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Thermal Performance of Sprayed Polyurethane Foam Insulation with Alternative Blowing Agents

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Résumé

Afin d'évaluer la performance thermique à long terme de six mousses de polyuréthane fabriquées avec le même polymère mais des agents de gonflage différents, on s'est servi d'une méthodologie d'essais utilisant des courbes résistance thermique-temps établies à partir de minces tranches de mousse, ainsi que la technique de mise à échelle visant à déterminer le temps de vieillissement en fonction de l'épaisseur de l'échantillon. Les agents de gonflage qui ont été employés sont : CFC-11 avec 0, 0,5, 1,0 ou 1,5 % d'eau et HCFC-123 ou HCFC-141b.

Sauf dans le cas des mousses fabriquées avec CFC-11 et 1,0 ou 1,5 % d'eau, pour lesquelles la résistance thermique a été plus grande que ne peut l'être pour des mousses de polyuréthane fabriquées avec HCFC-141b, les mousses de polyuréthane fabriquées avec CFC-11 et 0, 0,5 ou 1,0 % d'eau ont affiché une résistance thermique plus grande que celle des mousses fabriquées avec HCFC-123 ou HCFC-141b.

Thermal Performance of Sprayed Polyurethane Foam Insulation with Alternative Blowing Agents

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ABSTRACT: A test methodology that uses thermal resistance-time curves determined on thin slices of the foam and the scaling technique to relate aging time to the specimen thickness, was applied to evaluate long-term thermal performance of six polyurethane foams manufactured with the same polymer but different blowing agents. The blowing agents employed were: CFC-11 with 0, 0.5, 1.0 or 1.5% water and HCFC-123 or HCFC-141b.

Except for foams manufactured with CFC-11 and 1.0% or 1.5% of water, for which material inhomogeneity was higher than can be tolerated by the proposed test methodology, polyurethane foams manufactured with CFC-11 and 0.5% water, HCFC-123 and HCFC-141b showed long-term thermal performance similar to that of CFC-11 blown materials.

INTRODUCTION

IT IS CURRENTLY believed that chlorofluorocarbons (CFCs) used as blowing agents in gas-filled cellular plastics (foams) contribute to the depletion of earth's ozone layer. Therefore, as accorded in the Montreal Protocol, foam industry should be prepared to phase out the use of these blowing agents. The industry is left with very few options. As an interim solution the amount of CFC may be reduced by mixing with water (which reacts with the isocyanate part of the system to produce carbon dioxide as an additional blowing agent). In future, however, blowing agents other than CFC must be used. Two hydrochlorofluorocarbons (HCFC), viz. HCFC-123 and HCFC-141b have been identified as alternative blowing agents for polyurethane

foam insulation. During the past few years a number of research projects were initiated [1] to evaluate the effect of blowing agent replacement on selected aspects of performance of gas-filled foams. One such project was jointly undertaken by the Society of Plastics Industry and the Institute for Research in Construction, NRC Canada [2]. The objective of this project was to evaluate long-term thermal performance of sprayed polyurethane foams produced with alternative blowing agents and compare it to that of products blown with CFC-11.

A generic polyurethane spray [3] was developed to represent current commercial foam systems and six foam products were prepared by changing only the blowing agent. The blowing agents used were: CFC-11, HCFC-123, HCFC-141b, CFC-11 with 0.5% water, CFC-11 with 1.0% water and CFC-11 with 1.5% water. The foam blown with CFC-11 alone, i.e., that representing a typical, commercial, sprayed polyurethane foam, was called "Base 88." To compare various aspects of performance of these foams, the following measurements were carried out:

1. Thermal resistance
2. Change in thermal resistance with time
3. Water vapor permeance
4. Dimensional stability
5. Compressive strength
6. Nitrogen diffusion coefficient

The findings from the above investigation were presented in detail in a report submitted to the Society of Plastics Industry [3]. One important aspect of the investigation was the application of the slicing and scaling methodology [4,5] that predicts a design thermal resistance value for any required long-term period, e.g., 15 or 25 years. This paper briefly presents this test methodology and the results of its application to foams with the six blowing agents.

THE TEST METHODOLOGY

The test methodology employed in this work comprises two different types of thermal measurements:

1. To establish mean value and variability in the initial thermal resistance of the product
2. To establish time-dependent effects in thermal resistance of the product

The measurements of the first type are performed using the standard ASTM test methods C 518 or C 177 on full boards (or thick material layers) to test a large volume of the product. The measurements of the second type are performed on thin material layers (slices) and involve testing of the same

specimen over an extended period of time to establish the dependence of thermal resistivity on the aging period. Moreover, thermal resistivity (R -values) determined on these slices as a function of time are presented in relation to the initial R -values of the material (normalized curves).

Initial Thermal Resistivity

Usually the initial thermal resistivity is established from measurements on full thickness of the product using a large (e.g., 61 cm square) Heat Flow Meter (HFM) apparatus. In this work 25 mm thick test specimens were cut from the sample and tested on an HFM apparatus using ASTM C518 test method. As this measurement reflects the variability in the manufacturing of the material, appropriate statistical techniques must be applied to ensure that the mean value of thermal resistivity is representative of the product. Moreover, care must be taken to ensure that the age and aging conditions of these specimens were similar, e.g., all initial thermal resistivity measurements were, in this work, performed on specimens between 7 and 14 days old.

Slicing and Scaling Technique

Based on the assumption that the sprayed polyurethane foam sample may be regarded as a homogeneous material, one may test thin layers of material and extrapolate results to any desired thickness. Measurements on thin specimens shorten the duration of aging tests as well as reduce the variation in material properties within the test specimen. Hence the experimental part of the methodology includes preparation of thin test specimens, measurement of their thickness and determination of the changes in thermal resistivity with time, denoted here as "the aging curve." In the present investigation, aging took place under laboratory conditions.

Then, the aging curve established on a thin specimen can be transformed into that corresponding to the required material thickness by means of the scaling technique [4]. Application of the scaling technique is based on the following argument. Aging process is known to be largely due to the flow of components of air into the test specimens and outflow of the blowing agents. It is assumed that these transport processes are represented by simple diffusion equations; i.e., rate of diffusion is directly proportional to the gradient of partial pressure for each gas under consideration. Then, if one has two homogeneous specimens of thickness L_1 and L_2 , they will reach the same level of aging at time t_1 and t_2 which are proportional to the second power of these thicknesses. Therefore,

$$\frac{t_2}{t_1} = \frac{L_2^2}{L_1^2} \quad (1)$$

The material thickness in Equation (1) represents the characteristic path of the diffusion processes and is equal to the geometrical thickness less the thickness of the open cells at the specimen surface. Thus, two new concepts must be introduced. The "thickness of the destroyed surface layer" (TDSL) denotes an equivalent thickness of the open cells at the specimen surface, and the thickness L_1 or L_2 in Equation (1) is called "the effective thickness" (ET) of the material layer.

EFFECTIVE THICKNESS OF THE MATERIAL LAYER

There is a small fraction of open cells in each "closed cell foam." This fraction may be increased during the preparation of a thin layer for testing. For this reason, the concept of TDSL was introduced [6] as the difference between geometrical and closed-cell volumes divided by the surface area of the specimen. The ET is then calculated by subtracting twice the TDSL value from the measured average specimen thickness.

REFERENCE THICKNESS

Statistical considerations require that the aging curve representing a product is established on the basis of thermal resistivity-time curves determined for a number of specimens. The thickness of each of them varies from one another. While Equation (1) may be used to recalculate the aging curves from one thickness to another, it is more convenient to use the reference thickness as the basis of comparisons. Following the international proposal initiated by Sweden [5] the reference ET is chosen to be 10 mm.

REFERENCE AGING CURVE

Though it is assumed that foam samples are homogeneous, in practice this is never achieved. Hence, the aging curves calculated for the reference thickness using experimental data on several test specimens from the same sample, instead of being a unique and well-defined curve, will be a family of curves, with some displacement from one another. One way to arrive at a reference aging curve for the sample is to average all the calculated curves for the reference ET.

In this work, however, the curve with the lowest long-term thermal resistivity was chosen as the reference aging curve. This is, as will be evident later on, to guarantee a minimum thermal performance for the sample tested. Furthermore, if one has to compare the thermal resistivity of several foam samples it is convenient to normalize the results.

NORMALIZED REFERENCE AGING CURVE

To normalize the results, each value of thermal resistivity on the selected aging curve (reference aging curve) is divided by its initial thermal resistiv-

ity. Since the R -value of a thin slice changes rapidly, for all practical purposes, the initial R -values are determined on thick specimens prior to their slicing into thin layers. Thus, while the aging curve is determined on thin slices, the initial R -value which is used for normalization of these curves is determined on a thick slab.

There are two advantages in using the normalized aging curves:

1. In showing the relative significance of the air ingress stage of the aging process [7]
2. In reducing the error due to the so-called thickness effect in measurements of thermal resistivity performed on thin and thick specimens

In effect, the normalized reference aging curve together with the mean value of the initial thermal resistivity can be used to predict long-term thermal performance for any thickness of the foam.

PREDICTION OF LONG-TERM PERFORMANCE

The primary use of the reference aging curve is to recalculate it into an aging curve for material layer with the required thickness. For example, consider a spray foam applied to a thickness of 50 mm while the aging curves were experimentally determined over a six month period on 5 mm thick slices. First, when producing the reference aging curve which is related to 10 mm thick material layer, the time scale will be extended to cover aging for two years [as it follows from Equation (1): $(10 \text{ mm}/5 \text{ mm})^2 \times 0.5 \text{ year} = 2 \text{ years}$]. The reference aging curve contains information on aging for 50 years for a 50 mm thick foam sample [as it follows from Equation (1): $(50 \text{ mm}/10 \text{ mm})^2 \times 2 \text{ years} = 50 \text{ years}$]. It means that the information contained in the reference aging curve may be used to predict the thermal resistance of this 50 mm thick material layer at any selected time or the average thermal resistance or design R -value over any selected period, e.g., 5, 10, 15, 25 or even 40 and 50 years.

The design R -value (R_d) is defined as the average thermal resistance of the material layer over a specified aging period, which for practical purposes is expressed in days. Let the aging period be m days. If R_k is the thermal resistance on k^{th} day during the aging period, integrating from the first to the last day (m), one defines R_d as follows,

$$R_d = \frac{1}{m} \int_1^m R_k \cdot dk \quad (2)$$

Experimental investigations on several specimens [6,7], have shown that

during each of two aging stages, the process of aging can be experimentally approximated by Equation (3)

$$R_k = R_1 - A \log k \quad (3)$$

where R_1 is the R -value on the first day of the aging period under consideration and A is an empirical constant. The above equation can also be expressed as

$$\frac{R_k - R_m}{R_1 - R_m} = 1 - \frac{\log(k)}{\log(m)} \quad (4)$$

where R_m is the thermal resistances on the last day of the aging period. It can be shown (see Appendix 2) that the design R -value for the aging period, as defined by Equation (2), is numerically equal to the R -value on day d selected in such a manner that

$$d = m/2.72 \quad (5)$$

For an aging period of 25 years ($m = 9125$ days), according to Equation (5), the design thermal resistance R_d is reached on day $d = 3360$, i.e., after approximately nine years. Once again, if a foam sample of thickness 50 mm is considered, the 3360 days will correspond to $\{3360 \text{ days} \times (10 \text{ mm}/50 \text{ mm})^2\} = 134$ days on the reference aging curve. Thus the thermal resistivity on 134th day shown by the reference aging curve will correspond to the design thermal resistivity of the 50 mm foam sample for a period of 25 years. This is an invaluable piece of information on the long-term performance of the foam for designers and builders.

EXPERIMENTAL METHODS AND RESULTS

Each foam was sprayed at two industrial laboratories on a plywood substratum and a sample consisting of four boards with dimensions of 1.2 m $\times$ 1.2 m $\times$ 0.15 m was produced. These samples were sent to NRCC where from each material the following test specimens were prepared:

1. 600 mm square and 25 mm thick, four specimens
2. 300 mm square and 5 mm thick, four specimens
3. 300 mm square and 10 mm thick, eight specimens

These specimens were prepared in two stages. First, slabs slightly thicker than the required thickness were cut with a horizontal band saw. Then, to ensure smooth and parallel surfaces of the test specimen, both major surfaces of these slabs were ground with a Carborundum grinder. The thickness of each specimen was determined as an average from 16 readings on a micrometer gauge.

Through volumetric measurements [6] it was shown that the average TDSL was approximately 0.2 mm for all specimens. Hence the effective thickness was obtained by subtracting 0.4 mm (for two surfaces) from the measured average thickness of the specimen.

Initial thermal resistance of each material was determined with the help of a 600 mm square heat flow meter (HFM) apparatus within a few hours of the specimen's preparation. The results are listed in Table 1.

Aging curves for thin material layers were determined with a set of four HFM apparatuses that maintained identical hot and cold plate temperatures. The four 5 mm specimens were tested separately but the eight 10 mm specimens were tested as four pairs and each set of data was treated as an average for the pair. The thermal resistances of these thin specimens were continuously monitored for the first week. After the first week these specimens were removed from the apparatus and continued to age in the laboratory, up to six months. During this period, their thermal resistances were measured at pre-selected intervals. These measured values were used to establish the aging curve for the effective (corrected with two TDSL) thickness of the specimen. Subsequently, each aging curve was transformed into that for the reference thickness using the scaling technique [see Equation (1)].

Thus for each material eight aging curves were established. These aging curves, normalized by dividing each value by the initial thermal resistivity, are shown in Figures 1 to 6. As indicated earlier, the eight experimental curves for each foam sample form a series of curves. The upper and lower boundaries are marked in the figures and the lower boundary is referred to as the reference aging curve for the foam sample; it is assumed that this reference curve represents the lowest thermal resistivity for the respective foam sample.

Experimental data presented in Figures 1 to 4 clearly show that the refer-

Table 1. Average initial thermal resistivities, r_i , measured on 25.4 mm thick specimens and reported for each industrial laboratory separately as well as average for each product.

Blowing Agent	r_i (K·m/W)		
	Lab 1	Lab 2	Average
CFC-11	65.7	63.8	64.8
CFC-123	56.8	55.9	56.4
CFC-141b	57.8	54.5	56.2
CFC-11 + 0.5% Water	60.0	57.7	58.9
CFC-11 + 1.0% Water	55.3	55.9	55.6
CFC-11 + 1.5% Water	57.8	54.5	56.2

FIGURE 1. Normalized aging curves of 5 mm and 10 mm thick slices of SPUF blown with CFC-11.

FIGURE 2. Normalized aging curves of 5 mm and 10 mm thick slices of SPUF blown with HCFC-123.

FIGURE 3. Normalized aging curves of 5 mm and 10 mm thick slices of SPUF blown with HCFC-141b.

FIGURE 4. Normalized aging curves of 5 mm and 10 mm thick slices of SPUF blown with CFC-11 + 0.5% water.

FIGURE 5. Normalized aging curves of 5 mm and 10 mm thick slices of SPUF blown with CFC-11 + 1.0% water.

FIGURE 6. Normalized aging curves of 5 mm and 10 mm thick slices of SPUF blown with CFC-11 + 1.5% water.

Table 2. Average design thermal resistivity, R_d , for an aging period of 25 years and 50 mm thick foam layer.

Blowing Agent	R_d (K·m/W)
CFC-11	43.7
CFC-123	39.5
CFC-141b	39.5
CFC-11 + 0.5% Water	41.7

ence aging curve is a characteristic of the respective material. On the other hand, the scatter of data in Figures 5 and 6 indicates that the inhomogeneity of the materials is much higher than can be tolerated by the proposed methodology. It was also noticed that the densities of the materials in Figures 5 and 6 showed significant variations.

The data obtained on materials represented by Figures 1 to 4 were used to calculate, as outlined earlier [Equations (2) to (5)], the design thermal resistivities of those materials for an aging period of 25 years and are listed in Table 2.

CONCLUDING REMARKS

It was postulated in a review [8] that the evaluation of long-term thermal resistance of cellular plastics must include the overall thermal resistance as a function of time as well as the differences in material properties, which in turn may depend on manufacturing conditions or even on the location from which the sample was taken. It was ensured, therefore, that all foams manufactured for this research project were homogeneous enough in their cross-section [3] and that the manufacturing conditions were identical for all samples tested.

These measures permitted verification of the proposed slicing and scaling methodology [4] under conditions of material production existing in the industrial R&D facilities. The differences in material properties were assessed with measurements of:

1. The initial thermal resistance measured on large specimens from material manufactured, cut and tested in a repeatable manner
2. The normalized aging curve

These measures showed that some materials were homogeneous enough to permit the application of the proposed methodology (Figures 1 to 4), while inhomogeneity of two materials shown in Figures 5–6 precluded use of this technique.

Restricting our discussion to sprayed polyurethane foam products for which the selected manufacturing conditions were appropriate, i.e., materials blown with CFC-11, HCFC-123, HCFC-141b and CFC-11 + 0.5% water, one may conclude that slicing and scaling technique appears fully applicable. The measurements were performed on 5 and 10 mm thick layers to establish that use of such thin layers does not increase test imprecision and that the TDSL is correctly determined. It appears that use of two different slice thicknesses (such as 5 and 10 mm) should be continued.

It is also important to underline, as previously postulated [4], that the whole aging curve should be established instead of a single thermal resistivity value determined at a prescribed time. The reasons for this requirement are as follows:

1. To compare aging on slices with different thicknesses
2. To observe whether air ingress stage is completed in the considered period of aging [7]
3. To evaluate the scatter of aging curves determined on separate specimens

Comparison of normalized aging curves established on different specimens will confirm whether the scatter is small enough to permit the assumption of one curve as the material characteristic. This material characteristic may be of great importance for product R&D as it is correlated to gas-diffusion processes through the actual foam structure.

In conclusion, one may observe that:

1. Slicing and scaling technique appears fully applicable to sprayed polyurethane foam insulations.
2. Despite the higher initial thermal resistivity of Base 88, the long-term thermal performance of sprayed polyurethane foams blown with HCFC-123, HCFC-141b and CFC-11 + 0.5% water compares favorably with that of Base 88.

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APPENDIX 1: DEFINITIONS

- *Gas-filled cellular plastic (GFCP)*—A predominantly closed-cell cellular plastic (foam) containing a captive blowing agent with thermal insulating properties better than those of air.

- *Aging curve*—A dependence of thermal resistivity (inverse of thermal conductivity) on the period of aging. When dimensional changes occur in the specimen during the aging period, they shall be disregarded. Original specimen thickness shall be used in the calculations of thermal resistivity as a function of time. Aging curves may be determined on GFCP that are subjected to different environmental conditions. *In this work* aging took place under laboratory conditions.
- *Slicing and scaling technique*—A methodology to predict long-term thermal resistance of a GFCP layer from an aging curve experimentally determined on a thin material layer.
- *Thickness of destroyed surface layer (TDSL)*—An equivalent thickness of open cells on the specimen surface determined from a difference between geometric volume and such measured with a buoyancy method divided by the surface area.
- *Effective thickness (ET) of the closed-cell foam*—An average geometric thickness minus twice the TDSL value.
- *Reference thickness for aging curves*—An effective thickness of 10 mm.
- *Reference aging curve*—An aging curve selected from a number of them determined on the same GFCP product and recalculated to the reference thickness (10 mm). *In this work* the aging curve with lowest long-term thermal resistivity was chosen.
- *Normalized reference aging curve*—A reference aging curve divided by the value of the initial thermal resistivity from which this curve has been started, i.e., a dimensionless aging curve starting at 1.
- *Mean initial thermal resistivity*—A mean R -value from a large number of tests (as required by the statistical considerations) on specimens selected during a prescribed initial period of aging. These tests are performed on specimens with full thickness of the board or at least 25 mm thickness, if the GFCP product was proven to be homogeneous. *In this work* the measurements were performed between the 7th and 14th days of aging, on 25 mm thick specimens.

APPENDIX 2: THE RELATION BETWEEN THE PERIOD OF AGING AND THE DESIGN THERMAL RESISTANCE

Using a natural logarithm and defining the average R -value with the help of Equation (3), one obtains

$$R_{av} = \frac{1}{m} \int_1^m (R_1 - A \cdot \ln k) \cdot dk$$

and after integration

$$\frac{1}{m} \int_{R_1} R_1 - A(k \cdot \ln k - k) \int_1^m$$

further, substituting integration boundaries

$$1/m [R_1 \cdot (m - 1) - A(m \cdot \ln m - m) + A(1 \cdot \ln 1 - 1)]$$

and using approximation that $(m - 1)$ is equal to m , since m is large (long-term thermal resistance), one obtains

$$R_{av} = R_1 - A \cdot \ln m + A$$

R -value decays from the initial value—higher than R_{av} , to the final value lower—than R_{av} , there will always be a day d when the actual R -value (R_d) is equal to the average R -value. Requesting that $R_d = R_{av}$, one obtains

$$R_1 - A \cdot \ln d = R_1 - A \cdot \ln m + A$$

thus, $\ln (m/d) = 1$ and day $d = m/e$ where $e = 2.72$.

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