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IN-SITU AND THIN-SPECIMEN AGING OF EXPERIMENTAL POLYISOCYANURATE ROOF INSULATION FOAMED WITH ALTERNATIVE BLOWING AGENTS

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ABSTRACT: This paper reports apparent thermal conductivity (k) values from field and laboratory aging tests on a set of industry-produced, experimental polyisocyanurate (PIR) laminated boardstock foamed with hydrochlorofluorocarbons (HCFCs) as alternatives to chlorofluorocarbon (CFC). The PIR boards were blown with five gases: CFC-11, HCFC-123, HCFC-141b, and 50/50 and 65/35 blends of HCFC-123/ HCFC-141b.

The k-values were determined from 0 to 50°C (30 to 120°F) using techniques that meet ASTM C 1114 (Thin Heater Apparatus) and ASTM C 518 (Heat Flow Meter Apparatus). Results on laminate boards with facers provide an independent laboratory check on the increase in k observed for field exposure in the ORNL Roof Thermal Research Apparatus (RTRA). The observed laboratory increase in k was between 8% and 11% for a 240 day field exposure in the RTRA.

A thin-specimen aging procedure established the long-term thermal resistance of gas-filled foams. Thin specimens were planed from the industry-produced boardstock foams and aged at 24 and 65°C (75°F and 150°F) for up to 300 days.

An exponential dependency of k with the quantity (diffusion coefficient X time)$^{1/2}$/ thickness, provide effective diffusion coefficients for air components into the foams and blowing agent out of the foams. The foams blown with alternative blowing agents exhibited k-values 3 to 16% (average 9.4%) above CFC-11 foams under similar conditions.

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Field exposures were conducted on specimens under single ply EPDM membranes in the RTRA for over 400 days. Hourly averages of panel temperature and heat flux were analyzed to obtain as a function of mean temperature on a week by week basis. The k-values derived from the field data provided effective diffusion coefficients for air components in the foam, which were greater than those obtained from the thin-specimen aging procedure at 24°C (75°F) by 20 to 70%, but were less than the 65°C (150°F) aging values by 20 to 80%.

The relative performance of test specimens of HCFC-141b under a black and under a white membrane is reported. The field data analysis suggests that the percent increase in k over that of the foam blown with CFC-11 is, after one year of aging, 5.5% for HCFC-123 and 11.7% for HCFC-141b. This leads to the same ordering of foams as derived from the thin-specimen analysis.

KEYWORDS: polyisocyanurate foams, alternative blowing agents, thermal conductivity, thin-specimen aging, diffusion coefficients, field exposures, roof insulation, in-situ performance, gas-filled foams, CFC-11 blown foam, thin heater apparatus, heat flow meter apparatus, aging, roof thermal research apparatus

INTRODUCTION

In the mid-1980's, it was recognized that further increases in chlorofluorocarbon (CFC) concentrations in the upper atmosphere would lead to long-term damage to the ozone layer. International recognition of this fact culminated in the signing of the Montreal Protocol in 1987 by more than 50 industrialized and developing countries [1]. Domestic legislation [2,3] and the Protocol address the global impact of CFCs and outline a timetable for reduction of CFC consumption. If environmentally acceptable alternative gases and foams are not available, the estimated energy impact for building applications alone is between 0.65 and 1.5 quad/year [4].

The current effort is a cooperative industry/government program to establish the viability of alternative blowing agents that resulted from two workshops [5]. The project is directed by a Steering Committee of representatives of the sponsors and Oak Ridge National Laboratory. The purpose of the project is to determine if the performance of polyisocyanurate roof insulation foam boards blown with alternate agents differs from boards blown with CFC-11.

This paper describes apparent thermal conductivity (k) results obtained from field and laboratory tests during 1989 and 1990 on a set of experimental polyisocyanurate (PIR) laminate boardstock produced to evaluate the viability of alternative hydrochlorofluorocarbons (HCFCs) as blowing agents. All boardstock was manufactured from similar formulations that were not optimized for thermal performance. Commercial boardstock made in the future may differ in performance from this set. The PIR boards were prepared with CFC-11, HCFC-123, HCFC-141b, and 50/50 and 65/35 blends of HCFC-123/HCFC-141b.

OBJECTIVES

The cooperative project has two field tasks that are supported by two laboratory tasks. One field task is to monitor thermal performance of roof test panels of the boardstock installed in the ORNL RTRA and exposed to seasonal weather cycles. A second field task is to examine the behavior of roof panels for a range of installation conditions in the ORNL Roof Mechanical Properties and Foundations Research Apparatus (RMPFRA). The objective of the first laboratory task (Task A) is to establish the k of boardstock from 0 to 50°C (30 to 120°F) prior to installation and as a function of exposure time to field conditions in both the RTRA and the RMPFRA. The objective of the second laboratory task (Task B) is to establish k at 24°C (75°F) as a function of aging time at 24 and 65°C (75°F and 150°F) for specimens of three thicknesses sliced from the original boardstock.

Task A used two apparatuses that meet ASTM standards: the ORNL Unguarded Thin-Heater Apparatus (UTHA) (ASTM C 1114) [6] and the ORNL Heat Flow Meter Apparatus (HFMA) (ASTM C 518) [7]. The tests were conducted on rigid boardstock foam specimens that form the test panels to be exposed in the RTRA. Each RTRA panel was nominally 76 mm (3 in.) thick and consisted of two boards, 0.6 m x 0.6 m (24 in. x 24 in.), each nominally 38 mm (1.5 in.) thick, with an embedded heat flux transducer (HFT) at the board interface. The embedded HFTs were calibrated in-place to allow analysis of RTRA data.

For Task B, specimen aging at 24°C (75°F) was conducted under normal laboratory conditions and the 65°C (150°F) aging treatment was conducted in an environmental chamber. The ORNL HFMA was used to determine k (24°C) of the specimens as a function of aging time. The goals of Task B are to evaluate thin-specimen testing as an accelerated aging procedure and to provide a database to compare to RTRA and RMPFRA results with predictions of an aging model. Specimen characterization tests [8] were an integral part of Task B. Characterizations included cell size, preferred rise dimensions, cell wall thickness, fraction solid in the cell wall, and foam permeability to O_2, N_2, and blowing agent. These properties are needed for a model to predict the increase of k over the life of the rigid foam [9].

EQUIPMENT

ORNL Unguarded Thin-Heater Apparatus

The k of the RTRA panels and the calibrations of the embedded HFTs (Task A) were determined from 24 to 50°C (75 to 120°F) in the ORNL UTHA [6, 10, 11]. Initial tests were performed in a one-sided heat flow mode on specimens with GAF black-facers (0.64 mm. thick) in place, as recommended in ASTM C 1013 [12, 13]. The UTHA tests were performed in a two-sided heat flow mode operation for the RMPFRA panels.

The UTHA meets the requirements of ASTM C 1114-89 [6]. The UTHA is an absolute, longitudinal heat flow technique with less than 2% uncertainty for steady-state thermal resistance tests at mean temperatures from 20 to 50°C. This technique has been described in the literature [6, 10, 11].

In 1983 [11], tests were conducted on two standards from the National Institute of Standards and Technology (NIST). The ORNL results on the NIST Certified Transfer Standard were within 0.6% of NIST values at 303 K and 313 K (86 and 104°F). ORNL measurements from 22 to 60°C (72 to 140°F) on SRM 1450b yielded a maximum difference of 0.9% between measurements by the two laboratories at 297.13 K (75°F). These SRMs were retested in the UTHA in 1990 (see Table 1) and agreement with the NIST values was 0.3% for SRM 1451 and 1.1% for SRM 1450b. The UTHA k-values for the SRMs were fit to better than 0.3% by a linear function of temperature. Since all of these comparisons are within the most probable uncertainty of 1.2%, the UTHA k-values reported in this paper provide an accurate description of the temperature dependency of k of several materials.

TABLE 1 -- A comparison of ORNL UTHA and NIST results (1990)

Specimen	Mean Sample Temperature (K)	Sample Density (kg/m^3)	k-ORNL (W/m·K)	k-NIST (W/m·K)	Percent difference[a] (%)
Certified Transfer Standard, SRM 1451					
Two-sided	303.14	9.255	0.04829	0.04827	0.04
	313.14	9.270	0.05180	0.05166	0.27
SRM 1450b					
Two-sided (4 points)	297.13	127.0	0.03445	0.03485	-1.14

[a] 100·[(k-ORNL)-(k-NIST)]/(k-NIST).

ORNL Heat Flow Meter Apparatus

The thermal conductivity of the RTRA panels (Task A) were determined from 0 to 50°C (30°F to 120°F) in the ORNL HFMA [14]. This apparatus was also used to determine k (24°C) of the sliced, aging specimens (Task B). This comparative heat flow meter technique is designed to meet ASTM C 518, Configuration B: two transducers, both faces [7]. The ORNL HFMA is described in Ref. 15.

As specified by ASTM C 518, the HFT is calibrated with specimens of Standard Reference Materials, SRM 1450b and SRM 1451, to establish calibration factors as a function of specimen thickness and temperature prior to a measurement campaign. The apparatus uncertainty has been established to be less than ±5% by tests on identical specimens in the UTHA and in other C 518 apparatuses. The two standard deviation value (2σ) from a comparison to other C 518 apparatuses was 2.2% for planed polyisocyanurate boards [17]. This apparatus was used to test the RTRA panels as a two-board sandwich and to test the top and bottom boards at mean temperatures of 0, 15, 24, 32, and 50°C. All tests of RTRA panels were performed on specimens with the GAF facer in place, as recommended in ASTM C 1013 [12].

ORNL Roof Thermal Research Apparatus

Field comparisons were conducted on test specimens installed in the ORNL RTRA [18, 19]. It is a 3 m x 8.5 m conditioned building that can accommodate four 1.2 m x 2.4 m test specimens as sections on its roof. A cross-section of a test specimen is shown in Fig. 1. Each specimen is a 1.2 m x 1.2 m panel of two layers of 38 mm thick PIR foam insulation and a 1.1 mm black ethylene propylene diem monomer (EPDM) single-ply membrane. In addition, an HCFC-141b specimen was exposed under a white EPDM. The central 0.6 m x 0.6 m area of each specimen is used for thermal measurements and the 0.3 m perimeter is a passive thermal guard. A 50 mm square HFT [20] is mounted between the two layers of insulation in the center of the measuring section. Copper-Constantan thermocouples, all from the same spools to reduce relative errors, are placed at each boundary. Prior to installation, the initial values of k of the central area were measured and the HFTs were calibrated using the ORNL UTHA. The RTRA has a complete weather station for continuous monitoring of ambient conditions as well as solar and infrared radiation. This information and data from the thermocouples and HFTs are recorded at one-minute

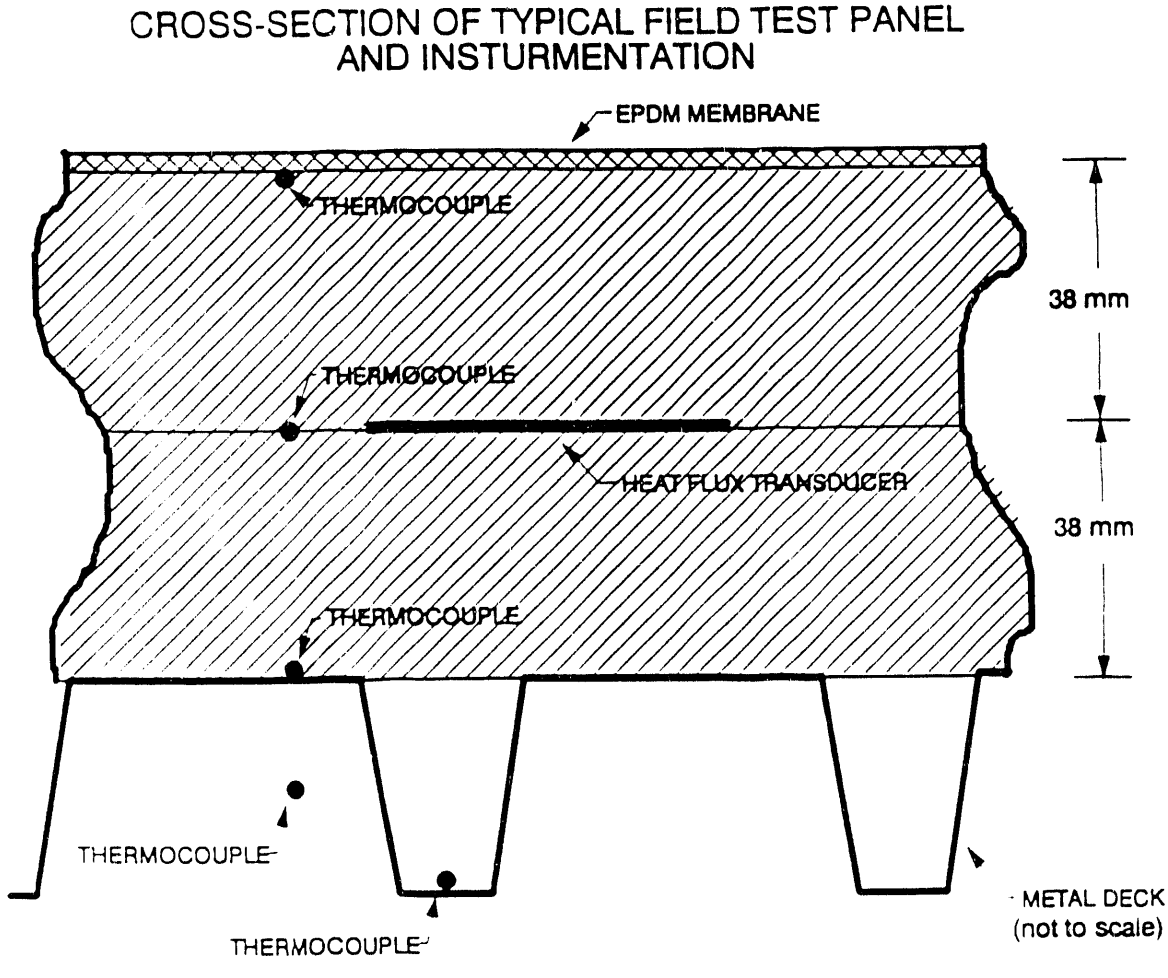

FIG. 1 -- Temperature and heat flux transducer placement in one of the test specimens of the RTRA.

intervals and averaged over an hour. Analysis of RTRA data is on a week-by-week basis using the computer program PROPOR (Properties - Oak Ridge) [21, 22].

For lightweight roof systems an adequate set size is 168 hourly data points for each input parameter. In this study, the PROPOR output consists of the k and dk/dT at the mean temperature of the specimen. For each specimen, independent calculations are carried out for the top board, the bottom board, and both boards combined (since each of these configurations have a different mean temperature each week).

SPECIMENS

Thermal performance tests have been conducted on two types of test specimens produced from the laminate boardstock manufactured by industry [23]: (1) panels for the RTRA and RMPFRA, and (2) thin specimens for aging at 24 and 65°C. The boardstock, blown with CFC-11, HCFC-123, or HCFC-141b, was produced in June 1989 and boardstock blown with 50/50 and 65/35 blends of HCFC-123/HCFC-141b was produced in December 1989.

The Task A test specimens were nominally 0.6 m x 0.6 m x 38 mm, with GAF black facers (0.6 mm thick) on each face for each type of blowing agent. Two of these specimens formed the central area of each 1.2 m x 1.2 m panels for the RTRA tests. A 50 mm x 50 mm x 3 mm slot was routed into the lower board to position the embedded HFT at the interface of the two boards.

Task B required 0.6 m x 0.6 m specimens of three thicknesses for each type of blowing agent. Three thicknesses, nominally 33, 19, and 10 mm were produced by planing the facer and foam from boardstock to produce one, two, and four specimens, respectively, for tests in the HFMA. The 33 and 19 mm thick specimens contained the boardstock centerline and the 10 mm thick specimen had the boardstock centerline as one face for the CFC-11, HCFC-123, and HCFC-141b boardstock. All of the blend specimens contain the boardstock centerline.

RESULTS

Characterization

Table 2 contains structural results obtained on the three boardstock foams produced in June 1989. The cells are elongated in the direction of boardstock production; cell wall thicknesses are between 0.3 and 0.5 μm; and the fraction solid in the cell wall is greater for the HCFC gases. Previous studies on foams blown with HCFC-123 and HCFC-141b show a similar increase in the fraction solid in the cell wall [8].

TABLE 2 -- Structural features of boardstock blown with CFC-11, HCFC-123, or HCFC-141b

	CFC-11	HCFC-123	HCFC-141b
Average distance between cell walls (mm)			
Parallel to facer	0.24	0.20	0.27
Perpendicular to facer	0.16	0.15	0.16
Cell wall thickness (μm)	0.30	0.40	0.53
Percent solid in cell wall	17	30	38

Task A. RTRA and RMPFRA Panel Thermal Conductivity

Figure 2 shows the temperature dependence of k as measured in the ORNL UTHA and the ORNL HFMA apparatuses for the Task A specimen blown with CFC-11. The panels for the other blowing agents showed a similar temperature dependence for k, i.e., a minimum k below 15°C (60°F), a nearly linear temperature dependence above 27°C (80°F), but a

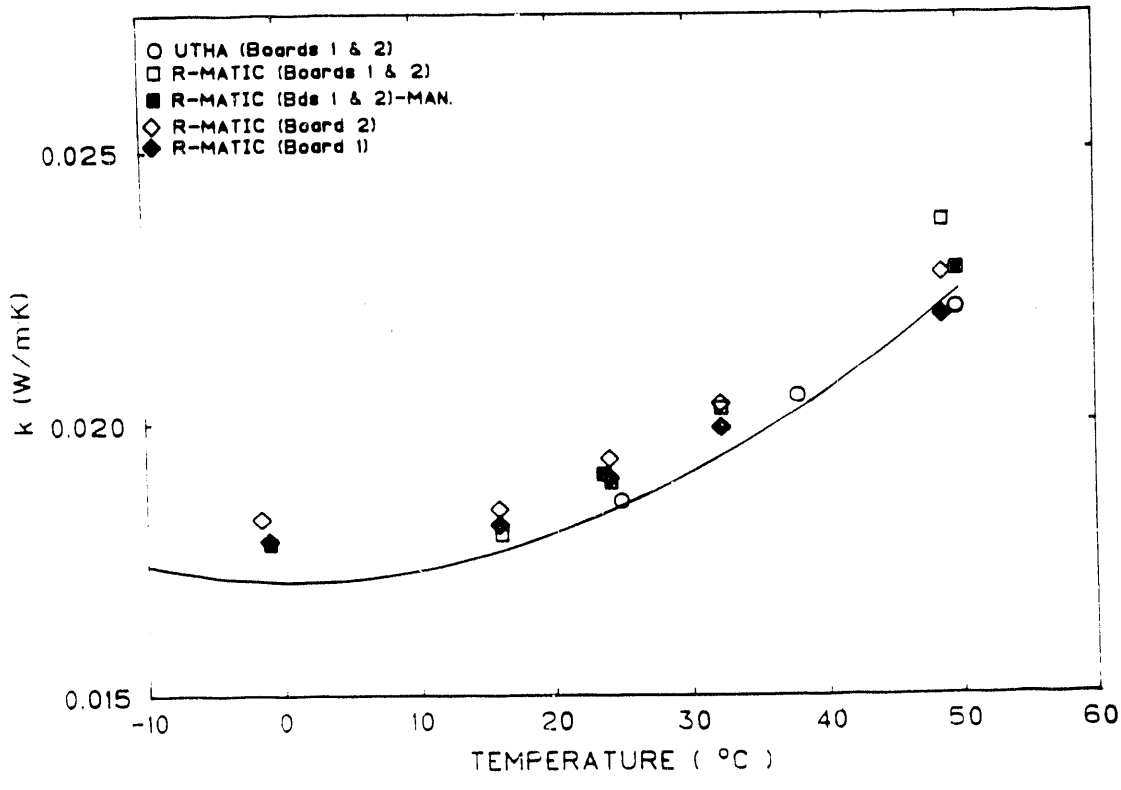

FIG. 2 -- The temperature dependency of the thermal conductivity of boardstock blown with CFC-11. Tested 65 days after manufacture. displacement in k that depended on blowing agent and age at the time

displacement in k that depended on blowing agent and age at the time of testing. ORNL/TM-11645 [15] provides additional results on all of the test materials. The k-values determined with the UTHA are lower than the k-values determined with the HFMA in the temperature range of overlap, but are within the experimental uncertainties for the two apparatuses. Because the UTHA is more accurate, our data analysis is weighted toward the UTHA k values. A least-squares fit was produced for both data sets. Each curve for the HFMA data showed a minimum and was displaced to lower k-values to produce agreement with the UTHA data from 27°C to 50°C. The resulting curve is shown in Fig. 2. Table 3 contains the k values (including the facers) as a function of temperature, specimen density, and specimen age.

TABLE 3 -- The thermal conductivity[a] of a boardstock blown with CFC-11, HCFC-123, HCFC-141b, and two blends 50/50 and 65/35 HCFC-123/HCFC-141b

Temperature	Thermal Conductivity (W/m·K)				
°C	CFC-11	HCFC-123	HCFC-141b	50/50	65/35
1. RTRA Panels					
Age, Days[b]	65	71	78	14	19
-4[b]	0.0179	0.0191	0.0211	-	-
4[b]	0.0169	0.0180	0.0195	-	-
10[b]	0.0171	0.0181	0.0194	-	-
24[b]	0.0185	0.0195	0.0206	0.0195	0.0196
38[b]	0.0205	0.0215	0.0227	0.0211	0.0212
43[b]	0.0212	0.0222	0.0234	0.0217	0.0218
Days[c]	334	336	340	-	-
24[c]	0.0212	0.0216	0.0255	-	-
Density, kg/m³					
Panel	44.5	44.5	43.5	46.3	44.5
Core	32.4	32.4	32.0	34.4	33.6
2. RMPFRA Panels					
Days	365	365	365	-	-
24	0.0221	0.0219	0.0238	-	-

[a] Includes GAF facer.
[b] Time since production when tested prior to installation in the RTRA.
[c] Includes 241 days of exposure in RTRA under black EPDM membranes.

The results shown in Table 3 describe the thermal performance of the respective panels just prior to their installation in the RTRA on August 28,1989. The RTRA panels blown with blends had nearly equal k (24°C) values at an age of 14 to 19 days. The other panels at an age of about 75 days show:

$$k(\text{CFC-11}) < k\ (\text{HCFC-123}) < k\ (\text{HCFC-141b})$$

After exposure in the RTRA for 241 days, 330 days after manufacture, the order relation remained, but the average k had increased 8.6% for CFC-11, 11.1% for HCFC-123, and 9% for HCFC-141b. Table 3 also contains k-values for the RMPFRA panels that were one year old when tested, and these values are greater than the RTRA panels by 12 to 20%.

Task B: Thin Specimens Aging at 24°C and 65°C

The ORNL HFMA was used to obtain k (24°C) values for planed specimens of three thicknesses being aged at 24°C and at 65°C. The tests were conducted periodically on stacks of one, two, or four thin specimens for aging times up to 300 days.

This provides a means to measure the two-stage diffusion process that causes foams to lose some of their insulating value. Without a barrier, air diffuses into the foam cells and the blowing agent diffuses out of the foam cells or goes into solution in the plastic. This process changes the cell gas composition, which changes the cell gas thermal conductivity and this changes the product thermal resistance. The thinner specimens show a more rapid change in k because of the shorter diffusion distance to the specimen centerline.

The results given in Table 4 for 24°C aging confirm the premise of the test procedure. Reference 15 provides results for aging at 65°C. Except for the initial values, the 54 k-values for the 24°C aging study (3 materials, 3 thicknesses, and 6 test times) show that for each thickness and time the order of the material k-values is:

$$k\ (\text{CFC-11}) < k\ (\text{HCFC-123}) < k\ (\text{HCFC-141b})$$

and for each material and time:

$$k\ (33\ \text{mm}) < k\ (19\ \text{mm}) < k\ (10\ \text{mm})\ .$$

The material order is the same as observed for the tests on the RTRA panels. These 54 k-values are smooth, monotonic functions of time divided by (thickness), t/h^2 [15, 16].

We have found that the non-linear behavior of the increase in k with time/(thickness)^2 can be described by two linear regions if one plots $\ell n\ 100\ k$ versus $t^{1/2}/h$. Empirically, k can be described by an exponential dependence on effective diffusion coefficient (D), time (t), and thickness (h):

$$k = k_o \exp((Dt)^{1/2}/h)\ , \tag{1}$$

where k_o is the initial thermal conductivity, then

$$\ell n\ k = \ell n\ k_o + (Dt)^{1/2}/h \tag{2}$$

if:
$$A = \ell n\ k_o$$
$$Y = \ell n\ k$$
$$X = t^{1/2}/h$$
$$B = D^{1/2}.$$

then:
$$Y = A + B\ X \tag{3}$$

TABLE 4 -- k (24°C) values for planed specimens aging at 24°C

		Thermal Conductivity, $W/m \cdot K \cdot 10^3$		
Age (days)	Boards	CFC-11	HCFC-123	HCFC-141b
3	1	18.29	19.78	20.52
	2	17.40	19.36	20.28
	4	19.07	20.35	21.77
17	1	17.87	19.82	20.94
	2	18.55	20.73	22.13
	4	20.05	21.85	22.79
51.5	1	18.97	20.52	21.81
	2	20.14	22.16	23.67
	4	21.67	23.40	24.45
106.5	1	19.07	21.12	22.21
	2	21.29	23.71	24.48
	4	22.50	24.25	25.01
190	1	20.45	21.95	23.16
	2	22.21	24.82	24.94
	4	22.96	24.65	25.49
290	1	20.77	24.44	23.44
	2	23.11	24.71	25.79
	4	23.48	25.41	26.19

Table 4 (cont'd)

		Thermal Conductivity, $W/m \cdot K \cdot 10^3$	
Age	Boards	50/50	65/35
2	1	19.76	19.92
	2	19.62	19.50
	4	21.17	20.94
42.5	1	20.97	21.43
	2	23.02	22.77
	4	25.36	25.31
74.5	1	21.95	22.76
	2	24.69	24.64
	4	26.14	26.02
127	1	21.74	22.63
	2	25.24	24.62
	4	26.66	26.12

Thus, if one measures the k of a foam product of thickness (h) as a function of aging time (t), then a plot of Y versus X should yield a straight line with slope B. A least square fitting of the data to Eq. 3 yields an intercept of $\ell n\ k_0$ and a slope of $D^{\frac{1}{2}}$.

Figure 3 shows the increase of k (24°C) (plotted as ℓn 100 k for convenience) as a function of $t^{\frac{1}{2}}/h$, ($days^{\frac{1}{2}}/mm$) for specimens of three

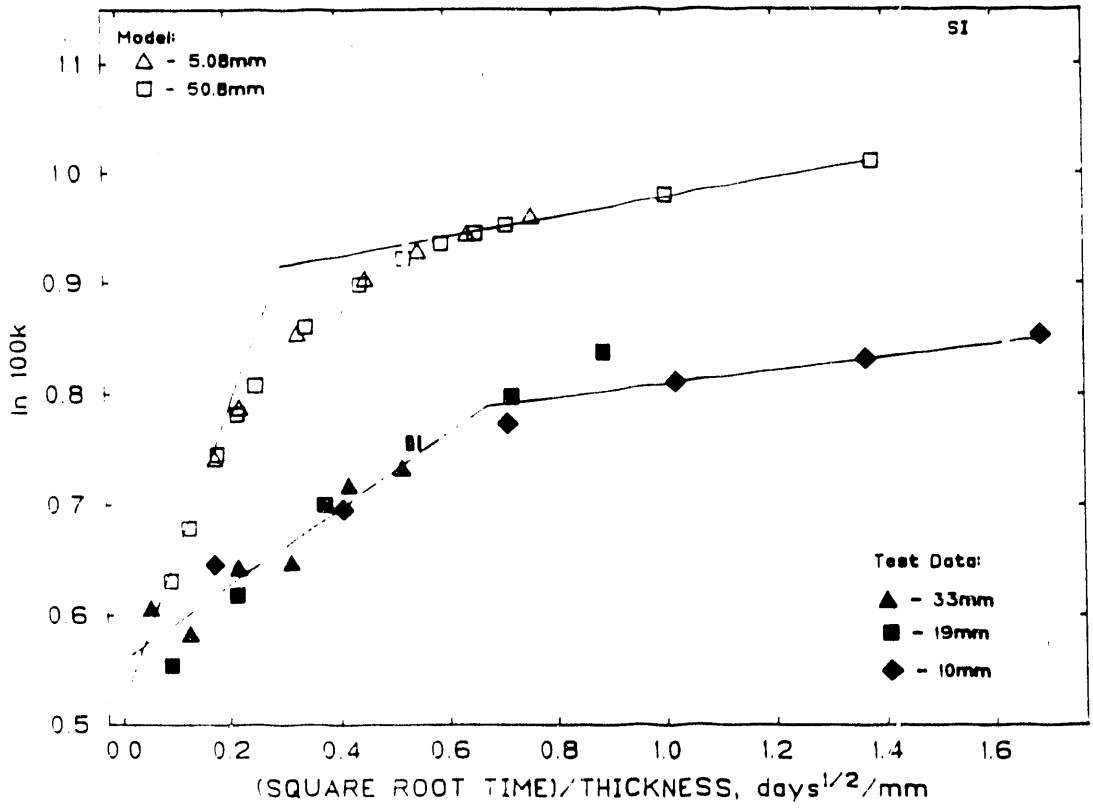

FIG. 3 -- Increase in k (24°C) for thin specimens of rigid board foamed with CFC-11 aging at 24°C compared to MIT model predictions.

thicknesses of foam blown with CFC-11 and aged at 24°C. The test data for the specimens of three thicknesses describe two linear regions of behavior, with an intermediate transition zone. The thin specimens reach larger values of $t^{1/2}/h$ than the thick specimens. We believe the first linear region should be associated with the increase in k due to the influx of air components and the second, lower slope, linear region should be associated with the loss of CFC-11 from the foam. The results of six tests up to 290 days after planing are similar to the MIT model predictions [9, 22] for 50.8 mm thick specimens aged for 5400 days ($t^{1/2}/h$ of 1.45) and 5.08 mm thick specimens aged for 15 days ($t^{1/2}/h$ of 0.76) at 24°C. The predictions are higher in k due to the model assumptions, but the behavior of k with $t^{1/2}/h$ is supportive of the test results.

Figure 4 includes the model predictions and the test data on three thicknesses of foam blown with CFC-11 aged at 65°C. The two linear region behavior occurs for aging at 65°C with larger values of k (24°C) that are closer to the model predictions. For example, at 65°C, the linear region extrapolations intersect near a value of $t^{1/2}/h$ of 0.25 days$^{1/2}$/mm, but at 24°C this intersection is near 0.55 days$^{1/2}$/mm. This result shows that, as expected, the diffusion of air components into the foam is faster at 65°C (339 K) than at 24°C (297 K).

We treated the model predictions for 24°C aging and the available test data for the foams blown with five gases aging at 24°C and at 65°C, as suggested by Eqs. 4 and 5 [15, 16]. The specific equations used were

$$\ell n\ k\ (\text{Region 1, Air}) = \ell n\ k_1 + (D_1\ t)^{1/2}/h \qquad (4)$$

$$\ell n\ k\ (\text{Region 2, Blowing Agent}) = \ell n\ k_2 + (D_2\ t)^{1/2}/h \qquad (5)$$

where k_1 is the projected initial k of the foam (Region 1), W/m·K,
k_2 is the intercept for Region 2, W/m·K,
D_1 is the effective diffusion coefficient for air components into the foam, cm^2/s, and
D_2 is the effective diffusion coefficient of the blowing agent out of the foam, cm^2/s.

Table 5 is a summary of the data fits obtained by the method of least-squares. The average deviation is less than 1% for all of the results, but is 2% for that of the blends in Region 2 and aging at 65°C. The square root of the B coefficients for the data fits provides the effective diffusion coefficients for Region 1 and Region 2. The resulting values for D_1, D_2, and the ratio, D_1/D_2, are given in Table 6. The effective diffusion coefficients derived from the aging tests are the same order of magnitude as the MIT program. The results for aging at 24°C show D_1 values near 1.5×10^{-8} cm^2/sec for the foams blown with individual gases and D_1 values near 2.5×10^{-8} cm^2/sec

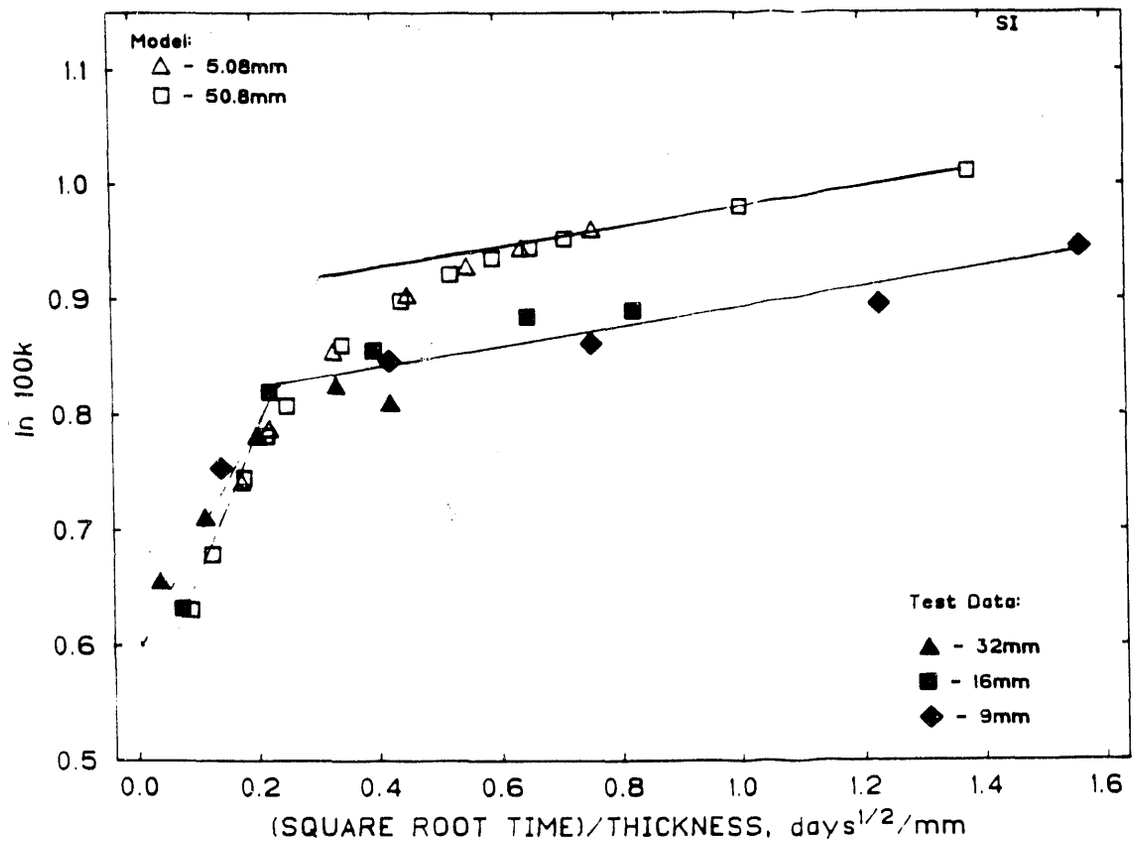

FIG. 4 -- Increase in k (24°C) for thin specimens of rigid board foamed with CFC-11 aging at 65°C compared to MIT model predictions.

TABLE 5 -- Summary of data fits by a least-squares method

Blowing Agent	Region	A	B x 10^6	Number of Data Points	Average Percent Deviation %	Intercept k W/m·K·10^3
Specimens aging at 24°						
CFC-11	1	0.5533	1.230	12	0.65	17.39
	2	0.7528	0.2063	6	0.39	21.23
HCFC-123	1	0.6445	1.218	12	0.41	19.05
	2	0.8454	0.1643	6	0.57	23.29
HCFC-141b	1	0.7021	1.1437	12	0.52	20.18
	2	0.8904	0.1329	6	0.54	24.36
50/50 Blend	1	0.6622	1.5692	8	0.63	19.39
	2	0.8599	0.3582	4	0.11	23.63
65/35 Blend	1	0.6575	1.6070	8	0.56	19.30
	2	0.8548	0.3462	4	0.60	23.51
Specimens Aging at 65°C						
CFC-11	1	0.5972	3.2829	6	0.64	18.17
	2	0.8025	0.3015	9	0.51	22.31
HCFC-123	1	0.6841	2.6102	6	0.57	19.82
	2	0.8463	0.3087	6	0.40	23.31
HCFC-141b	1	0.7429	2.7556	6	0.49	21.02
	2	0.9087	0.2641	9	0.83	24.81
50/50 Blend	1	0.6419	2.9327	6	0.08	19.00
	2	0.8320	0.7231	6	1.75	22.98
65/35 Blend	1	0.6699	2.7907	6	0.64	19.54
	2	0.8333	0.7013	6	1.88	23.01

TABLE 6 -- Effective diffusion coefficients derived from aging tests, cm^2/sec D_1 (air components), D_2 (blowing gas)

	$D_1 \times 10^8$	$D_2 \times 10^{10}$	D_1/D_2
MIT Model (CFC-11)	18.78	9.23	203.5
24°C			
CFC-11	1.51	4.26	35.4
HCFC-123	1.48	2.70	54.8
HCFC-141b	1.31	1.77	74
50/50	2.46	12.83	19.2
65/35	2.48	11.99	21.5
65°C			
CFC-11	10.78	9.09	118
HCFC-123	6.81	9.53	71
HCFC-141b	7.59	6.98	109
50/50	8.60	52.3	16.4
65/35	7.79	49.2	15.8

for the blends. The D_1 values for aging at 65°C are three to seven times larger than the D_1 (24°C aging) as would be expected for temperature dependent diffusion processes. The results for aging at 24°C show D_2 values significantly lower than the D_1 values. The D_2 values range from 1.8 to 4.3 x 10^{-10} cm^2/sec for the individual gases and are above 12 x 10^{-10} cm^2/sec for the blends. The D_2 values for aging at 65°C are 2 to 4 times larger than the D_2 values for aging at 24°C. Some evidence exists that the D_2 values for the foams aging at 24°C are lower for the more complex alternative blowing agents, i.e., D_2 (HCFC-123 and HCFC-141b) < D_2 (CFC-11). The D values are a clear reflection of the foam structural properties and the diffusing species and may be a guide to optimizing boardstock. If one plots the D_1 and D_2 values obtained at 24°C (297 K) and 65°C (338.6 K) as a function of 1/T (K), then the activation energy for Region 1 is near 8000 cal/mol and for Region 2 is near 6000 cal/mol [15].

The constants, A, given in Table 5 can be used to obtain the intercept values k_1 and k_2 for Regions 1 and 2 given in Table 5. The values of k_1, the initial k of the foam, can be used to compare the impact of the blowing agents before any aging occurred. The 24°C aging results show that the order of k_1 values from low to high are: CFC-11, HCFC-123 (9%), 50/50 (11%), 65/35 (11%), and HCFC-141b (15%), where the value in parentheses is the percent increase in k over that of the foam blown with CFC-11. The 65°C aging results suggest a slightly different order.

It has been suggested [25, 26] that the k of a 32 kg/m^3 fresh (unaged) foam can be calculated by adding the blowing gas k and a constant term, 0.01053 W/m·K, to represent the solid and radiation conduction contribution to k. Table 7 shows that this type calculation overestimates k_1, but the percent difference between k (calculated) and k_1 is less than 10%. An alternative calculation is to subtract k(gas) from k_1 and associate the difference with

k(solid) + k(radiation). Table 7 shows that the result of doing so yields an average value of 0.0092 W/m·K for the prototypical foams being tested in this project.

TABLE 7 -- Calculated k (24) for unaged foams for various gases

Gas	Gas k $W/m \cdot K \cdot 10^3$	Foam k Calculated $W/m \cdot K \cdot 10^3$	% Difference[a]	k_1-k(g) $W/m \cdot K \cdot 10^3$	E m^{-1}
CFC-11	8.22	18.75	-7.8	9.17	1716
HCFC-123	10.48	20.91	-9.8	8.67	1831
HCFC-141b	10.10	20.63	-2.2	10.08	1575
50/50	10.24	20.77	-7.1	9.15	1772
65/35	10.28	20.81	-7.8	9.02	1745
				9.22	1728

[a] $100 \times [k_1 - k\ (calculated)]/k_1$.

If half of the solid and radiation contribution is assigned to the radiation contribution [25], and if this is described by the Rosseland approximation [27, 28],

$$k(Rad) = \frac{16}{3} \times \frac{\sigma n^2 T^3}{E} \qquad (6)$$

where σ is the Stefan-Boltzmann constant,
n is the index of refraction, and
E is the extinction coefficient, m^{-1},

then the E-values in Table 7 can be computed from Eq. 6. The average E-value is 1728 m^{-1}. Theory predicts a value of about 2950 m^{-1} for a foam density of 31.2 kg/m^3 and a cell size of 0.2 mm [24]. This E-value would be obtained if 30% of k(radiation) + k(solid) had been associated with k(radiation) instead of 50%.

The accelerated aging test results from this study can be used to predict the thermal resistivity (r, where r=1/k) at 24°C as a function of aging time at 24°C or 65°C for 38.1 mm (1.5 in.) thick unfaced prototypical foam boards. Table 8 shows initial and predicted r-values for one, two, and five years, which correspond to values of $t^{1/2}/h$, $(days)^{1/2}/mm$, of 0.50, 0.71, and 1.12, respectively, for a 38.1 mm thickness. The predicted r-values decrease with time at 24°C, but exceed 39.5 m·K/W for the individual gas blowing agents tested. This value exceeds the minimum stabilized r-value of 38.8 for unfaced PUR or PIR foams stated by the Society of the Plastics Industry assessment [29]. All of the values reported in Table 8 are interpolated values. The results given in Table 8 show that thin specimen aging is a promising accelerated aging procedure and provides a positive response to the key question of this program: the blowing agents tested in these prototypical experimental boards exhibit a long-term thermal performance at 24°C that is within 7 to 15% (average 11.6%) of that obtained by CFC-11 under similar conditions.

TABLE 8 -- Predicted thermal resistivity at 24°C for unfaced 38.1 mm thick prototypical foam boards aged at 24°C and at 65°C

	Initial		Aging Time, Years		
Blowing Agent	k_1[a]	r[b]	1 r(1)[c]	2 r(2)	5 r(5)
At 24°C					
CFC-11	17.39	57.50	47.84	45.13	44.02
HCFC-123	19.05	52.49	43.75	41.46	40.70
HCFC-141b	20.18	49.55	41.74	39.93	39.31
50/50	19.39	51.71	41.11	39.31	38.43
65/35	19.30	41.81	40.70	39.59	37.92
At 65°C					
CFC-11	18.17	55.04	42.85	42.08	40.56
HCFC-123	19.82	50.45	40.97	40.21	38.76
HCFC-141b	21.01	47.60	40.84	38.13	36.95
50/50	19.00	52.63	39.03	37.44	34.32
65/35	19.54	51.18	39.10	37.58	35.53

[a] $W/m \cdot K \cdot 10^3$.
[b] $m \cdot K/W$
[c] Number in parentheses indicates years.

ROOF FIELD EXPOSURE

Weekly k-values collected for a 67-week period from September 1989 until December 1990 are shown in Fig. 5 for the two-board test systems containing CFC-11, HCFC-123, and HCFC-141b boards under the black EPDM. These k-values are reported at the combined mean board insulation temperatures experienced during that week and varied from 5°C (41°F) in the winter of 1989-90 to 32°C (89°F) in the summer of 1990. The average annual temperature during the first complete annual cycle of field exposure was 21°C (70°F). After 538 days from production the relative ordering of performance of all three boards remains the same. Although the performance difference between the CFC-11 and HCFC-123 specimen was insignificant for several summer months in 1990.

Weekly k-values collected for a 43-week period from January 1990 until December 1990 are shown in Fig. 6 for the HCFC blends. These boards were not exposed to the same (temperature/time) conditions as the other three boards, so the relative comparison to CFC-11 is a bit misleading. The slight dip in k-value, shown for the CFC-11 specimen does not occur for the blends because these boards were installed after the period of cold temperatures, which occurred in winter of 1989-90. The mean temperatures of the 50/50 and 65/35 blend (HCFC-123/HCFC-141b) boards were higher during their initial aging period than that of the exposure for the CFC-11 boards. There is a very small thermal performance difference between the two blends for most of the summer which occurred between days 150 and 300.

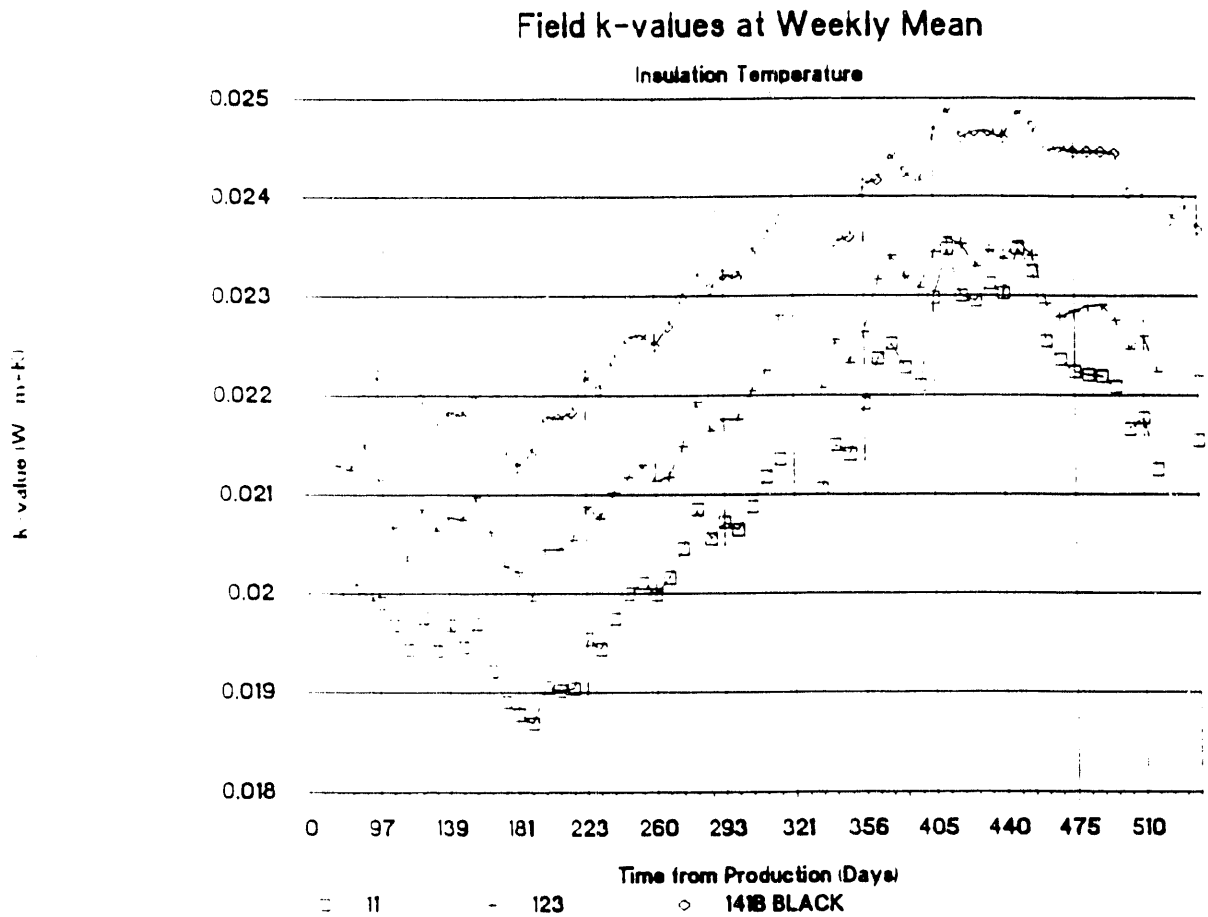

FIG. 5 -- Field k-values as a function of time since production for individual gases under black EPDM.

A second objective of the field testing of HCFC blown roof insulation boards is to determine if roof application practices might be identified which would reduce the rate of thermal aging. The testing of HCFC-141b under both a black and white membrane was designed to determine if a reduction of in-service mean insulation temperature would reduce the rate of thermal aging. Figure 7 shows the k-value of the HCFC-141b test specimens under a white and a black EPDM membrane for a 445 day field exposure. The k-value is displayed for a mean insulation temperature of 24°C. The temperature effect on RTRA k-values was adjusted using the k-value temperature relationship developed by steady state laboratory measurements of the test specimens prior to installation on the RTRA shown in Fig. 2. The equation used to fit the laboratory data was used to derive an adjustment factor as a function of mean insulation temperature. This relationship was then used to convert the weekly measured k-values at actual experienced mean weekly insulation temperature to k-values at 24°C. This procedure assumes that the shape of the k-value versus mean insulation temperature remains constant for the full field testing period. Prior to field insulation the two stacks of HCFC-141b boards were tested in the laboratory. The two stacks were measured at an age of 77 days.

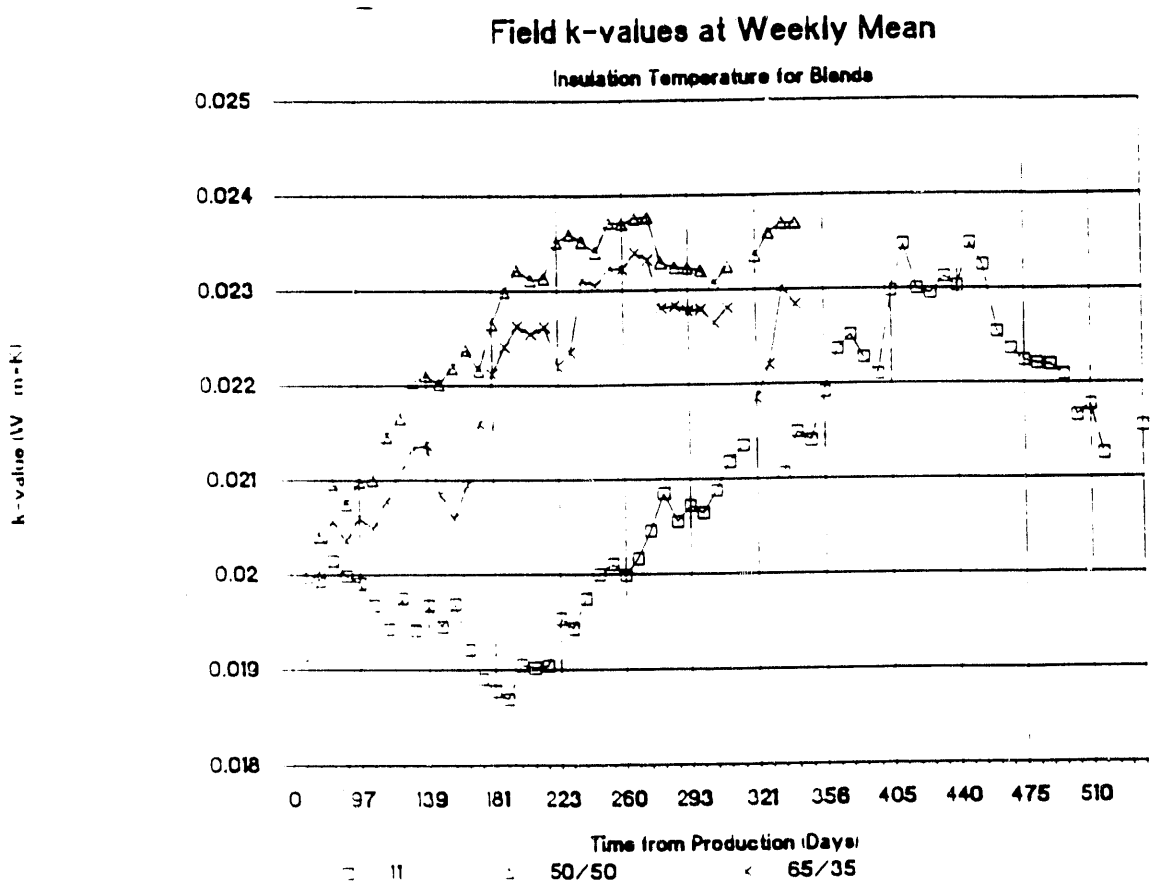

FIG. 6 -- Field k-values as a function of time since production for blends under black EPDM.

There was no significant difference in k-value as shown in Fig. 7. Throughout the first winter season (1989-90) it appeared that the HCFC-141b boards under the white membrane did age less than the HCFC-141b under the black membrane. However, in the spring the field k-values measured the same. This was supported by the laboratory measurements shown in Fig. 7 around 336 days of age, which also showed an insignificant to small difference between these two sets of test specimens. Throughout the summer of 1990, there appears to be no difference in thermal performance between the specimens under the black and white membranes. For one complete annual cycle the mean roof insulation temperature for both test boards combined, under the white membrane was 18°C (64.1°F), 3.3°C (6°F) less than the insulation under the black membrane 21.2°C (70.2°F). The maximum weekly mean temperature of the top board only under the black membrane was 34°C (93°F) compared to 26.7°C (80°F) for the top board under the white membrane. Surprisingly after the summer cooling season a difference in field thermal performance measurements reappears. In one week the boards under the black membrane rise in k-value and in the opposite direction of about the same magnitude the boards under the white membrane appears to have improved. Just after this unusual behavior, which replicates that which occurred in the transition from cooling to heating in 1989, the test specimens were checked in the laboratory. The laboratory measurements are remarkably consistent with the field data throughout the summer but do not suggest a difference in k-value at the time of measurement. It appears as if the field measurement

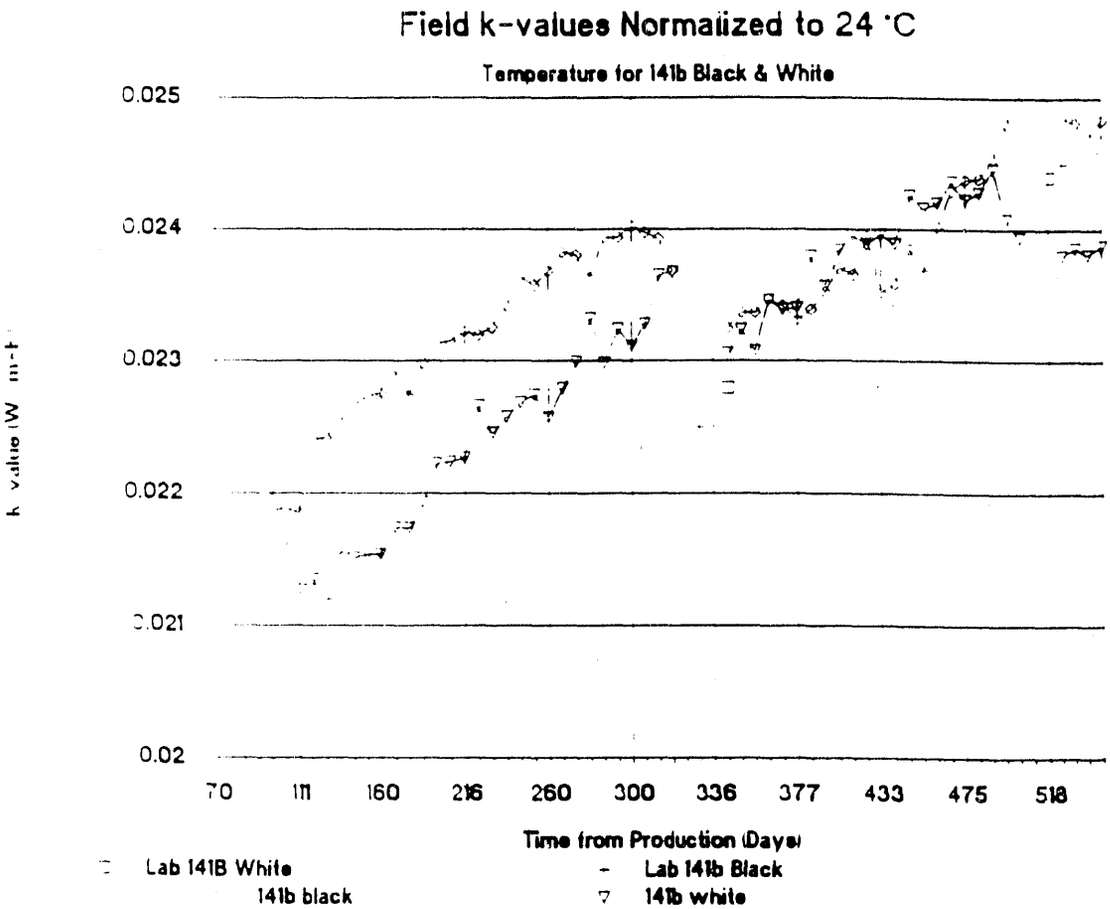

FIG. 7 -- Field k-value, normalized to 24°C, for HCFC-141b under black and white EPDM.

procedure is sensing a phenomena which is occurring only during heat flow primarily in the upward direction that is not reflected when removed from the dynamic roof environment. The field measured difference between the k-value shown for the last month between the HCFC-141b test boards under the black and white membrane is less than 4%.

The data sets shown in Fig. 7 illustrate a linear relationship between k-value (24°C) and age for the first 8-10 months of field exposure. This observed relationship can be used to derive the effective air component diffusion coefficients for field-installed foams. Using the empirical equation described in Eq. 3.

Figures 8, 9, and 10 show the increase of field measured k (24°C) (plotted in the same form as the laboratory slicing data) as a function of $time^{0.5}$/thickness ($days^{1/2}$/mm) for the two-board combination of the CFC-11, HCFC-123, HCFC-141b black, and HCFC-141b white. The linear fit leads to reasonably high R^2 for several cases (0.85, 0.92; 0.97, 0.97) for CFC-11, HCFC-123, HCFC-141b black, and HCFC-141b white. After about 9-10 months of field exposure, the k-values do suggest a leveling out for 1.5-in. thick specimens just as predicted from the aging model and slicing analysis.

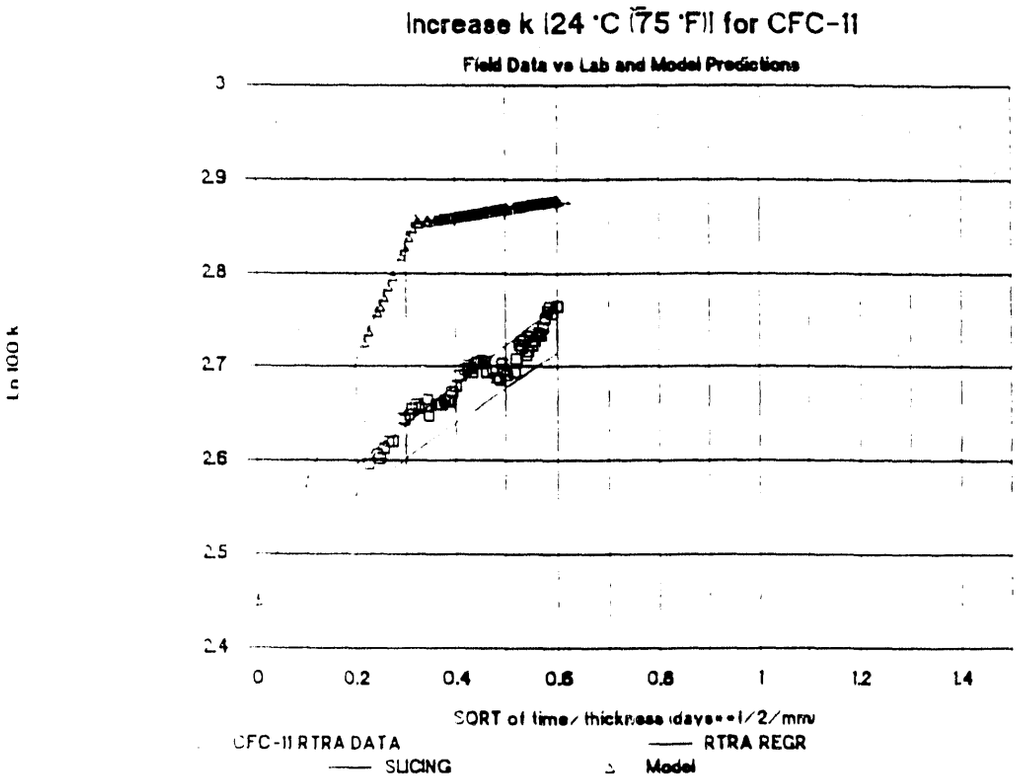

FIG. 8 -- Increase in k for CFC-11.

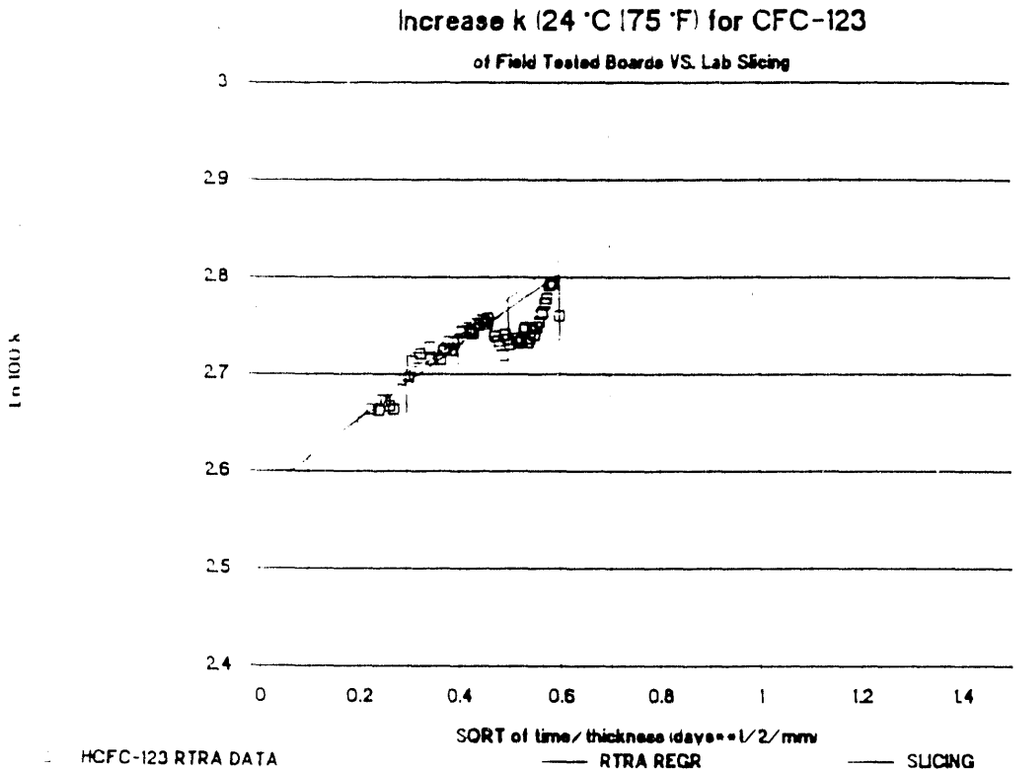

FIG. 9 -- Increase in k for CFC-11.

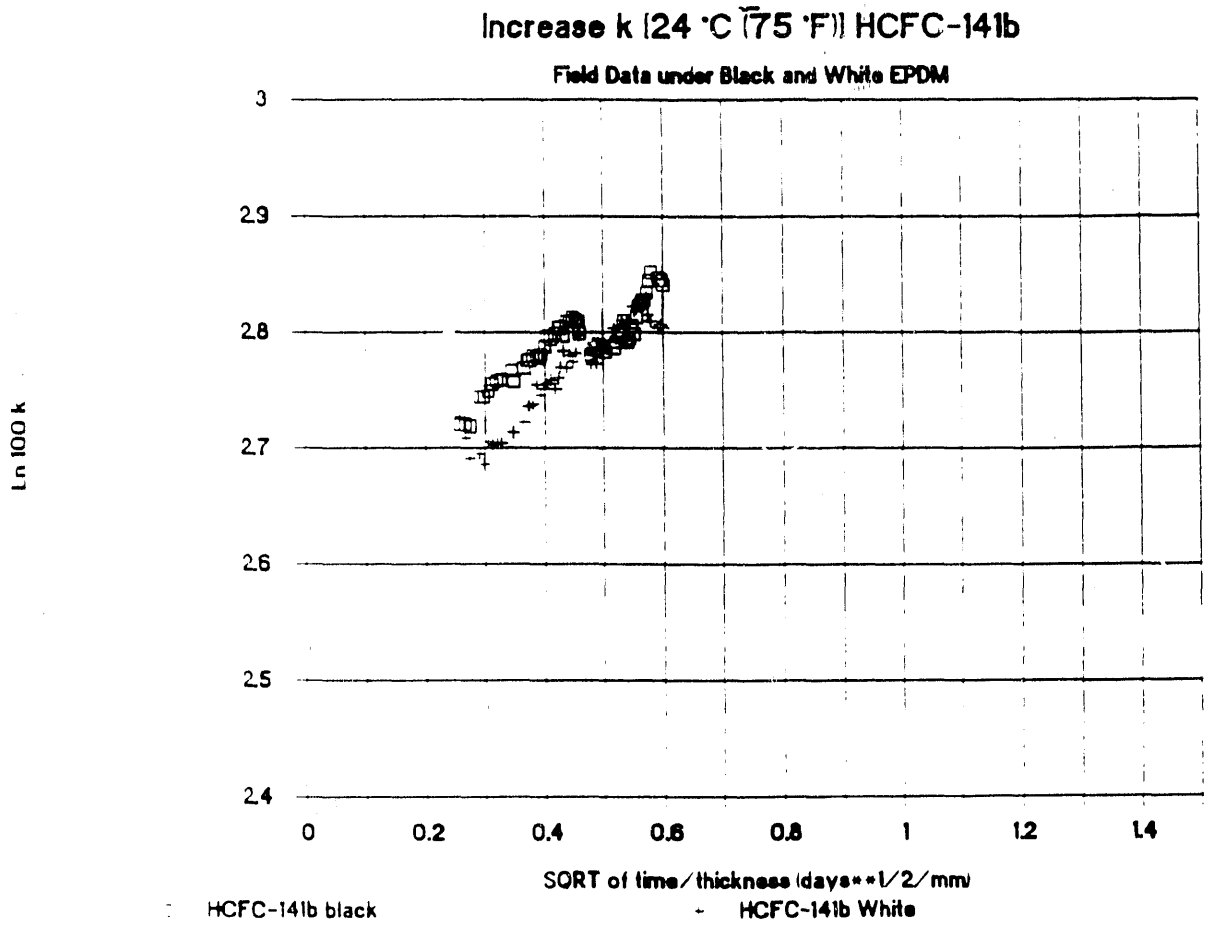

FIG. 10 -- Increase in k for HCFC-141b.

The resulting effective air component diffusion coefficients obtained from the field data are shown in Table 9 and compared with the values derived from the slicing analysis, aged at 24°C and 65°C.

The effective air diffusion coefficients appear to be higher in the faced field specimens than laboratory measurements on the unfaced sliced specimens conditioned at 24°C by 20 to 71% for those specimens under the black membrane. The same HCFC-141b boards under a white membrane lead to a much higher value than that under the black membrane. The air diffusion appears to be lower in the field specimens under the black membrane than in laboratory specimens condition at 65°C by 20 to 80%. However, the diffusion coefficient derived from the HCFC-141b boards under the white membrane is actually 21% higher than even the diffusion coefficient derived at 65°C conditioned samples.

Table 10 contains the field data at one year of age for all four faced 1.5 inch thick test specimens, the k-values at 24°C are compared to predictions based on laboratory slicing made after less than 200 days of actual laboratory aging at 24°C and 65°C. The slicing predictions were within ±1.5% of the field data normalized to 24°C for laboratory specimen conditioned at 24°C and where 10% above the field data when the sliced specimens were conditioned at 65°C. The comparison suggests that conditioning the boards at 24°C is reasonable for predicting aging behavior for roof application.

TABLE 9 -- Effective diffusion coefficients derived from RTRA data and slicing tests in laboratory, 10^{-8} cm^2/sec for the air components, D_1

	CFC-11 Black EPDM	HCFC-123 Black EPDM	HCFC-141b Black EPDM	HCFC-141b White EPDM
RTRA Data	2.21	1.96	2.58	4.02
Steady-State Lab (24°C)	1.52	1.64	1.51	1.51
% Difference	+45%	+20%	+71%	+166%
Steady State Lab (65°C)	10.78	6.81	3.33	3.33
% Difference	-80%	-71%	-23%	21%

TABLE 10 -- Field k-value at 24°C for faced 1.5-inch thick prototypical foam boards using field data compared to unfaced slicing analysis prediction values conditioned at 24°C and at 65°C, for an age of one year (365 days)

	k-value w/m·k			
	CFC-11 Black EPDM	HCFC-123 Black EPDM	HCFC-141b Black EPDM	HCFC-141b White EPDM
Field data	0.02127	0.02219	0.02338	0.02345
Steady state (24°C)	0.02118	0.02189	0.02372	0.02372
% Difference*	-0.4%	-1.3%	+1.5%	+1.19%
Steady state (65°C)	0.02357	0.02457	0.02557	0.02557
% Difference*	+10.8%	+10.7%	+9.4%	+9.0%

$$\frac{100 \times R\ (Lab) - R\ (Field)}{R\ (Field)}$$

CONCLUSIONS

This paper reports k-values on a set of industry-produced, experimental polyisocyanurate laminated boardstock foams blown with five gases: CFC-11, HCFC-123, HCFC-141b, and 50/50 and 65/35 blends of HCFC-123/ HCFC-141b. The k-values were determined from 0 to 50°C using the ORNL Unguarded Thin-Heater Apparatus and the ORNL Heat Flow

Meter Apparatus. The test results on panels with facers provide an independent laboratory check on the increase in k observed for a 241-day field exposure in the ORNL Roof Thermal Research Apparatus. The observed laboratory increase in k was between 8% and 11%.

Test results are reported on a thin-specimen accelerated aging procedure to establish the long-term thermal resistance of gas-filled cellular foams. These thin specimens were planed from the industry-produced boardstock foams and aged at 24°C and 65°C for up to 290 days. The resulting k-values were correlated with an exponential dependency on (diffusion coefficient x time)$^{1/2}$/thickness and provided effective diffusion coefficients for air components into the foam and blowing agent out of the foam. This accelerated aging procedure is a promising technique and was used to predict the five-year thermal resistivity of the foams. Aging at 24°C and at 65°C showed the foams blown with alternative blowing agents exhibited a thermal performance within 3 to 16% (average 9.4%) of that obtained by CFC-11 under similar conditions.

Field thermal performance at one year of age was within ±1.5% of predictions from laboratory aging. Field tests in the RTRA for up to 67 weeks yielded effective diffusion coefficients for air components that were higher than those of the thin aging tests. After about 40 weeks of field exposure, air component diffusion has neared completion for specimens blown with individual blowing agents. The field performance of specimens blown with two blends were similar to each other after 25 weeks of exposure. Field exposure of HCFC-141b specimens under black and white EPDMs yielded no difference in relative thermal performance.

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