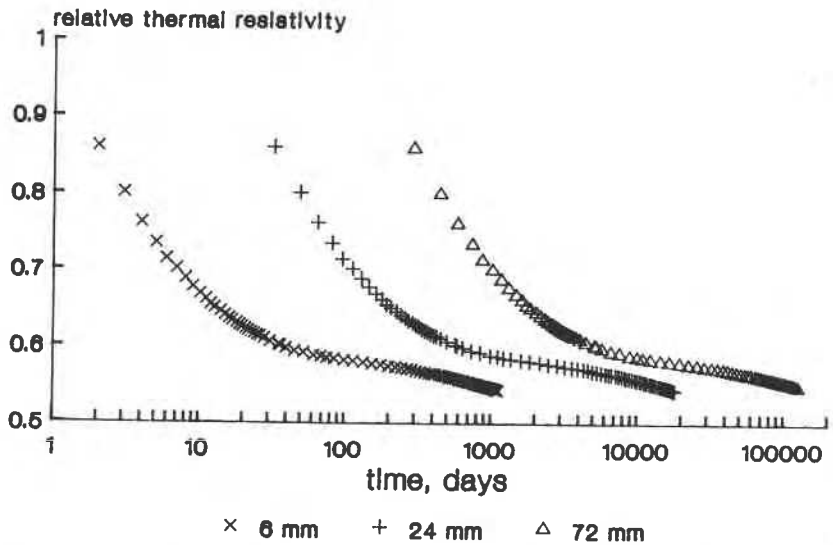

FIGURE 3. Thermal resistivity of an ideal material, calculated from the model [3] for three material thicknesses.

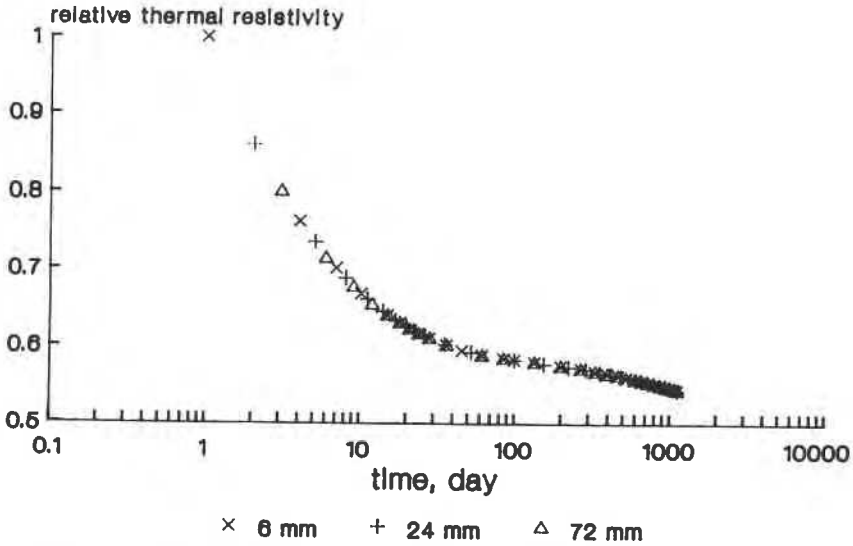

FIGURE 4. Thermal resistivity of an ideal material in relation to the logarithm of scaled time. Six-mm layer selected as reference thickness.

Using $Fo = 0.005$ and nitrogen diffusion coefficient of $0.2 = 10^{-11}$ m²/s one obtains approximately two days of aging for a 10-mm thick specimen. Thus, the first two days of aging were disregarded. The same aging curves as those shown in Figure 3 are now presented against scaled time, with a 6-mm layer used as the reference. In multiplying the actual time by the scaling factor, S_L, all layers follow the same aging curve (Figure 4). It may be seen that use of the scaling factor compensates for differences in specimen thickness.

4.2. Homogeneous and Uniform Material: Core of Extruded Polystyrene

Specimens for testing of thermal resistance were prepared in two stages. They were cut with a horizontal bandsaw, then mounted for polishing with a carborundum grinder that moves across the surface of the stationary specimen. This grinding process destroyed some of the cells, increasing the fraction of open cells in the layer adjacent to the surface. Thus, the thickness of the layer that actually participates in the diffusion process is smaller than the geometrical thickness. The destroyed surface layer (TDSL) was approximated by comparing the geometrical volume to the volume determined by means of a gas-burette (air displacement method). For extruded polystyrene, the TDSL was 0.18 mm and the actual thickness of each layer was corrected by 0.36 mm.

Figure 5 shows the aging of four layers cut from the middle of the extruded polystyrene board, using the thinnest layer as the reference for scaling factors. As explained in Figure 3, the results of measurements from the first two days of aging of the thin specimen were discarded. Thermal resistivity measured on 10, 12 and 19-mm thick specimens is shown in relation to the logarithm of scaled time. Since an extruded polystyrene sometimes exhibits small differences in initial thermal resistivity at different locations of the cross-section, it was not surprising that the average value of initial thermal resistivity differed for layers with different thicknesses. (These differences could, perhaps, be attributed to differences in the initial CFC pressure).

The slopes of each of the aging curves shown in Figure 5 are similar indicating that aging rate is practically the same for specimens with different thicknesses. Similar agreement was shown by Sandberg [5], who used scaling factors for aging of extruded polystyrene with different thickness.

4.3. Non-Uniform Material

4.3.1. Core and Surface Layers of Extruded Polystyrene Board

Another extruded polystyrene product was sliced into layers with 5- and 10-mm nominal thickness. The actual thickness of each layer was corrected by subtracting 0.36 mm. Aging curves, recalculated to 5-mm thick layers, for material cut from different locations in the cross-section are shown in

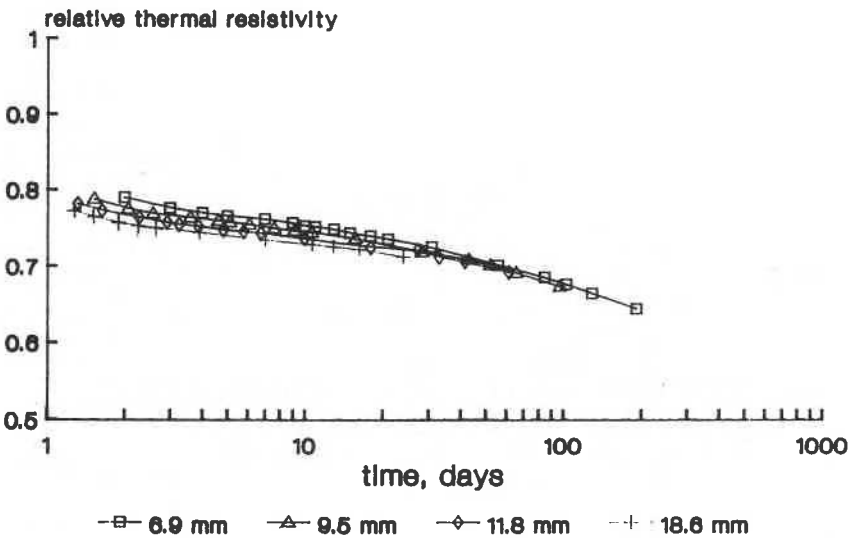

FIGURE 5. Core of extruded polystyrene, product "B", against logarithm of scaled time.

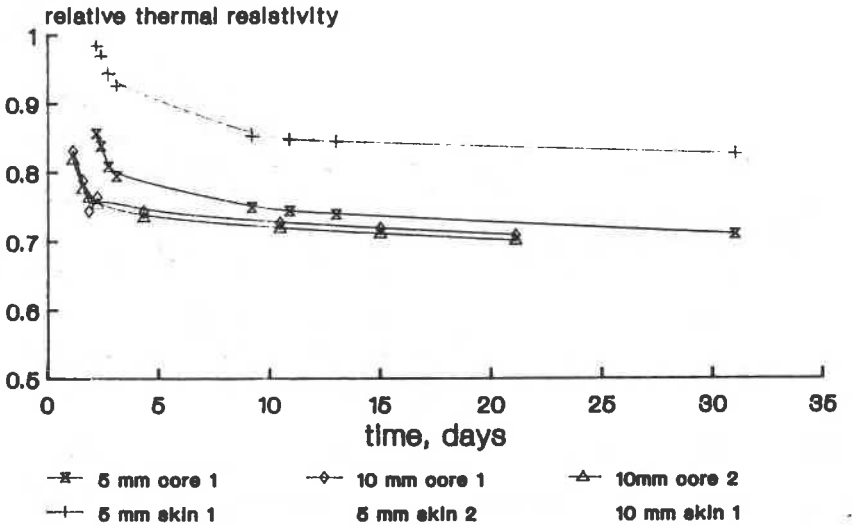

FIGURE 6. Surface and core layers of extruded polystyrene, product "A3", against logarithm of scaled time.

Figure 6. All specimens display two stages of aging, although the first stage (thermal drift) is very short.

Regardless of the large differences in thermal resistivity shown in Figure 6, there is good agreement between the rates of aging at each stage, as demonstrated by the similarity in the slopes of the aging curves. The actual values of thermal resisitivity differ, however. Whereas specimens of the same thickness show agreement when cut from one location, there are large differences for core and surface layers. This difference for core and surface layers may be associated with a density variation in the cross-section of the material. (Density change may affect both the initial CFC content and the extinction of radiative heat transfer.) The average density of the core layers was 34.8 kg/m³. The density of 5-mm thick surface layers was 39.4 kg/m³. Thicker 10-mm surface layers showed an intermediate density of 37.9 kg/m³.

4.3.2. Core and Surface Layers of Polyisocyanurate Board

Another test series comprised four 8.45-mm thick slices of PIR board after 400 days of storage under laboratory conditions; but they were cut from different locations in the cross-section, i.e., from the middle core and layers adjacent to surfaces. (The original surfaces of the board were removed before the slices were cut.)

Figure 7 shows thermal resistivity as a function of logarithm of time for the four slices. Again, thermal resistivity was expressed in dimensionless

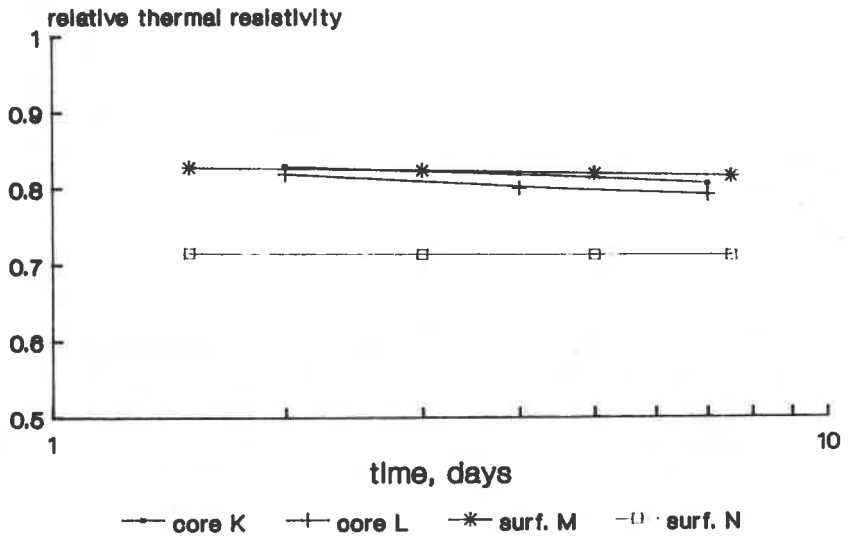

FIGURE 7. Surface and core layers of polyisocyanurate, against logarithm of time (no scaling applied).

form (fraction of the initial thermal resistivity value). Since all layers had identical thickness, no scaling was applied. Results were obtained for three specimens, two cores and one layer adjacent to the surface that showed good agreement. They follow the same aging curve. The other surface layer, however, displays a different aging curve, and after 400 days appears to have reached a plateau level. Although differences in thermal resistance of full-thickness specimens cut from this PIR board were very small (standard deviation of 1.1%) [3], one may infer a significant difference in material structure at one of the board surfaces, as documented by the difference in aging rate.

5. DISCUSSION

The scaling factor, S_f, depends on both the effective diffusion coefficients and the second power of thickness. If the aging rates of the surface and core layers are similar, the scaling factor, S_L, will depend on only the ratio of the second power of the thickness. In this case, and providing that aging history is known, scaling is easy to apply. Such cases are shown in Figures 4 and 5.

The non-uniform or layered structure of the material often prohibits use of this simple scaling factor S_L. Figure 6 shows that aging of core and surface layers of extruded polystyrene differ. Figure 7 shows the existence of a spatial variability in PIR board. One specimen from the surface layer shows thermal performance much worse than the average for the board. Results

obtained on other specimens from this surface (not reported in this paper) confirm the existence of a systematic difference between both material surfaces. Scaling factors cannot be used in these cases.

Thus, use of scaling factors is limited to homogeneous and uniform materials. As most of the currently manufactured board products are laminated and layered, more research is required to extend the application of scaling factors to layered materials. Such an extension appears to be possible as soon as methods of determining initial CFC concentration and effective diffusion characteristics of skins and core material are developed.

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