# STUDY OF BONDING RATE OF POLYMERIC FILMS WITH A VIEW TO DEVELOPMENT OF SIMULATION PROCEDURE FOR CONTROL OF HEAT SEALING CYCLE

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# ABSTRACT

## STUDY OF BONDING RATE OF POLYMERIC FILMS WITH A VIEW TO DEVELOPMENT OF SIMULATION PROCEDURE FOR CONTROL OF HEAT SEALING CYCLE

by David Pegaz

The lack of direct relationship between the heated bar temperature, dwell time and pressure used traditionally in heat seal strength prediction and control has been realized and in order to overcome it, application of heat transfer theory to computation of bonding interface temperatures has been attempted in the past.

This study includes detailed investigation of the deviations between the temperatures computed under several Initial and Boundary Value Problem Models and the experimentally obtained temperatures, identifying the causes for these deviations and determining a suitable model which can be used for the computations and whose results are independent of the heat sealer construction.

Several theories of bonding mechanisms in heat sealing are reviewed and on the basis of these, variables such as viscosity, fluidity and diffusivity ratio are computed in addition to temperature at the bonding interface.

A large number of bond strength determinations of heat seals, made under varying conditions, are tabulated and their plot results in a set of indifference curves believed to be characteristic of the material used.

Within the range studied, the effects of pressure increase on the bonding rate are observed with the conclusion that pressure in excess of approximately 1.6 p.s.i. slows the bonding process.

The indifference curves show clearly the transient nature of the bonding process proving the fallacy of associating a given temperature with a fixed level of bond strength.

Finally, a simulation procedure is proposed, by means of which control of heat sealing conditions is possible and heat seal strength resulting from such conditions is predictable.

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By

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# INTRODUCTION

Heat sealing, a process by which polymeric materials are bonded by application of heat and pressure, is used extensively in a large variety of fabricating methods in the manufacture of widely different products, from raincoats to packages. There is hardly any package made from flexible packaging materials in which heat sealing has not been used.

In spite of this wide use and the success with which this technique has been employed, the exact nature of the heat seal bond formation is not completely understood. Suppliers and manufacturers of polymeric films usually report material heat sealing temperature ranges of 30 to 150 degrees F, depending on the type of heatsealer employed, where the reported temperatures refer to the hot bar of the heatsealer. Since the temperatures of the film at the bonding interface are lower than either the lower or the upper limits of the heat sealing temperature range reported, the useful information obtained from such a specified range is rather limited and reduces to the following:

a. The lowest temperature at which heat sealing is possible is lower than the low level of the reported heat sealing temperature range.

b. The highest temperature, a heatsealer heated bar can have before degradation of the film occurs, corresponds to the upper level of the range.

In order to gain more information on the film's heat sealing properties, users and researchers have tabulated heatsealer temperatures, pressures and dwell times against heat seal strength values in order to determine the best heat sealing conditions. The main disadvantage of these results is that they are valid only for the experimental heat sealing system. No conclusive information can be obtained from them for general application.

An improvement in the approach to determination of the best heat sealing conditions was achieved by the use of heat transfer theory, which enables the computation of the bonding interface temperature, so making the relationship of the tabulated values of heat seal strength to the temperature levels more meaningful.

The validity of the computed temperatures depends heavily on the degree of analogy between the physical model, the heatsealer and the film, and the mathematical model used in representing it in the computational scheme. In the language of mathematicians the computational scheme is referred to as a Boundary Value Problem whose accuracy in representation of the real case depends on the propriety of assumptions describing the Initial and Boundary conditions of the real system.

A recent development of surface temperature measuring made it possible to investigate the agreement between the bonding interface temperatures, calculated by means of different computational models, and experimentally determined temperature.

In this project the writer attempted, after investigation of the reliability of the above mentioned surface temperature measuring method, to determine the most suitable mathematical model possessing the necessary characteristics of adaptability so that a wide variety of heatsealers with different physical constructions could be accommodated. Further, various proposed bonding mechanisms of polymers were surveyed, with a view to possible application to heat sealing. Borrowing from studies conducted on the phenomena of adhesion and autohesion of high polymers, the mechanism of diffusion was found to be most applicable. Viscosity, fluidity and diffusivity were added to temperature as the variables of interest and their computed values were used in interpretation of heat seal strength results obtained in a series of tests with sealed polypropylene.

Although the results do not lead to an easily expressible formula, an attempt was made to construct a procedure by means of which a heat seal strength can be predicted or, if a strength level is predetermined, the conditions at which the seal has to be made can be found.

Since the conclusions of this study deal with the strength - temperature - time relationship, the above procedure can be used in determination of any one of the three variables, given the desired values for the other two.

There is some indication of the relationship between heat sealing conditions and changes in barrier properties of polymeric films used in packaging, an area into which a further study is presently contemplated at the Michigan State University School of Packaging.

The outcome of the above study together with the results discussed in this paper could be combined into a design evaluation program for flexible packages by means of which optimal design characteristics could be found using the heat seal strength and the permeability or gas transmission rate as constraints.

The writer believes that charts of bond strength vs. time with temperature, viscosity, fluidity indifference curves should be added by the film manufacturers to other physical and chemical data characterizing each particular material and should be made available to every user who would then obtain useful information more meaningful than the customarily reported heat sealing range.

# BACKGROUND

Throughout the literature dealing with heat sealing, attempts can be found to define formally the phenomenon. The definitions imply their author's opinion on the nature and mechanism of bond forming.

Scarpa (1)[*], in his paper on joining plastics with ultrasonics, views the surface of the solids to be bonded as having peaks, or asperites, that tower over adjacent valleys by several hundreds to many thousands of atomic diameters. When two such surfaces are put in contact, only the tips of their peaks touch. In ultrasonic bonding the rapid and powerful impacts cause a flatening of the surface peaks and, on microscopic level, cause the material to flow to either side of each peak. This increases the contact area and also forces the plastic surfaces to move not only vertically but laterally as well. The friction of this motion generates intense heat at the bonding interface which not only contributes to the tendency of the plastic to flow, so further increasing the area of contact, but also, within a few microns thickness of the interface, stress-relieves the new bonds as they form. These bonds make up the adhesive properties accounted for in general by the Van der Waal's forces of interatomic or intermolecular attraction, including interaction of polar groups and hydrogen bonding.

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[*] Numbers denote the source as listed in List of References on Page 74 .

This explanation places clearly the bonding process into the realm of electrochemical phenomena, where the pressure and heat serve only as means of achieving the necessary interatomic and intermolecular proximity for the forces of attraction to take hold.

A different mechanism of bonding is implied by Stuwe (2), who maintains that fusing of thermoplastic polymers is obtained by heating the surfaces of the two layers to be bonded up to a melting temperature while subjecting them to such a pressure that molecular groups near the surfaces flow one into another. He further points out that this temperature has to be below the decomposition temperature of the material bonded, concluding that the temperature distribution at the bonding interface is of particular importance.

Stuwe's approach seems to attribute the formation of a bond to thermal motion of the molecular chains, or parts of them, which interlock across the interface. He does not though, discuss the nature of the forces keeping them in place which may be either rheological only (mechanical) or Van der Waal's type forces of attraction.

Voyutskii (3), in his paper on the adhesion and autohesion of polymers, takes a critical look on the theories by which at the present the adhesion of polymer to polymer can be explained. Making use of experimental results of a number of Russian researchers, he examined the adsorption, electrical and diffusion theories as related to adhesion of polymers, noting in particular whether the theories were supported or refuted.

The adsorption theory regarding adhesion as a purely surface process explains the strong bond formation as a result of the action of variety of molecular forces. This theory, however, has serious

shortcomings. First, while the work of peeling may be as great as $10^4 - 10^6$ erg/cm$^2$, the work required to overcome the molecular forces is only $10^2 - 10^3$ erg/cm$^2$.

Second, the work of peeling an adhesive bond depends on the rate of separation, whereas the work expended on overcoming the molecular forces should not depend on the rate at which the molecules separate.

Third, the adsorption theory cannot explain the good adhesion between non-polar polymers.

To explain the facts which do not come within the framework of the adsorption theory, an electrical theory of adhesion has been proposed (4). In this theory the adhesive-substrate system is viewed as condenser coated with a double electrical layer formed by the contact of the two substances of different nature. During delamination of the bond a potential difference develops, increasing to a certain limit, until discharge occurs.

Although this theory conveniently accounts for the rate of separation dependence of work of peeling, a number of factors limit its application to the mutual adhesion of high polymers (5).

First, high polymers are able to bond together strongly even when there are no electrical effects during lamination.

Second, in the case of high-polymers that are dielectrics, it is difficult to assume that electron transfer occurs to any considerable extent from one polymer to another.

Third, the more similar the nature of the polymers, the greater is their adhesion, while by the above theory, a reverse dependence should have been observed, because the more similar the phases brought into contact, the smaller should the potential difference become.

Fourth, if adhesion were determined only by the formation of a double electrical layer, adhesion between polymers filled with carbon black would be impossible because such mixtures are good conductors.

It seems then, that the electrical theory of adhesion is applicable only to polymers which are mutually insoluble. For mutually soluble polymers which are non-polar the electrical mechanism is impossible and the formation of bond is probably due to interweaving of the molecular chains of the surface layers as a result of their mutual diffusion. The formation of bond between polar polymers may give rise to electrical layers, but if the molecular chains or their segments are capable of intense thermal motion there will be cross-linking of both layers as a result of diffusion. As the time of contact increases, the effect of diffusion becomes more and more important because of the elimination of the surface of contact and the increase in depth of penetration. The above conclusion has been also reached by Morozova and Krotova in experimental study of the relative rate of electrical and diffusion processes in the adhesion of polymers (6).

The diffusion theory explains adhesion, like autohesion, to consist of diffusion of chain molecules or their segments which form the strong bond between the two layers. Autohesion strength has been found to depend on viscosity of the bonding polymers in which, the lower their viscosity the more easily self-diffusion will occur. Support for viewing autohesion as a diffusion process has been obtained by experimentally determined relationships between the autohesive strength and the time of contact at constant temperature of

polyisobutylene (7, Figure 1), and between the autohesive strength and the temperature with constant contact time (7, Figure 2). In the time-of-contact function the strength at first increases rapidly but slows down tending toward a definite limit. The strength as a function of temperature shows, over the temperature range studied, an exponential relationship. Both the above characteristics seem to be in accord with the diffusion nature of the process. One additional finding reported by Voyutskii (7) supports the notion that molecular chain segments of approximately equal magnitude participate in the diffusion process. This finding shows the molecular weight independence of the computed activation energy of autohesion for polyisobutylene, which indicates the identical nature of kinetic units participating in diffusion.

The diffusion approach also conveniently accounts for the difference in bonding rates and temperatures of polymers with diverse molecular structures. The ease of bonding can be now related to the relative flexibilities of polymer chains which decrease by inclusion of bulky sidegroups, unsaturations, and polar groups. The same relationship can be observed in the characteristic viscosities of different polymers or in their characteristic glass-transition temperatures (8).

Experimental studies of heat-seal bond strength were traditionally attempting to view the bond strength as a function of heated bar temperature, heat-sealing time and pressure. This resulted in efforts to improve the experimental heatsealer design so as to increase the degree of control over the three variables--temperature, time and pressure (9, 10). Results of tests conducted even on these improved laboratory heatsealers could not be, however, correlated either with similar results obtained on another laboratory heatsealer, or with results obtained on actual production equipment. This lack of correlation has been caused mainly by two factors. One, the temperature of the heatsealer heated bar is no indication of the temperature which the bonding interface experiences and which is determined in addition to the temperature gradient (heated bar - initial temperature) by the heat transfer properties of the total heat sealing assembly. Two, the heat sealing time, referred to as dwell time, while it represents the length of time for which the outer side of the heat sealing assembly is exposed to the heated bar temperature, does not give any information on the temperature-time function at the bonding interface.

The shortcomings which prevented the meaningful interpretation of heat sealability studies of various materials were realized and the use of heat transfer theory for computation of the bonding interface temperatures were proposed by several writers (2, 11, 12).

Mathematically, the determination of the temperature at the bonding interface involves the solution of the classical heat transfer equation formulated on the basis of Fourier's Fundamental Law of Heat Conduction (13). This solution has to satisfy the Initial and Boundary Conditions by means of which the particular model, whose solution is being sought, is characterized. The Initial and Boundary Conditions influence greatly the computed results which would be only as good as is the accuracy of representation of the actual model, the heat sealing assembly, in terms of mathematically expressed Boundary Conditions.

Several mathematical models of heat sealing assemblies are found in literature (2, 11), but to this writer's knowledge, no extensive studies of heatsealability using the computed bonding interface temperatures were undertaken to this day, partly because of lack of confidence in the accuracy of representation of the actual system and the unavailability of direct measurement method.

A recently developed method for measurement of temperatures of thin plastic films (14) has made it possible to compare the temperature functions computed under different Boundary Value assumptions with the actually measured one.

The determination of the temperature functions under the different models involves considerable amount of computation, hence the use of a digital computer was found to be necessary. The models, described in detail in Appendix I, have shown significant deviations in computed solutions from the experimentally determined temperature function, with the exception of Model IV whose computed function almost coincides with the measured one, due to its flexibility of adaptation with which the physical characteristics of the experimental heatsealer have been accommodated.

It is this flexibility which makes possible the study of temperature - time - heat seal strength relationship independent of the heatsealer construction and enables the observation of the transient nature of temperature, viscosity, fluidity, and on the temperature dependent diffusivity at the bonding interface.

For experimental determination of the heat seal bond strength a method was employed, which represents a combination and modification of two test methods used in determination of bond strength of heat seals and adhesives (9, 15) and ASTM method D 832-61T for testing the tensile properties of thin plastic sheeting. The experimental procedure described in detail under EXPERIMENTAL on Page 32, was chosen for two reasons. One, the writer believes that it is sufficiently instrumentally controlled to give interlaboratory reproducibility. Two, the manner of loading is the same as used in ASTM method D 832-61T for tensile properties determination, so allowing the test results to be viewed in terms of the tested material's tensile, yielding, and breaking characteristics.

The material used throughout the experimental part of this study, polypropylene, was selected because of its relatively high melting point which enabled the testing of the surface temperature measurement method at temperatures up to 250 degrees F, considerably higher than previously tested (14), and because it is considered to be somewhat more difficult to heat seal than polyethylene or PVC.

A few links are still missing in the process of application of the diffusion theory to derivation of a quantitative bond strength prediction scheme, either by using a flux diffusing across a surface, or by use of formulas derived by Vasenin (16) on the basis of physiochemical considerations. This should not hinder the utilization of information obtained in this study for production control or design optimizing procedures. Both of the above mentioned approaches require the knowledge of the number and the depth of penetration of the diffusing molecular chain segments. To the writer's knowledge, no such experimental study with polymers has been undertaken to this time, although, some mthods applicable to polymers have been employed in other material science areas such as metallurgy (17). One such method involves the use of radioactively - labeled molecules which make it possible to observe the rate of diffusion of one component in a two-component system of uniform chemical composition. The observation could be done by autoradiography or any other available radioactivity detection method. The experimentally determined concentration function could then be used in computation of diffusion flux which should be directly related to bond strength.

Even without the availability, at the present, of the clearly defined relationship, the results of this study enabled the plotting of a set of temperature - viscosity - fluidity - diffusivity indifference curves, which the writer believes to be characteristic of the particular material tested. Since the bonding characteristic of any given material is influenced by any number of factors related to formulation and manner of fabrication, the results of this study could not hold for polypropylene films in general. However, if a family of indifference curves were added to the properties which a film manufacturer makes available to users, and which are specified for each formulation, these could be then used in determining the necessary heat sealing apparatus settings which yield a desired bond strength level.

A relationship between heat sealing temperature, dwell time and loss of barrier properties was observed (18, 19) and a study of it is now being contempleted at the Michigan State University School of Packaging. Results of this study, provided some definite relationship will be found, would lend themselves to be used in conjunction with the bond strength curves in design optimizing method by means of which the bond strength - barrier properties affecting variables could be determined, hence the heat sealing apparatus settings. Also, given the apparatus settings, the strength and the barrier properties of the package could be predicted.

# THEORETICAL

The following is the discussion of the relevant theories related to the various aspects of this study.

## Adhesion - Diffusion

Vasenin (16), in his classification of adhesion phenomena by type of bond and mechanism of adhesion, characterizes diffusion as follows:

| | |
|---|---|
| Nature of the forces | Intermolecular |
| Magnitude of the forces | $10^{-6} - 10^{-8}$ |
| Energy of the bond, erg/bond | $10^{-14} - 10^{-16}$ |
| Mechanism of formation of a bond | Interpenetration of macromolecules and of parts of them as a result of diffusion. |
| Main parameters influencing the mechanism of adhesion | Intersolubility and mobility of the macromolecules. |
| Mechanism of failure of the adhesion combination | Extraction of terminal portions of macromolecules with slight depth of interpenetration and rupture of chemical bonds with deeper penetration. |
| Field of applicability | Intersoluble polymers in a physical state allowing retention of a certain mobility of the molecular chain. |

On the basis of this characterization, Vasenin discusses the theoretical implications which lead to a quantitative interpretation, all of which are quoted below.

"Diffusion Phenomena in Adhesion.

> 'The question of the part played by diffusion in the phenomena of sticking arose in connection with the investigation of the autohesion of high polymers (2, 22, 25, 26*). It was established that autohesion depends on the time of contact, temperature, the molecular weight, the chemical nature, and the physical state of the polymer. The influence of these factors is easily explained by the diffusion theory of autohesion. Thereafter diffusion conceptions were used for explaining the adhesion of high polymers. The mechanism of formation of the adhesive bond may be represented as follows (2).'
>
> 'In the contact of two polymers as a result of thermal motion there occurs an interpenetration of the chains, governed by the diffusion of the terminal or median segments of the macromolecules. The portions of the macromolecules which have diffused are retained in the polymers by intermolecular forces. The strength of the adhesion combination which is formed is proportional to the number of molecular chains intersecting the interface and to the depth of penetration of the macromolecules. The latter depends upon the time of contact, the external conditions, the chemical nature of the polymers and their physical state. The applicability of diffusion conceptions to real system is determined by the fulfillment of the following set of conditions (2):
>
> (1) the thermodynamic condition:
>
> $$\Delta Z_{sol} = (\Delta H_{sol} - T\Delta S_{sol}) < 0$$
>
> the condition of intersolubility of the polymers, and (2) the kinetic conditions.'
>
> 'The macromolecules or portions of them must possess sufficient mobility to effect a process of interpenetration. The mobility may be increased by raising the temperature or by mixing the polymers with low molecular weight substances.'

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[*] References throughout the quoted text are Vasenin's.

'The role of diffusion in adhesion is confirmed by many experimental facts:

(1) a connection has been found between the inter-solubility and adhesion of polymers (27, 28);

(2) there has been established experimentally the disappearance of the phase boundaries in the contact of mutually soluble components (29);

(3) there has been demonstrated an increase in adhesion on increasing the duration of contact (30): this being a necessary but not adequate condition for the applicability of diffusion concepts to real adhesive-substrate system. In adhesion to porous bodies and also with the adsorption mechanism of formation of an adhesive bond as a result of decreasing flow and adsorption of polymers we also observe an increase in adhesion with increasing time of contact;

(4) an increase in adhesion has been established with increasing temperature of contact (30), but its exponential character cannot, of course, be used as an unambiguous proof of the diffusion mechanism of the formation of the adhesion bond, since the same type of dependence of adhesion on temperature is observed also with adhesion to porous bodies, and with adhesion due to chemisorption of an adhesive on a substrate;

(5) with increasing molecular weight of polymer (3) there is a reduction in the number of terminal segments capable of diffusion, therefore with increasing molecular weight the adhesion decreases;

(6) the rate of diffusion depends upon the magnitude and shape of the diffusing molecules (32); the greater the dimensions of the molecules and the more short side branchings they have the lower the rate; it has been established experimentally (33) that in the adhesion of polybutadiene to cellophane, increasing the number of butadiene groupings in the 1, 2 position reduces the adhesion; sufficiently long side-branchings play the part of terminal segments and increase the adhesion;

(7) the flexibility of the macromolecules has an exceptionally great importance in the interdiffusion of polymers (33) but the quantitative influence of this factor on adhesion is very difficult to investigate; it is established for instance that increas -

ing the number of styrene groupings in a butadien-styrene copolymer reduces the adhesion as a result of the reduction in the flexibility of the chains.'

'Quantitative Interpretation.'

'A quantitative interpretation of these experimental facts is a difficult matter. Its complexity lies in the impossibility of varying only one of the factors which influences adhesion while maintaining the others constant. For instance, in investigation the effect of structural peculiarities of macromolecules on adhesion, one must note that diffusion is altered, not only by the geometrical form, but also by the flexibility of the macromolecule. In addition, constant molecular weight of the polymer must be maintained otherwise this too will affect the adhesion.'

'Adhesion of High Polymers.'

'To predict adhesion quantitatively it is necessary to solve three problems (34): 1) the nature and magnitude of the intermolecular forces governing adhesion: 2) the number of macromolecular chains intersecting the interface in the formation of a bond: 3) the depth n of penetration of macromolecules.'

'In the first problem we must realize that if the molecules of a high polymer diffuse mutually to a depth corresponding to the length of a segment of a macromolecule with a degree of polymerization below 200-600, then destruction of the bond takes place with sliding of the chains. If the portion which has diffused is greater, then destruction of the bond occurs by chemical bond rupture of the macromolecules. The critical value of the length n (i.e., $n_{cr}$) of the molecule in terms of groups of atoms, at which an alteration in the mechanism of failure occurs, is determined by the ratio:

$$f_i \cdot n_{cr} = f_{ch}$$

where $f_i$ is the force of intermolecular interaction of the atom group; $f_{ch}$ the force required for rupture ofa chemical bond. Then the conditions under which there is effected one or the other mechanism of destruction of contact will be:

$f_i \cdot n < f_{ch}$ the mechanism of chain slip; (6)

$f_i \cdot n > f_{ch}$ the mechanism of rupture of chemical bonds.' (7)

'Corresponding to conditions (6) and (7) the adhesion strength depends upon the depth of penetration only to $n_{cr}$. The adhesion strength for condition (6) is equal to:

$$F = \sum_{1}^{2} f_i \, Q \cdot n$$

while for condition (7)

$$F = \sum f_{ch} \, Q$$

where $\sum$ indicates the need to take into account the interdiffusion of the components.'

'To solve the problem of the magnitude of the intermolecular forces it is possible to make use of the theory of intermolecular interaction. For viscous flow we may also make use of the relationship determining the friction force for each group of atoms in the form (35):

$$f_i = \nu mv/_2$$

where m is the mass of interacting groups; $\nu$ the frequency of interaction; v the rate of movement of a molecule (the rate of extraction).'

'The number of molecular chains intersecting the interface may be determined with the provisos that 1) only the terminal segments of the macromolecules diffuse; 2) the polymers are not treated so that the molecules become oriented; 3) the macromolecules are non-regular and give random, non-ordered arrangement of the chains.'

'In 1 g.mol of any given substance there are N molecules occupying a volume $V_M$ . The number of particles in one cu. cm. is $N/V_M$ ; the number of terminal segments in 1 cu. cm. is equal to 2 $N/V_M$ while for 1 sq. cm. it is $(2N/V_M)^{2/3}$ or

$$Q = (2N\rho/M)^{2/3} \qquad (8)$$

here $\rho$ and M are the density and molecular weight of the polymer. Equation (8) determined the statistical mean number of terminal segments in 1 sq. cm. As the process proceeds the number of segments intersecting the interface increases by diffusion of the more deeply placed terminal segments and the possible diffusion of middle portions of the chain. The role of the latter in the adhesion of high polymers is not yet explained.'

'The most complex task is the determination of the depth of penetration of the macromolecules. It may be solved on the basis of Fick's second law. A special feature of the problem is the dependence of the coefficient of diffusion of a macromolecule upon its length. Since the length of the diffusing portion of a macromolecule increases as the adhesion bond is formed, the coefficient of diffusion also changes with time.'

'Problems of this type are considered in a number of text books on diffusion. Making use of standard methods an approximate solution can be found. For the depth of penetration of macromolecules in terms of groups of atoms, we obtain the equation (34):

$$n = \frac{\pi^{\frac{1}{2}} K_D{}^{\frac{1}{2}} t^{(1-\beta)/2}}{2l \cos \alpha/2} \tag{9}$$

where l is the length of the C.C bond; $\alpha$ the angle complementary to the valence angle; $\beta$ a constant determining the alteration of the coefficient of diffusion with time; and $K_D$ a constant characterizing the properties of the diffusing molecules and the diffusion medium. In particular, $K_D$ depends upon the flexibility of the diffusing macromolecules and the temperature of contact.'

'For condition (6) taking into account (8) and (9) the adhesion strength

$$F = \frac{\pi^{\frac{1}{2}}(2N)^{2/3}}{2l \cos \alpha/2} \left[\sum_{1}^{2} f_i \left(\frac{\rho}{M}\right)^{2/3} K_D{}^{\frac{1}{2}}\right] t^{(1-\beta)/2} \tag{10}$$

Equation (10) is correct for $n < n_{cr}$ or $t < t_{cr}$, as determined by

$$t_{cr} = \left[\frac{2l\cos \alpha/2}{\pi^{\frac{1}{2}} K_D{}^{\frac{1}{2}}} \frac{f_{ch}}{f_i}\right]^{2/(1-\beta)}$$

For condition (7) $t > t_{cr}$:

$$F = (2N)^{2/3} \sum_{1}^{2} f_{ch} \left(\frac{\rho}{M}\right)^{2/3}$$

> 'Since for polymers in the viscous and high-elastic states $n_{cr}$ is quite large ($f_i << f_{ch}$) it is possible to make use of (10) over a wide time range. According to this equation the adhesion strength is a parabolic function of the time of contact, inversely proportional to the 2/3 power of the molecular weight and proportional to $K_D^{\frac{1}{2}}$ characterizing the mobility of the molecular chains.'
>
> 'The diffusion theory of adhesion is faced with the problems: 1) of direct experimental proof of diffusion of macromolecules or of their portions in adhesion phenomena; of quantitative investigation; 2) of the influence of the molecular weight and the part played by the diffusion of the middle portion of the macromolecules in the adhesion of polymers; 3) of the temperature dependence of the adhesion for the determination of the energy of activation of the process and its comparison with the energy of activation of the diffusion processes in polymers; 4) of the dependence of adhesion on the chemical nature of the adhesive and the substrate (36).'"

## Diffusion

Diffusion is a movement of molecules or atoms under the influence of a concentration gradient, leading to an equalization of concentration within a single phase (20).

In macromolecules, diffusion is closely related to Brownian motion and it could be said that the molecules or particles of a substance diffuse because of their Brownian motion (21).

Transfer of heat by conduction is also due to random molecular motions, and there is an obvious analogy between the two processes. This was recognized by Fick, who first put diffusion on a quantitative basis by adopting the mathematical equation of heat conduction derived earlier by Fourier. The mathematical theory of diffusion in isotropic substances is therefore based on the hypothesis that the rate of transfer of diffusing substances through unit area of a

section is proportional to the concentration gradient measured normal to the section, i.e.

$$F = - D \frac{dC}{dx}$$

where F = rate of transfer per unit area of section

C = concentration of diffusing substance

x = the space coordinate measured normal to the section

D = diffusion coefficient.

In diffusion involving dilute solutions D can be considered as constant, but in diffusion in high polymers, it depends very markedly on concentration (17).

For unsteady-state conditions there will be an accumulation of diffusible matter in a unit volume, and the concentration at any point within a solid will vary with time, hence $\frac{dC}{dt} \neq 0$. The increase in the amount of substance within a volume element bounded by two parallel planes $P_1$ and $P_2$, having a unit area and located at the distances x and x + dx, will be equal to the difference in the flux $F_x$ and $F_x + dx$ shown in Figure 1.

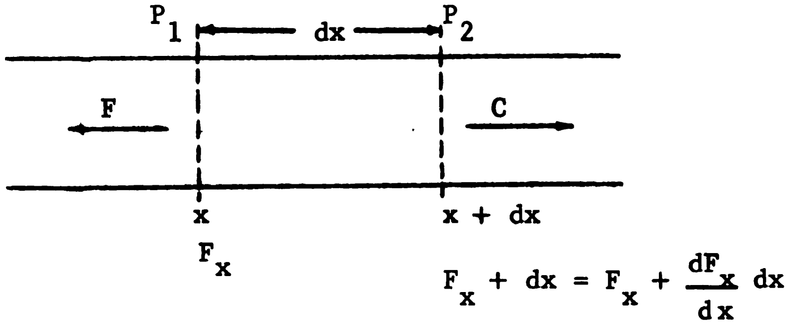


Figure 1 Schematics for Derivation of Fick's Law

Thus, the flux at x + dx is

$$F_x + \frac{dF_x}{dx} dx = - D \frac{dC}{dx} - \frac{d}{dx} ( D \frac{dC}{dx} ) dx$$

and at x is

$$F_x = - D \frac{dC}{dx}$$

The difference between the two is

$$\frac{dF_x}{dx} = - \frac{d}{dx} D \frac{dC}{dx} \quad .$$

Since $\frac{dF_x}{dx}$ is equal to the negative rate of concentration change, i.e.,

$$\frac{dF_x}{dx} = - \frac{dC}{dt}$$

we obtain the Second Fick's law

$$\frac{dC}{dt} = D \frac{d^2C}{dx^2}$$

Where D is assumed to be constant at constant temperature.

The diffusion coefficient is a measure of the ease with which molecules move within solid. We know, however, that the movement of molecules or their segments within high-polymer is related directly to the thermal agitation, hence the diffusion coefficient is strongly temperature dependent and varies with temperature according to the Arrhenius-type equation:

$$D = D_o e^{-\frac{E}{RT}}$$

where D = diffusion coefficient

$D_o$ = a constant (in diffusion of gases taken as the diffusion coefficient at infinitely high temperature)

E = activation energy

R = gas constant

T = temperature in $^oK$ .

Fick's law cannot be directly applied to diffusion of high polymers because of the complex nature of the diffusion process. The diffusion flux there will probably depend not only on temperature and concentration, but also on the change of concentration gradient and on the nature and extent of the "grain" boundaries (20).

## Viscosity

A characteristic property which determines the flow of any fluid is its viscosity. Viscosity may be defined as internal friction or as the resistance of a fluid during flow (20). It is more exactly defined as the ratio of the shearing stress to the rate of shearing, i.e.,

$$\eta = \frac{S}{D}$$

where $\eta$ = the viscosity

$S$ = shearing stress

$D$ = rate of shearing (22).

Viscosities are usually reported in terms of poises. A fluid has a viscosity of 1 poise if steady tangential force of 1 dyne produces a relative velocity of 1 cm per sec between two parallel planes of area 1 cm$^2$, separated by a distance of 1 cm and immersed in the fluid.

The reciprocal of viscosity is a measure of fluidity which may be loosely viewed as the ease of flow of a substance. Formally defined

$$R = \frac{D}{S}$$

where R = fluidity

S = shearing stress

D = rate of shearing .

It can be seen that $R = \frac{1}{\eta}$ and if $\eta$ is in poises, R is in rhe.

$$\text{rhe} = \text{poise}^{-1}$$

Some liquids may be supercooled to form glasses without crystalization taking place as the temperature is lowered. In such materials the viscosity changes at so great a rate that over a small temperature interval the character of the material changes from a liquid to a rigid solid or glass. Many other properties of a substance also show a dramatic change in this glass transition region. The nature of glass transition is not completely understood and in the thermodynamic sense should not be called a transition. Polymers have glass transition, and from the effect of these transitions it is convenient to treat the phenomenon as a true transition.

A polymer above glass transition temperature $T_g$ , may exhibit viscous, viscoelastic or elastic behavior, depending on the molecular weight or other characteristics related to geometry.

Below the $T_g$ , molecular motion is frozen in. At $T_g$ the polymer has expanded to the extent that there is enough free volume available in the material for molecular motion to begin. Molecular segments occasionally have room enough to jump from one position to another with respect to their neighbors at this temperature. At $T_g$ the viscosity of a large number of polymers is roughly $10^{13}$ poises.

For normal liquids well above $T_g$ , the viscosity $\eta$ may generally be represented as a function of temperature by

$$\eta = \eta_o e^{\frac{\Delta H}{RT}}$$

where $\eta_o$ = constant representing the viscosity at infinite absolute temperature

$\Delta H$ = activation energy of viscous flow

R = gas constant

T = temperature in $^oK$

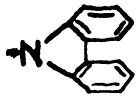
For high polymers between $T_g$ and 100 degrees above $T_g$, it has been found that the viscosity may be more accurately approximated by

$$\log \left[\frac{\eta_T}{\eta_{T_g}}\right] = \frac{-17.44(T-T_g)}{(51.6+T-T_g)}$$

where T and $T_g$ are in $^oK$, $\eta_{T_g}$ taken as $10^{13}$ poises (23).

Glass transition temperature $T_g$, is related closely to chemical structure of a polymer. The most important factors determining the value of $T_g$ seem to be the flexibility of the polymer chain and such chain flexibility related factors as steric hindrance and bulkiness of the side groups attached to the backbone chain. Bulkiness of a side group increases the glass transition temperature $T_g$, as can be seen on the following illustration (Figure 2) in which the increasing side group bulkiness is compared with the resulting $T_g$.

Figure 2 Polymer Sidegroups vs. $T_g$

| Polymer | Side group | $T_g$ (°C) |
|---|---|---|
| Polyethylene | $-H$ | $-120$ |
| Polypropylene | $-CH_3$ | $-10, -18$ |
| Polystyrene |  | 100,105 |
| Polyvinylcarbazole |  | 208 |

## Heat Conduction

When different parts of a body are at different temperatures heat flows from the hotter part to the cooler.

The heat transfer occurs by three possible methods: 1) conduction, 2) convection, and 3) radiation. In solids, convection is absent altogether and radiation negligible, hence conduction is the applicable method.

The basic law which quantitatively defines heat conduction is attributed to Fourier. The one-dimensional form of the Fourier law states that the Quantity of heat dQ donducted in the x-direction of a homogenous solid in time dt is a product of the conducting area A normal to the flow path x, the temperature gradient $\frac{dT}{dx}$ along this path, and a property K of the conducting material known as "thermal conductivity", i.e.,

$$\frac{dQ}{dt} = -KA\frac{dT}{dx} \qquad (13).$$

The one-dimensional heat conduction equation can be now derived as follows:

Assume two parallel planes of unit area each, within a homogenous circular rod with associated flow rates per unit time across them $f_1$ and $f_2$.

If no heat flow takes place through the curved surface the rate of heat increase in the section between the two planes is $f_1 - f_2$ .

Also, if T is the average temperature in the section, dx is the distance between the planes, and $\rho$ and C the density and specific heat respectively, the rate at which the section gains heat is equal to $\rho C dx \frac{dT}{dt}$ , i.e.,

$$(f_1 - f_2)dt = \rho C dx \frac{dT}{dt} dt$$

Making use of Fourier Law for unit area

$$\frac{dQ}{dt} = - K \frac{dT}{dx}$$

the rate of gain of heat through the plane at x can be expressed as

$$dQ_x = - K \frac{d}{dx}\left(\frac{dT}{dx}\right) dt$$

and through the plane at x + dx, as

$$dQ_{x+dx} = - K \frac{d}{dx}\left(\frac{dT}{dx} + \frac{dT}{dx} dx\right) dt$$

The total increase in internal energy within the section will then equal

$$dE = (f_1 - f_2)dt = \rho C dx \frac{dT}{dt} dt$$

and

$$dQ_x = dQ_{x+dx} + dE$$

Substituting the already derived expression for $dQ_x$ and $dQ_{x+dx}$, we obtain:

$$-K\left(\frac{d^2T}{dx^2}\right)dt = -K\left(\frac{d^2T}{dx^2}\right)dt \; -K\left(\frac{d^2T}{dx^2}\right)dxdt + \rho C dx\frac{dT}{dt}dt \quad .$$

Rearranging and cancelling we obtain the final relationship:

$$K\frac{d^2T}{dx^2} = \rho C\frac{dT}{dt} \quad .$$

Assuming that for the temperature interval of interest the conductivity, density, and heat capacity of a material stay constant, we group the constants and define a new one called thermal diffusivity, i.e.,

$$\frac{K}{\rho C} = \alpha$$

Hence, to determine the temperature within a solid at point x and time t, a solution has to be found which satisfies the <u>One-dimensional Heat Conduction Equation</u> as follows:

$$\frac{d^2T}{dx^2} = \frac{1}{\alpha}\frac{dT}{dt} \quad .$$

## Initial and Boundary Conditions

Before a particular solution of conduction can be found, it is necessary to determine the formulae which will express the Initial and Boundary Conditions which the temperature satisfies. These are the mathematical statements of hypotheses founded on the experimentally or otherwise obtained knowledge of the characteristics of the system under consideration.

Assumptions that in the interior of a solid temperature is a continuous function of distance and time, and that the same holds for first and second differential coefficients with regard to distance and for the first differential coefficient with regard to time, do not hold at the boundary of a solid and at the instant at which flow of heat is supposed to start.

Initial and Boundary Conditions arising in the mathematical theory of Conduction of Heat are as follows:

a. Initial conditions.

> The temperature throughout the body is supposed given arbitrary at the time coordinate zero.
>
> $$T = f(x) \text{ at } t = 0$$

b. Prescribed surface temperature.

> This temperature may be constant, or a function of time, or position, or both. For example,
>
> $$T = c, \text{ at } t = 0 \text{ and } x = 0 \quad .$$

c. No flux across the surface.

> The surface is perfectly insulated.
>
> $$\frac{dT}{dx} = 0 \quad .$$

d. Linear heat transfer at the surface. The "radiation" boundary conditions.

> If the flux across the surface is proportional to the temperature difference between the surface and the surrounding medium, so that it is given by
>
> $$H(T - T_0)$$

where $T_o$ is the temperature of the medium and H is a constant, the boundary condition is then

$$K\frac{dT}{dx} + H(T - T_o) = 0$$

or

$$\frac{dT}{dx} + h(T - T_o) = 0 \text{ , where } h = \frac{H}{K}$$

as h approaches zero this tends to condition "c" and as h approaches infinity it tends to condition "b".

The quantity H has been called the "Outer" or "Surface" Conductivity. It is often simpler to specify the surface thermal resistance per unit area, $R = \frac{1}{H}$ .

e. The surface of separation of two media of different conductivities $K_1$ , and $K_2$ .

Let $T_1$ and $T_2$ denote the temperatures in the two media. Then the flux over the surface of separation is

$$K_1\frac{dT_1}{dx} = K_2\frac{dT_2}{dx} \quad ,$$

and if at the surface of separation the temperatures of the two media are the same we have also

$$T_1 = T_2$$

This assumption will be valid only for solids in intimate contact. In other cases the rate of transfer between the two surfaces may be proportional to their temperature difference, so that

$$-K_1\frac{dT_1}{dx} = H(T_1 - T_2) \quad . \qquad (24).$$

The above list is by no means exhaustive and the notation used has been adjusted to One-dimensional problems.

## EXPERIMENTAL

The following is a description of all experimental phases of this study arranged in the order in which they were made.

### Thermistor Calibration

The method and equipment have been described in detail in (14), and this particular study was undertaken mainly to extend the temperature interval of measurement up to 250°F.

The thermistor was a one mil metalized Hercules Powder Pro-Fax polypropylene bi-axially oriented film, cut to eight inches by one inch on a Model CDC 25 Precision Sample Cutter made by Thwing - Albert Instrument Company.

The ability to measure surface temperature by the aluminized film thermistor rests on the temperature dependence of specific resistance of the thin aluminum layer which has to be sufficiently linear in order to enable interpretation of the voltage function detected on the oscilloscope screen.

Kahlbaum (25), reports the constants for pure aluminum to be used in the temperature-resistivity relationship.

$$r_t = r'_t \left[ 1 + \alpha(t-t')10^{-3} + \beta(t-t')^2 10^{-6} \right]$$

where $r_t$ for a solid = resistance at temperature t of a specimen which at $t_R$ has a length of 1 cm, and a uniform transverse sectional area of 1 $cm^2$.

for $t_1 \leq t \leq t_2$

$t' = 0^{\circ}C$ $\qquad$ $\beta = 1.80$

$r'_t = 2.62 \times 10^{-6}$ ohm-cm. $\qquad$ $t_1 = -80^{\circ}C$

$\alpha = 4.46$ $\qquad$ $t_2 = 400^{\circ}C$

The values of resistivity associated with temperatures from 100°F to 250°F were computed and appear in Table 1, Appendix II, page 86. Their plot can be seen in Figure 3 which shows the almost linear relationship. Although one might expect some "strain gage" effect in the thin aluminum layer due to the expansion of the polypropylene substrate caused by increase in temperature (26), this proved to be non-significant as shown by linearity of the measurements over the entire experimental temperature range (Table 2). Also the varying levels of loading of the thermistor during the calibration from .22 to 3.32 p.s.i. did not have any noticeable effect.

The resulting calibration data are tabulated in Table 2, Appendix II, page 87, and the resulting plot of the line drawn through the points is shown in Figure 4.

The calibration was carried out with the oscilloscope vertical sensitivity setting of 0.02v/division, hence, from the slope of the line in Figure 4 the sensitivity of the thermistor was determined as 1.008 mv/deg °F.

Next, a temperature-time function was generated and its trace recorded on the storage mode of the oscilloscope screen, from which by the use of the above derived sensitivity factor it was converted into degrees °F. In this procedure the thermistor was placed with

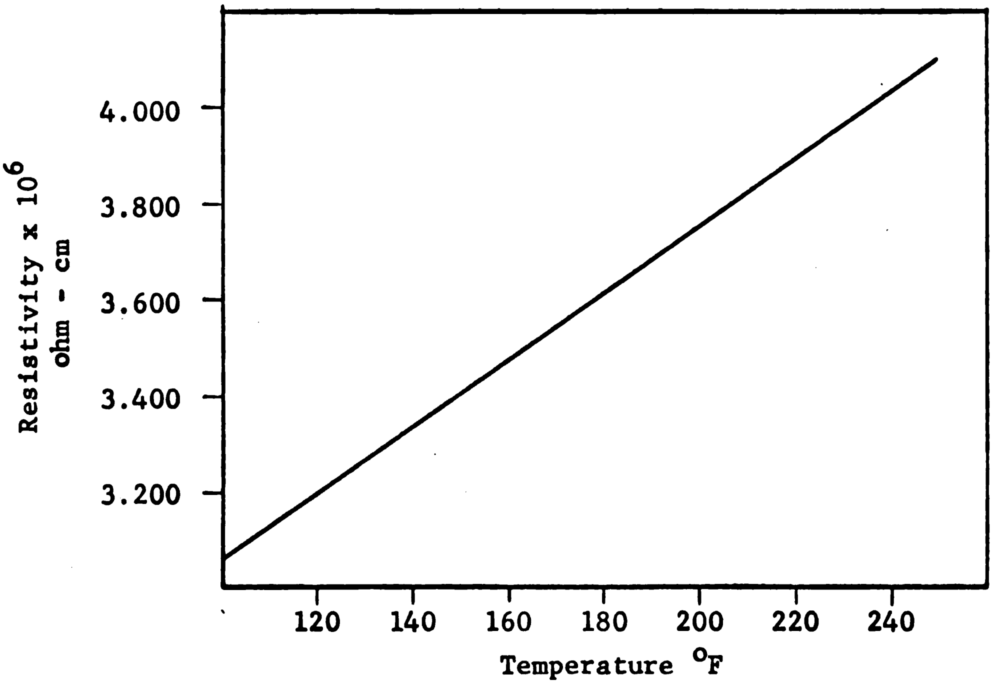


Figure 3

Plot of Resistivity vs. Temperature (Computed)

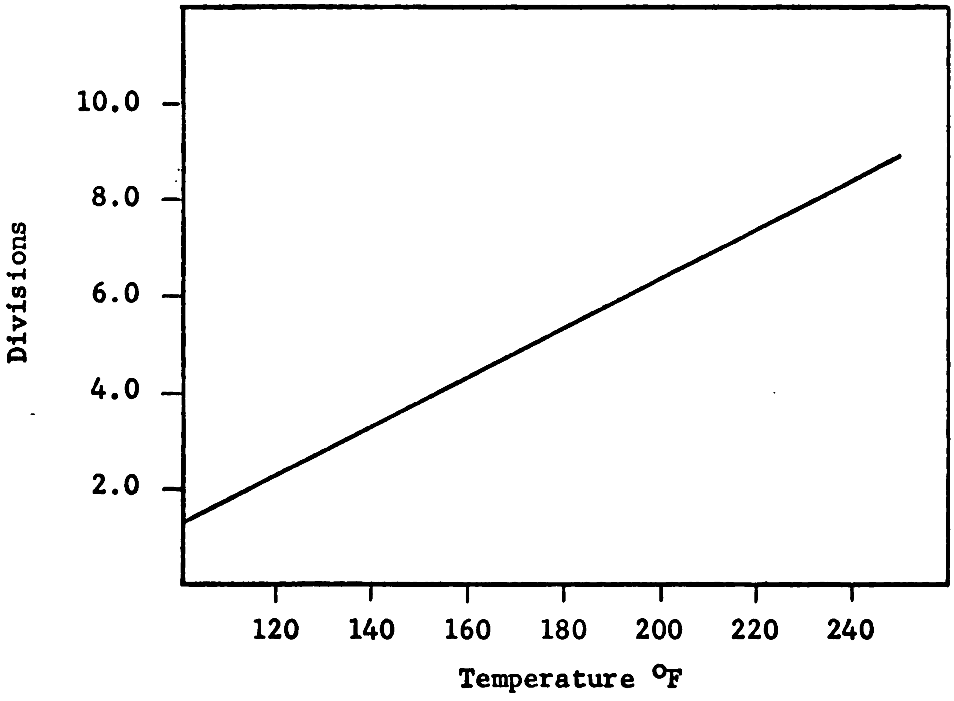


Figure 4

Plot of Thermistor Calibration Data

the metalized side away from the teflon coated bar of the heat-sealer so that the temperature sensing plane was in the position where the bonding interface would be positioned, had the film been actually heat sealed. The initial temperature $T_o$ was measured as 80°F and the heated bar temperature $T_h$ used 250°F. The temperature function was traced for two seconds. The two second period was selected after attempts to observe longer interval in which the oscilloscope trace showed a very steep rise within the first second, which made it difficult to interpret the trace with reasonable accuracy. These experimentally determined temperatures appear in Table 3, page 88, Appendix II, and are represented graphically in Figure 5.

The excellent agreement between the calibration values of the thermistor at five different loading levels, .22 to 3.32 p.s.i., leads to the conclusion that the effects of pressure in heat sealing must be of rheological nature only, since it does not affect, within the range studied, the heat transfer properties of the film.

## Comparison of Computed Temperature Functions

Three different approaches were used in determining the temperature-time function at the bonding interface of heat sealed film. Each of the three approaches was used for polypropylene (11), but the inconsistency in choice of common parameters made the results incomparable, so making an estimate of the relative importance of various details impossible.

Each one of the approaches involves finding of a solution which satisfies the One-dimensional heat conduction equation

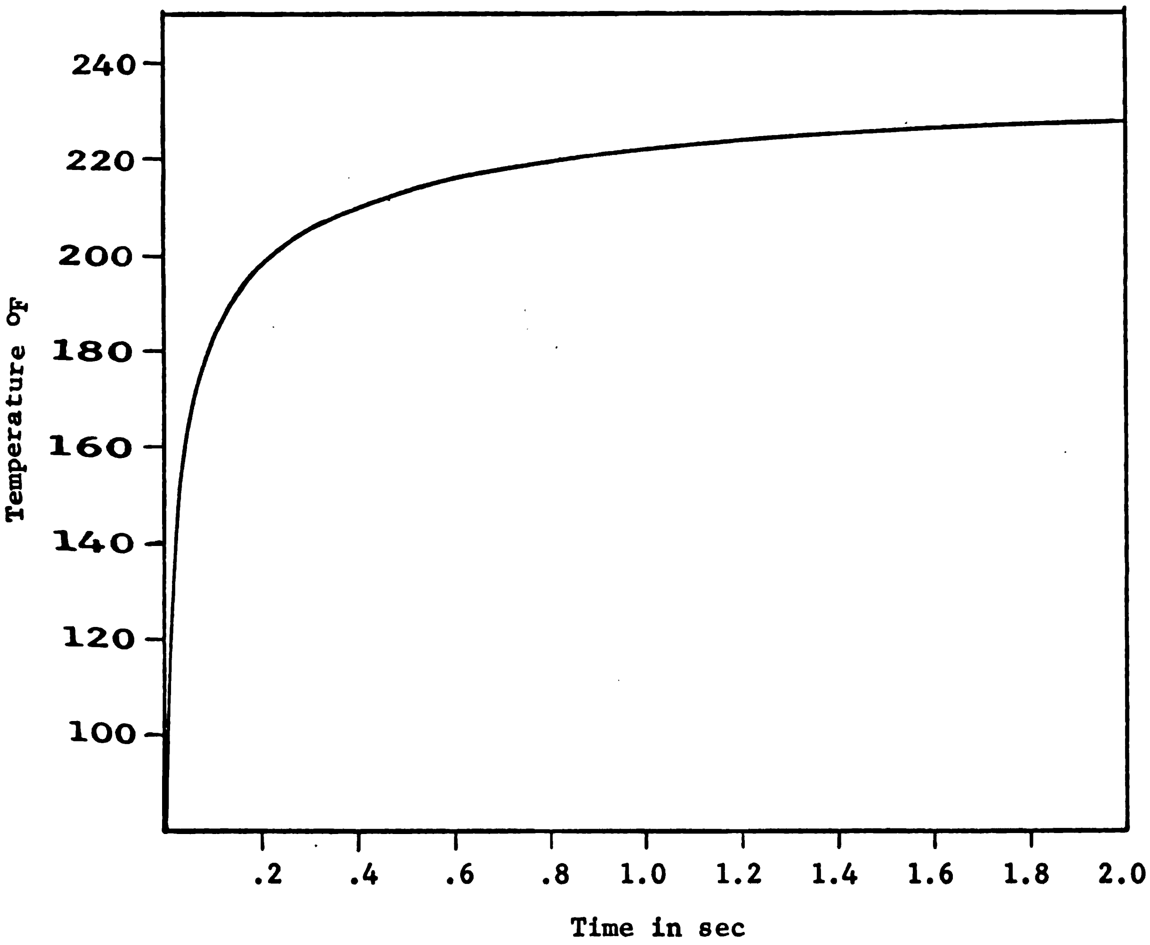


Figure 5

Temperature Function Obtained
By Use of a Thermistor

$$\frac{d^2T}{dx^2} = \frac{1}{\alpha}\frac{dT}{dt}$$

and the Initial and Boundary Conditions by means of which the heat sealing assembly is modeled.

The mathematical statement of each of the problems, the schematic representation of the geometry, and the solution to the problem appear in Appendix I.

An alternate procedure for determining computationally the bonding interface temperature is the use of a finite-difference method for transient heat conduction. One method in particular is the Crank-Nicholson Approximation which combines the Forward Difference and Backward Difference Approximation methods and which roughly states that the temperature at a point within a solid at a point in time is the average of the temperatures at either side of the point at the previous time. This method and a computer program for its use in solving One-dimensional transient heat-conduction problems are described in detail in (14). The schematic representation of the problem geometry is shown in Appendix I, pages 82-84.

For each of the computational procedures a program in Fortran was written and the temperature functions of the bonding interface were generated on CDC 3600 computer, under equal conditions as follows:

Material: Polypropylene

Thickness: .001 inch

Heated Bar Temperature, $T_h$: 250°F

Initial Temperature, $T_o$: 80°F

Time Interval: 4 sec

Bonding Interface Position, x: .001 inch

*Contact Resistance Gap-Air Layer, L: .0001 inch

Case 1 - Finite thickness of polypropylene in intimate contact with heated and unheated metal bars.

The temperature function computed by means of Equation I, Appendix I, page 77, is tabulated in Appendix II, Table 4, page 88. Its plot (Figure 6) shows clearly the effect of bare metal in contact with the film on the temperature at the interface of interest. It can be observed that after a very short time the temperature rises to a constant level which is the average value of the two boundary temperatures, i.e.,

$$T = \frac{T_h + T_o}{2}$$

It shows clearly the need for back-up thermal insulation in heat sealing situation involving heated and unheated bars.

Case 2 - Semi-infinite slab of polypropylene in intimate contact with heated metal bar.

Equation 2, Appendix I, page 79, tabulated in Appendix II, Table 5, page 89, and plotted in Figure 7, shows that a much higher temperature can be obtained at the interface of interest, when the bare metal, acting as a heat sink, is removed. For mathematical purposes, the film material is viewed as a semi-infinite slab. Kavesh (11) maintains that in heat sealing, approximately the same temperature-

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*Where applicable.

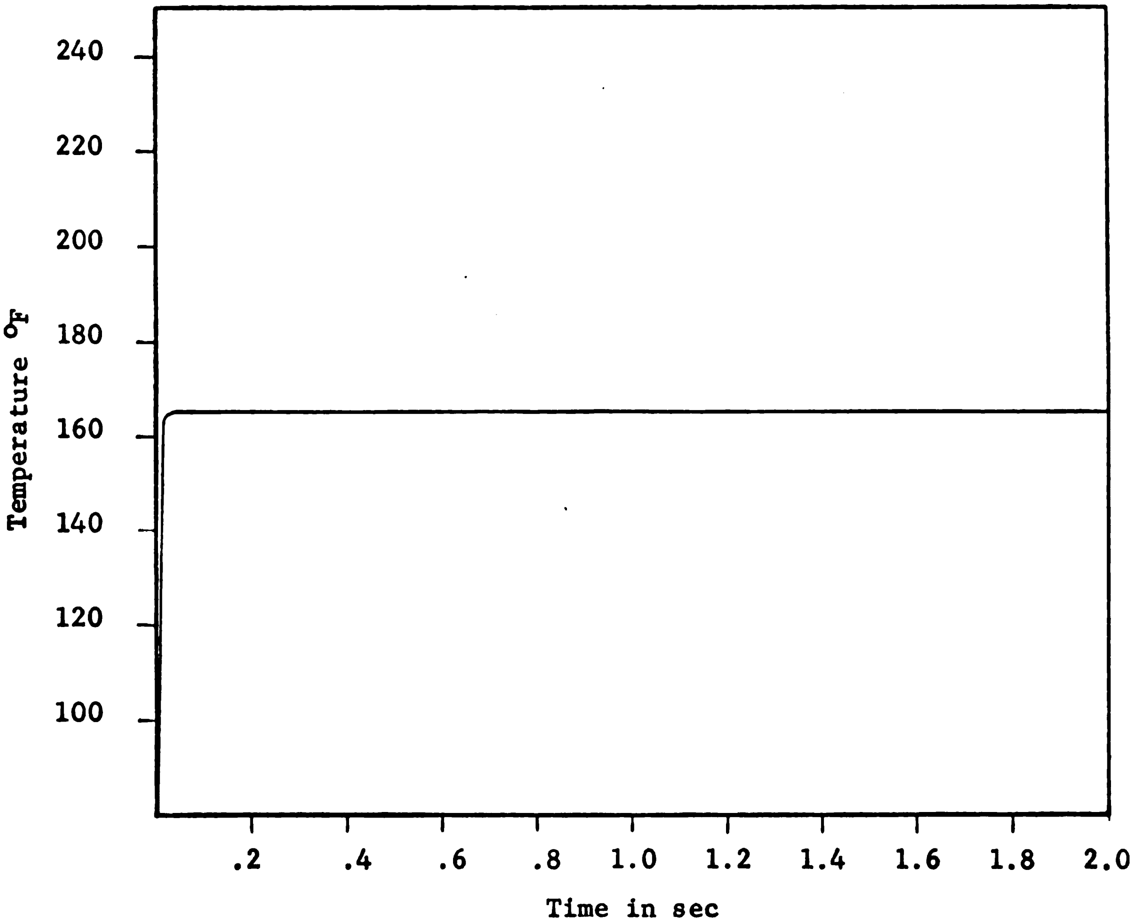


Figure 6

Temperature Function Obtained
By Computation Under Model I

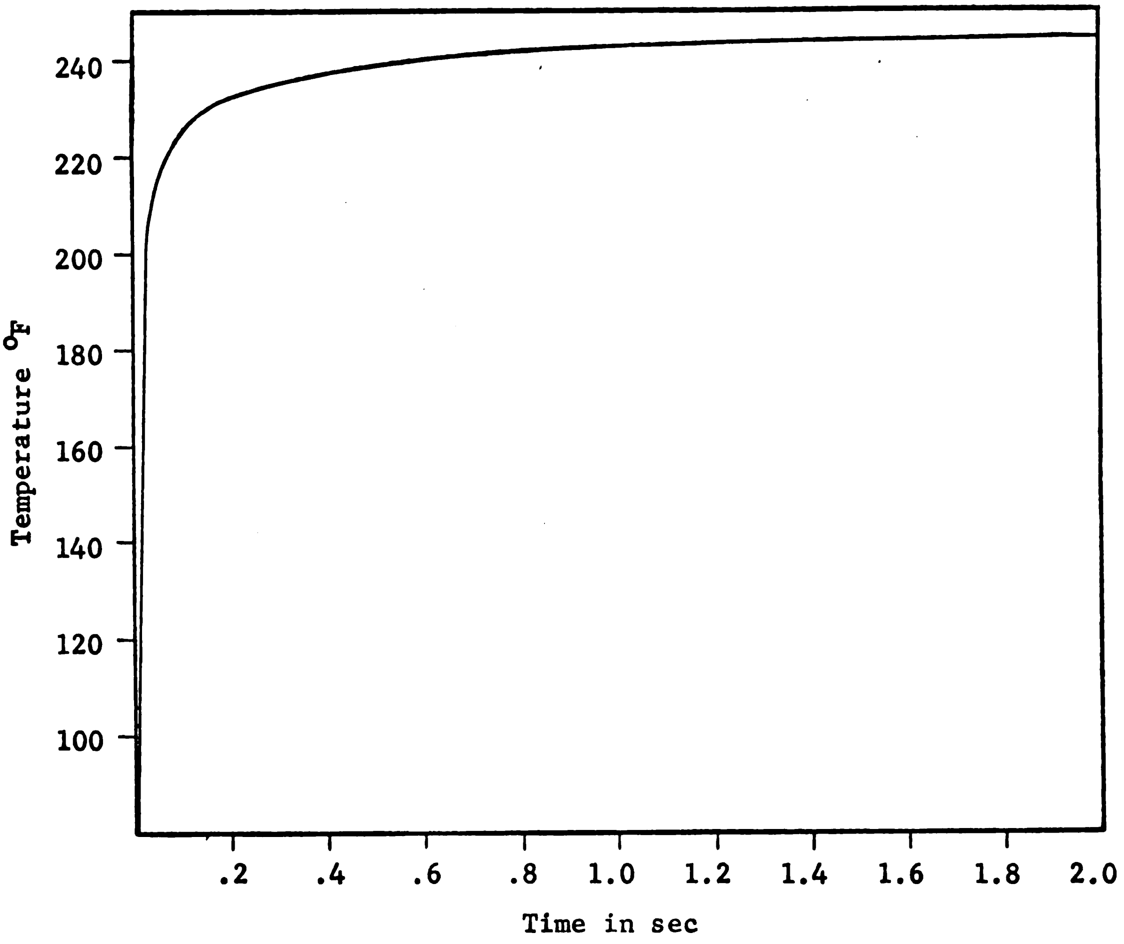


Figure 7

Temperature Function Obtained by
Computation Under Model II

time function can be maintained if the semi-infinite slab is replaced by insulating material backing whose thickness and thermal conductivity properties satisfy the following relationship:

$$\frac{K_i}{L} = \frac{QA^{-1}}{(T(x,t)-T_o)}$$

where $K_i$ = thermal conductivity of the insulating material.

$L$ = thickness of the insulating material.

$T(x,t)$ = computed temperature at interface at x and at time t.

$T_o$ = initial temperature.

$QA^{-1}$ = heat flow per area at the interface x and time t.

$QA^{-1}$ is obtained by,

$$QA^{-1} = K \frac{(T_h - T_o)}{\sqrt{\pi \alpha t}} e^{-\frac{x^2}{4\alpha t}}$$

Since we deal here with a transient situation, t has to be selected as some finite value. Kavesh (11), used t = 1 second.

Case 3 - Semi-infinite slab of polypropylene separated from heated metal bar by an air layer.

"Contact" or "Surface" Resistance to heat flow is present in heat sealing arrangements in which pressure cannot be applied due to resiliency of backing, as found in heat sealing of overwraps of soft goods, and to a lesser degree in all other heat sealing applications, due to causes not completely understood. This Contact Resistance is for computational purposes viewed as a thin layer of gas, air, separating the solid of heat source and the solid of heat transfer. Kavesh (11), treats the air layer and the plastic film as a two-

component system, in which only the conductive properties of air are considered; although, the transfer of heat through the air may involve convection and radiation to a significant degree.

In his article, Kavesh did not include rigorously stated hypotheses of the problem and the solution given by him does not seem to fit the problem described. Furthermore, the tabulated values of the temperature function seem to indicate a mistaken use of air layer thickness of 0.001 inch, instead of 0.0001 inch as discussed in his text.

Equation 3, Appendix I, page 80, represents a solution to the problem of conduction of a two-component system, which has been formulated in detail in Appendix III, pages

The tabulated values of Equation 3 appear in Appendix II, Table 6, page 89, and their plot is given in Figure 8.

As can be seen, the inclusion of an intermediate layer with lower conductivity results in bonding interface temperature function which rises slower, and within the time interval of two seconds, reaches lower level than the function obtained from Equation 2 (Figure 7).

For the two-component system the same treatment for insulating material determination, described under Case 2, is proposed by Kavesh. It has to be noted, however, that the insulation requirement is computed from a flux determined at a finite point in time, and only the thermal conductivity of the insulating material is considered. It is to be expected that the actual temperature function will deviate from the one determined by Equation 3, due to the effects of the

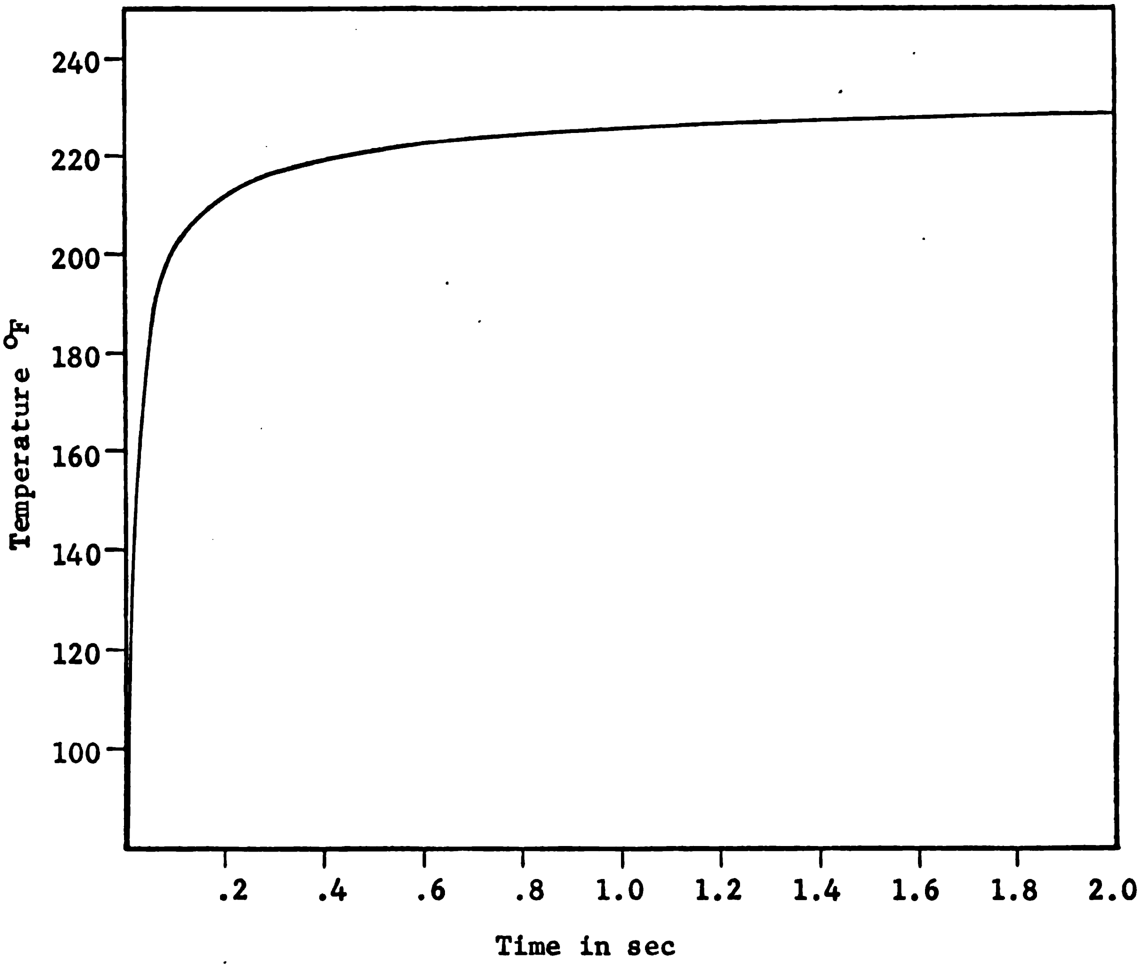


Figure 8

Temperature Function Obtained by
Computation Under Model III

density and the heat capacity of the insulation, which would cause it to act as a heat sink at first, and as a heat barrier later. This has been shown in Figure 9.

Case 4 - Finite thickness of polypropylene separated from unheated metal bar by finite thickness of insulating material and from heated metal bar by an air layer and Teflon coating.

In this procedure, the bonding interface temperature is obtained by solving a system of difference equations of the type 4 - a and 4 - b, shown in Appendix I, page 83. The program for solving the multi-component heat conduction system has been originally written by Dr. J. Beck of Michigan State University's College of Engineering for problems involving heat transfer through metals. It was adapted for use with thin plastic films, and it's main advantage lies in the ability of this program to take into account all the thermal characteristics of the components of which a heat sealing assembly is made. The contact resistance effect is approximated by the ratio of thermal conductivity of air to the thickness of the air layer, used as a coefficient at the appropriate interface.

The temperature function resulting from replacement of a semi-infinite slab of plastic by a finite thickness of insulating material was computed and its values appear in Appendix II, Table 7, page 90 . The plot of this function is shown in Figure 9, where the effects of finite thickness of insulating material can be observed on curve B arrived at by Equations 4 - a and 4 - b, superimposed on curve A obtained by Equation 3.

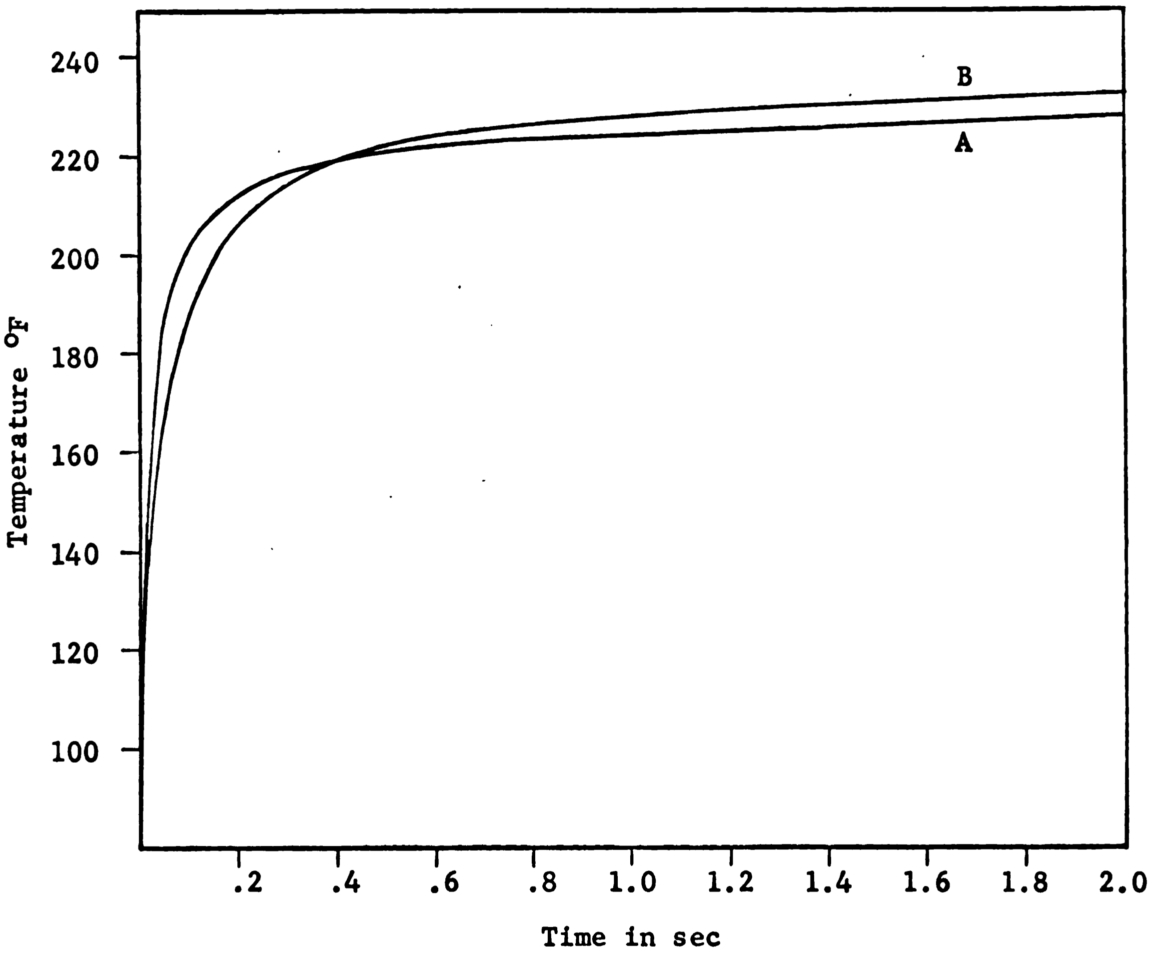


Figure 9

Temperature Functions Under Assumption of Semi-infinite Solid (curve A) vs. Finite Thickness of Insulating Material (curve B)

Additional factors seem to affect the bonding interface temperature. Under the assumptions of all cases discussed, the heated bar temperature surface is at $T_h$. In actual heat sealing assemblies the metal bar is coated with a layer of Teflon which, on the laboratory heat sealer used, had the thickness of .001 inch. In addition, the heated bar temperature control is maintained by means of a sensing device, possibly thermocouple, positioned some distance away from the surface. In accounting for these factors, the Crank-Nicholson Approximation method used in Beck's program showed its main strength.

Since the actual thickness of the air layer involved in contact resistance cannot be measured, and its being a uniformly thick layer is doubtful, it was necessary to determine its approximate value by rather indirect procedure.

The computational program was adapted to incorporate all the components of the heat sealing assembly, including the Teflon coating, the distance from the surface to the thermocouple, the thickness and type of insulating material, et cetera, and the temperature functions were then generated using three different values of contact resistance representing 0.0001, 0.0002 and 0.0004 inches of air layer.

These functions tabulated in Appendix II, Table 8, page 90, and plotted Figure 10, were then compared with the function obtained experimentally by means of thermistor, shown in Figure 5.

The applicable contact resistance for polypropylene was found to be the one corresponding to an air layer of 0.0001 inches.

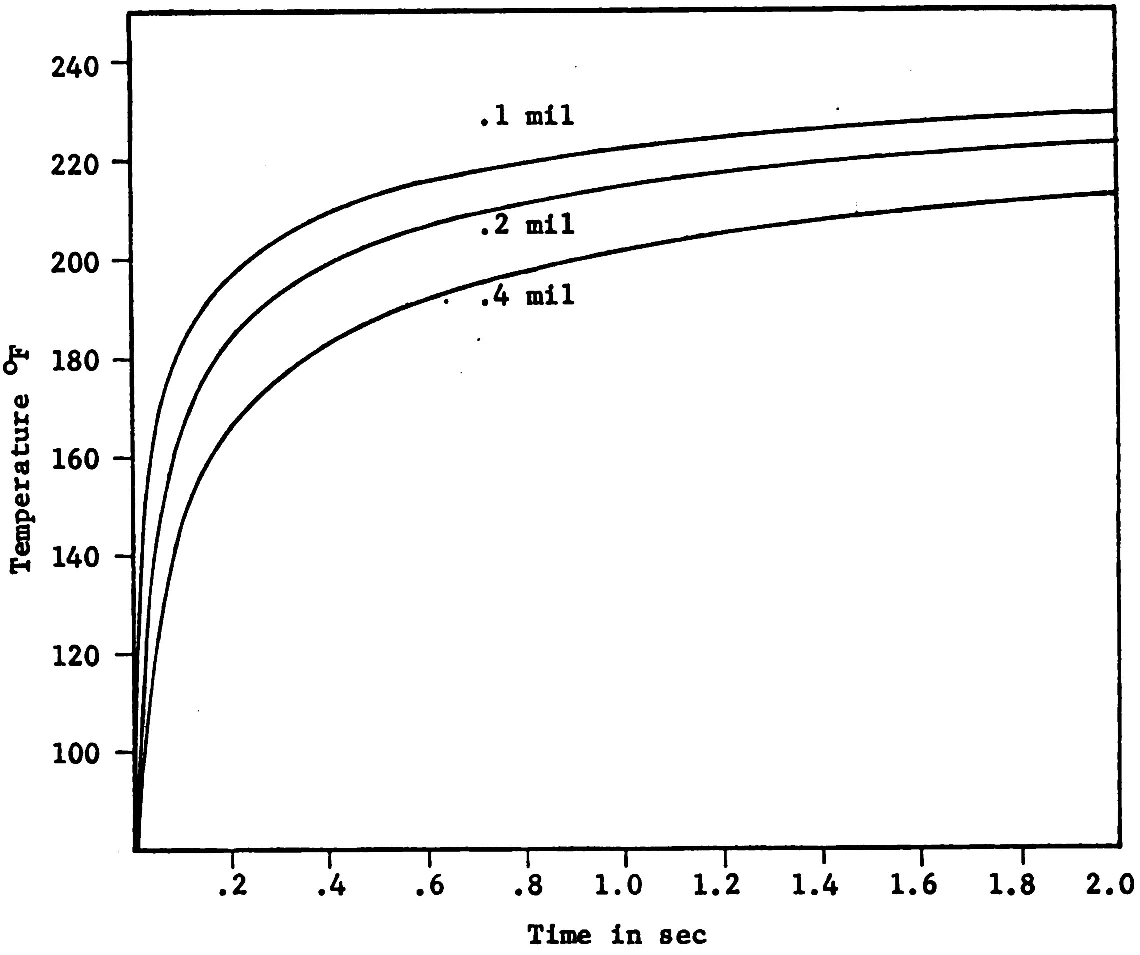


Figure 10

Temperature Functions Computed by Model IV for Contact Resistance Corresponding to .1, .2 and .4 Mil Air Layer

The good agreement between the computed and measured functions over the temperature range studied, 80 - 250°F, seems to support the conviction that, with enough care taken in translating the physical features of the heat sealing assembly into the computational parameters, the Beck's program can be used for simulation of the thermal changes at the bonding interface in heat sealing.

The importance of taking into consideration all the physical features of a heat sealing assembly is best illustrated by observing the differences in functions generated under the assumption, 1) metal surface of the heated bar has the temperature $T_h$, and 2) the metal is coated with .001 inch thick Teflon and $T_h$ is the temperature at the thermocouple located 1/8 of an inch away from the surface (Figure 11).

## Heat Seal Bond Strength Determination

The following is a description of the procedure employed for compilation of bond strength values of heat seals prepared under varying conditions of temperature, time, and pressure to be studied with respect to the computationally determined bonding interface temperature and with respect to viscosity, fluidity, and diffusivity ratio which depend on temperature.

Heat Sealer - Laboratory heat sealer modeled after Olin Mathieson Chemical Corporation design (10), used with a control consol equipped with a temperature controller, electric timer and a pressure gage.

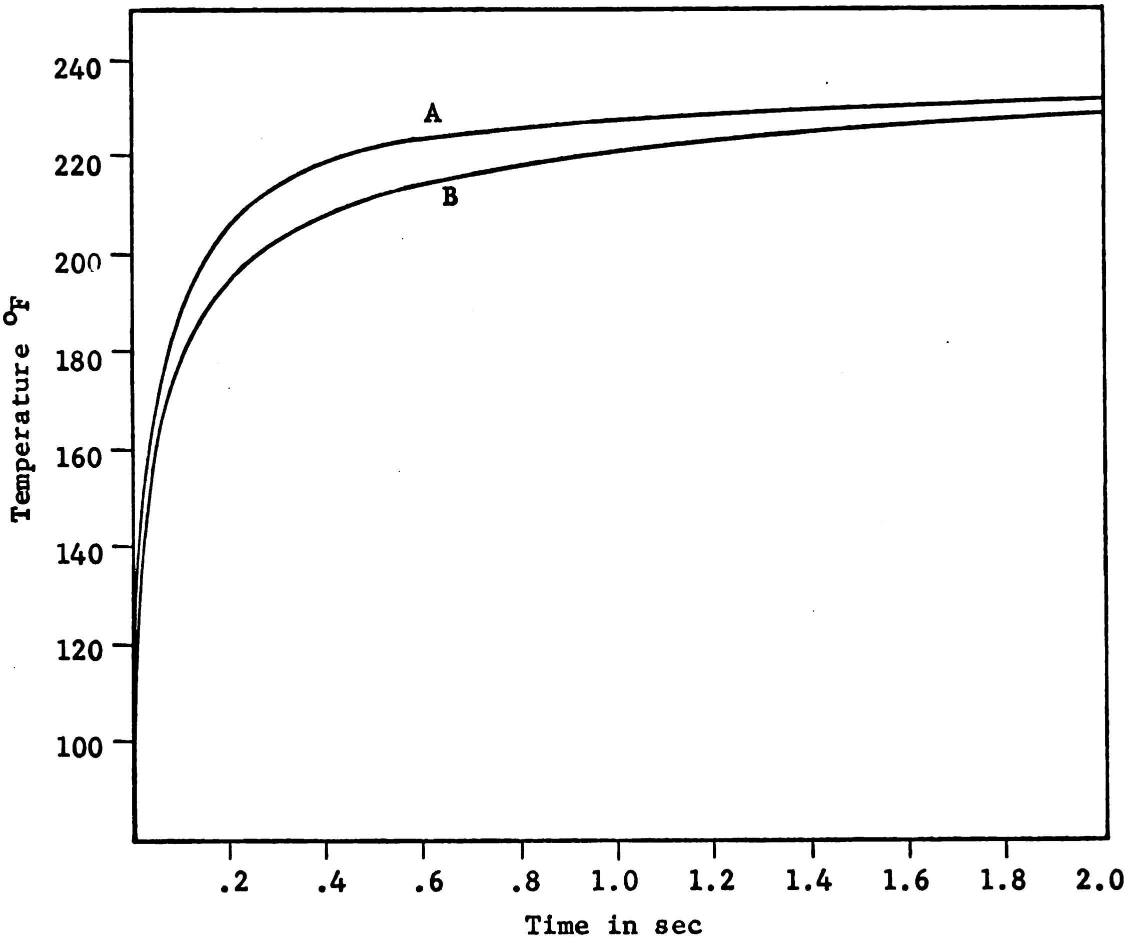


Figure 11

Temperature Functions Computed Under Assumption of Metal Surface at $T_h$ (curve A) vs. Metal Coated With 1 Mil Teflon and $T_h$ is 1/8 in. Away From Metal Surface (curve B)

The heat sealer is in the form of a table 8½ inches by 4 inches, with an opening at the center to admit the heated bar, located at a position below the sealer plane. An air piston moves the bar to a sealing position ¼ inch above the plane. The sealing bar is made of aluminum with a surface coating of approximately .001 inch thick Teflon. The temperature sensing element, thermocouple, is imbeded in the metal bar at an 1/8 inch distance from its surface, and the bar is heated by means of a 150 watt heating element mounted in its lower part. The sealing surface has the dimensions of one inch width and 6½ inches length.

The sealing pressure is determined by dead weight consisting of an aluminum base plate 4 inches by 6½ inches wide and long respectively, weighing 21.94 ounces on which lead plates, each weighing approximately one pound, can be added.

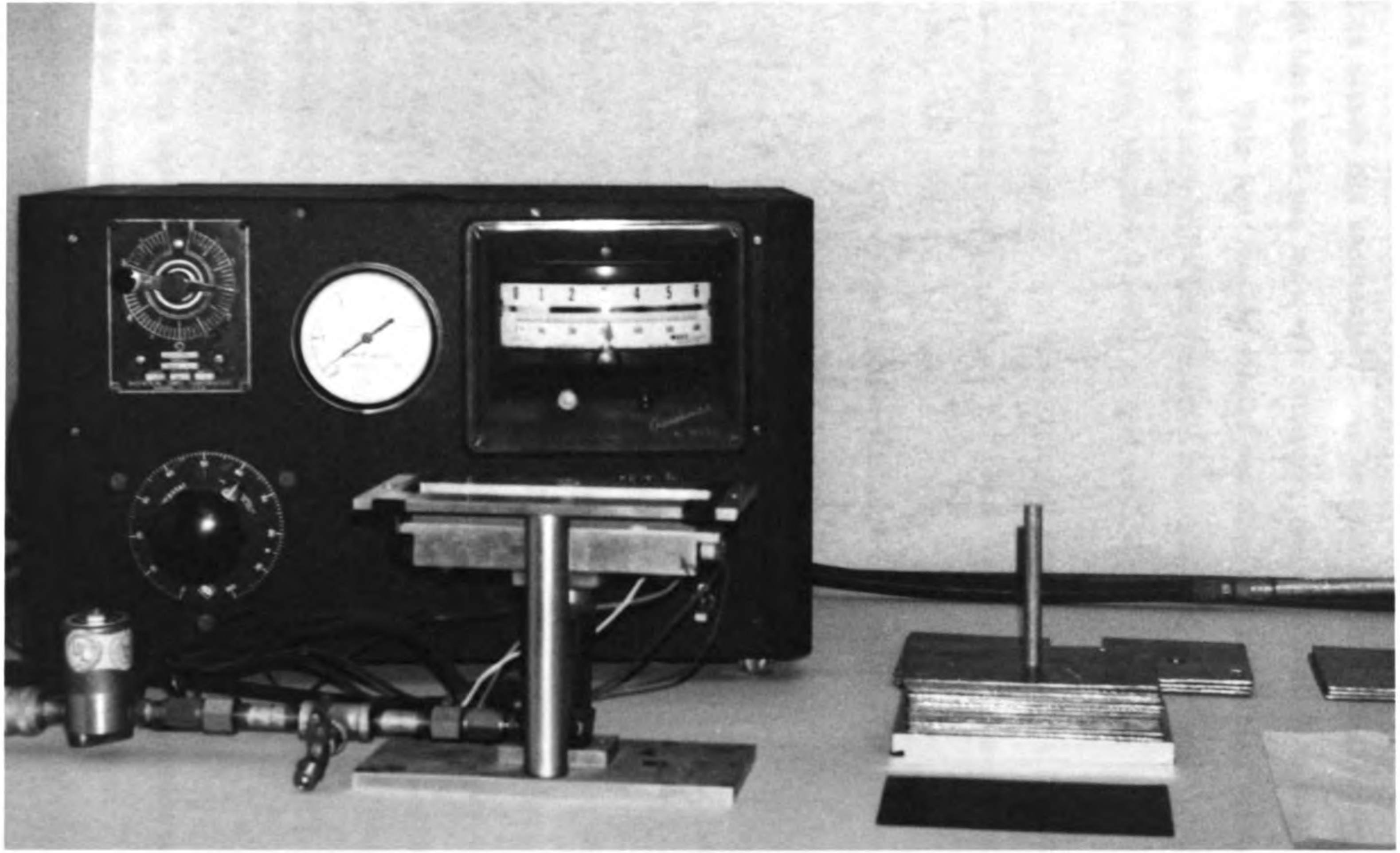
The sealing time, dwell time, is controlled by a timer which is started by means of a microswitch adjusted so that the timer is connected at the instant at which the sealing bar first touches the film. The timer, at the end of the pre-set interval, actuates a solenoid valve which releases the air pressure to the air piston, causing the sealing bar to retract at the end of the sealing cycle.

The thermal insulation used was a flat piece of commercial neoprene 1/16 inch thick, 4 inches by 6½ inches.

The entire heat sealing set-up is shown in Figure 12.

Film Samples - Samples of polypropylene film, OLEFANE-A made by Avisun Corporation, approximately 2 mil thick, were cut to 3 inches

Figure 12 Experimental Heat Sealing Set-up



by 9 inches. Each sample was folded in half and heat sealed by placing it across the heatsealer table in such a manner that a one inch wide heat seal was made approximately one inch away from the folded edge. This heat sealed sample was then cut to testing dimensions, one inch wide and five inches long, with the inner edge of the seal, fin-type, located exactly in the center.

*Tensile Strength Tester* - For bond strength determination, a Baldwin - Emery SR-4, Testing Machine, Model FGT, made by Baldwin-Lima-Hamilton Corporation, was used.

As a sensor, a Load Cell Model LB2TC±10-350, with ± 10 pounds range, made by Statham Instruments Corporation, was mounted on the stationary beam of the testing machine.

The transient features of the heat seal failure were recorded on a Baldwin Stress-Strain Recorder, Model MA 1B.

A general view of the testing equipment is shown in Figure 13.

In order not to limit excessively the effective range of the load cell, a special set of film grips made of light-weight materials, aluminum and magnesium, were constructed. To prevent slipping, the gripping faces were coated by a layer of polyvinyl acetate, Elmer's Glue, approximately 1/48 inch thick. A close-up illustration of the grips in actual use is shown in Figure 14.

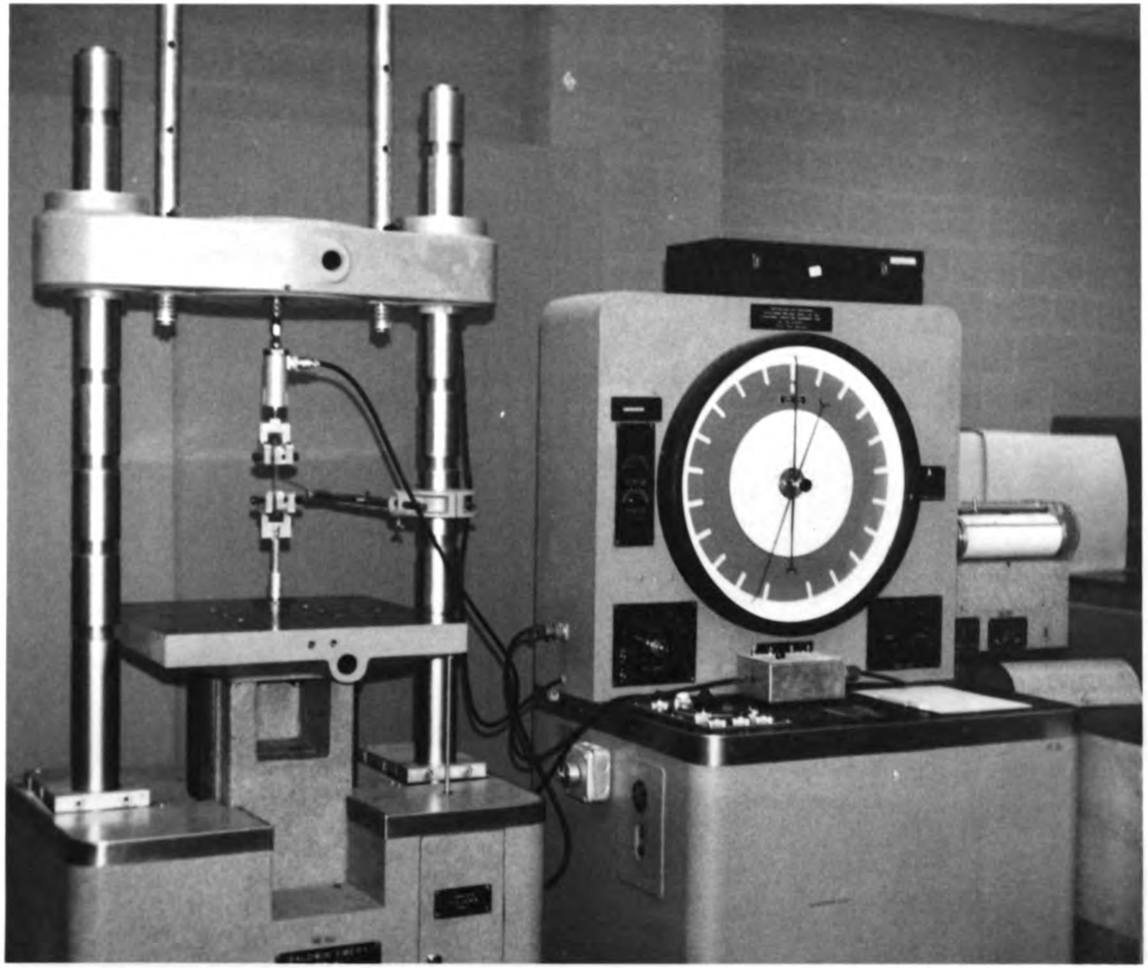
Procedure - Ten heat seal test samples were made for each combination of heated bar temperature, pressure and dwell time listed below:

Heated bar temperature $T_h$-°F: 300, 315, 320, 325, 337.5

Pressure levels - p.s.i.: .68, 1.15, 1.61, 2.09, 2.55, 3.47, 3.94

Dwell times - sec: .2, .5, 1.0, 1.5, 2.0, 3.0, 4.0

Figure 13 General View of Bond Strength Testing Apparatus



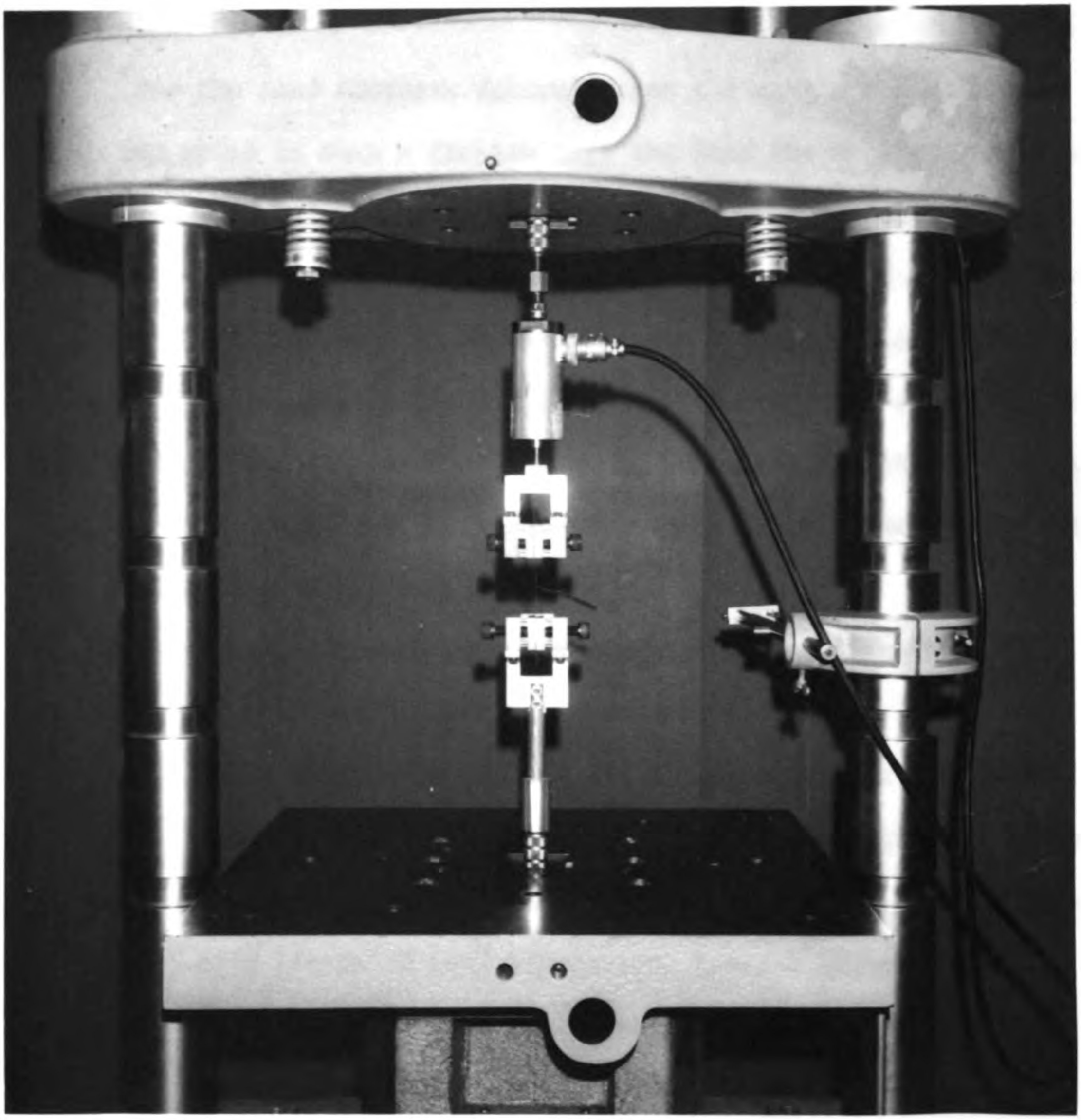


Figure 14 Lightweight Film Grips in Use

The polypropylene film samples were conditioned to 73°F and 50% R.H. before heat sealing and allowed to return to these conditions for a period of not less than 30 and not more than 45 minutes between completion of the seal and its actual use in bond strength determination.

For the bond strength determination the sample was clamped into the grips in such a fashion that the load was applied perpendicularly to the seal plane, T-test or peel-test. The conditions of testing employed were those specified under Method A of ASTM D 882-61T for Tensile Properties of Thin Plastic Sheeting, i.e., Initial Grip Separation of 2 inches and Rate of Grip Separtion of 20 inches per minute. Ten samples prepared under identical conditions of heat sealing were tested and their results averaged. All the samples were cut in the Machine Direction.

The failure of each bond was recorded on the Stress-Strain recorder and the record interpreted as follows:

a) Bonds failing by peeling had a characteristic record as shown in Figure 15, curve A. The bond strength was taken as the mean value of the Stress-Strain curve over the entire length of bond separation, i.e., 2 inches.

b) Bonds failing by tear or break at the inner edge of the seal had a characteristic record as shown in Figure 15, curve B. The bond strength was taken to be the Breaking Factor of the ASTM D 882-61T Method, i.e., the maximum force recorded at the point of failure, reported for the width of the sample.

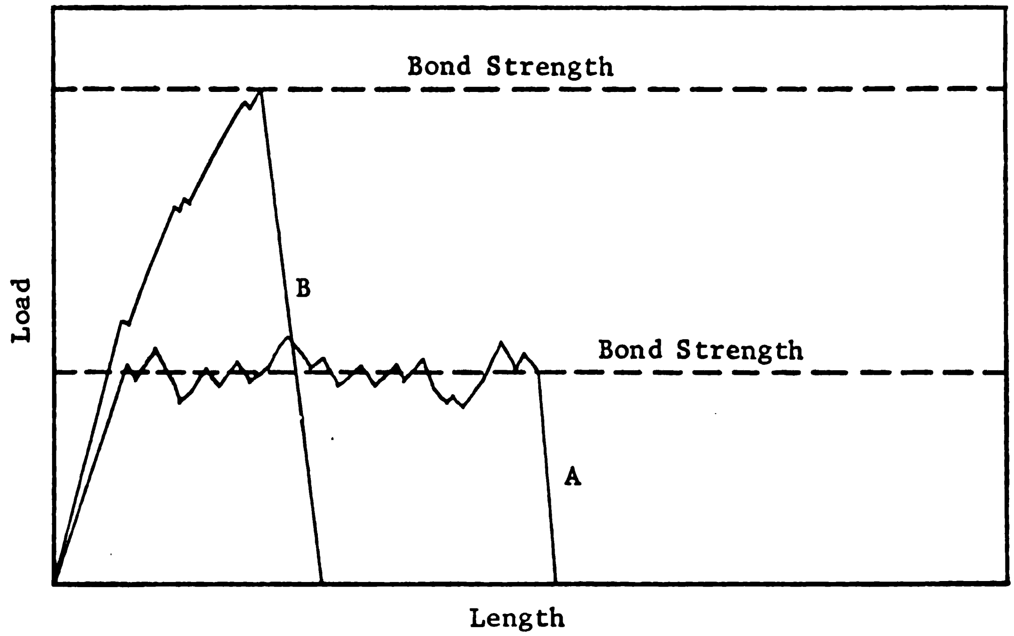


Figure 15

Bond Failure Record, Curve A - Peeling, Curve B - Break

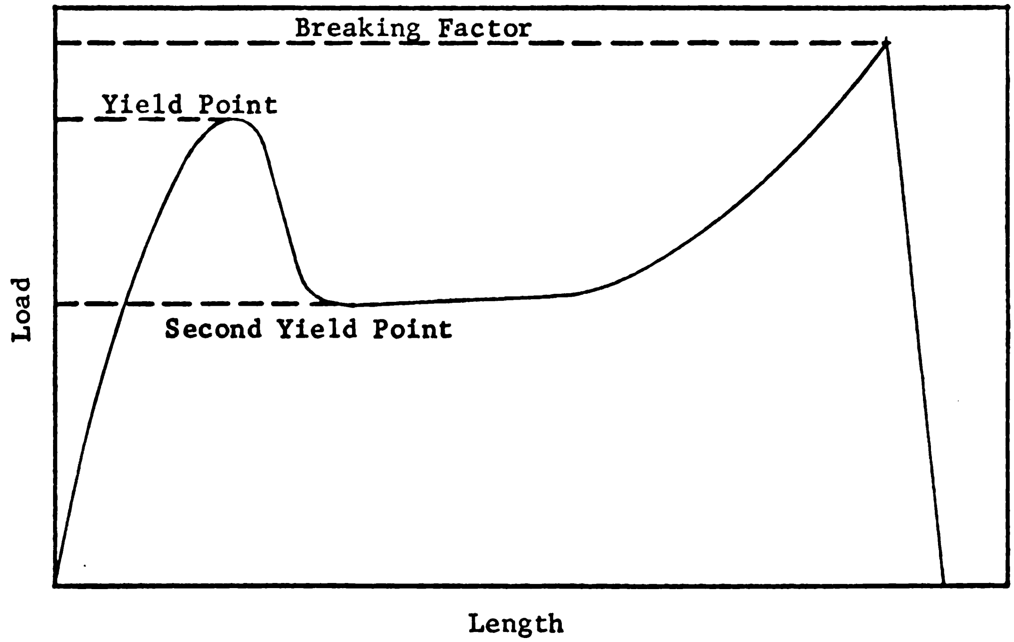


Figure 15a

Stress Strain Curve for 2 Mil Polypropylene

The bond strength results were identified by their respective heat sealing conditions and are tabulated in Appendix II, Table 9, pages 91-3 , in which the values are the averages of ten tests and are reported for each temperature $T_h$, dwell-time, and pressure combination.

In order to relate the bond strength results to the properties of the material used, ten samples of the film were cut in the Machine Direction and their tensile properties determined under the same conditions used for the bond strength tests. The characteristic Stress-Strain curve is shown in Figure 15a and the average values of ten tests were found to be as follows:

Film thickness: .00206 inches
Sample width: one inch
Yield point: 7.76 pounds per inch
Second yield point: 5.87 pounds per inch
Breaking factor: 11.26 pounds per inch
Elongation at break: 532%

The Beck's computational model was then adapted to simulate the heat sealing system used in preparation of the seals tested and the temperature functions at the bonding interface were generated. The computed temperatures appear in Appendix II, Table 10, page 94, and are plotted in Figure 16.

Each computed temperature was used within the same computational model to determine the values of viscosity, fluidity, and the diffusivity ratio $D/D_o$. The following formulae were used:

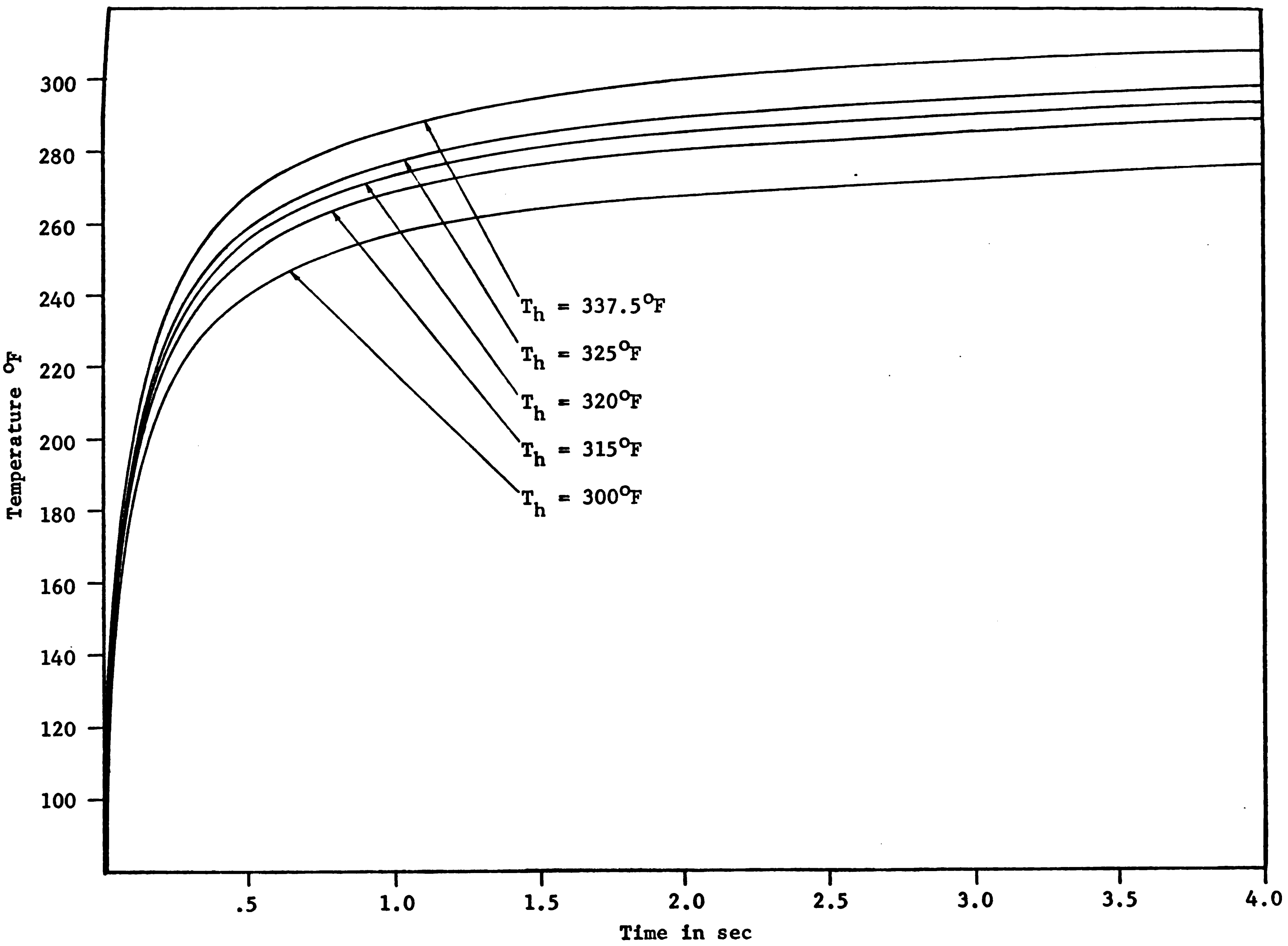


Figure 16

Temperature Functions Resulting From Different $T_h$ Values

1) $$\eta_T = \eta_{T_g} \times 10^{-\frac{17.44(T-T_g)}{51.6+T-T_g}}$$

where $\eta_T$ = viscosity in poises

$\eta_{T_g} = 10^{13}$ poises

$T, T_g$ = temperatures in $^{\circ}K$

2) $$R = \eta_T^{-1}$$

where R = fluidity in rhe

3) $$D/D_o = e^{-\frac{E}{RT}}$$

assuming that $D/D_o$ at $T_g$ = .0001

$E = 48.08$ cal/mole$^{-1}$

$R = 1.987$ cal deg$^{-1}$

$T_g = 263^{\circ}K$

The resulting functional values are tabulated in Appendix II, Tables 11, 12, 13, pages 94-5 , and their plotted curves are shown in Figures 17, 18, 19.

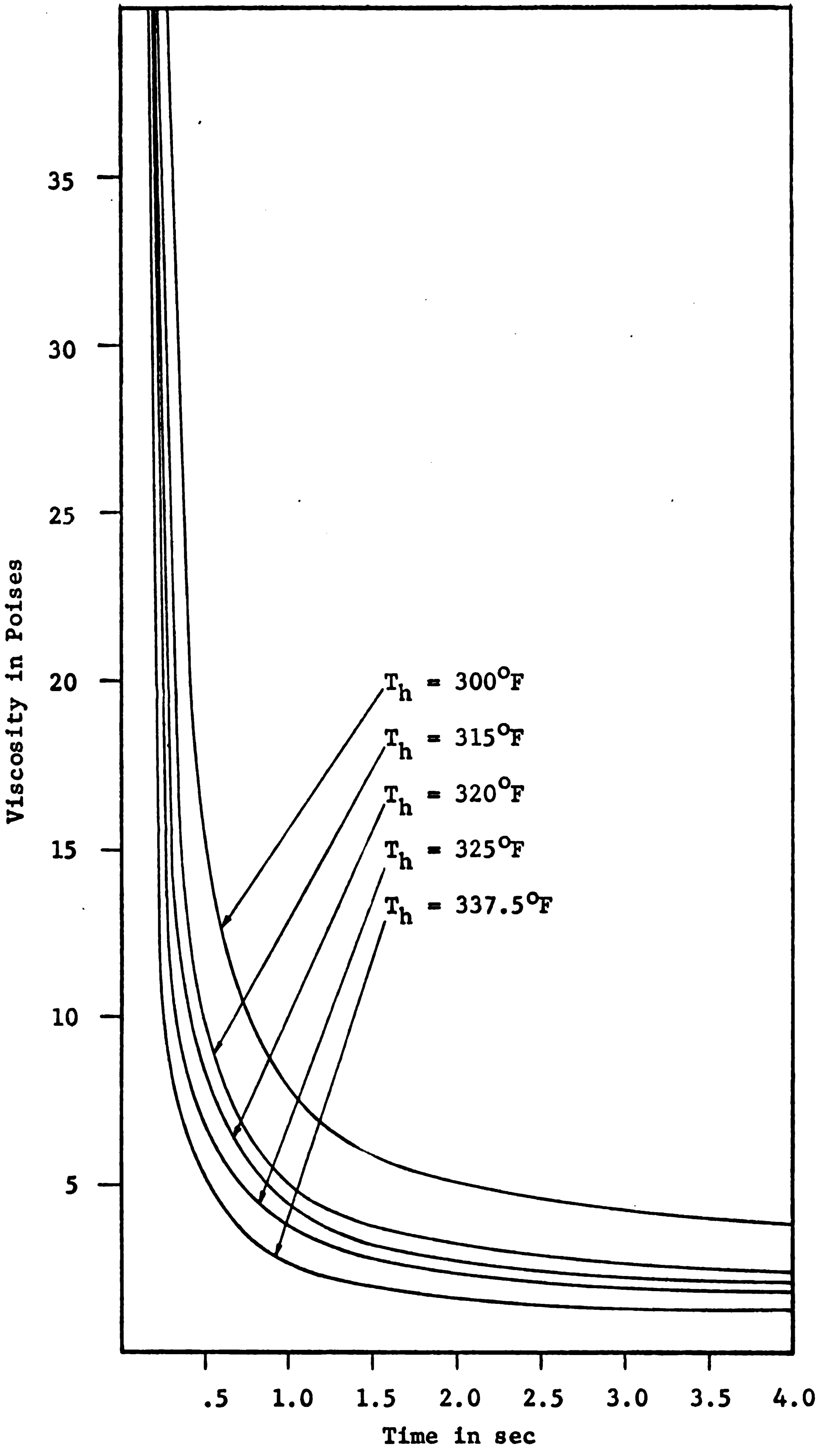


Figure 17

Viscosity Functions Resulting From Different $T_h$ Values

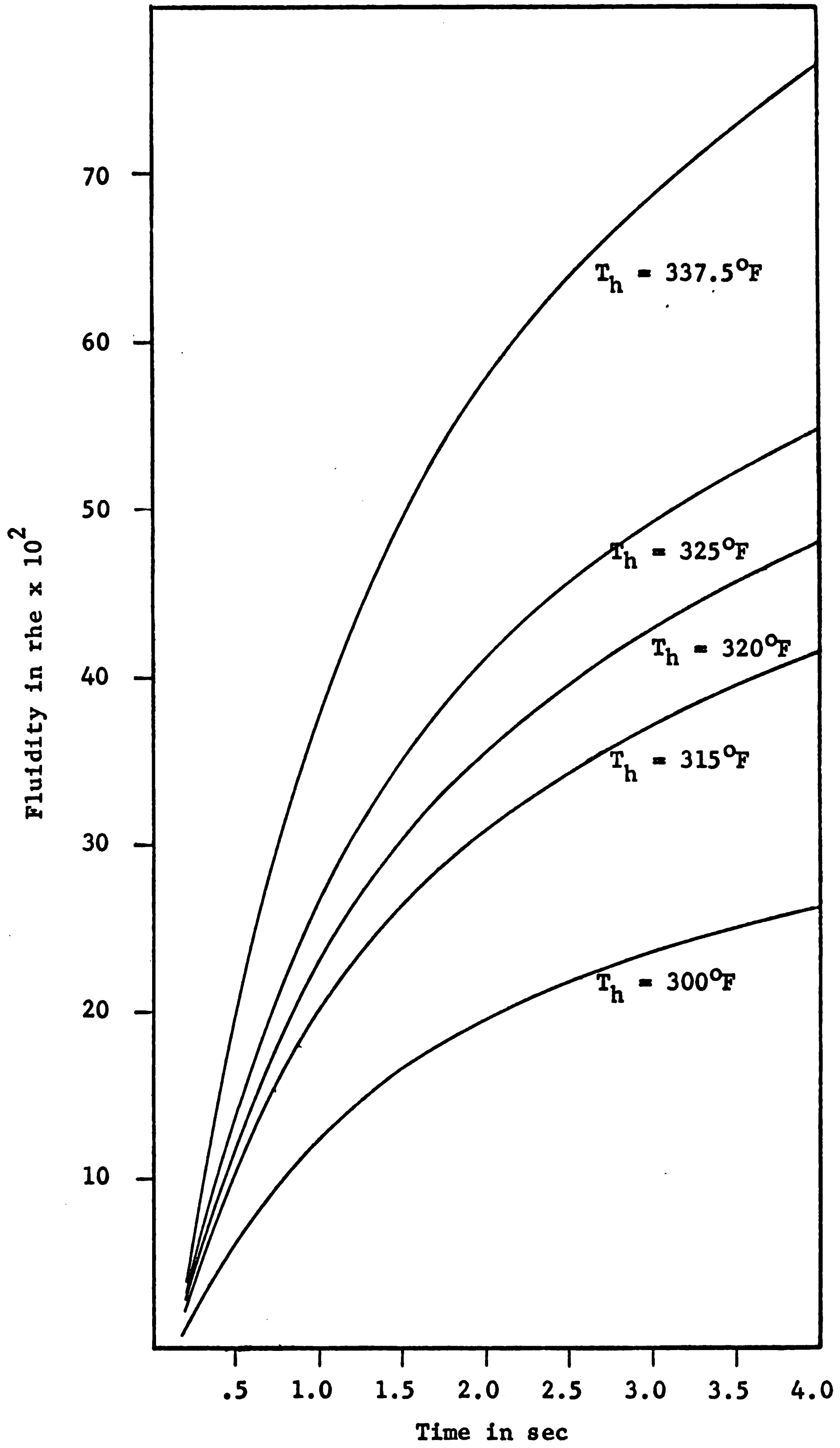


Figure 18

Fluidity Functions Resulting From Different $T_h$ Values

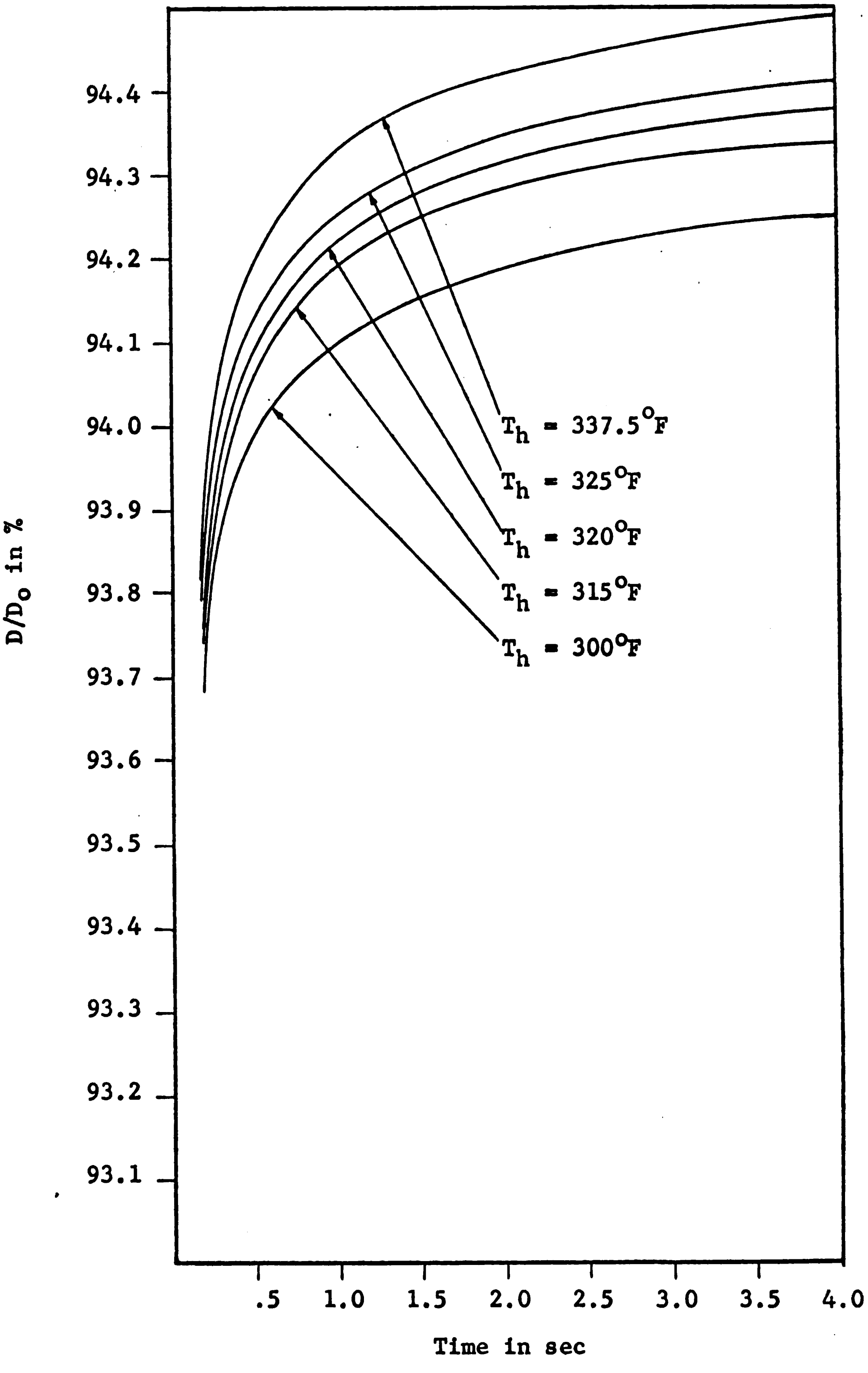


Figure 19

Diffusivity Ratio Functions Resulting
From Different $T_h$ Values

## ANALYSIS AND CONCLUSIONS

Simulation Model - Comparison of the temperature functions (Figures 6, 7, 8, 10) makes obvious the importance of including all the physical features of the heat sealing assembly in the computational method employed in simulation of the transient temperature behaviour at the bonding interface. Since the Beck's approach enables not only the manipulation of parameters according to the heat sealing system modeled, but also its solution agrees closely with the experimentally determined temperature function, it is ideally suited for studies of heat sealing. Its use does away with the effects of heatsealer construction variations which made until now any interlaboratory correlation and reproducibility impossible.

Contact Resistance - By the rather indirect method of contact resistance determination, Akers (14) found 0.0002 inch to be an appropriate value for contact resistance for polystyrene. For polypropylene the appropriate contact resistance was determined to correspond to 0.0001 inch. Noting that both the polymers possess the same basic molecular chain structure and vary only in the type and size of the side groups, phenyl and methyl respectively, the contact resistance seems to be related to the bulkiness of these side groups in manner similar to the glass transition temperature. Extension of this line of reasoning leads then to expectation of a much lower

contact resistance to be present in heat sealing of polyethylene which has a smaller hydrogen in place of methyl and phenyl groups of the above mentioned polymers.

Effect of Pressure - In Figure 20 the experimentally determined bond strength resulting from the same temperature settings and varying pressure levels are plotted. Each curve is associated with different pressure. It can be observed that initially the bond strength values rise with increase in pressure which indicates that some amount of pressure is necessary to overcome a kind of "bonding surface resistance". This resistance may be related to the thermal contact resistance although other factors are necessarily at play, since the thermal contact resistance did not require an increase in pressure to overcome it, or rather increase in pressure did not eliminate it (Table 2).

With further increase in pressure the bond strength values tend to become lower, or the rate of bond strength growth slows down as if the process of bond formation, diffusion, were laboring against some restrictive force. Based on speculation only, this could be viewed as a mass flow of the softened material which, because of resistance offered to it in the perpendicular direction due to higher viscosity in locations closer to the unheated side of the heat sealing assembly, tends to be forced by the pressure to move in parallel to the plane of bonding with the result of lesser contribution to the bond strength assumed here to depend on the number and depth of molecular segments penetrating across the plane of initial separation. The adverse effects of excessive pressure were discussed by several writers and remedial changes in heatsealer die profile were made, which sought

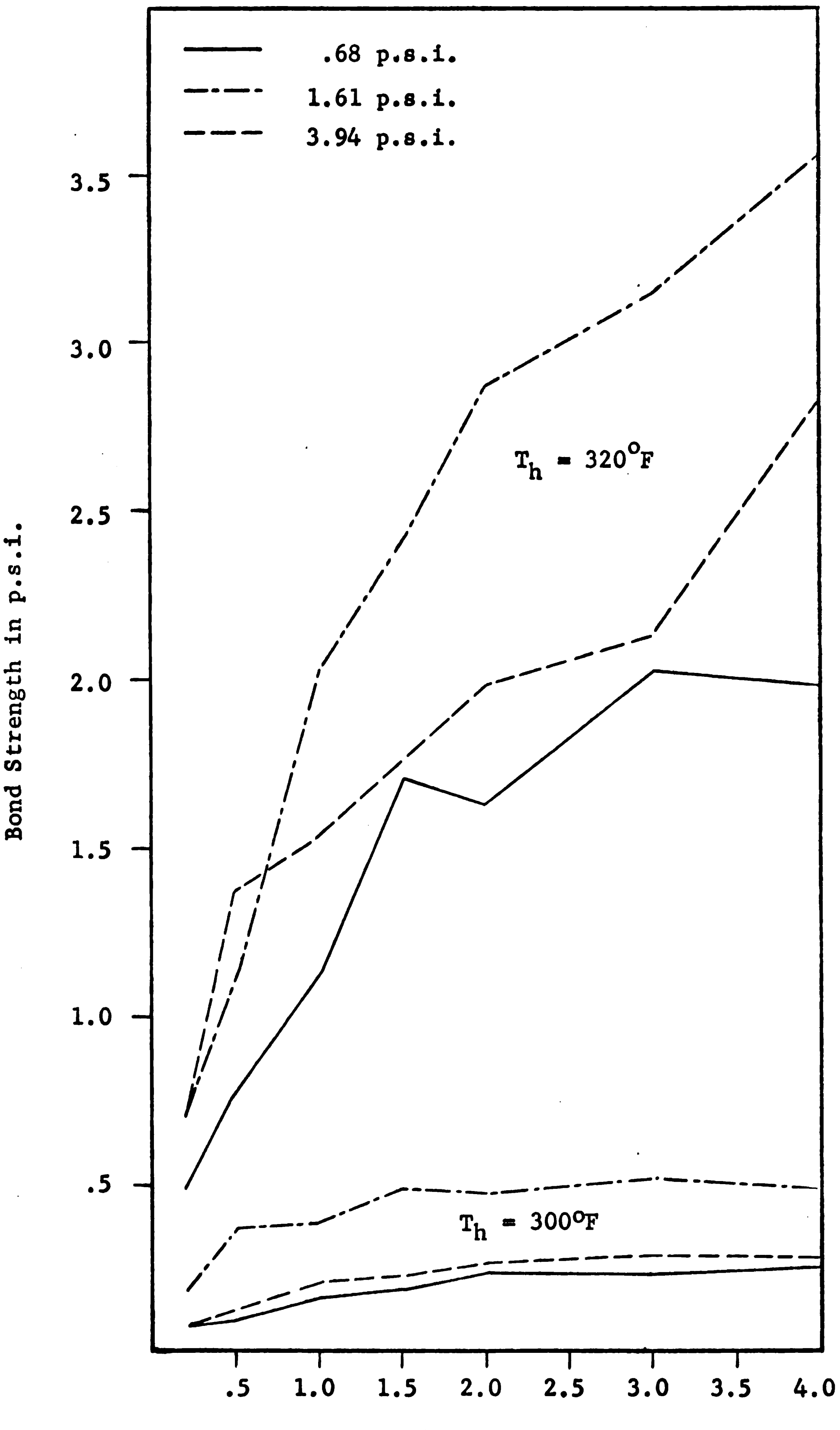


Figure 20

Bond Strength vs. Dwell Time for Two $T_h$ and Different Pressures to minimize the formation of bead by "extrusion" of molten material from the heat seal area, or to prevent "thinning" of the heat sealed area, in particular, the border line at which the heat sealed and un-heat sealed areas meet. Both the bead and the thinning cause a weakening of the material strength properties at the edge of the seal. Exactly at this edge failure occurs when the strength of a seal is tested which leads often to the erroneous conclusion that the bond is stronger than the material itself.

On the basis of the quite limited amount of experimental evidence obtained during this study, this writer is inclined to conclude that pressures in excess of some minimal required level are not only unnecessary but also detrimental both to the bonding rate, speed at which a bond is formed, and to the resulting bond strength.

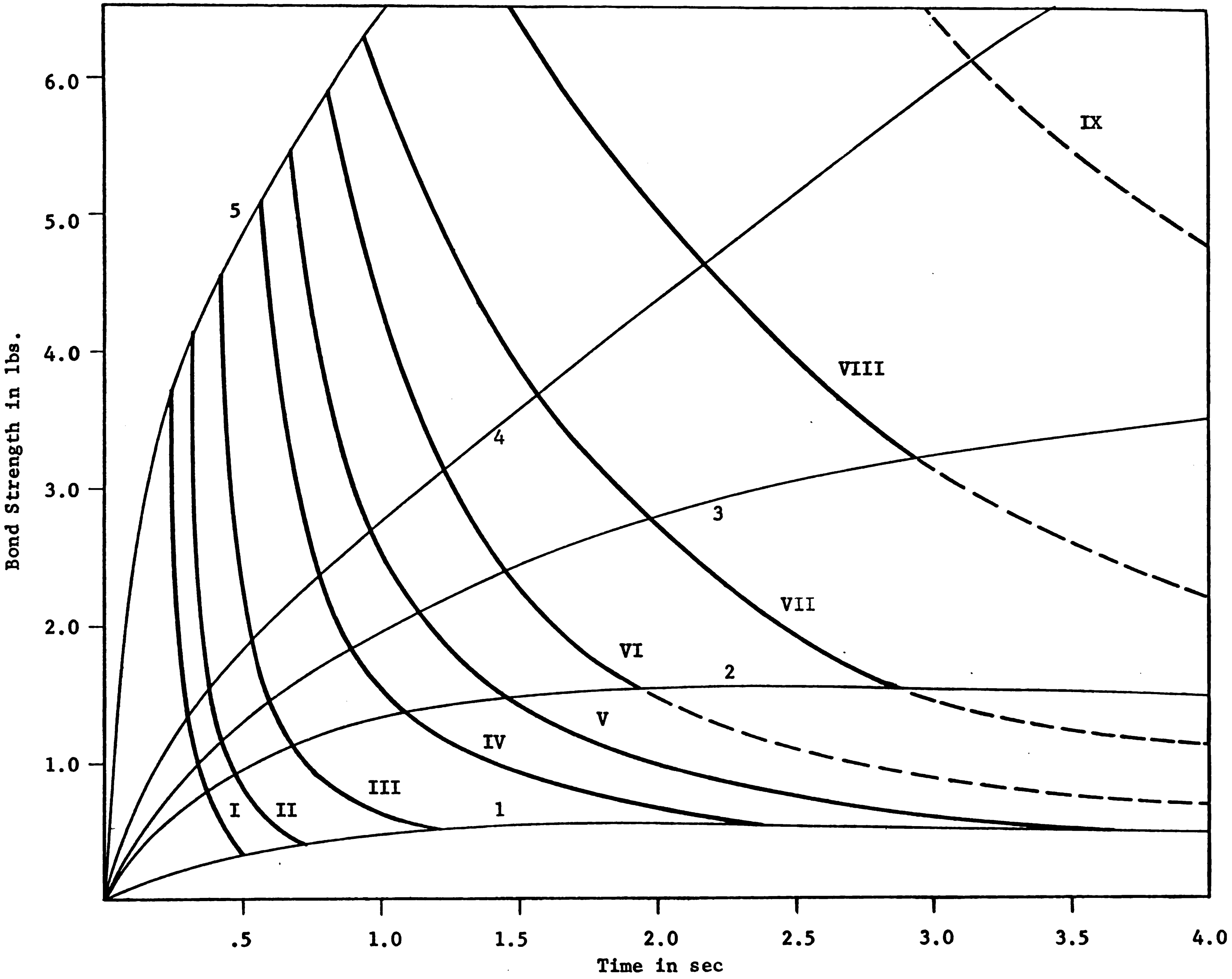
Bonding Rate - Plotting the bond strength values from Table 9, in Appendix II, for single pressure level and all $T_h$ results in a group of curves (Figure 21) from which interesting observations can be made. The bond strength as represented by curve 1 and 2 reaches maximum after approximately 2 seconds and starts slowly to decline with prolonged dwell times. This could be due to two distinct mechanisms. First, a number of low molecular weight polymer chains involved in bond formation in the early stages of the process moves on extended heating completely across the plane of the bonding interface, hence eliminating their contribution to the total bond strength. Second, during the initial period of heat sealing, segments and terminal portions of the molecular chain move across the plane of separation with the result of stressing some from the surface distant portions of the same molecular chain. Upon extended heating the molecular chain

Figure 21

Bond Strength vs. Dwell Time for Different $T_h$ Values

1 - Bond strength for $T_h$ of 300°F
2 - Bond strength for $T_h$ of 315°F
3 - Bond strength for $T_h$ of 320°F
4 - Bond strength for $T_h$ of 325°F
5 - Bond strength for $T_h$ of 337.5°F

I - Constant temperature of 240°F
II - Constant temperature of 250°F
III - Constant temperature of 260°F
IV - Constant temperature of 270°F
V - Constant temperature of 275°F
VI - Constant temperature of 280°F
VII - Constant temperature of 285°F
VIII - Constant temperature of 290°F
IX - Constant temperature of 295°F


6.0
5.0
4.0
3.0
2.0
1.0
Bond Strength in lbs.
.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
Time in sec
5
4
3
2
1
I
II
III
IV
V
VI
VII
VIII
IX

is allowed to achieve a configuration of least stress, or equilibrium of forces, which may result in total retraction of some, and partial retraction of others, to the bond contributing segments.

Each of the curves, 1 through 5, in Figure 21 represents the bond strength results obtained for each of the five initial temperatures $T_h$ for which we also have the computed temperature, viscosity, fluidity and diffusivity ratio $D/D_o$ functions (Figures 16, 17, 18, 19). It is now possible to plot on these bond strength curves the points of common value of the variables which, when connected by a smooth curve, give rise to a set of level curves or, since no three-dimensional relationship is intended to be implied, indifference curves, each representing a constant temperature, viscosity, fluidity and $D/D_o$. The indifference curves are identified on Figure 21 by Roman numerals I through IX. The shape of the curves seems to indicate a hyperbolic or exponential relationship between the variables considered above and the experimentally determined bond strength values.

The form these indifference curves could take are as follows:

Hyperbolic Curve -- $Y = A(X-C)^{-n}+B$

Exponential Curve -- $Y = Ae^{n(x-c)}+B$

Combination of Exponential Curves -- $Y = A_1e^{-bx} \pm A_2e^{-cx} + D$

The shape of the indifference curves clearly shows that the basically static approach of associating temperature directly with bond strength by use of steady state temperature profiles is not applicable. The transient features of the temperature, viscosity, fluidity and diffusivity ratio, in particular, the rise rates resulting from the heat sealing assembly's initial temperature gradient, are the

determining factors in bonding. In other words, the greater the thermal "shock", the better or stronger bonds are obtained. For example, if the bonding interface temperature of 270°F is arrived at in 0.6 second, a bond strength of 5.1 pounds per inch results. If it takes, however, 2.4 seconds to obtain the same temperature, 270°F, a bond strength of only 0.5 pounds per inch is obtained.

Although an explanation of this behaviour on the basis of some fundamental properties of the material involved has not been found, a practical use of the indifference curves, which this writer believes to be characteristic for each of the materials used in heat sealing, is possible.

Since heat seal strength is only one aspect of a flexible package's performance, the other being its barrier properties, no attempt was made, at this stage, to devise a simulation system for design optimatization. Until results of investigation of heat sealing conditions effect on the deterioration of flexible packaging materials barrier properties will be available, a procedure for quality control of heat sealing only, can be proposed.

Utilization of such a procedure presumes two factors. First, that the manufacturers supply together with other pertinent technical data, such as thermal conductivity, heat capacity, and possibly the appropriate contact resistance, a set of indifference curves developed for each of the materials they produce. Second, that the user, package fabricator, has access to, and ability to use a digital computer.

Assuming that both the conditions are satisfied, the user adapts the Beck's computational model to conform to the physical

features of the heat sealing situation, by assigning appropriate values to the relevant parameters, such as thermal and physical constants of the film or films, the thickness of each, and similar constants for the components of the heatsealer in question. By selecting a feasible heated bar temperature $T_h$, he then generates a temperature function whose values enable the determination of the expected bond strength.

It is obvious that such procedure could be used for a variety of purposes, from finding the proper temperature $T_h$ setting when the dwell time is invariable due to a fixed production speed of a machine and a desired level of bond strength is required, to determination of the dwell time necessary to achieve a given bond strength under fixed $T_h$, or for purely predictive purposes.

Care should be exercised not to be misled into regarding the indifference curves as the $T_h$ temperature which has no direct relationship with the bond strength.

If the simulation procedure were left entirely to a computer, the indifference curves would have to be stored in the computer memory which can be done by a system of ordered variable pairs (y,x) for each of the indifference curves, from which an intermediate value of y can be determined by means of any number of interpolating methods such as the Newton's Interpolation Method of Descending Differences.

$$f(x+n) = (1+\Delta)^n f(x)$$

$$= f(x)+n\,\Delta' f(x)+\frac{n(n-1)\,\Delta''}{2!}f(x)+\frac{n(n-1)(n-2)\,\Delta'''}{3!}f(x)+\cdots$$

where n = fraction of dx

$$\Delta' f(x) = f(x+dx)-f(x)$$

$$\Delta'' f(x) = \Delta' f(x+dx)-\Delta' f(x)$$

$$= f(x+2dx)-2f(x+dx)+f(x)$$

$$\Delta'''f(x) = \Delta''f(x+dx)-\Delta''f(x)$$

$$= f(x+3dx)-3f(x+2dx)+3f(x+dx)-f(x)$$

Another possibility is offered by representing each of the indifference curves by a power series which, because of the recognizable hyperbolic characteristic, will take the form,

$$y = A_0 + \frac{A_1}{x} + \frac{A_2}{x^2} \quad \frac{A_3}{x^3} + \ldots\ldots + \frac{A_{n-1}}{x^{n-1}} \quad .$$

Although the derivation of coefficients $A_n$ is rather lengthy, the final formulae offer the advantage of direct evaluation of the bond strength value y for any given x within the empirically defined interval.

The expressions for the indifference curves of Figure 21 are given below.

$$y_{240} = 10.029 - 18.329x^{-1} + 13.112x^{-2} - 4.487x^{-3} + .744x^{-4} - .046x^{-5}$$

$.250 \leq x \leq .500$

$$y_{250} = -235.499 + 720.552x^{-1} - 925.100x^{-2} + 647.910x^{-3} - 268.407x^{-4} + 66.317x^{-5} + .556x^{-6}$$

$.325 \leq x \leq .675$

$$y_{260} = 3110.0 - 19465.9x^{-1} + 52438.8x^{-2} - 79355.2x^{-3} + 73742.0x^{-4} - 43064.9x^{-5} + 15426.3x^{-6}$$

$.400 \leq x \leq 1.200$

$$y_{270} = 7.596 - 51.278x^{-1} + 143.934x^{-2} - 202.829x^{-3} + 155.754x^{-4} - 61.160x^{-5} + 9.564x^{-6}$$

$.500 \leq x \leq 2.300$

$$y_{275} = 42.0 - 583.5x^{-1} + 3342.2x^{-2} - 10120.7x^{-3} + 17507.0x^{-4} - 17243.6x^{-5} + 8921.3x^{-6} - 1860.2x^{-7}$$

$.650 \leq x \leq 3.450$

$$y_{280} = -.2564 + 4.6141x^{-1} - 7.4826x^{-2} + 13.4433x^{-3} - 5.7655x^{-4}$$

$.800 \leq x \leq 3.200$

$$y_{285} = 17.26 - 187.86x^{-1} + 789.25x^{-2} - 1510.52x^{-3} + 1376.90x^{-4} - 478.93x^{-5}$$

$1.000 \leq x \leq 3.500$

$$y_{290} = -7.971 + 64.309x^{-1} - 133.708x^{-2} + 146.174x^{-3} - 62.848x^{-4}$$

$1.200 \leq x \leq 3.200$

$$y_{295} = -4.562 + 57.554x^{-1} - 96.538x^{-2} + 68.275x^{-3}$$

$2.800 \leq x \leq 4.000$

*Recommendations for Future Study* - The known relationship between the bulkiness of side groups of polymers having identical backbone molecular structures and the $T_g$, as well as, the observation of the indirectly determined contact resistance which seems to follow a similar pattern, points out the possibility of existence of bulkiness-contact resistance relationship. As more metalized polymeric films become available, the resistance-molecular structure relationship should be studied with view on determining the contact resistance levels applicable for groups of films possessing similar structures.

Further, if quantitative methods are to be employed in heat seal bond study and bond strength prediction, the investigation of mechanisms and rates of the diffusion process are necessary. As already mentioned, radiography could be used in determining not only the

concentration and concentration gradient of the interdiffusing polymers but also in establishing the depth of penetration during bonding. The effects of pressure could be also studied by the above techniques. The results may have considerable consequences, such as lightweight construction and thus higher speeds of packaging machinery, if pressure is found to affect adversly the bonding rate or the depth of penetration of interdiffusing films.

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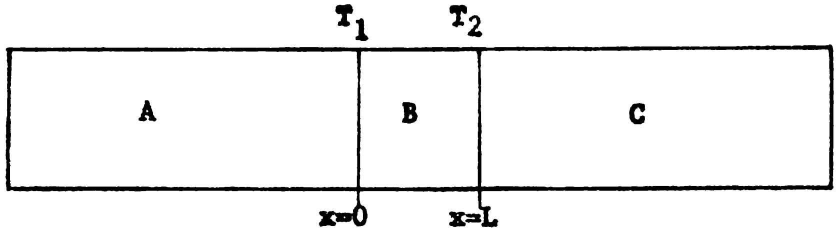
# APPENDIX I Models

# APPENDIX I

## Model I

Figure 22: Schematic Representation of Heat Sealing Assembly for Model I.



A = heated metal bar

B = film

C = unheated metal bar

Problem:

$$\frac{dT}{dt} = \alpha \frac{d^2T}{dx^2} \quad ; \; 0 \leq x \leq L$$

$$T_1 > T_2$$

$$T = T_1 = T_h, \text{ at } x = 0, \; t > 0$$

$$T = T_2, \qquad \text{at } x = L, \; t > 0$$

$$T = T_o = \text{constant at } t \leq 0$$

Solution: Equation I

$$T(x,t) = T_1 + (T_2 - T_1)\frac{x}{L} + \frac{2}{\pi}\sum_{1}^{\infty}\frac{T_2 \cos n\pi - T_1}{n} \sin\frac{n\pi x}{L} e^{-\frac{\alpha n^2 \pi^2 t}{L^2}}$$
$$+ \frac{2T_o}{\pi}\sum_{1}^{\infty} \sin\frac{n\pi x}{L} e^{-\frac{\alpha n^2 \pi^2 t}{L^2}} \left[\frac{1-\cos\pi n}{n}\right]$$

where $T(x,t)$ = temperature at x and time t

$L$ = total thickness of film

$T_h$ = temperature of heated bar

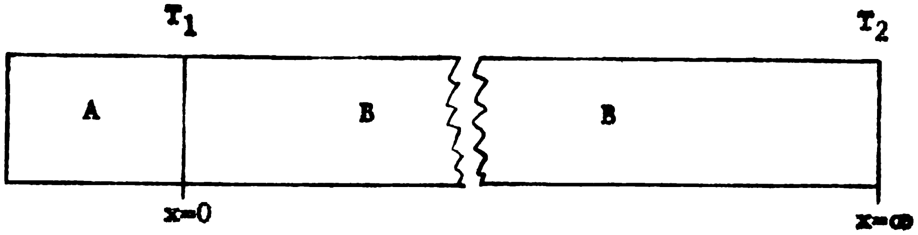
$T_o$ = initial film temperature

$\alpha$ = thermal diffusivity of film

$n$ = index of summation.

## Model II

Figure 23: Schematic Representation of Heat Sealing Assembly for Model II.



A = heated metal bar

B = film ( semi-infinite slab ).

Problem:

$$\frac{d^2T}{dx^2} - \frac{1}{\alpha}\frac{dT}{dt} = 0;\ 0 \leq x \leq \infty$$

$$T_1 > T_2$$

$$T = T_1 = T_h,\ \text{at } x = 0,\ t > 0$$

$$T = T_2 = T_o,\ \text{at } x = \infty,\ t \geq 0$$

Solution: Equation II

$$T(x,t) = T_1 + (T_2 - T_1)\frac{2}{\sqrt{\pi}}\int_0^{z=\frac{x}{2\sqrt{\alpha t}}} e^{-z^2}\,dz$$

$$\text{where } \frac{2}{\sqrt{\pi}}\int_0^{z} e^{-z^2}\,dz = \text{erf}(z) = \frac{T_1 - T(x,t)}{T_1 - T_2}$$

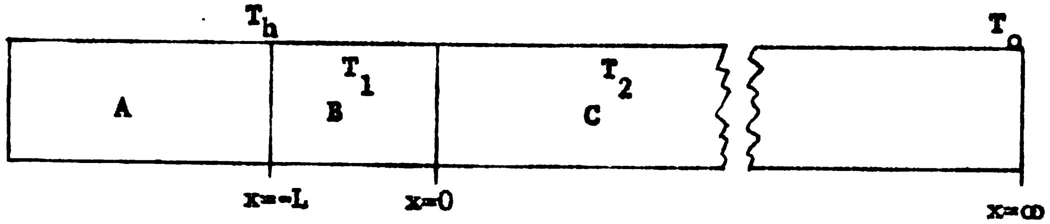
$T_h$ = temperature of heated bar

$T_o$ = initial temperature of the film

$\alpha$ = thermal diffusivity of film.

## Model III

Figure 24: Schematic Representation of Heat Sealing Assembly for Model III.



A = heated metal bar

B = air layer

C = film ( semi-infinite slab ) .

Problem:

$$\frac{d^2T_1}{dx^2} - \frac{1}{\alpha_1}\frac{dT_1}{dt} = 0 \;;\quad -L < x < 0,\ t > 0$$

$$\frac{d^2T_2}{dx^2} - \frac{1}{\alpha_2}\frac{dT_2}{dt} = 0 \;;\quad x > 0,\ t > 0$$

$$K_1\frac{dT_1}{dx} = K_2\frac{dT_2}{dx} \quad \text{at } x = 0,\ t > 0$$

$$T_1 = T_2 \quad \text{at } x = 0,\ t > 0$$

$$T_h > T_o$$

Solution: Equation III

$$T_2(x,t) = T_o + \frac{2(T_h - T_o)}{1 + \sigma}\sum_{0}^{\infty}\beta^n \operatorname{erfc}\frac{(2n + 1)L + bx}{2\sqrt{\alpha_1 t}}$$

where $T_2(x,t)$ = temperature within the film

$T_h$ = heated bar temperature.

$T_o$ = initial temperature

$K_1, K_2$ = thermal conductivities of air and film

$\alpha_1, \alpha_2$ = thermal diffusivities

$$b = \sqrt{\frac{\alpha_1}{\alpha_2}}$$

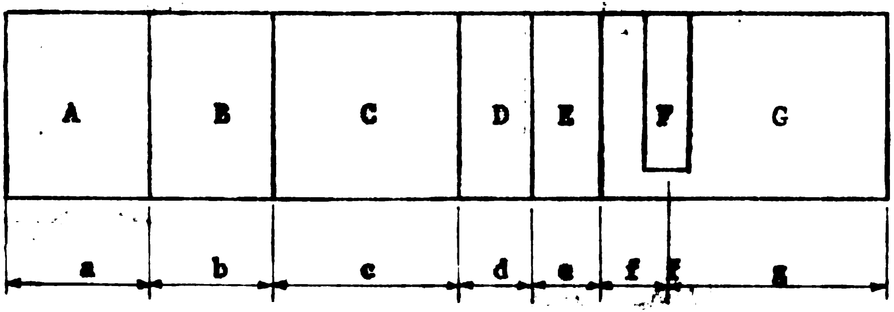
$$\sigma = \frac{K_2 b}{K_1}$$

$$\beta = \frac{\sigma - 1}{\sigma + 1}$$

$$\mathrm{erfc} = 1 - \mathrm{erf}$$

## Model IV

Figure 25: Schematic Representation of Heat Sealing Assembly for Model IV.



A = unheated metal bar

B = insulating material

C = film

D = air layer

E = Teflon coating

F = thermocouple

G = heated metal bar

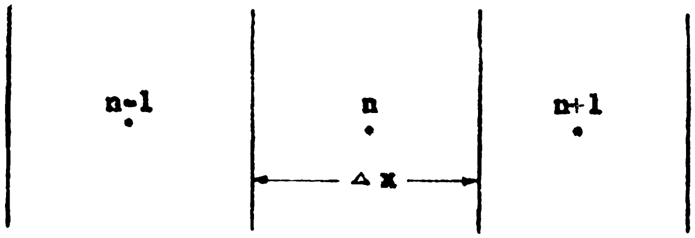
a,b,c,d,e,f,g = thicknesses of the components.

Problem: The problem still is to solve the heat conduction equation

$$\frac{d^2T}{dx^2} - \frac{1}{\alpha}\frac{dT}{dt} = 0 .$$

The approach used, however, is the Crank-Nicholson Method of Mean Finite Differences Approximation which employs the "heat balance", i.e., heat increase in a section = heat in - heat out. The solution is obtained from a system of simultaneous difference equations given below.

Figure 26: Nodal Scheme for Internal Nodes.



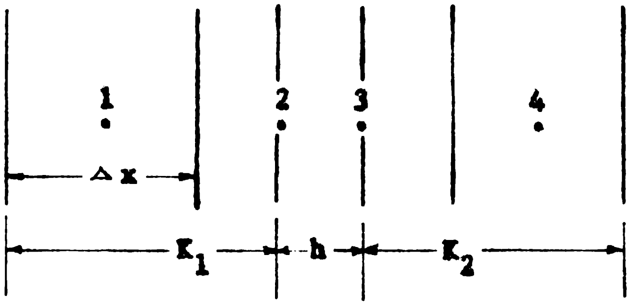
Equation 4-a.

$$\frac{1}{2}\left|\frac{K_1}{\Delta x}\right| \left|T^{m}_{n-1} - T^{m}_{n}\right| + \frac{1}{2}\left|\frac{K_1}{\Delta x}\right| \left|T^{m-1}_{n-1} - T^{m-1}_{n}\right|$$

$$-\frac{1}{2}\left|\frac{K_1}{\Delta x}\right| \left|T^{m}_{n} - T^{m}_{n+1}\right| - \frac{1}{2}\left|\frac{K_1}{\Delta x}\right| \left|T^{m-1}_{n} - T^{m-1}_{n+1}\right|$$

$$= \rho_1 C_1 \frac{\Delta x}{2} \frac{\left|T^{m}_{n} - T^{m-1}_{n}\right|}{\Delta t}$$

Figure 27: Nodal Scheme for Interface Nodes.



Equation 4-b.

$$T_1^{m}\left|K_1 - \frac{\lambda_2 \rho_1 C_1 (\Delta x_1)^2}{\Delta t}\right| - T_2^{m}\left|K_1 + 2h\Delta x_1 + \frac{\lambda_1 \rho_1 C_1 (\Delta x_1)^2}{\Delta t}\right|$$

$$+ T_3^{m}\left|2h\Delta x_1\right|$$

$$= T_1^{m-1}\left|K_1 - \frac{\lambda_2 \rho_1 C_1 (\Delta x_1)^2}{\Delta t}\right| - T_2^{m-1}\left|K_1 - \frac{\lambda_1 \rho_1 C_1 (\Delta x_1)^2}{\Delta t}\right|$$

for material with $K_1$.

where $T$ = temperature

$\Delta t$ = time increment

$\Delta x$ = distance between nodes

$n$ = node index

$m$ = time index

$\rho i$ = density of ith material

$C_i$ = heat capacity of ith material

$K_i$ = thermal conductivity of ith material

$h$ = contact resistance ($h = \frac{K_{air}}{L}$).

$L$ = thickness of air layer

$\lambda_1$ = boundary approximation constant

$\lambda_2 = 1 - \lambda_1$

# APPENDIX II TABLES

# APPENDIX II

TABLE 1 Computed Aluminum Resistivity as a Function of Temperature

| Temperature Deg. °F | Resistivity x $10^6$ ohm - cm |
|---|---|
| 100 | 3.068 |
| 110 | 3.135 |
| 120 | 3.202 |
| 130 | 3.270 |
| 140 | 3.338 |
| 150 | 3.406 |
| 160 | 3.475 |
| 170 | 3.543 |
| 180 | 3.613 |
| 190 | 3.682 |
| 200 | 3.752 |
| 210 | 3.822 |
| 220 | 3.892 |
| 230 | 3.962 |
| 240 | 4.033 |
| 250 | 4.104 |

TABLE 2 Thermistor Temperature Calibration Data for Varying Loading Levels at Sensitivity Setting of .02 Volts per Division

| Temperature Deg. °F | Loading Levels in lb./sq. in. | | | | |
|---|---|---|---|---|---|
| | .22 | .99 | 1.77 | 2.54 | 3.32 |
| 100 | *1.32 | | 1.36 | | 1.42 |
| 105 | | 1.52 | | 1.46 | |
| 110 | 1.79 | | 1.90 | | 1.88 |
| 115 | | 2.19 | | 2.00 | |
| 120 | 2.32 | | 2.43 | | 2.39 |
| 125 | | 2.68 | | 2.47 | |
| 130 | 2.78 | | 2.87 | | 2.70 |
| 135 | | 3.09 | | 2.99 | |
| 140 | 3.34 | | 3.41 | | 3.25 |
| 145 | | 3.52 | | 3.49 | |
| 150 | 3.80 | | 3.86 | | 3.95 |
| 155 | | 4.00 | | 4.10 | |
| 160 | 4.36 | | 4.22 | | 4.30 |
| 165 | | 4.50 | | 4.60 | |
| 170 | 4.80 | | 4.71 | | 4.89 |
| 175 | | 5.15 | | 5.20 | |
| 180 | 5.41 | | 5.22 | | 5.50 |
| 185 | | 5.67 | | 5.58 | |
| 190 | 5.82 | | 5.70 | | 5.93 |
| 195 | | 6.18 | | 6.06 | |
| 200 | 6.20 | | 6.30 | | 6.38 |
| 205 | | 6.45 | | 6.60 | |
| 210 | 6.84 | | 6.88 | | 6.74 |
| 215 | | 7.19 | | 7.21 | |
| 220 | 7.39 | | 7.29 | | 7.42 |
| 225 | | 7.71 | | 7.60 | |
| 230 | 7.78 | | 7.89 | | 7.92 |
| 235 | | 8.00 | | 8.15 | |
| 240 | 8.36 | | 8.45 | | 8.30 |
| 245 | | 8.56 | | 8.70 | |
| 250 | 8.82 | | 8.83 | | 8.98 |

*Tabulated entries represent divisions on oscilloscope screen.

TABLE 3 Temperatures Determined Experimentally

Thermistor Tickness = .001 inch
$T_o = 80^oF$, $T_h = 250^oF$

| Time Sec | Temperature at .001 inch away from Heated Bar |
|---|---|
| 0.00 | 80.0 |
| 0.05 | 170.5 |
| 0.10 | 182.5 |
| 0.30 | 206.5 |
| 0.50 | 214.0 |
| 0.75 | 219.2 |
| 1.00 | 222.1 |
| 1.50 | 225.5 |
| 2.00 | 228.5 |

TABLE 4 Temperatures Computed by Equation 1

$T_o = 80^oF$ , $T_h = 250^oF$

| Time Sec | Temperature at $x$ = .001 inch Deg. $^oF$ |
|---|---|
| 0.000 | 80.0 |
| 0.002 | 115.2 |
| 0.004 | 142.1 |
| 0.006 | 154.5 |
| 0.008 | 160.2 |
| 0.010 | 162.8 |
| 0.012 | 163.9 |
| 0.014 | 164.5 |
| 0.018 | 164.9 |
| 0.020 | 165.0 |
| $\infty$ | 165.0 |

TABLE 5 Temperatures Computed by Equation 2

$T_o = 80^oF$ , $T_h = 250^oF$

| Time Sec | Temperature at x = .001 inch Deg. °F |
|---|---|
| 0.00 | 80.0 |
| 0.02 | 197.3 |
| 0.04 | 212.3 |
| 0.06 | 219.1 |
| 0.08 | 223.1 |
| 0.10 | 225.9 |
| 0.20 | 232.9 |
| 0.60 | 240.1 |
| 1.00 | 242.4 |
| 1.40 | 243.5 |
| 1.80 | 244.3 |
| 2.20 | 244.9 |

TABLE 6 Temperatures Computed by Equation 3

$T_o = 80^oF$ , $T_h = 250^oF$, L = .0001 inch

| Time Sec | Temperature at x = .001 inch Deg. °F |
|---|---|
| 0.00 | 80.0 |
| 0.02 | 164.6 |
| 0.04 | 183.7 |
| 0.06 | 192.8 |
| 0.08 | 198.4 |
| 0.10 | 202.2 |
| 0.30 | 216.4 |
| 0.60 | 222.2 |
| 1.00 | 225.3 |
| 1.50 | 227.3 |
| 2.00 | 228.5 |

TABLE 7 **Temperature Computed by Equation 4 with Finite Thickness of Insulation**

$T_o = 80^\circ F$, $T_h = 250^\circ F$, $L = .0001$ inch

| Time Sec | Temperature at x = .001 inch Deg. °F |
|---|---|
| 0.00 | 80.0 |
| 0.02 | 144.5 |
| 0.06 | 170.6 |
| 0.10 | 187.1 |
| 0.20 | 207.8 |
| 0.40 | 220.4 |
| 0.60 | 224.7 |
| 0.80 | 227.3 |
| 1.00 | 229.1 |
| 1.50 | 231.9 |
| 2.00 | 233.6 |

TABLE 8 Temperature Computed by Equation 4 with Finite Thickness of Insulation and .001 inch Teflon Coating

$T_o = 80^\circ F$ , $T_h = 250^\circ F$

| Time | Temperature at x = .001 inch in deg. °F | | |
|---|---|---|---|
| Sec | L = .0001 inch | L = .0002 inch | L = .0004 inch |
| 0.00 | 80.0 | 80.0 | 80.0 |
| 0.10 | 179.8 | 164.9 | 144.7 |
| 0.20 | 197.1 | 184.4 | 164.8 |
| 0.30 | 204.9 | 193.8 | 175.4 |
| 0.50 | 213.3 | 203.9 | 187.6 |
| 0.70 | 217.9 | 209.7 | 194.9 |
| 0.90 | 221.1 | 213.6 | 199.9 |
| 1.00 | 222.3 | 215.1 | 201.9 |
| 1.50 | 226.4 | 220.4 | 209.1 |
| 2.00 | 228.9 | 223.6 | 213.5 |

TABLE 9 Bond Strength in Pound per Inch Tabulated for Each Heat Sealing Time and Pressure Combination and Varying Heated Bar Temperature $T_h$.

| $T_h$ Deg. °F | Pressure p.s.i. | Heat Sealing Time in Sec | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | .2 | .5 | 1.0 | 1.5 | 2.0 | 3.0 | 4.0 |
| 300 | [1].68 | .07 | .10 | .17 | .19 | .24 | .23 | .26 |
| | 1.15 | .12 | .13 | .15 | .21 | .20 | .21 | .24 |
| | 1.61 | .18 | .37 | .39 | .49 | .47 | .52 | .49 |
| | 2.09 | .14 | .22 | .29 | .36 | .35 | .34 | .34 |
| | 2.55 | .13 | .18 | .22 | .28 | .31 | .33 | .32 |
| | 3.02 | .09 | .19 | .23 | .26 | .29 | .28 | .30 |
| | 3.47 | .09 | .14 | .19 | .23 | .26 | .29 | .27 |
| | 3.94 | .08 | .15 | .21 | .23 | .27 | .29 | .28 |
| 315 | .68 | .27 | .64 | .86 | .98 | .92 | .88 | .91 |
| | 1.15 | .54 | .96 | .94 | .99 | 1.08 | .98 | 1.11 |
| | 1.61 | .59 | .84 | 1.18 | 1.51 | 1.56 | 1.48 | 1.51 |
| | 2.09 | .51 | .78 | .98 | 1.21 | 1.32 | 1.36 | 1.37 |
| | 2.55 | .41 | .80 | .98 | 1.18 | 1.43 | 1.46 | 1.43 |
| | 3.02 | .40 | .63 | .97 | 1.22 | 1.38 | 1.22 | 1.37 |
| | 3.47 | .39 | .57 | .92 | 1.06 | 1.36 | 1.28 | 1.41 |
| | 3.94 | .37 | .57 | .76 | .98 | 1.26 | 1.39 | 1.32 |

[1] Entries in lb/in are averages of ten samples

TABLE 9 (Continued)

| $T_h$ Deg. °F | Pressure p.s.i. | Heat Sealing Time in Sec |  |  |  |  |  |  |
|---|---|---|---|---|---|---|---|---|
|  |  | .2 | .5 | 1.0 | 1.5 | 2.0 | 3.0 | 4.0 |
| 320 | [i] .68 | .49 | .77 | 1.12 | 1.71 | 1.63 | 2.02 | 1.98 |
|  | 1.15 | .68 | .96 | 1.46 | 1.94 | 1.86 | 2.63 | 2.98 |
|  | 1.61 | .70 | 1.12 | 2.01 | 2.40 | 2.86 | 3.14 | 3.55 |
|  | 2.09 | .81 | 1.21 | 1.96 | 2.08 | 2.47 | 2.83 | 3.27 |
|  | 2.55 | .76 | 1.23 | 1.86 | 1.98 | 2.36 | 2.56 | 3.07 |
|  | 3.02 | .72 | 1.11 | 1.84 | 2.03 | 1.98 | 2.36 | 2.86 |
|  | 3.47 | .74 | 1.07 | 1.37 | 1.91 | 2.13 | 2.27 | 2.94 |
|  | 2.94 | .68 | 1.13 | 1.53 | 1.77 | 1.98 | 2.13 | 2.81 |
| 325 | .68 | .81 | 1.30 | 1.40 | 1.90 | [ii] - | - | [iii] 5.73 |
|  | 1.15 | 1.12 | 1.70 | 2.30 | 2.46 | - | - | 6.19 |
|  | 1.61 | 1.23 | 1.70 | 2.51 | 3.51 | - | 6.12 | - |
|  | 2.09 | 1.70 | 2.23 | 2.13 | - | - | 6.86 | - |
|  | 2.55 | .90 | 2.30 | 2.10 | 3.12 | - | - | 6.92 |
|  | 3.02 | .86 | 1.42 | 2.16 | 2.96 | - | - | 6.13 |
|  | 3.47 | .89 | 1.56 | 1.96 | 3.26 | - | - | 6.27 |
|  | 3.94 | .83 | 1.32 | 2.03 | 3.01 | 3.41 | - | 5.26 |

---

[i] Entries in lb/in are averages of ten samples.

[ii] Unable to determine because of too eratic record.

[iii] Failure not at the heat seal edge.

TABLE 9 (Continued)

| $T_h$ Deg. °F | Pressure p.s.i. | Heat Sealing Time in Sec | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | | .2 | .5 | 1.0 | 1.5 | 2.0 | 3.0 | 4.0 |
| 337.5 | [i] .68 | 2.13 | 2.96 | 3.47 | [iv] M | | | |
| | 1.15 | 3.27 | 4.03 | 6.31 | M | | | |
| | 1.61 | 3.48 | - | 6.43 | M | | | |
| | 2.09 | 3.04 | 3.84 | 6.03 | M | | | |
| | 2.55 | 2.64 | 3.19 | 5.96 | M | | | |
| | 3.02 | 3.18 | 3.29 | 5.76 | M | | | |
| | 3.47 | 3.21 | - | 5.22 | M | | | |
| | 3.94 | 3.56 | - | 4.96 | M | | | |

[i] Entries in lb/in are averages of ten samples.

[iv] Film melted.

TABLE 10 Temperatures at Bonding Interface Computed for .002 inch Polypropylene Film with Varying Heated Bar Temperatures $T_h$.

| Time Sec | Interface Temperature - °F. | | | | |
|---|---|---|---|---|---|
| | $T_h$=300°F | $T_h$=315°F | $T_h$=320°F | $T_h$=325°F | $T_h$=337.5°F |
| 0.00 | 80.0 | 80.0 | 80.0 | 80.0 | 80.0 |
| 0.04 | 149.5 | 154.1 | 155.7 | 157.2 | 161.1 |
| 0.08 | 173.6 | 179.8 | 181.9 | 184.0 | 189.2 |
| 0.12 | 188.2 | 195.4 | 197.9 | 200.3 | 206.3 |
| 0.16 | 199.4 | 207.4 | 210.1 | 212.8 | 219.4 |
| 0.20 | 208.4 | 217.0 | 219.9 | 222.7 | 229.9 |
| 0.50 | 240.9 | 251.7 | 255.3 | 258.8 | 267.8 |
| 1.00 | 256.8 | 268.6 | 272.5 | 276.5 | 286.4 |
| 1.50 | 263.7 | 276.0 | 280.1 | 284.2 | 294.4 |
| 2.00 | 267.9 | 280.5 | 284.7 | 288.9 | 299.3 |
| 3.00 | 273.0 | 285.9 | 290.2 | 294.5 | 305.3 |
| 4.00 | 276.0 | 289.1 | 293.5 | 297.9 | 308.8 |

TABLE 11 Viscosities at Bonding Interface of .002 inch Polypropylene Film Computed with Varying Heated Bar Temperature $T_h$.

| Time Sec | Viscosity in Poises | | | | |
|---|---|---|---|---|---|
| | $T_h$=300°F | $T_h$=315°F | $T_h$=320°F | $T_h$=325°F | $T_h$=337.5°F |
| 0.00 | -- | -- | -- | -- | -- |
| 0.04 | 6,451.4 | 4,156.4 | 3,605.9 | 3,135.1 | 2,230.1 |
| 0.08 | 809.8 | 507.3 | 463.6 | 376.7 | 263.6 |
| 0.12 | 282.6 | 175.8 | 151.1 | 130.2 | 90.8 |
| 0.16 | 137.0 | 85.1 | 73.1 | 62.9 | 44.0 |
| 0.20 | 80.4 | 49.9 | 42.9 | 37.0 | 25.9 |
| 0.50 | 15.4 | 9.7 | 8.3 | 6.9 | 5.1 |
| 1.00 | 7.9 | 4.9 | 4.3 | 3.7 | 2.6 |
| 1.50 | 5.9 | 3.8 | 3.3 | 2.8 | 2.0 |
| 2.00 | 5.1 | 3.2 | 2.8 | 2.4 | 1.7 |
| 3.00 | 4.2 | 2.7 | 2.3 | 2.0 | 1.4 |
| 4.00 | 3.8 | 2.4 | 2.1 | 1.8 | 1.3 |

TABLE 12 Fluidity at Bonding Interface of .002 inch Polypropylene Film Computed with Varying Heated Bar Temperatures $T_h$.

| Time Sec | Fluidity in Rhe x $10^2$ | | | | |
|---|---|---|---|---|---|
| | $T_h$=300°F | $T_h$=315°F | $T_h$=320°F | $T_h$=325°F | $T_h$=337.5°F |
| 0.00 | -- | -- | -- | -- | -- |
| 0.04 | 0.016 | 0.024 | 0.028 | 0.032 | 0.045 |
| 0.08 | 0.124 | 0.197 | 0.229 | 0.265 | 0.379 |
| 0.12 | 0.354 | 0.569 | 0.662 | 0.768 | 1.101 |
| 0.16 | 0.730 | 1.175 | 1.368 | 1.588 | 2.275 |
| 0.20 | 1.244 | 2.002 | 2.330 | 2.704 | 3.868 |
| 0.50 | 6.472 | 10.331 | 11.985 | 13.857 | 19.622 |
| 1.00 | 12.737 | 20.194 | 23.369 | 26.947 | 37.914 |
| 1.50 | 16.746 | 26.454 | 30.576 | 35.237 | 49.397 |
| 2.00 | 19.658 | 30.999 | 35.805 | 41.207 | 57.688 |
| 3.00 | 23.740 | 37.338 | 43.088 | 49.544 | 69.196 |
| 4.00 | 26.457 | 41.545 | 47.917 | 55.066 | 76.802 |

TABLE 13 $D/D_0$ at Bonding Interface of .002 inch Polypropylene Film Computed for Varying Heated Bar Temperatures $T_h$.

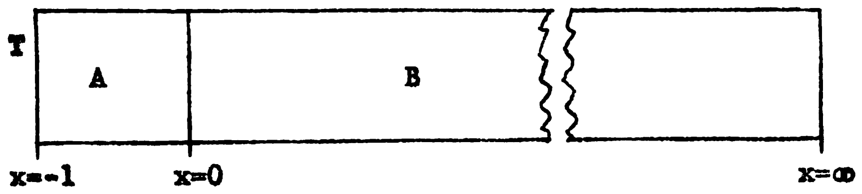
| Time Sec | $D/D_0$ in % | | | | |
|---|---|---|---|---|---|
| | $T_h$=300°F | $T_h$=315°F | $T_h$=320°F | $T_h$=325°F | $T_h$=337.5°F |
| 0.00 | -- | -- | -- | -- | -- |
| 0.04 | 93.10 | 93.15 | 93.16 | 93.18 | 93.22 |
| 0.08 | 93.35 | 93.41 | 93.43 | 93.45 | 93.51 |
| 0.12 | 93.50 | 93.57 | 93.59 | 93.61 | 93.67 |
| 0.16 | 93.60 | 93.68 | 93.70 | 93.73 | 93.79 |
| 0.20 | 93.69 | 93.76 | 93.79 | 93.81 | 93.88 |
| 0.50 | 93.99 | 94.06 | 94.09 | 94.13 | 94.19 |
| 1.00 | 94.10 | 94.19 | 94.22 | 94.25 | 94.33 |
| 1.50 | 94.15 | 94.25 | 94.28 | 94.31 | 94.39 |
| 2.00 | 94.19 | 94.28 | 94.31 | 94.35 | 94.42 |
| 3.00 | 94.23 | 94.32 | 94.36 | 94.39 | 94.46 |
| 4.00 | 94.25 | 94.35 | 94.38 | 94.41 | 94.49 |

# APPENDIX III Derviation of Model III

## APPENDIX III

### Derivation of Problem and Solution of Conduction of Heat in Composite Solids (Model III)

Figure 28: Schematic Representation of Composite Solid.



A = air layer

B = semi-infinite slab of film material.

Given: $\alpha_a$, $\rho_a$, $C_a$, $K_a$, $T_a$, and $\alpha_f$, $\rho_f$, $C_f$, $K_f$, $T_f$ to represent the diffusivity, density, heat capacity, conductivity, and temperature, with subscript a for air in region $-1 < x < 0$, and with subscript f for film material in region $x > 0$.

The differential equations to be solved are:

III-1 $$\frac{d^2T_a}{dx^2} - \frac{1}{\alpha_a}\frac{dT_a}{dt} = 0 \; ; \quad -1 < x < 0, \quad t > 0$$

III-2 $$\frac{d^2T_f}{dx^2} - \frac{1}{\alpha_f}\frac{dT_f}{dt} = 0 \; ; \quad x > 0 \; , \quad t > 0$$

Observing that $T_a = T_o + v_a$

$T_f = T_o + v_f$

$T = T_o + V$

where $T_o$ = initial temperature of the system

$v_a$ = variable temperature in air layer

$v_f$ = variable temperature in film material

T = constant temperature of the heated bar at $x = -1$

V = constant temperature gradient between $x = -1$ and $x = 0$.

We can now treat the whole problem as a solid with zero initial temperature and $x = -1$ kept at constant temperature V for $t > 0$.

The differential equations to be solved become:

III-1a $$\frac{d^2 v_a}{dx^2} - \frac{1}{\alpha_a}\frac{dv_a}{dt} = 0 \; ; \quad -1 < x < 0 \, , \; t > 0$$

III-2a $$\frac{d^2 v_f}{dx^2} - \frac{1}{\alpha_f}\frac{dv_f}{dt} = 0 \; ; \quad x > 0 \, , \; t > 0$$

Boundary conditions are:

III-3 $$K_a \frac{dv_a}{dx} = K_f \frac{dv_f}{dx} \; ; \quad x = 0 \, , \; t > 0$$

III-4 $$v_a = v_f \; ; \quad x = 0 \, , \; t > 0$$

Applying Laplace transformation to III-1a and III-2a yields the subsidiary equations:

III-5 $$\frac{d^2 \bar{v}_a}{dx^2} - q_a^2 \bar{v}_a = 0 \; ; \quad -1 < x < 0$$

III-6 $$\frac{d^2 \bar{v}_f}{dx^2} - q_f^2 \bar{v}_f = 0 \; ; \quad x > 0$$

III-7 where $q_a = (p/\alpha_a)^{\frac{1}{2}}$ , $q_f = (p/\alpha_f)^{\frac{1}{2}}$

These have to be solved with:

III-8 $$K_a \frac{d\bar{v}_a}{dx} = K_f \frac{d\bar{v}_f}{dx} \; ; \quad \bar{v}_a = \bar{v}_f \, , \text{ at } x = 0$$

III-9 $\bar{v}_a = \frac{V}{p}, \quad x = -l$

III-10 $\bar{v}_f \longrightarrow 0, \quad \text{as } x \longrightarrow \infty .$

A solution of III-5 which satisfies III-9 is:

III-11 $\bar{v}_a = \frac{V}{p} \cosh q_a (l+x) + A \sinh q_a (l+x),$

and a solution of III-6 which satisfies III-10 is:

III-12 $\bar{v}_f = Be^{-q_f x} .$

Substitution of III-11 and III-12 into III-8 gives us the unknowns A and B, and using the notation

III-13 $b = (\alpha_a/\alpha_f)^{\frac{1}{2}}, \quad \sigma = \frac{K_f b}{K_a}, \quad \beta = \frac{\sigma-1}{\sigma+1}$

we get

III-14 $$\bar{v}_a = \frac{V(\cosh q_a x - \sigma \sinh q_a x)}{p(\cosh q_a l + \sigma \sinh q_a l)}$$

III-15 $$\bar{v}_f = \frac{V}{p(\cosh q_a l + \sigma \sinh q_a l)} e^{-q_f x} .$$

Expressing the hyperbolic functions in III-15 in terms of negative exponentials, we get:

III-16 $$\bar{v}_f = \frac{2V}{p(1+\sigma)\left[1-\beta e^{-2q_a l}\right]} e^{-bq_a x - q_a l}$$

$$= \frac{2V}{(1+\sigma)p} \sum_{0}^{\infty} \beta^n e^{-q_a((2n+1)l+bx)} .$$

Again using Laplace transform as follows:

$$\text{III-17} \qquad \bar{v}(p) = \frac{e^{-qx}}{p} \longrightarrow v(t) = \operatorname{erfc} \frac{x}{2\sqrt{\alpha t}} ,$$

we get

$$\text{III-18} \qquad v_f = \frac{2V}{1+\sigma} \sum_{0}^{\infty} \beta^n \operatorname{erfc} \frac{(2n+1)l + bx}{2\sqrt{\alpha_a t}} .$$

Similarly

$$\text{III-19} \qquad v_a = V \sum_{0}^{\infty} \beta^n \left[ \operatorname{erfc} \frac{(2n+1)l + x}{2\sqrt{\alpha_a t}} - \beta \operatorname{erfc} \frac{(2n+1)l - x}{2\sqrt{\alpha_a t}} \right].$$

Since we are interested in temperatures within the film material, we substitute the relations $T_f = T_o + v_f$, and $T = T_o + V$ into III-18, to obtain the desired solution:

$$\text{III-20} \qquad T_f(x,t) = \frac{2(T-T_o)}{1+\sigma} \left[ \sum_{0}^{\infty} \beta^n \operatorname{erfc} \frac{(2n+1)l + bx}{2\sqrt{\alpha_a t}} \right] + T_o .$$

The solution is now complete.