The Measurement of 'Aged Thermal Conductivity' of Factory Produced Insulation Boards

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ABSTRACT

The new European standard, EN13165, for factory made rigid polyurethane foam products used as thermal insulation boards for buildings, was published in May 2001 [1]. While CE markings have already been possible for over a year, it is compulsory from March 2003 onwards. At this time the former national standards will have to be withdrawn. Extensive effort has been dedicated to the definition of a method to declare aged thermal conductivity, which is described in Annex C of this standard. Quality data on long-term thermal conductivity (lambda) values are quite rare, especially for foams with blowing agents such as pentane which have been used commercially for only about 10 years. As a result, the design of the above norm was based mainly on calculations and modelling. A lambda ageing predictive model (Agesim), developed by Huntsman Polyurethanes, was used extensively to enable the norm design.

The intention of EN13165, Annex C is to provide an estimate of the average lambda value during 25 years of use under operational conditions. The board producer can choose between two basic routes to come to this value. The first route is called the fixed increment method. This is based on initial lambda measurement followed by the addition of a fixed increment, which depends on blowing agent, board thickness and diffusion tightness of the facing. A test showing that the product has a normal ageing behaviour (called normality test) needs to be passed before using this fixed increment method. A second route is called accelerated ageing method. It is based on a lambda measurement after storing the board for 25 weeks at 70C, followed by the addition of a safety increment. This safety increment can be reduced, depending on the outcome of an acceleration test.

Within this paper, some initial experimental results with EN13165 are presented. Special attention has been given to new experimental test conditions, such as the 25 weeks at 70C ageing and the acceleration test, which were not used previously. This paper shows what range of declared lambda values are to be expected using this new method. These declared lambda values are then compared to 25-year predictions using the Huntsman Polyurethanes lambda ageing predictive model. In this way, this paper provides a first in-depth evaluation of the validity of the new European Standard for declaring aged thermal conductivity.

INTRODUCTION

Polyurethane (PUR) and polyisocyanurate (PIR) factory made insulation boards are used in commercial, industrial and residential buildings throughout Europe. The combination of low thermal conductivity, good strength at low density and ability to adhere at various facing materials give PUR/PIR boards a significant share of the total insulation market. The most distinctive feature of PUR/PIR insulation boards compared to other insulation materials is its low thermal conductivity (lambda). This low lambda value results from the fine cell structure, low density of the foam and the presence of an insulating gas within the foam cells. However, the gas composition in the foam cells can change over time, leading to a phenomenon, known as lambda ageing. Air entering the foam cells has a much higher thermal conductivity than the insulating gases initially present.

When trying to define an aged lambda value starting from a lambda value of a fresh board, the basic question arises which method to use. Possible methods include experimental and modelling methods, or alternatively simple increments to the initial lambda value. Experimental methods are designed to accelerate the diffusion of gases (physical blowing agent, air, CO_2). This can be done via ageing the foam at a higher temperature than the operational one. Another experimental option is to cut the foam board into thin slices and thus reduce the diffusion distance. The ageing processes will be a lot faster compared to the real (thicker) product. Of course, the main question with these experimental methods is the necessary time at accelerated ageing conditions to be relevant for the long term ageing at operational conditions. In some particular cases, experimental techniques are not suitable to accelerate the ageing process e.g. the ageing of aluminium foil faced board can not be studied via thin slicing. As mentioned above, modelling techniques [2] are an alternative to the experimental methods. These models calculate changes in foam cell gas composition and lambda over time. Although results can be quite accurate, these models require an extensive set of input parameters, which are subject to a very careful experimental determination. For this reason, these models are not suitable to serve as a norm for the industry.

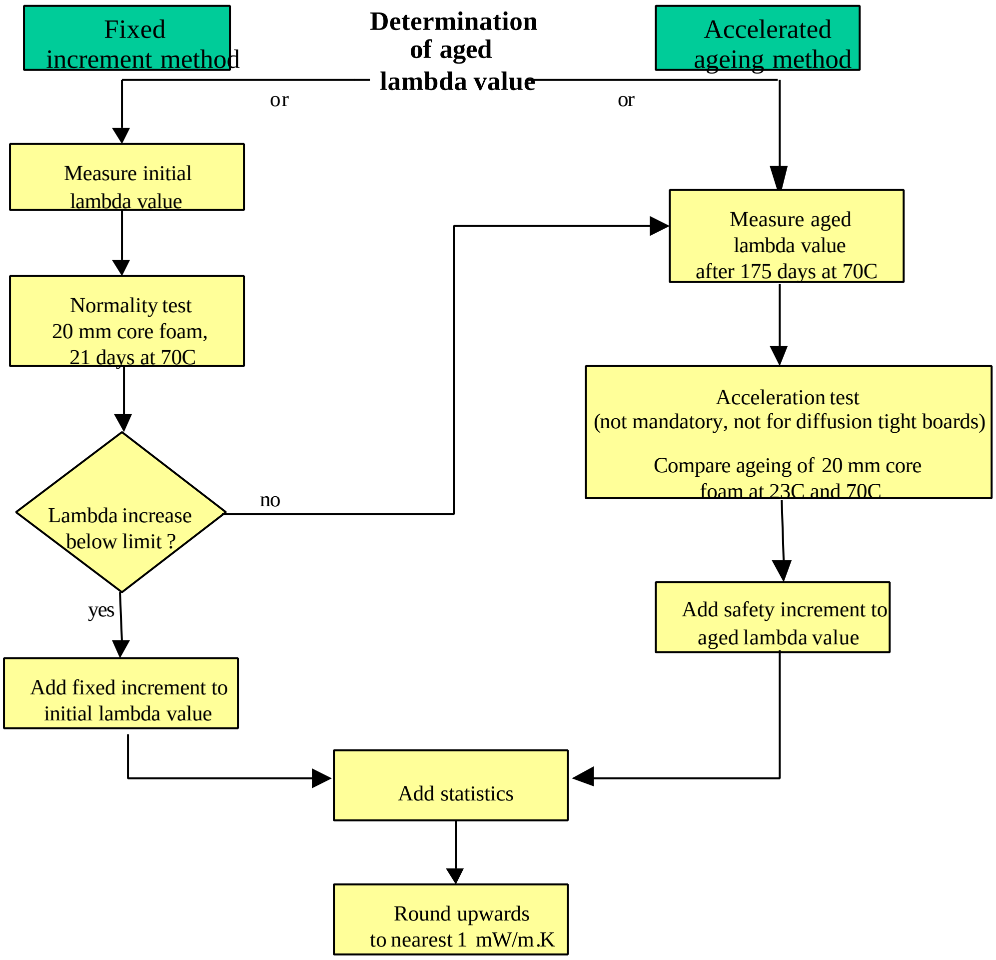
The declaration of an aged lambda value by trying to estimate the lambda changes during the expected economic lifetime, makes a lot of sense. In fact, this has been done in Europe for quite some time although methods have been different in the various countries. These methods were based on ageing the foam at elevated temperatures for well defined times, as well as adding fixed increments to the initial lambda value. In the US, ageing of boards for half a year at room temperature has been used as a method in the past. Currently the US boardstock industry is in the process of switching towards a Canadian method known as S770 [3], which is essentially an accelerated ageing at room temperature of thin slices of the original product while taking into account the possible diffusion retardation by foam skins and facings [4]. Within the whole process of harmonising the European standards, a uniform method to declare aged lambda values for insulation boards, was required. The final method is described in EN13165 – Annex C.

CEN TC88/WG6/EN13165 – Annex C

Within CEN, the European Committee for Standardisation, a separate technical committee (called TC88) is present which deals with all normative issues related to ‘thermal insulating materials and products’. Within TC88, several working groups (WG) have been formed which deal with specific insulation materials or specific applications. WG6 deals specifically with factory made polyurethane foam products used as thermal insulation for buildings. The different test methods as defined in this WG6 are described in a European Standard called EN13165, published in May 2001. During the design of this standard, a lot of attention was paid to the method used for the measurement of aged thermal conductivity, which is described in Annex C of the standard. The final method is the result of an extensive discussion between board producers, raw material suppliers and test institutes. In view of all the effort spent at the design of Annex C, it currently serves as the basis for other working groups dealing with other applications of PUR, when designing a method to measure aged thermal conductivity. This is another reason why the detailed experimental evaluation of this Annex C and a comparison with a modelling program is so crucial.

Although all details of Annex C can be read in the official standard EN13165, a short summary of the method is given here (see figure 1). The basic intention of Annex C is to declare an aged lambda value, which represents the time-weighted lambda value over a period of 25 years. Due to the specific shape of the lambda – time curve, this 25 year time-weighted lambda value is typically reached after about 8 years, although exceptions are possible. On the basis of this definition, two possible methods have been defined, which are called ‘the fixed increment method’ and ‘the accelerated ageing method’. The ‘fixed increment method’ is basically a measurement of the initial lambda followed by a lambda increment, which depends on blowing agent type, board thickness and diffusion characteristics of the facing. Since this ‘fixed increment method’ does not include an experimental evaluation of the ageing characteristics of a board, it was decided that another small test (called the normality test) was required to exclude boards with unacceptable ageing behaviour. The normality test is ageing of 20 mm core foam for 21 days at 70C. The lambda increase during this period should not be more than a certain limit which depends on the kind of blowing agent used (6.0 mW/m.K for pentane blown products). In case this normality test is not passed, the user is then obliged to use ‘the accelerated ageing method’. This method consists basically of a lambda measurement after ageing the full product including its facings for 25 weeks at 70C. A safety increment still needs to be added to this lambda value to account for the fact that the ageing during 25 weeks at 70C might be less than the targeted 25-year time-weighted lambda value. However, if it can be proven via another test called the ‘acceleration test’ that the ageing at 70C is enough of an acceleration compared to room temperature, the safety increment can be omitted (or reduced) for diffusion open boards. After obtaining an aged lambda value with the fixed increment method or the accelerated ageing method, statistics needs to be added, followed by a rounding upward of the obtained lambda value to the nearest 1.0 mW/m.K.

Figure 1. Determination of aged lambda according to EN13165



EXPERIMENTAL

Eight insulation boards, produced by 4 different European producers, have been incorporated in this study. All boards were pentane blown. There were six boards with diffusion open facings and two boards with diffusion tight facings. The diffusion open boards had thickness' ranging from 30 mm to 120 mm; the diffusion tight boards from 30 mm to 50 mm. Lambda measurements (according to EN13165) have been carried out by the board producers; cell gas analysis of fresh and aged foam samples as well as determination of diffusion coefficients has been carried out by Huntsman. The Huntsman lambda ageing simulation software, called Agesim, has been used to fit experimentally measured short-term lambda vs. time curves as well as to make 25-year lambda predictions. The cell gas analysis technique as well as Agesim are described below.

Cell gas analysis

The cell gas content was analyzed quantitatively using a modified gas chromatograph [5]. Small foam cylinders (0.9 cm diameter, 2-3 cm length) were drilled out of the foam samples using a cork bore. These cylinders were subsequently crushed and ground by a special device, which collects escaping gases into an injection system. The device was preheated at 100C to allow vaporization of all the blowing agents present in the foam cells. Blowing agent absorbed in the foam matrix is not released during this short crushing period. Subsequent injection on a gas chromatograph allows for quantitative and qualitative analysis of all gases. Calibration is done via measuring the retention time and response area of a well-defined amount for every relevant gas. Based on the weight of the original foam cylinder, its density and closed cell content, the gas composition can be expressed as a weight % or as a cell gas pressure.

Lambda Ageing Simulations: Agesim

As discussed earlier, the change in cell gas composition over time and related change in gas thermal conductivity (thermal ageing) is quite a complex phenomenon. Different gases have different diffusion rates (CO_2 > air > BA) which will also depend on foam morphology and temperature. Moreover, blowing agents can undergo condensation and partially dissolve in the foam matrix. The exact calculation of cell gas composition and gas thermal conductivity (k-gas) as a function of time and position in the foam is done within the Huntsman lambda ageing software, called Agesim. The first step in the calculation is to define an initial cell gas composition for a given foam board. This can be calculated on the basis of the formulation or, even better, be measured on the basis of the above described cell gas analysis technique. Initial gas thermal conductivity is calculated on the basis of the Wassiljewa equation (Lindsay-Bromley calculation of gas interaction parameter) [6].

After defining an initial cell gas composition, several ageing modules are available. For rectangular boards, a choice between a one dimensional (1D) and a three dimensional (3D) ageing routine is possible. The 3D ageing routine is based on an analytical solution of Fick's law. Diffusion takes place in 3 directions in a rectangular block at uniform temperature. In each of the three orthogonal directions, a uniform effective diffusion coefficient needs to be chosen. At each of the six faces, a diffusion tight facing is possible. The output consists of cell gas pressure and k-gas as a function of time for a given position in the block. In the 1D ageing routine, the foam board is divided in small elements, which have a well-defined temperature and diffusion coefficient for all gases. If the foam board is subjected to a temperature gradient, diffusion coefficients vary in each element, depending on the temperature of this element. If facings act as diffusion barriers, the exact diffusion coefficient of each gas in the facing can also be specified. Condensation is also assessed for each element, again depending on its temperature. In each time step, every element will undergo diffusion with its neighbor elements. After each time step, the gas-liquid equilibrium is first reassessed and k-gas is subsequently calculated. This procedure (diffusion – condensation – k-gas) is then repeated until the complete time period of interest is covered. The main output consists of graphs of (position-averaged) cell gas pressure and k-gas as a function of time on the one hand and cell gas pressure and k-gas as a function of position at certain times. Compared to the 3D ageing routine, this 1D ageing routine gives a more realistic calculation of the ageing process, especially for foams which are subjected to a large temperature gradient or foams with skin/facings which retard diffusion significantly. At the other hand, the 1D routine is limited to diffusion processes taking place in one direction and some of the input parameters (D_{eff} and thickness of skin/facing) are not easy to determine. In this paper, only the 3D ageing routine has been used while in another recent publication [7], the 1D routine was used.

Effective Diffusion Coefficient Determination

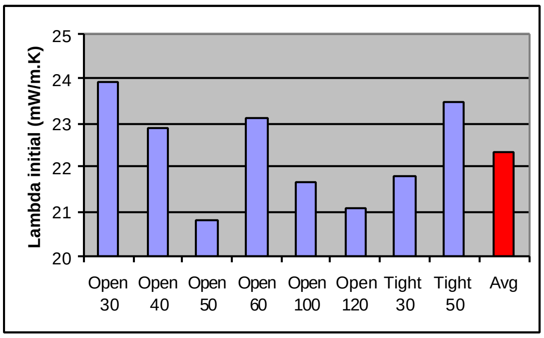
Effective diffusion coefficients (D_{eff}) and their temperature dependence are a very critical input parameter for obtaining a successful thermal conductivity ageing simulation. Two basic methods, both requiring cell gas analysis, are used at Huntsman Polyurethanes to determine diffusion coefficients. In the first method, a relatively thin lambda block (e.g. 20 x 20 x 3cm) is aged at a well-controlled temperature. By performing cell gas analysis before the gas has reached its equilibrium value, the D_{eff} of air and CO_2 can easily be determined via Agesim. This method is not suitable for determining D_{eff} of blowing agents as they diffuse very slowly. Instead, small cylinders are aged for a given time at a fixed temperature, and cell gas analysis performed. By taking into account the specific geometry of these cylinders, D_{eff} of blowing agents is calculated. The temperature dependence of D_{eff} is described by an Arrhenius type of equation, after measuring D_{eff} at at least two temperatures, generally at 23C and 70C [5].

RESULTS

Fixed increment method

As explained earlier on, the 'fixed increment method' requires first a measurement of the initial lambda at 10C. These results are given in graph 1.

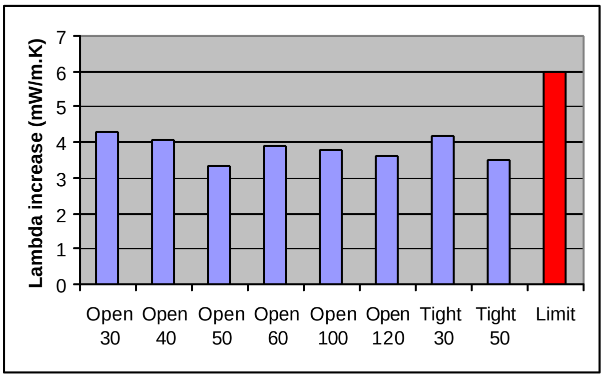
Graph 1. Initial lambda of 8 different boards involved in this study.



Initial lambda values of the 8 boards involved were between 20 and 24 mW/m.K with an average initial thermal conductivity slightly above 22 mW/m.K. It must be noted that all of these boards were blown with n-pentane (or a mixture n-pentane/i-pentane) but none was blown with cyclopentane, which is known to have the lowest gas thermal conductivity of the three pentanes. In graph 1, boards are referred to on the basis of the diffusion open/tight nature of the facing and the thickness in mm. The same terminology will be used here onward.

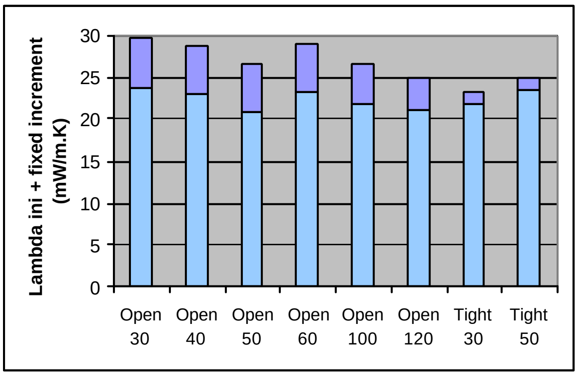
Before adding fixed increments to these initial thermal conductivity values, the normality test needs to be checked. The measured lambda increase after 3 weeks ageing of 20 mm core foam at 70C is given in graph 2.

Graph 2. Lambda increase during normality test.



The lambda increase during this normality test should not be more than 6.0 mW/m.K for pentanes. As can be seen in graph 2, none of the 8 boards show a lambda increase even close to this limit value. It is also quite remarkable that the lambda increases, manufactured and measured by different board producers, are very similar. In any case, the addition of fixed increments to the initial lambda values is allowed for all 8 boards. This is shown in graph 3.

Graph 3. Initial lambda value + fixed increment.

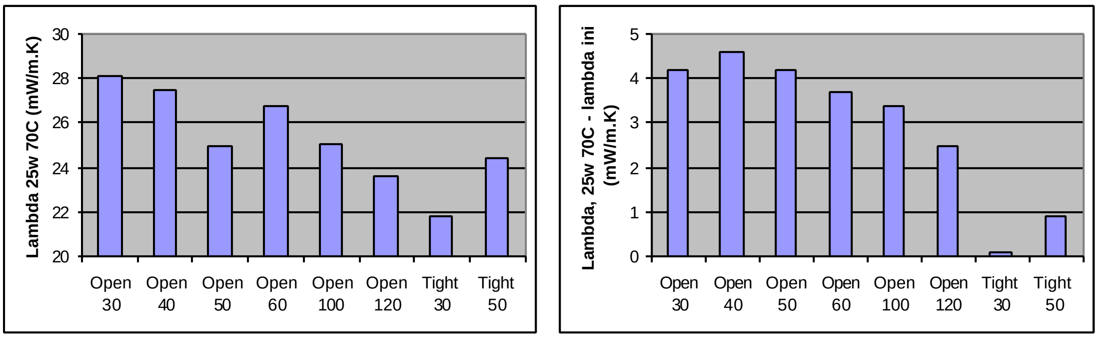


The highest fixed increments are to be used for diffusion open products with a thickness less than 80 mm (5.8 mW/m.K for pentane) while the 100 mm and 120 mm diffusion open products have slightly reduced fixed increments (4.8 and 3.8 mW/m.K respectively). This is based on the fact that diffusion processes will be slower with increasing board thickness. For the diffusion tight products, where gas changes will only occur at the edges, the fixed increments are, very low. The resulting lambda values, after adding fixed increments to the initial lambda values, range between 25 to 30 mW/m.K for the diffusion open boards and 23 to 25 mW/m.K for the diffusion tight boards.

Accelerated ageing method

As explained earlier, the basic measurement within the accelerated ageing method is an ageing of the full product including its facings for 25 weeks at 70C. This has been carried out on all the eight boards used in this study. The resulting lambda, measured at 10C on the full product is given in graph 4 (left). The lambda increase during this 25 weeks ageing at 70C is given in graph 4 (right).

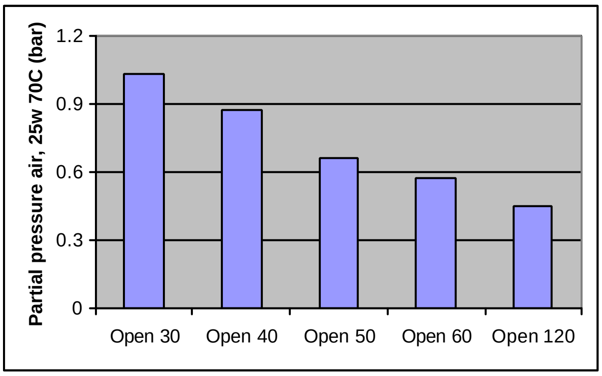
Graph 4. 25 weeks at 70C: lambda value at 10C (left) and lambda increase at 10C (right)



It is interesting to compare the lambda increases of the different boards during the 25 weeks ageing at 70C. As expected, the lambda increase for the 2 diffusion tight boards is very small. For the diffusion open boards, the expected slower lambda ageing for thicker boards is clearly visible in graph 4 (right).

It is valuable to know the partial pressure of air for the different diffusion open boards after ageing for 25 weeks at 70C. When the boards are freshly made, they typically have an air partial pressure of 0.02 - 0.05 bar. When the diffusion open boards are aged at 70C, air partial pressure will gradually increase till it reaches atmospheric pressure (~ 1 bar). The measurement of the air partial pressure was carried out for most of the diffusion open boards and it is given in graph 5.

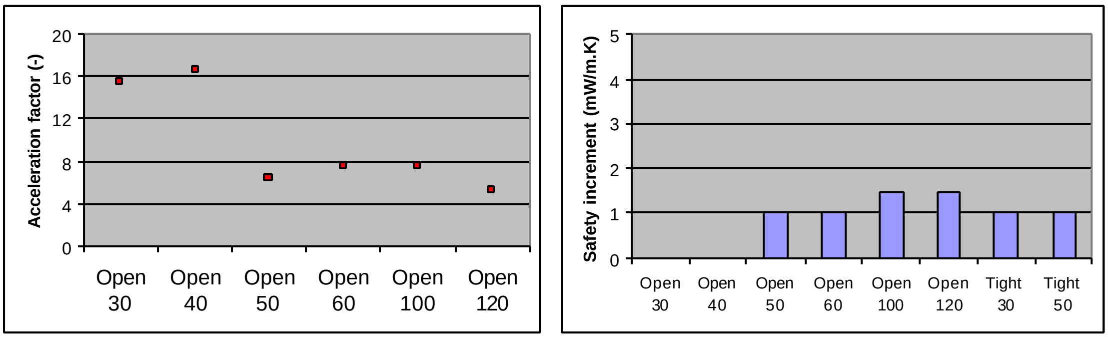
Graph 5. Air partial pressure of diffusion open boards after 25 weeks at 70C



The boards with a thickness of 30 and 40 mm have clearly reached the equilibrium partial pressure for air. It must be noted that, even after a board has reached a partial air pressure of 1 bar at 70C, upon cooling to room T for cell gas analysis, only 0.87 bar pressure will remain based on ideal gas pressure – temperature relation. The fact that the 30 mm board has a measured pressure of about 1 bar is probably due to extra ageing at room T caused by delays in cell gas analysis after taking the specimen out of the oven. However, the main trend in graph 5 is the decreasing air partial pressure as a function of board thickness. The 120 mm diffusion open board has only reached half of its equilibrium air pressure during the 25 weeks at 70C.

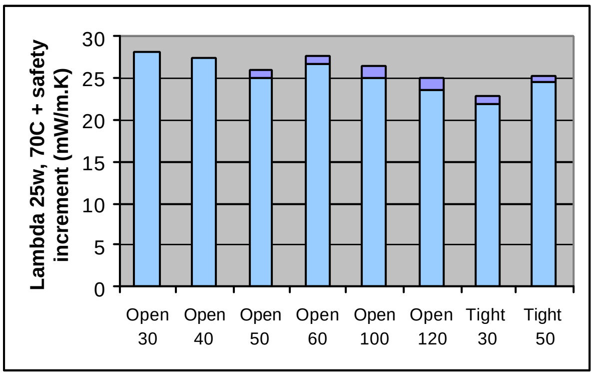
After carrying out this 25 weeks at 70C ageing, a safety increment still needs to be added. The acceleration test offers the possibility to reduce this safety increment for diffusion open boards. This acceleration test has been carried out for the six diffusion open boards used in this study. The result is given in graph 6 (left). It can be seen that 2 out of the 6 boards have an acceleration factor higher than 12, which means that the safety increment can be set to zero. None of the boards has an acceleration factor between 8 and 12, which would mean a reduction of the safety increment. The 4 boards having an acceleration factor lower than 8 can not benefit from a reduction in safety increment. The resulting safety increment, which does not only depend on the outcome of the acceleration test but also on the board thickness, blowing agent and facing type, can be seen in graph 6 (right).

Graph 6. Acceleration factor for diffusion open boards (left). Safety increment (right).



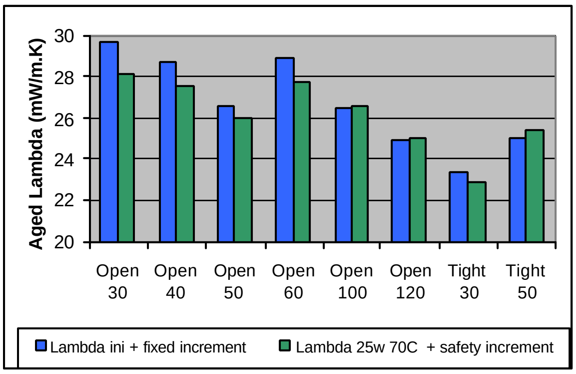
The safety increments can now be added to the lambda after 25 weeks at 70C. This is shown in graph 7. Lambda values between 23 and 25 mW/m.K are obtained for the diffusion tight boards and between 25 and 28 mW/m.K for the diffusion open ones. As it was the case for the fixed increment method, statistics still need to be added to the obtained lambda values.

Graph 7. Lambda 25 weeks at 70C + safety increment.



It is now possible to compare the outcome of both methods (fixed increment and accelerated ageing). The user always has the possibility to choose between both methods. The comparison between the fixed increment and accelerated ageing resulting lambda values is given in graph 8. Again, statistics are not added yet.

Graph 8. Comparison between fixed increment and accelerated ageing method.



For the thick (> 80mm) diffusion open and the diffusion tight boards, both methods give a very similar result. Although we can not generalise this conclusion based on these limited results, it looks like the extra effort of the accelerated ageing method compared to the fixed increment method does not result in a better end result for the boards mentioned. On the other hand, for the thin (< 80 mm) diffusion open boards, the accelerated ageing method is clearly giving a lower aged lambda value. The final declared lambda value could be 1-2 mW/m.K better by doing the extra effort of the accelerated ageing method for these boards.

Comparison with modelling expectations

After comparing the declared aged thermal conductivity by each of the two methods, a pertinent question is "how the resulting value correlates with the time averaged lambda value over 25 years?" All boards used in this study were produced recently and lambda ageing data do not go further than 1 year at the moment. For this reason, it was decided to try to simulate a 25-year ageing curve with Agesim, the Huntsman lambda ageing simulation software. For all boards, a cell gas analysis of the fresh board was done as it is one of the crucial input parameters for the lambda ageing simulation.

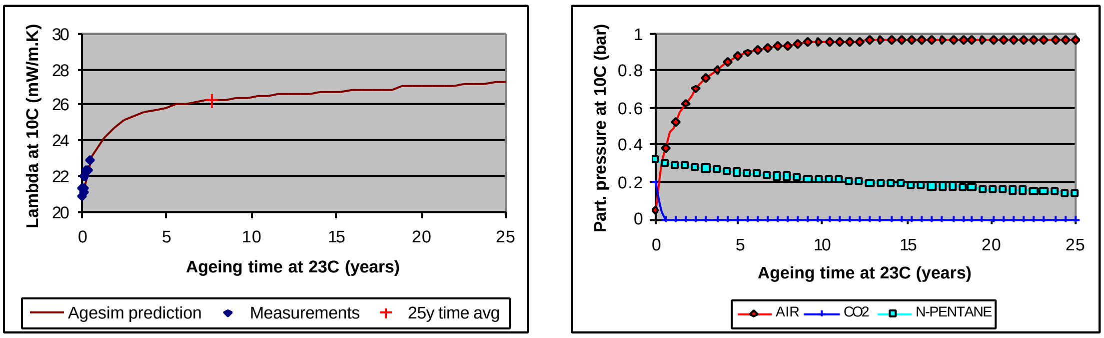
For the diffusion open boards, the other crucial input parameter is the effective diffusion coefficient of air. This parameter will be different for the core foam and the foam close to skin/facing. Although the option to consider separate diffusion coefficients for core/skin is present within our software, the experimental determination of both is not straightforward. For this reason, an overall diffusion coefficient of air has been determined. Since earlier calculations had shown that the exact value of the diffusion coefficients of CO_2 and pentane are less critical with respect to the 25 year lambda-time curve, the default values based on earlier measurements were used.

For the diffusion tight boards, diffusion takes place in the perpendicular direction compared to a diffusion open board. For these boards, the diffusion coefficient of air and CO_2 was determined experimentally. Again, overall diffusion coefficients were determined, without differentiating between foam and skin/facing.

After obtaining these input parameters, the existing short-term lambda-time curves (25 weeks at 70C, normality test, acceleration test) have been fitted. After this evaluation, exactly the same input parameters and k-gas model have then been used for the 25 year at room temperature simulation.

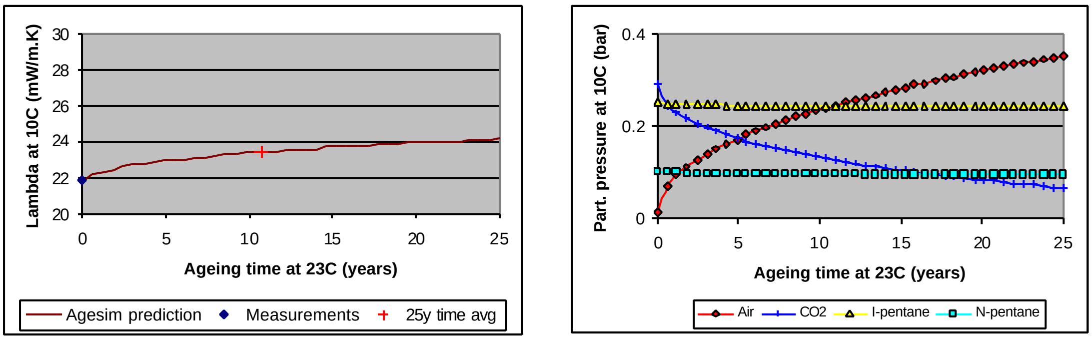
An example of a 25-year simulation is given in graph 9 for a diffusion open board. The left part contains the expected lambda changes, the right part the expected changes in cell gas composition.

Graph 9. Simulation for 50 mm diffusion open board. Lambda (left) and partial pressure (right) evolution.



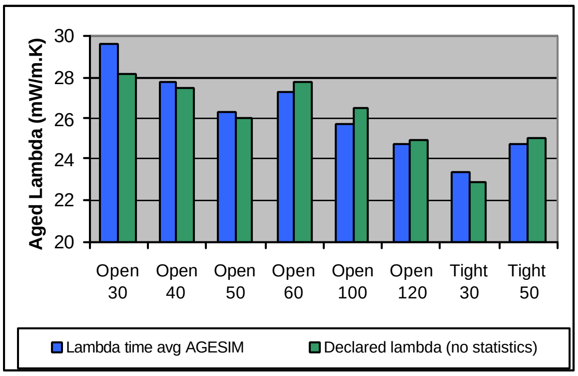
It can be seen from graph 9 that the time averaged lambda value is reached after about 8 years. The lambda measurements obtained during the first year are also indicated on the graph. The CO_2 has completely left the foam after a very short time while the air reaches its saturation level after ± 10 years. Of course these conclusions are valid only for this specific board. Another example is given in graph 10 for a 30 mm diffusion tight board.

Graph 10. Simulation for 30 mm diffusion tight board. Lambda (left) and partial pressure (right) evolution.



Diffusion processes are a lot slower than for the diffusion open board of graph 9. CO_2 is not leaving the board completely during its economic lifetime and air is far from reaching its equilibrium pressure. Diffusion of pentane is negligible. The 25 year time averaged lambda value is reached in a later stage (~12 years), due to the different shape of the lambda-time curve. After this type of work had been carried out for all 8 boards, a comparison can now be made between the simulated 25-year time averaged lambda value and the declared lambda value as obtained in EN13165. For the latter, the lowest lambda value obtained by the 2 different methods is chosen since this is also possible in reality. The comparison is given in graph 11.

Graph 11. Comparison between 25-year time averaged lambda value (Agesim simulation) and declared lambda value without statistics (EN 13165)



It can be seen from graph 11 that there is a good agreement between the simulated and declared lambda value in general. In most cases, the differences between both values is less than 1 mW/m.K. In view of the complexity of the EN13165 and the lambda ageing simulations, the differences as observed in graph 9 are considered as small. Moreover, there is a good balance between slight overestimation's and slight underestimation's by the simulations.

CONCLUSIONS

The following conclusions can be made:

1) In general, the evaluation of EN13165, Annex C on 8 European insulation boards has been very positive and supportive for the new norm.
2) Declared lambda values (without statistics) in the range 25-28 mW/m.K for the diffusion open boards and 23-25 mW/m.K for diffusion tight boards have been measured. Lower values are expected to be possible for fully optimised boards.
3) The 'fixed increment method' and the 'accelerated ageing method' yield a similar declared lambda value for thick (>80 mm) diffusion open and diffusion tight boards. For thin (<80 mm diffusion open boards), the accelerated ageing method gives a 1-2 mW/m.K better result.
4) The comparison between the declared lambda value and the simulated 25-year time averaged lambda value by Agesim, yields very similar lambda values for the 8 boards of this study.

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BIOGRAPHIES

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Kristof joined Huntsman Polyurethanes in 1998 after receiving his Ph.D. degree in Polymer Chemistry from the University of Leuven - Belgium (KUL). He has been working on various fundamental issues of rigid foam such as lambda aging simulations. He is currently a project leader within the Rigid Foam Core Technology Team and is based at Everberg.

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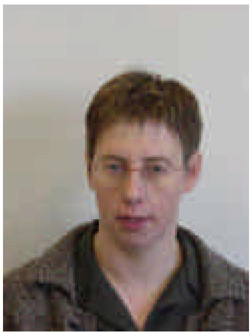
Sachchida joined Huntsman Polyurethanes in 1987 after receiving his Doctor of Science degree in Materials Science and Engineering from MIT. He has worked in a wide variety of application areas of polyurethane. He is currently a PU Application Development Associate in the Construction Group of the Huntsman Polyurethanes Rigid Foam Regional Development Center in West Deptford, NJ.

Marleen Baes



Marleen joined the Chemical Research Centre of Shell in Louvain-la-Neuve in 1992 after receiving her Ph.D. degree in applied biologic sciences from the University of Leuven - Belgium (KUL). She was project leader for the applicational development activities and technical service in the field of rigid polyurethane laminates.
Since the end of 1999 she was transferred to Huntsman Polyurethanes in Everberg. She is currently a marketing officer in the Huntsman Polyurethanes Rigids department and is mainly involved with market extension activities and issue management.