ON THE PREDICTION OF THE RADIATION TERM IN THE THERMAL CONDUCTIVITY OF PLASTIC FOAMS

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***Abstract*– The radiative contribution to thermal conductivity in plastic foams is studied through two different approaches. Both consist in close forms derived from the heat transfer equation governing the intensity of radiation in foams, where scattering can be treated as isotropic. While one approach is based on the solution of the equation for a continuous medium, the other one is based in the solution for a discrete medium. Predictions are contrasted for typical values of thermal properties of foams, and a possible explanation for the found differences is proposed.**

***Keywords*– thermal conductivity; physical properties of foams; thermal radiation.**

I. INTRODUCTION

Heat is transferred in foams through four distinct mechanisms: convection in the gas phase, conduction along the solid polymer, conduction through the gas within the cells, and thermal radiation. Conduction through the gas is the principal mechanism, but radiation is also important, especially for low-density foams. There is a consensus regarding the contributions due to the first three mechanisms to the total thermal conductivity. The modeling of the thermal radiation, on the contrary, continues to be controversial (Collishaw and Evans, 1994; Sirdeshpande and Khanpara, 1993).

The radiative contribution to the thermal conductivity in plastic foams is usually studied through two basic different approaches. On one hand, Glicksman developed an expression using the mean extinction coefficient of the foam (Schuetz and Glicksman, 1984; Glicksman, 1994). This will be referred as the continuous medium approach (CMA). On the other hand, Williams and Aldao (1983) considered a model that takes into account reflection and transmission of radiation through a stack of parallel layers perpendicular to the direction of heat flow. This will be referred as the discrete medium approach (DMA).

The present work was motivated by a previous work (De Micco and Aldao, 2004) where we found that the values of k_r determined with the CMA have a tendency to be much lower than those experimentally found. Conversely, the values for k_r determined with the DMA present a very small average error. That work was based on the foams studied by Almanza *et al.* (2000) who reported the thermal conductivity for polyolefin foams manufactured by means of a high-pressure nitrogen gas solution process. Resulting materials are excellent to check models as they present a much simpler morphology than usual foams.

After presenting the two approaches that lead to expressions for the radiative contribution to thermal conductivity, we address two main points. First, we contrast the resulting thermal conductivity due to radiation using the two described models with recently published experimental results, expanding a recent paper (De Micco and Aldao, 2004). Second, we present a way to figure out the transmissivity of a stack of parallel plates. Thus, we show the inherent limitations in measuring the extinction coefficient using an infrared spectrometer and discuss the appropriateness of applying the Rosseland diffusion approximation.

II. THE TWO APPROACHES FOR THE RADIATIVE THERMAL CONDUCTIVITY

In the first approach to determine the radiant heat flux through a cellular material, the foam is treated as a continuous and isotropic medium. Fortunately, in the case of commercial foams, it is possible to simplify the equation of radiative heat transfer for an absorbing, emitting, and scattering medium. In particular, properties are considered wavelength independent (Glicksman *et al.*, 1987). Thus, for an optically thick foam, the diffusion model for radiant transport can be easily applied. With these assumptions, the Rosseland equation for radiant heat transfer can be derived:

$$q_r = -\frac{16\sigma\overline{T}^3}{3K}\frac{d(Temperature)}{dx}, \qquad (1)$$

where K is the mean extinction coefficient, σ the Stefan-Bolzmann constant, and $\overline{T}$ the mean absolute temperature. The extinction coefficient is related to the radiation lost due to scattering and absorption per unit distance in a given material. The radiant thermal conductivity is then given by (Glicksman, 1994)

$$k_r = \frac{16\sigma\overline{T}^3}{3K}. \qquad (2)$$

The second approach consists in applying the equation of heat transfer for each cell wall that constitutes the foam, taken into account the discreteness of the medium. Williams and Aldao (1983) solved a simplified model consisting in a stack of parallel plates with a separation distance equal to the cell size. In doing it, cell walls are considered partially transparent, partially reflecting, and partially absorbing. Thus, the same phenomena are included in both the continuous and discrete medium analysis.

To determine the radiant thermal conductivity, heat fluxes in each cell are figured out by applying a method known as the net radiation method (Siegel and Howell, 2002). The trick consists in considering fluxes that include all possible origins: emitted, reflected, and transmitted contributions. For the *i* plate, see Fig. 1, we can write

$$S_i^+ = TS_{i-1}^+ + (1-T)S_{i+1}^-, \tag{3}$$

where T is the fraction of the incident radiation that in steady state reaches the other side of a cell wall -the net transmittance-, (1-T) is reflected back, and S_i^+-for example- is the flux leaving plane i to the right.

The discrete medium approach leads to a closed form for the net radiation through a shield formed by a series of plates that can be partially transparent. (interestingly, it can be shown that the net radiation through a structure formed by in-line three-dimensional cells is the same than that for the stack of plates.) The expression for the radiant thermal conductivity obtained is

$$k_r = \frac{4\sigma \bar{T}^3 L}{1 + n(1/T - 1)}, \tag{4}$$

where L is the foam thickness, and n the number of plates. It is important to remark that the net transmittance is defined as the fraction of the incident radiation that in steady state reaches the other side of a plate due to partial transparency and re-emission of the absorbed energy. T is given by

$$T = \frac{(1-r)(t+1)}{2(1+rt)}. \tag{5}$$

T depends on r, the fraction of the incident radiant energy reflected by each gas-solid interface, and on t, the fraction of radiant energy transmitted through the solid membrane. r can be related to the refraction index w as

$$r = [(w-1)/(w+1)]^2, \tag{6}$$

and t is given by the Bouguer's law

$$t = \exp(-aL_s), \tag{7}$$

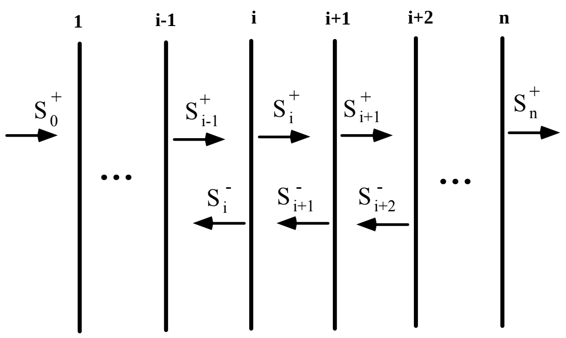

Fig. 1. Scheme of radiant energy through a set of *n* solid plates used to determine the radiant thermal conductivity in a plastic foam.

where a is the absorption coefficient and L_s the cell wall thickness.

III. RESULTS AND DISCUSSION

To compare theoretical values with the experimental measurements of k, the thermal conductivity due to conduction must be subtracted from the measured value of conductivity (convection can be neglected because of the cell sizes). This parallels the analysis of Almanza *et al* (2000), who based their analysis on the work of Schuetz and Glicksman (1984). We focus on the nine foams studied by Almanza *et al* (2000) that were based on low-density polyethylene for which all the relevant parameters are reported.

Our findings are very surprising. The values for k_r determined with the CMA have a tendency to be much lower than those experimentally found, with an average error of ~50%. Conversely, the values for k_r determined with the DMA present an average error of only ~4%. These results indicate that while the DMA reproduces the experimental results very well, the CMA strongly underestimates the radiant thermal conductivity.

To determine K in applying Eq. 1, the transmissivity is usually measured with an infrared spectrometer over the spectral range for which there is substantial radiant energy emitted, from 5 to 30 microns (foams are approximately gray) (Schuetz and Glicksman, 1984; Almanza *et al.*, 2000). This measurement is carried out for several slices with different thickness in the range 0.5-3 mm [slices with widths in the smaller range 0.5-1.5 mm were used by Schuetz and Glicksman (1984) and by Glicksman (1994)]. The extinction coefficient then follows from a plot of the ratio of the transmitted to the incident beam intensity, S/S_0, vs. thickness,

$$S/S_0 \propto \exp(-KL). \tag{8}$$

We showed recently (De Micco and Aldao, 2004) the radiant thermal conductivity as a function of thickness for one of the samples studied by Almanza *et al.* (2000). We found that the determined extinction coefficient can result much larger than that corresponding to the thicker foams used to measure the thermal conductivity. Thus, an error can be traced to the way that the extinction coefficient is measured. In particular, it is erroneously assumed that the extinction coefficient is independent of thickness. Eq. 8 would be only applicable in cellular materials if the number of cells were sufficiently high. In contrast, the measurements are carried out with samples having too low a thickness for Eq. 8 to be valid.

In order to check the validity of the CMA in studying the behavior of a cellular material, we will apply next the net radiation method in the conditions in which the extinction coefficient is determined. Now, the fraction of the radiation absorbed at plates does not have to be considered, so the fraction transmitted, T, and the fraction reflected, R, do not sum one. The stack of plates is depicted in Fig. 1 and the net radiation fluxes satisfy the following relationships

$$S_i^+ = TS_{i-1}^+ + RS_{i+1}^- , \quad (9)$$

$$S_{i+1}^- = RS_i^+ + TS_{i+2}^- , \quad (10)$$

$$S_{i+1}^+ = TS_i^+ + RS_{i+2}^- . \quad (11)$$

From Eqns. (9)-(11) a recurrent relation can be obtained

$$\frac{S_i^+}{S_{i-1}^+} = \frac{1}{C - \frac{S_{i+1}^+}{S_i^+}} , \quad (12)$$

where C is given by

$$C = \frac{1 + T^2 - R^2}{T} . \quad (13)$$

In this case, since re-emission of the absorbed energy is not included, the net transmittance T is the fraction of the incident radiation that in steady state reaches the other side of a plate due only to partial transparency. Then T is given by

$$T = \frac{t(1-r)^2}{1 - r^2 t^2} . \quad (14)$$

The fraction of the incident radiation flux, R, that is reflected can be calculated with

$$R = \frac{r\left[1 + t^2(1-2r)\right]}{1 - r^2 t^2} . \quad (15)$$

The experimental curve used to determine the extinction coefficient, transmission as a function of foam thickness, can be determined theoretically by means of Eq. 12. To start the recurrent relation, we need to consider that for the last plate $S_n^+ / S_{n-1}^+ = T$. A typical value for the refractive index is w=1.6 and then, using Eq. 6, r=0.053. Glicksman (1994) states that the transmissivity of a 2 μm thick film is about 0.8. Thus, with the help of Eqs. 7 and 14, the absorption coefficient a is found to be 0.58 μm^{-1}. Similar values were proposed by Almanza *et al.* (2000).

According to Glicksman, commercial foams present cell walls in the order of 0.5 μm thick. Therefore, with Eq. 7 we can determine t=0.971. Thus, for a typical foam having r=0.053 and t = 0.971, we can figure out, using Eqs.14 and 15, that T=0.873 and R= 0.098. Next, we can calculate C=2.0075 using Eq. 13. Finally, the recurrent relation given by Eq. 12 can be applied to determine the radiation flux as a function of the number of plates, *i.e.* the sample thickness.

In Fig. 2, the resulting transmission as a function of the number of plates is shown. It can be noted that the slope is not constant. This implies that Eq. 8 is not applicable for foams with a low number of cells and the measured slope is not the same that the one that corresponds to the thick foam under study. To analytically quantify the error involved, we can determine the values of the extinction coefficients for a stack of n plates corresponding to the limit cases of n=1 and n=∞:

$$K_1 = -\ln(T)$$

$$K_\infty = -\ln\left(\frac{C - \sqrt{C^2 - 4}}{2}\right) . \quad (16)$$

For the example given above, K_1/K_∞=1.57, which implies an error that can be as large as 57 % in the determination of the extinction coefficient. (The same result is obtained figuring the extinction coefficients out as seen in Fig. 3.)

So far, expanding our previous work (De Micco and Aldao, 2004), we have presented the problems originated in the determination of the extinction coefficient. We have also assumed that the Rosseland equation is appropriate to predict the thermal conductivity of foams. In what follows we raise some concerns in this respect.

The Rosseland equation represents a very useful approximation to treat radiation since, even for simple one-dimensional geometries, finding the heat flux can become quite hard. Practically, to find the radiant thermal conductivity implies the determination of the extinction coefficient. As discussed above, K is measured from the decrease in the intensity of an external radiation as it traverses samples of different thickness. What we have developed, Eqs. 12-15, represents the analytical result for this type of measurement applied to a set of parallel plates that are partially transparent, partially reflecting, and partially absorbing.

In Fig. 3 we compare the resulting thermal conductivity for the DMA and the CMA in the limit of an infinitely thick foam, *i.e.* for a very large number of plates. For the DMA, we applied Eq. (4) in the limit n→∞. For the CMA, we used Eq. (2) where K is determined with Eq. (16). A typical value of r was adopted (0.053) and k_r is plotted as a function of t. At first sight, results are striking. As expected, both approaches lead to monotonous functions of t. However, k_r for the DMA presents a finite thermal conductivity for t=0 and t=1 while for the CMA k_r becomes null for t=0 and infinite for t=1.

A null value for t implies an opaque cell wall. Nevertheless, the net transmittance need not be null as the absorbed energy is re-emitted backward and forward and thus the DMA predicts a smaller but finite

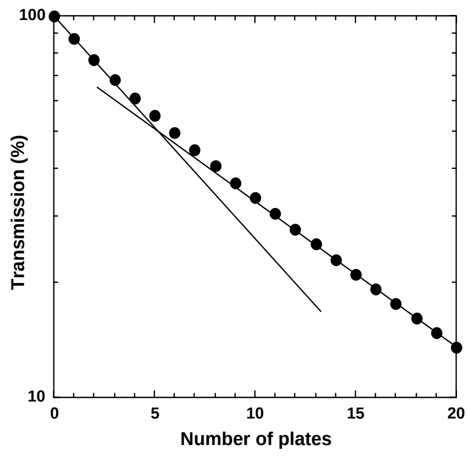

Fig. 2. Transmission as a function of the number of plates for r=0.053 and t = 0.971. Straight lines show the slopes corresponding to a very low and very high number of plates. The slopes differ by a factor of 1.57.

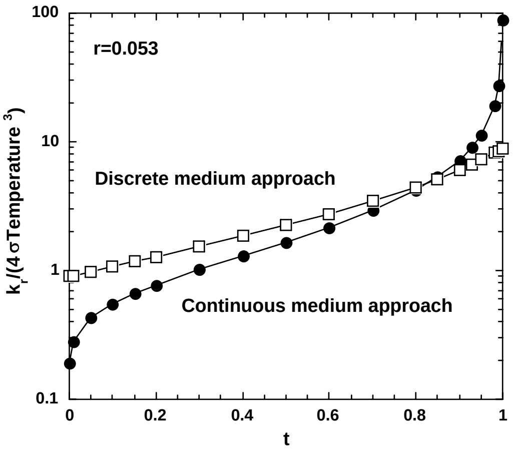

Fig. 3. Radiant thermal conductivity as function of t for r=0.053 determined with the discrete and the continuous medium models.

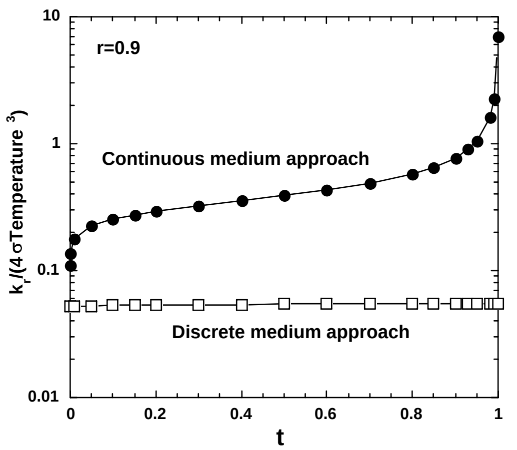

Fig. 4.. Radiant thermal conductivity as function of t for r=0.9 determined with the discrete and the continuous medium models.

value for k_r. Since an opaque wall implies that an external radiation does not traverse samples of any thickness, the extinction coefficient for the CMA is infinite implying that k_r is null.

A unity value of t means that cell walls do not absorb radiation. In this case, the DMA predicts a finite value for k_r as multiple reflections reduce heat transfer. Conversely, the CMA predicts an infinite value for the thermal conductivity because there is no manner for radiation to extinct.

The outcomes for the CMA and the DMA diverge more and more with r. Indeed, in Fig. 4, we present the thermal conductivity for a very large value of r equals to 0.9. As above, with the CMA k_r becomes null for t=0 and infinite for t=1. Now, the models predictions are remarkably different.

The above findings indicate that the Rosseland equation can lead to very wrong results in describing radiation in cellular materials. The Rosseland equation approximation was meant to be applied far from boundaries. The present problem consists of a stack of plates where boundaries play a central role. We conclude that the Rosseland approximation has been used where it is not appropriate because it is outside its domain of validity.

IV. CONCLUSIONS

In agreement with Almanza *et al.* (2000), we found that the geometric model of Aldao-Williams fits better the experimental results for the thermal conductivity of plastic foams. An explanation for these findings is suggested. Essentially, we propose that slices used to determine the extinction coefficient are not thick enough to apply the diffusion limit approximation and, as a consequence, the values obtained for K with thin slices cannot be used for thicker samples. We derived a way to analytically determine the extinction coefficient for a stack of parallel plates that show that the transmission does not necessarily depend exponentially on the sample thickness. The reciprocal of the extinction coefficient is 1 mm or less, much smaller than the overall dimensions of commercial foams. Thus, one is prone to model the transfer process as a diffusion process. However, we found that the discrete character of the foams cannot be overlooked. Furthermore, we found that, in general, the Rosseland equation is not a good approximation to describe heat transfer in foams because of the discrete character of the material.

NOMENCLATURE

a Adsorption coefficient of the plastic
k_r Radiant thermal conductivity
K Mean extinction coefficient
L Foam thickness
L_s Thickness of a solid membrane
n Number of plates
q_r Radiant heat transfer
r Fraction of the incident energy reflected by each solid-gas interface
R Fraction of the incident radiation flux reflected
S_i^+ Heat flux leaving plane i to the right
S_i^- Heat flux leaving plane i to the left
T Net transmittance
t Fraction of energy transmitted through a solid membrane
$\overline{T}$ Mean absolute temperature
w Refractive index of the plastic
x direction x

Greek symbols

σ Stefan-Boltzmann constant

Subscripts

i component leaving plane i

Superscripts

\+ component leaving to the right
\- component leaving to the left

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