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MOTION OF CONTINUOUS FIBERS THROUGH A NEWTONIAN RESIN FOR HIGH FIBER VOLUME FRACTION*

L. N. Hjellming** and J. S. Walker***

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** Graduate Research Assistant, Department of Theoretical and Applied Mechanics. Current address: Department of Mechanical Engineering and Engineering Science, University of North Carolina at Charlotte, Charlotte, NC 28223

*** Professor, Department of Mechanical Engineering

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Thermal Curing Cycles for Composite Cylinders with Thick Walls and Thermoset Resins

L. N. Hjellming
Department of Mechanical Engineering and Engineering Sciences
University of North Carolina, Charlotte, NC 28223
and
J. S. Walker
National Center for Composite Materials Research
University of Illinois, Urbana, Illinois 61801

ABSTRACT

This paper presents autoclave temperature histories to cure thick-section composite cylinders with graphite fibers and thermoset resins. The two cylinders considered have wall thicknesses of 0.15 m and 0.3 m. The first objective is to achieve a consolidation period when the resin viscosity is everywhere relatively low for a reasonably long period of time. For the 0.15 m thickness cylinder, the viscosity drops below 15 Pa · s for 4.1 hrs.; for the 0.3 m thickness cylinder, the viscosity drops below 30 Pa · s for 3.4 hrs. The second objective is to complete curing with the constraint that the temperature never exceeds 180 C at any point in the composite. This objective is achieved by limiting the autoclave temperature after the consolidation period. The peak interior temperature is very sensitive to small changes in the autoclave temperature during the final stages of curing.

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I. INTRODUCTION

This paper considers possible autoclave thermal curing cycles for long cylinders which have thick walls, which are composed of graphite fibers and thermoset resins, and which have no significant curing before placement in the autoclave. We consider composite cylinders made from Hercules 3501-6 resin because Lee, Loos and Springer [1] present equations for the rate of curing and viscosity of this resin as functions of degree of cure and temperature. We consider two cylinders with wall thicknesses of 0.15 m and 0.3 m, because comparable thicknesses may be appropriate for certain applications involving compressively loaded, thick-section composite cylinders.

We use the thermal-chemical model developed by Loos and Springer [2] for the coupled problem of the heat conduction through a composite section and the exothermic curing process which is temperature dependent. Loos and Springer [2] treat the thermal problem for graphite fiber-thermoset resin plates with thicknesses up to 0.021 m. For these thin plates, they conclude that the temperature can be kept uniform through the section at each time if the autoclave temperature is increased sufficiently slowly. Calius and Springer [3] apply this thermal-chemical model to filament-wound cylinders with thicknesses up to 0.032 m and threat thermal cycles which produce non-uniform temperatures. Both Loos and Springer [2] and Calius and Springer [3] compare the predictions of their thermal-chemical model with experimental measurements, and these comparisons validate their model for both uniform and non-uniform temperatures. Our objective is to use their model to identify optimal autoclave temperature histories for cylinders with walls which are so thick that strongly non-uniform temperatures are inevitable.

The viscosity μ of thermoset resins decreases with increasing temperature T and increases with increasing degree of cure α ($\alpha = 0$ corresponds to uncured resin and $\alpha = 1$ corresponds to completely cured resin). The viscosity of uncured resin at T = 20 C is too high for any significant resin flow to occur. At the beginning of the curing cycle, the autoclave temperature is increased and the resin viscosity decreases dramatically. This low viscosity permits the consolidation of the fibers and the elimination of excess resin. For filament wound cylinders, the consolidation force may be provided by the fiber tension alone or supplemented by an additional compressive force. The period of low viscosity is relatively brief because the resin cures quickly at the high autoclave temperatures.

At the beginning of the curing cycle, the autoclave must be heated in order to raise the temperature of the composite above 20 C and to initiate curing. However, curing is an exothermic process. Once the curing rate reaches a critical values, then the autoclave must be cooled in order to maintain a constant autoclave temperature. Without this cooling, the exothermic heat would cause the composite temperature to rise dramatically, and such high temperatures degrade the fibers and the resin. Loos and Springer [2] arbitrarily choose 180 C as the maximum permissible temperature to avoid materials degradation, and we use the same limit here.

In curing thick-section composites, a number of problems arise because of the low thermal conductivity of the composite. When we first raise the autoclave temperature, the inside and outside surfaces become hot, the local viscosity decreases and curing begins here. However, the central interior region, midway between the inner and outer surfaces, remains cold because it takes a long time for any heat to reach the interior by conduction. Therefore, the interior viscosity remains high during the period when the surface viscosity first decreases and then increases because of curing. Some consolidation occurs at the outside surface, but nowhere else. By the time the interior viscosity is sufficiently low to permit local resin flow, the outside surface resin has gelled, so that it prevents the escape of any interior resin. Therefore there is no significant consolidation or elimination of excess resin.

Once the interior becomes sufficiently hot for a significant rate of curing, the exothermic heat cannot reach the outside and inside surfaces by thermal conduction. Therefore, the interior temperature rises independently of the autoclave temperature. As the interior temperature rises, the rates of curing and of exothermic heat release also increase, which causes the interior temperature to rise even faster. Interior temperatures quickly reach extremely high values and severely damage the interior region.

We illustrate these effects with our first cylinder which has an inside radius $r_1 = 0.45$ m and a wall thickness $L = 0.15$ m (the outside radius $r_2 = 0.6$ m) and with two autoclave temperature cycles for curing thin-section composites. Loos and Springer [2] consider an autoclave temperature T_a which rises from 20 C to 177 C at a rate of 2.8 C/min. and which is then held constant at $T_a = 177$ C. For their plate with 0.021 m thickness, this cycle allows consolidation and involves a peak interior temperature of 180 C. For our first cylinder, the degree of cure at the inside and outside surfaces α_s has reached 0.46 when the interior temperature T_i (midway between the inside and outside surfaces) has only reached 32 C. By the time $T_i = 64$ C, $\alpha_s = 0.88$. Once the interior becomes somewhat hotter, it cures in a few minutes, and T_i reaches a peak of 358 C at 112 minutes after the start of the cycle.

The controllable variable is the autoclave temperature as a function of time, $T_a(t)$. The first objective for each thick-section composite cylinder is to achieve a consolidation period during which the viscosity is everywhere relatively low for a reasonably long period of time. The second objective is to achieve complete curing of the entire cylinder with T_i never exceeding 180 C. The second objective involves controlling the rate of curing in the interior, and this objective influences the first objective since curing rate depends on the entire history. The temperature during the consolidation period is limited because a higher temperature would initiate a rapid increase of the interior temperature and curing rate which could not be controlled by any subsequent autoclave temperature. We treat two cylinders where the second is twice the size of the first, i.e., the second cylinder has an inside radius $r_1 = 0.9$ m, a thickness $L = 0.3$ m, and an outside radius $r_2 = 1.2$ m.

We neglect the effects of consolidation on the thermal problem. In other words, we assume that the thermal diffusivity κ, the total heat of reaction per unit mass H_R, and the outside radius r_2 are constant throughout the entire curing process. The error here depends on what happens to the excess resin after it is squeezed out of the composite during consolidation. If the excess resin accummulates in a porous bleeder cylinder around the composite cylinder, then the present model may not be far off. The heat of reaction of the excess resin is still released, and the exothermic heat from the interior must still be conducted through the layer of excess resin in the bleeder cylinder. The position where $T = T_a$ at $r = r_2$ is effectively the outside surface of the excess resin in the bleeder cylinder, rather than the outside surface of the consolidated composite cylinder. On the other hand, if the excess resin is somehow removed as soon as it reaches the outside surface of the composite cylinder, then the present model may be far off after consolidation begins. Loos and Springer [2] treat the thermal and consolidation problems simultaneously and include the effects of consolidation on the thermal problem.

Our present purpose is to demonstrate the feasibility of curing thick-section composites with controlled autoclave temperature histories, $T_a(t)$. By neglecting the effects of consolidation on the thermal problem, we are making the second objective of keeping T_i below 180 C during the curing period more difficult to achieve. Consolidation would decrease H_R and increase κ, so that less exothermic heat would be released in the interior and its conduction to the inside and outside surfaces would be easier. Therefore, our second objective is easier to achieve in reality than in our conservative model. Our second purpose is to illustrate the critical role played by the thermal conductivity of the composite in determining acceptable autoclave temperature histories. Following Loos and Springer [2], we use the formula for the thermal conductivity of a composite presented by Springer and Tsai [4].

2. PROBLEM FORMULATION

We assume that the cylinder is sufficiently long that axial thermal conduction is negligible. We also assume an axisymmetric temperature. Therefore, the temperature T and the degree of cure α are only functions of the radial position r and of time t. The heat equation governing the temperature is

$$\frac{\partial T}{\partial t} = \frac{\kappa}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r} \right) + \frac{H_R}{C} \frac{\partial \alpha}{\partial t} \tag{1}$$

for $r_1 \leq r \leq r_2$ and for $0 \leq t < \infty$. The thermal diffusivity $\kappa = k/\rho C$, where k, ρ and C are the thermal conductivity, density and specfic heat of the composite. The law of mixtures is used for ρ, C and H_R, while the approximate formula of Springer and Tsai [4] is used for k. We assume that the fiber volume fraction is 0.53 throughout the curing cycle. The fiber volume fraction would normally increase to a value such as 0.7 during consolidation. This change would increase κ and decrease H_R/C, making it easier to keep the temperature below 180 C during the final stage of curing. Using the physical constants presented by Loos and Springer [2], we get $\kappa = 3.348 * 10^{-7}$ m^2/s, and $H_R/C = 230$ K. The quantity H_R/C represents the temperature increase due to adiabatic release of the heat of reaction.

We use the modified Arrhenius equation for the rate of curing presented by Lee, Loos and Springer [1],

$$\frac{\partial \alpha}{\partial t} = (K_1 + K_2 \alpha) (1 - \alpha) (B - \alpha), \qquad \text{for } \alpha \leq 0.3, \tag{2a}$$

$$\frac{\partial \alpha}{\partial t} = K_3 (1 - \alpha), \qquad \text{for } \alpha > 0.3, \tag{2b}$$

where

$$K_n = A_n \exp (- \Delta E_n / R T), \qquad \text{for } n = 1, 2, 3. \qquad (2c)$$

The pre-exponential factors are $A_1 = 2.101 \star 10^9 \text{ min}^{-1}$, $A_2 = -2.014 \star 10^9 \text{ min}^{-1}$, and $A_3 = 1.96 \star 10^5 \text{ min}^{-1}$. The activation energies ΔE_n, divided by the universal gas constant R, are $\Delta E_1/R = 9706$ K, $\Delta E_2/R = 9357$ K, and $\Delta E_3/R = 6807$ K. Here T is the absolute temperature, while the constant B = 0.47.

We assume that the thermal conductivity of the mandrel is much greater than that of the composite, so that

$$T = T_a(t), \qquad \text{at } r = r_1. \qquad (3)$$

If there is a porous bleeder cylinder around the composite, we assume that the thermal conductivity of the bleeder filled with air is much greater than the conductivity of the composite, so that

$$T = T_a(t), \qquad \text{at } r = r_2. \qquad (4)$$

The initial conditions are

$$T = 20 \text{ C} = 293 \text{ K}, \qquad \alpha = 0, \qquad \text{at } t = 0. \qquad (5)$$

We numerically integrate the equations (1,2) forward in time using a fourth-order Runge-Kutta scheme with a time step $(\Delta t) = 0.2 - 1.0$ minutes. The values of T and α at each time t are kept at 20 equally spaced grid points between r_1 and r_2 , and central differences are used for the radial derivatives in the equation (1). For the time step from t_1 to $t_1 + (\Delta t)$, the Runge-Kutta scheme involves computing the values of

$\partial T/\partial t$ and $\partial\alpha/\partial t$: (1) based on values of T and α at t_1, (2) based on a first estimate of the values of T and α at $t_1 + 0.5\ (\Delta t)$, (3) based on a second estimate of the values of T and α at $t_1 + 0.5\ (\Delta t)$, and (4) based on an estimate of the values of T and α at $t_1 + (\Delta t)$. The four sets of time derivatives are combined to obtain the change in T or α at a particular r for this time step. Since the radial derivatives in the equation (1) involve two neighboring values of T, each of the four steps in the Runge-Kutta scheme must be completed at all radial positions before the next step begins. We have checked the accuracy of this scheme by using 40 equally spaced grid points between r_1 and r_2 and by varying the time step (Δt).

3. CYLINDER WITH 0.15 m THICKNESS

For the consolidation period, we want to raise the temperature throughout the section while keeping the degree of cure at every radius as low as possible. For a given autoclave temperature history $T_a(t)$, and at each time in the curing cycle, we plot α_{max}, the maximum degree of cure at any radius, versus T_{min}, the minimum temperature at any radius. During the first stage of any curing cycle, the maximum degree of cure occurs at the outside and inside surfaces and the minimum temperature occurs at the central interior position midway between these surfaces, so that $\alpha_{max} = \alpha_s$ and $T_{min} = T_i$. As the exothermic heat accummulates in the interior, the interior temperature T_i rises to the surface temperature, which is also the autoclave temperature, so that $T_i = T_a$, at $t = t_{eq}$. At this time, the interior degree of cure α_i is still less than α_s. For the second brief stage immediately after $t = t_{eq}$, the interior has the highest temperature and the lowest degree of cure, so that $T_{min} = T_a$ and $\alpha_{max} = \alpha_s$. With the higher temperature, the interior cures quickly and α_i soon catches up to α_s. For the final stage in the curing cycle, $\alpha_{max} = \alpha_i$ and $T_{min} = T_a$. For $t < t_{eq}$, the cycle requires a heat input, while for $t > t_{eq}$, there is a heat efflux from the cylinder.

The solid lines in Figure 1 are the plots of α_{max} versus T_{min} for three autoclave temperature histories with T_a rising at 2 C/min. from 20 C to T_{ao} and then remaining constant at $T = T_{ao}$. The three curves correspond to T_{ao} = 60 C, 70 C, and 80 C. Our objective for the consolidation period is to produce a plot which is as low and as far to the right as possible, i.e., to minimize α_{max}, while maximizing T_{min}. The cycle with T_{ao} = 60 C provides a low curve until $T_i = T_a$, but then T_{min} remains at 60 C while curing continues slowly. The cycle with T_{ao} = 80 C cures the outside and inside surfaces quickly, so that it gives a high curve, even though it reaches T_{min} = 80 C before turning up. The initial rate of temperature increase, namely 2 C/min., is not important here. We varied this rate from 1 C/min. to 20 C/min. for these three values of T_{ao} and the various results are indistinguishable from those presented in Figure 1.

We wish to find an optimal autoclave temperature history which generates an envelope below and to the right of all possible plots of α_{max} versus T_{min}, where the three solid lines in Figure 1 represent three typical plots. We have found such an optimal envelope by trial and error through twenty more curing cycles. The plot of α_{max} versus T_{min} for this optimal autoclave temperature history is given by the dashed line in Figure 1. Our trial-and-error search successively revealed characteristics of the optimal cycle which guided the next stages of the search. With a knowledge of the characteristics of this optimal cycle, an optimal cycle for a different cylinder can be discovered without a lengthy trial-and-error search.

The solid curves for $T_{ao} = 70$ C and 80 C are nearly straight, but the curve for $T_{ao} = 60$ C begins with a small slope and then curves up sharply as T_{min} approaches 60 C. We generate a tangent off of the $T_{ao} = 60$ C curve where it curves up sharply at approximately $T_{min} = T_i = 55$ C and $\alpha_{max} = \alpha_s = 0.04$. We contrast the part of the $T_{ao} = 60$ C curve which we use with three alternate choices. The first alternative is to follow the $T_{ao} = 60$ C curve until $T_{min} = T_i = 60$ C, at $t = t_{eq}$, and then to raise T_a, so that $T_a = T_i$ throughout the next stage of the cure. This gives a nearly uniform temperature during this next stage. With this alternative, μ_s, the viscosity at the inside and outside surfaces, is greater than μ_i, the interior viscosity, because $\alpha_s > \alpha_i$, even though $T_i = T_a$. To compensate for the fact that $\alpha_s > \alpha_i$, we want T_a to be slightly higher than T_i after we leave the $T_{ao} = 60$ C curve. Thus, we leave this curve when $T_a = 60$ C and $T_i = 55$ C, and we raise T_a at a rate which keeps it slightly above T_i throughout the next stage of the curing cycle. Therefore, our curing cycle gives a more uniform viscosity at each time than the first alternative cycle. The second alternative would be to start on a $T_{ao} = 50$ C curve rather than on the $T_{ao} = 60$ C. The $T_{ao} = 50$ C curve is only slightly below the $T_{ao} = 60$ C curve, and, of course, turns up at $T_{min} = 50$ C. If we generate the best tangent off of the $T_{ao} = 50$ C curve, it ends up lying on top of the tangent off of the $T_{ao} = 60$ C curve. Therefore, this alternative gives a slightly lower α_{max} for $T_{min} < 55$ C, but the same α_{max} for $T_{min} > 55$ C. Since the viscosity is still relatively high for $T < 55$ C, this alternative does not provide any significant improvement.

On the other hand, it dramatically increases the curing time, so that there is a large penalty for negligible benefit. The third alternative is to begin the cycle with a temperature spike. For example, we could raise T_a from 20 C to 80 C at 2 C/min., then lower T_a from 80 C to 60 C at -2 C/min., and finally hold T_a = 60 C until time to follow the tangent. For all such spiked histories, the interior temperature T_i rises faster, but the surfaces cure much faster during the spike. The penalty in increased α_{max} is greater than the benefit in increased T_{min}, and the curves for initially spiked histories all lie above the corresponding curves shown in Figure 1.

Once we leave the T_{ao} = 60 C curve, we must choose dT_a/dt, the rate of increase of the autoclave temperature, and T_{ac}, the second constant temperature which follows this slow increase in T_a. For this part of the cycle, we consider only linear increases in T_a from 60 C to T_{ac}, and then constant $T_a = T_{ac}$. The value of T_{ac} is determined by the second objective of keeping T_i below 180 C throughout the curing cycle. This objective is discussed below, but it turns out that T_{ac} = 83 C for this cylinder and for the present assumptions.

We leave the T_{ao} = 60 C curve when T_i = 55 C and T_a = 60 C, and we want to pick the rate of increasing T_a from 60 C to 83 C We have considered seven values of dT_a/dt and we found that the rate which gives the lowest value of $\alpha_{max} = \alpha_s$ for each T_{min} is the one for which T_i and T_a both reach 83 C simultaneously. If dT_a/dt is larger than this rate, then T_a reaches 83 C before T_i, so that the surfaces are curing faster, while the interior is colder. If dT_a/dt is smaller than this rate, then T_i passes 83 C before T_a reaches 83 C. In this case, μ_i is considerably less than μ_s, so consolidation is limited by the surface viscosity which can be reduced by raising T_a, i.e., by using a higher values of dT_a/dt. In our trial and error, we chose various values of dT_a/dt and we integrated until $T_i = T_a$ at some temperature T_{ac}. We then held $T_a = T_{ac}$ and integrated until T_i peaked. Each value of dT_a/dt gave a different temperature at which T_i caught up to T_a and thus a different T_{ac}. We found that $dT_a/dt = 0.1$ C/min. = 6 C/hr. gave T_{ac} = 83 C, which led to a peak T_i of 180 C, which we discuss below.

We use the formula for viscosity presented by Lee, Loos and Springer [1],

$$\mu = \mu_\infty \exp \left[\frac{U}{R\,T} + K\,\alpha \right], \qquad \text{for } \alpha < 0.5, \qquad (6)$$

where $\mu_\infty = 7.93 * 10^{-14}$ Pa · s, U/R = 10,920 K, and K = 14.1. The equation (6) is only valid for $\alpha < 0.5$, i.e., before gellation. The surface viscosity μ_s and interior viscosity μ_i as functions of time for our optimal autoclave temperature history are presented in Figure 2. The curve for μ_s has two slope discontinuities: the first at t = 3.7 hrs. when we leave the T_{ao} = 60 C curve and begin to increase T_a at 6 C/hr., and the second at t = 7.5 hrs., when we stop increasing T_a and hold it at T_{ac} = 83 C. Our optimal curing cycle achieves a viscosity which is everywhere less than 20 Pa · s for 4.6 hrs. and which is everywhere less than 15 Pa · s for 4.1 hrs.

Once we know the viscosities as functions of time, then a consolidation model for thick-section composites can determine the consolidation force required to achieve a given final fiber volume fraction. If the fiber tension alone cannot provide sufficient force, then an additional compressive force is needed. Such an additional force can be provided by bagging the composite cylinder and the surrounding porous bleeder cylinder and by increasing the autoclave pressure p_a above the bag pressure p_b. The bag pressure must be relatively high to prevent growth of water-vapor voids when T_i climbs to 180 C [5] and the autoclave pressure must be even higher because $p_a - p_b$ is the consolidation pressure exerted on the outside of the bleeder cylinder by the bag.

The peak value of T_i is very sensitive to small changes in T_{ac}, the constant autoclave temperature for $t > t_{eq}$, where $t_{eq} = 7.5$ hrs. for our optimal cycle. The value $T_{ac} = 83$ C gives a peak $T_i = 180$ C at $t = 13.6$ hrs. For $T_{ac} = 85$ C, T_i peaks at 201 C at $t = 12.1$ hrs., and for $T_{ac} = 82$ C, T_i peaks at 167 C at $t = 14.5$ hrs. Such a sensitivity indicates the need for a control which makes small adjustments of T_a during the final stage of the curing cycle. We should monitor the total heat efflux from the cylinder or the rate of curing at some interior position. If the heat efflux or interior rate of curing begin to rise sharply, then the curing is too fast for the escape of the exothermic heat, and we should lower T_a slightly. If the heat efflux or interior rate of curing begin to decrease, then the curing is too slow, and we should increase T_a slightly. Such a control would compensate for variations between nearly similar cylinders. After the interior reaches $\alpha_i = 1.0$, we can raise T_a to complete the curing of the inside and outside surfaces quickly. We arbitrarily choose to raise T_a from 83 C to 163 C at 2 C/min.

The autoclave temperature history T_a, which is also the temperature of the inside and outside surfaces, is presented in Figure 3a. The interior temperature T_i is presented in the same figure. The period for

4.6 hrs. < t < 9 hrs., when T_a and T_i are relatively close and are both above 60 C, is the consolidation period. After t = 9 hrs., T_i is considerably above T_a, so that the interior is loosing heat to the surfaces and dT_i/dt decreases, but stays positive. As the slowed rise in T_i continues, the curing rate increases exponentially. The increased exothermic heat release overwhelms the thermal conduction to the surfaces, and T_i shoots up to its peak at 180 C. The degrees of cure at the surfaces α_s and at the interior α_i are presented in Figure 3b. One remaining difficulty is that the interior cures completely while the surfaces have only reached $\alpha_s = 0.25$. The maximum difference between α_i and α_s can be reduced by setting a lower limit on the peak value of T_i, which reduces the value of T_{ac}. However, this step dramatically increases the total time to cure the cylinder.

4. CYLINDER WITH 0.3 m THICKNESS

The solid lines in Figure 4 are the plots of α_{max} versus T_{min} for four autoclave temperature histories with T_a increasing from 20 C to T_{ao} at 1 C/min., and then staying constant at $T_a = T_{ao}$. The curves correspond to T_{ao} = 50 C, 60 C, 70 C, and 80 C. If we follow the guidelines presented in the previous section, we obtain an optimal curing cycle represented by the dashed line (e) in Figure 4. A slightly lower α_{max} versus T_{min} curve could be obtained by following the T_{ao} = 50 C curve until $T_i = T_a$ at $t = t_{eq}$, and then increasing T_a so that $T_a = T_i$ during the period following $t = t_{eq}$. However, α_s would be considerably larger than α_i, so that μ_s would be much larger than μ_i, even though $T_i \doteq T_a$. Therefore, we leave the T_{ao} curve when $T_i = T_{min}$ = 41 C and $\alpha_{max} = \alpha_s = 0.045$ at t = 9.36 hrs. As we raise T_a from this point, we are improving the uniformity of the viscosity, even though the α_{max}

versus T_{min} curve is not the lowest possible one. Following the previous guidelines, we raise T_a at 0.02 C/min. until $T_i = T_a = 63.2$ C at t= 20.2 hrs. We then hold $T_a = T_{ac} = 63.2$ C for t > 20.2 hrs., and T_i peaks at 180 C at t = 38.0 hrs.

There are two problems associated with this "optimal" curing cycle. First, it takes a long time at the beginning of the cycle for the interior temperature to begin to rise. During this long period, the surfaces are curing, so that α_s has grown to 0.045 when T_i has only reached 41 C. The second problem is that T_a must stop at $T_{ac} = 63.2$ C to keep the peak value of T_i below 180 C. This T_{ac} is too small to reduce the surface viscosity to an acceptable value, especially because α_s has reached 0.109 in the 20.2 hours needed for T_a to reach 63.2 C. The results of these two problems are demonstrated by the plots of μ_s and μ_i for this "optimal" cycle, which are presented in Figure 5. The surface viscosity barely dips below 50 Pa · s before T_a reaches 63.2 C and μ_s begins to rise sharply. At $t = t_{eq} = 20.2$ hrs., $T_a = T_i = 63.2$ C, but $\alpha_s = 0.109$, while $\alpha_i = 0.065$, so that $\mu_s = 47.1$ Pa · s, while $\mu_i =$ 26.2 Pa · s. For the 0.15 m thickness cylinder, the first stage of curing gave a slight elevation of α_s above α_i, and we compensated for this difference by leaving the $T_{ao} = 60$ C curve when $T_a = 60$ C and $T_i =$ 55 C. Here we attempt to make the same compensation by leaving the $T_{ao} = 50$ C curve when $T_a = 50$ C and $T_i = 41$ C, but this step is insufficient to compensate for the large difference between α_s and α_i.

For the 0.15 m thickness cylinder, we considered only monotonically increasing autoclave temperature histories, except for the histories with initial spikes which were worse than the monotonic histories. Here the $T_{ac} = 63.2$ C needed to keep T_i below 180 C for a monotonic history is simply too low to compensate for the high value of α_s. The only alternative is to raise T_a above 63 C in order to reduce μ_s during the consolidation period, and then to decrease T_a to whatever value is necessary to prevent the interior temperature T_i from exceeding 180 C during the final stage of curing.

We have considered ten possible autoclave temperature histories which (1) follow the $T_{ao} = 50$ C curve until t = 9.36 hrs., (2) then rise at $(dT_a/dt)_1$ from T_{ao} to T_{ap}, the peak autoclave temperature, (3) then decrease at $(dT_a/dt)_2$ from T_{ap} to T_{ac}, and (4) finally remain constant at $T_a = T_{ac}$. Of the histories considered, the best is the one for the curve (f) in Figure 4, with $(dT_a/dt)_1 = 0.04$ C/min., $T_{ap} = 76.4$ C at t = 20.2 hrs., $(dT_a/dt)_2 = -0.2$ C/min., and $T_a = T_{ac} = 52.4$ C for t > 22.2 hrs. For consolidation, we want to make T_{ap} as high as possible and delay the temperature decrease as long as possible. However, both of these steps make it more difficult to prevent T_i from exceeding 180 C. Once the interior begins to cure too fast, it is impossible to control by lowering T_a. If we continue the increase in T_a at $(dT_a/dt)_1 = 0.04$ C/min. to $T_{ap} = 78.8$ C at t= 21.2 hrs., and then bring T_a down to $T_a = 50$ C, the peak in T_i is 215 C at t = 26 hrs. The decrease in T_a is simply too late to stop the rapid rise in T_i. Once we begin to decrease T_a from T_{ap} to T_{ac}, we want to do it as fast as possible. This decrease in T_a abruptly ends the consolidation period, because μ_s quickly shoots up past 100 Pa · s, so that the objective of lowering T_a is to get to T_{ac} as soon as possible to control T_i. We have chosen $(dT_a/dt)_2 = -0.2$ C/min. with the assumption that the autoclave is only capable of cooling a large, long, massive composite cylinder at a rate comparable to this.

The interior viscosity μ_i and surface viscosity μ_s for our optimal, non-monotonic autoclave temperature history are presented in Figure 6. These values are clearly an improvement over those presented in Figure 5. Before μ_s barely dipped below 50 Pa · s, while now μ_s almost reaches 25 Pa · s. The viscosity is everywhere below 30 Pa · s for 3.4 hrs. and below 35 Pa · s for 4.2 hrs. For the 0.3 m thickness, we do not achieve the uniformity of viscosity that we achieve for the 0.15 m thickness cylinder, as presented in Figure 2. Here μ_i has not even reached its minimum before we must decrease T_a and end the consolidation period. However, the reason μ_i is still decreasing at $t = 20.2$ hrs. is that T_i is rising rather quickly, so that the interior curing is becoming progressively more difficult to control. We have only considered histories with a linear temperature increase from 50 C to T_{ap}, followed immediately by a linear temperature decrease from T_{ap} to T_{ac}. A non-linear temperature increase or a plateau at constant T_{ap} might improve the uniformity of the viscosity slightly, but a significant improvement does not appear to be possible.

The autoclave or surface temperature T_a and the interior temperature T_i for our optimal, non-monotonic autoclave temperature history are presented in Figure 7a. Again we raise T_a to 163 C after $\alpha_i = 1.0$ at $t = 30.5$ hrs., in order to complete the surface curing quickly. The consolidation period is again the period when T_i and T_a are relatively close and both are above 55 C. The surface and interior degrees of cure are presented in Figure 7b. The values of α_s and α_i are further apart here than in Figure 3b. The interior completes curing when α_s has only reached 0.18.

5. CONCLUSIONS

We have shown that it is possible to achieve: (1) a consolidation period with a viscosity which is everywhere relatively low for a reasonable period of time, and (2) a complete cure with the temperature never exceeding 180 C. The different results for the cylinders with 0.15 m and 0.3 m thicknesses illustrate that there is no simple scaling law for thickness. The characteristic time for thermal conduction through the section with thickness L is L^2/κ, which is 18.67 hrs. and 74.67 hrs. for our two cylinders. The characteristic time for curing is independent of L and varies as exp $(\Delta E/RT)$. We must decrease T as we increase L in order to maintain a constant ratio of these two characteristic times. However, then the viscosity increases with L because T is decreasing, so that consolidation becomes difficult. As L increases, we do decrease T but not enough to keep the ratio of the characteristic times the same. Therefore, curing is occurring faster than thermal conduction for the thicker cylinder. If all times scaled with the characteristic conduction time, then it would take four times as long to cure the second cylinder than the first. Figures 3 and 7 show that it only takes twice as long with our cycles. With curing being faster than thermal conduction, the degree of cure is higher which makes the viscosity higher for the thicker cylinder. The Figures 2 and 6 show that the consolidation times are roughly comparable, but the viscosity of the thicker cylinder is roughly twice that of the thinner one during their consolidation periods. Therefore we have as much time to squeeze the excess resin out of a section which is twice as thick and has twice the viscosity. We can expect the consolidation force required to remove excess resin to vary roughly as L^2.

If a cylinder with a thick wall is wound with considerable excess resin and is then consolidated to reach the desired fiber volume fraction, the fibers may wrinkle during consolidation and curing. Fiber wrinkles may decrease the strength of the final cylinder, particularly in compression. Therefore, a cylinder may be wound with very little excess resin and with a fiber volume fraction of 0.65 - 0.75. The consolidation period is less important, but our second objective of limiting the maximum interior temperature is still important. The present model applies to this process with little resin removal if we increase the fiber fraction. This change increases the value of κ and decreases the value of H_R, so that complete cure can be achieved in a much shorter period of time with higher autoclave temperatures and with smaller peak differences between T_a and T_i.

We have neglected any temperature differences through the mandrel at r_1 and through the bleeder and bag at r_2. In reality, the mandrel for a large cylinder must be quite thick and may have a significant temperature difference. Similarly, if the bleeder and bag are made from materials with low thermal conductivities, then they too may have a significant temperature difference. Given a particular process, the temperature drops across the mandrel and bag/bleeder could easily be included by solving the heat equation (1) with $H_R = 0$ and the appropriate values of κ in the mandrel and in the bag/bleeder. These additional thermal barriers clearly make both of the present objectives more difficult to achieve.

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CAPTIONS FOR FIGURES

Figure 1. Plots of α_{max} versus T_{min} for the cylinder with 0.15 m thickness and for four autoclave temperature histories, $T_a(t)$. For the solid lines, T_a increases from 20 C to a value T_{ao} at 2 C/min., and then T_a is held constant at $T_a = T_{ao}$: (a) $T_{ao} = 60$ C, (b) $T_{ao} = 70$ C, and (c) $T_{ao} = 80$ C. For the dashed line: (1) T_a increases at 2 C/min. from 20 C at $t = 0$ to 60 C at $t = 20$ mins., (2) $T_a = 60$ C for 20 mins. $\leq t \leq 3.7$ hrs., (3) T_a increases at 0.1 C/min. or 6 C/hr. from 60 C to 83 C, and (4) $T_a = 83$ C for $t \geq 7.5$ hrs.

Figure 2. Viscosity at the inside and outside surfaces μ_s and at the interior position midway between these surfaces μ_i as functions of time t for the cylinder with the 0.15 m thickness and for the optimal autoclave temperature history corresponding to the dashed line in Fig. 1.

Figure 3. Temperatures and degrees of cure as functions of time for the 0.15 m thickness cylinder and for the optimal curing cycle.
(a) Autoclave or surface temperature T_a and interior temperature T_i.
(b) Surface degree of cure α_s and interior degree of cure α_i.

Figure 4. Plots of α_{max} versus T_{min} for the cylinder with 0.3 m thickness and for six autoclave temperature histories, $T_a(t)$. For the solid lines, T_a increases from 20 C to a value T_{ao} at 1 C/min. and then remains constant at $T_a = T_{ao}$: (a) $T_{ao} = 50$ C, (b) $T_{ao} = 60$ C, (c) $T_{ao} = 70$ C, and (d) $T_{ao} = 80$ C. The two dashed lines coincide with the solid line (a) for $0 \leq t \leq 9.36$ hrs. For the dashed line (e), T_a rises from 50 C at $t = 9.36$ hrs. to 63.2 C at $t = 20.2$ hrs. with 0.02 C/min., and then remains constant at $T_a = 63.2$ C for $t \geq 20.2$ hrs. For the dashed line (f), T_a rises from 50 C at $t = 9.36$ hrs. to 76.4 C at $t = 20.2$ hrs. with 0.04 C/min., then T_a decreases to 52.4 C at

at t = 22.2 hrs. with -0.2 C/min., and finally T_a remains constant with $T_a = T_{ac} = 52.4$ C for t ≥ 22.2 hrs.

Figure 5. Surface viscosity μ_s and interior viscosity μ_i versus time for the 0.3 m thickness cylinder and for the curing cycle (e) in Figure 4.

Figure 6. Surface viscosity μ_s and interior viscosity μ_i versus time for the 0.3 m thickness cylinder and for the curing cycle (f) in Figure 4.

Figure 7. Temperatures and degrees of cure as functions of time for the 0.3 m thickness cylinder and for the curing cycle (f) in Figure 4.

(a) Autoclave or surface temperature T_a and interior temperature T_i.

(b) Surface degree of cure α_s and interior degree of cure α_i.

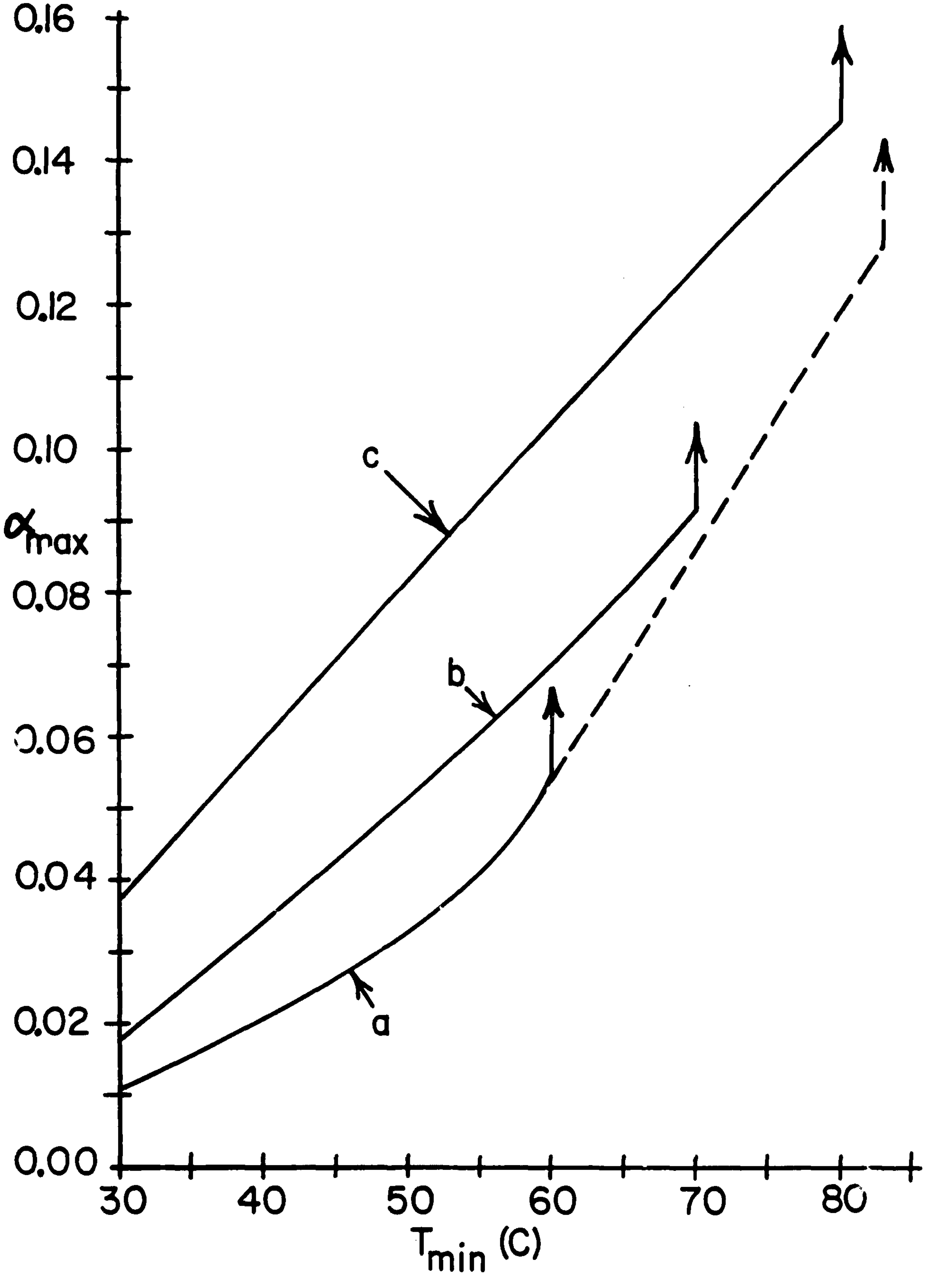


Figure 1

FIG 1. H TELLMING & WALKE

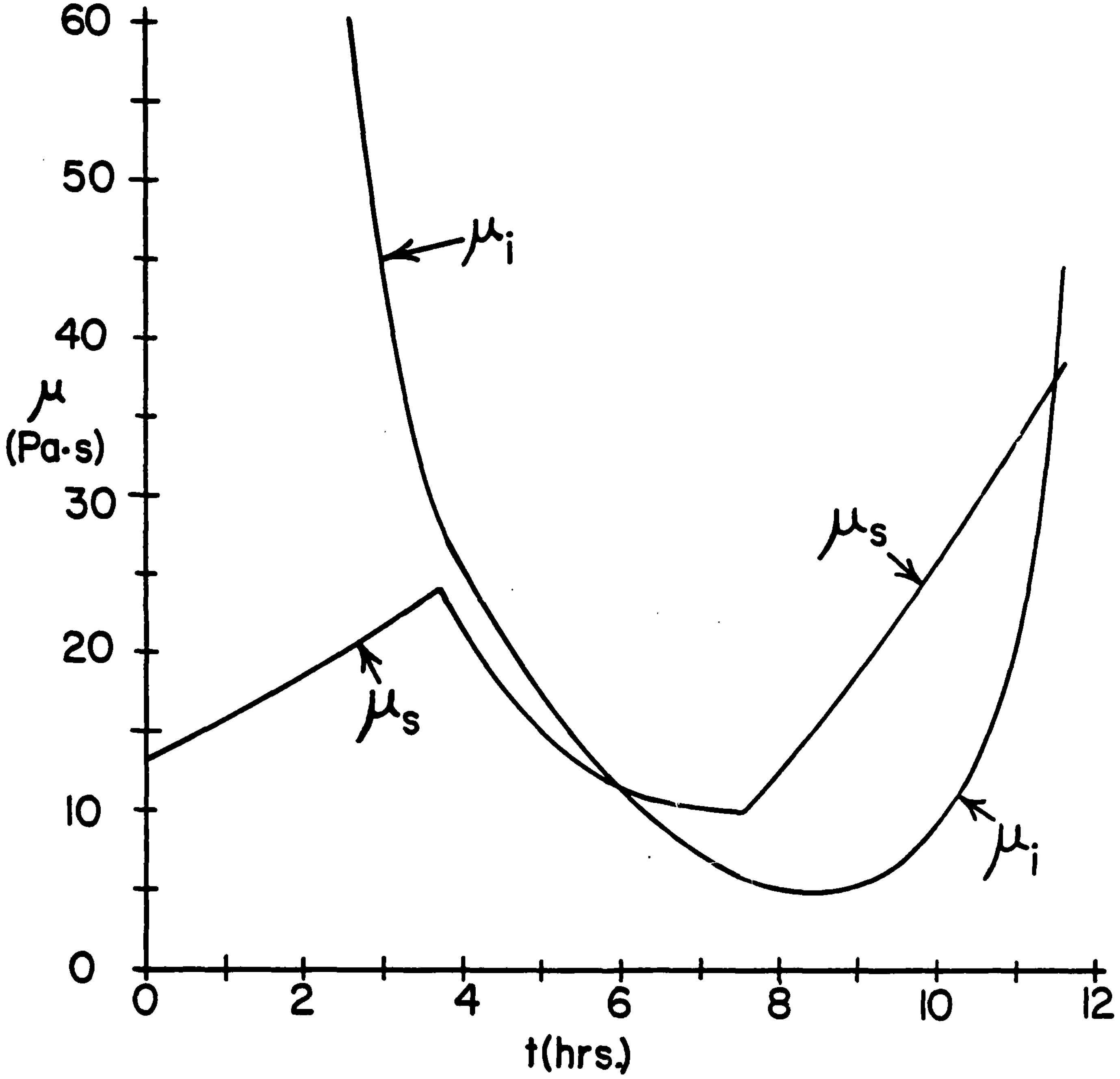


Figure 2

FIG. 2 - HJELMING + WALKER

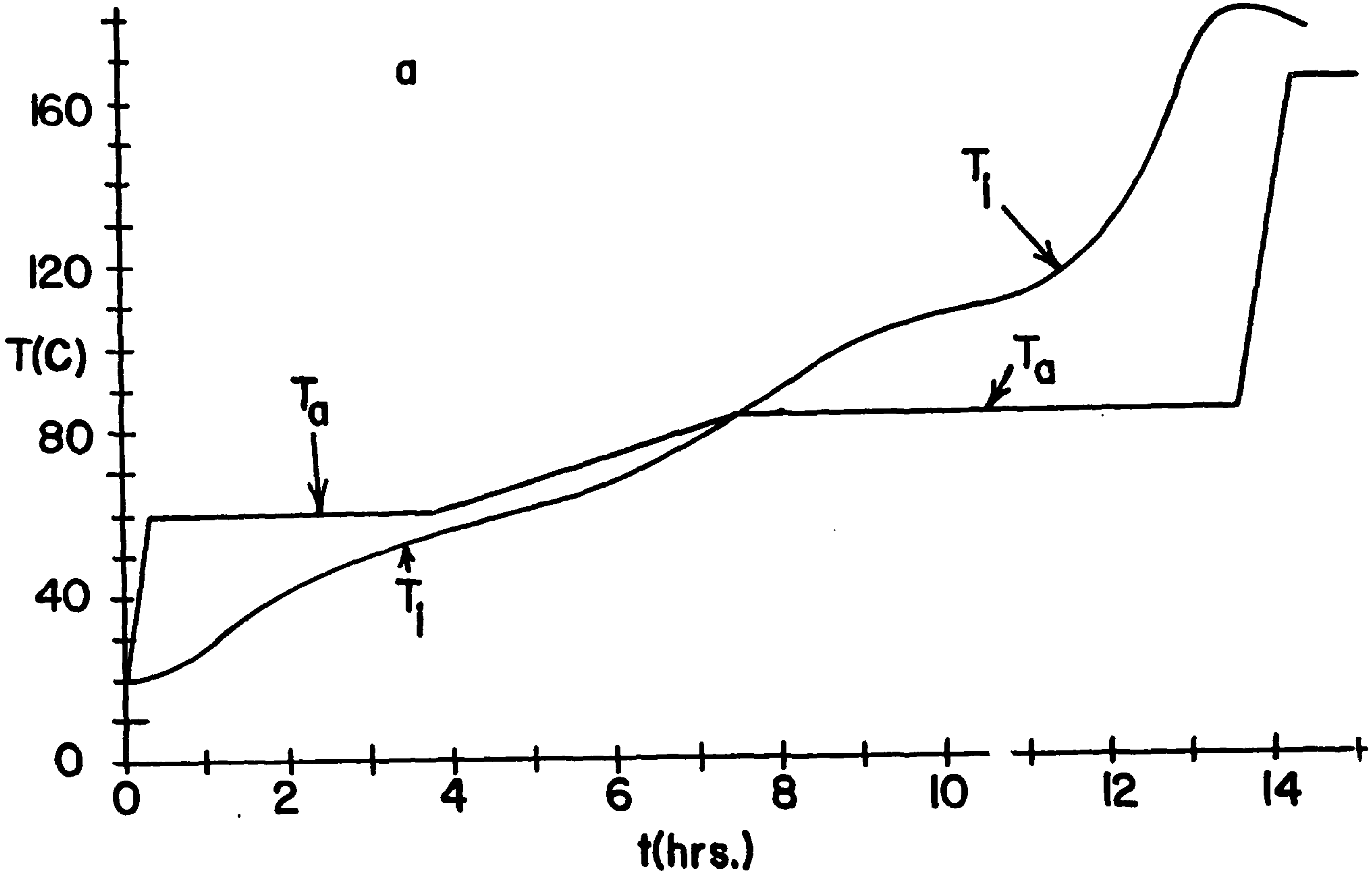
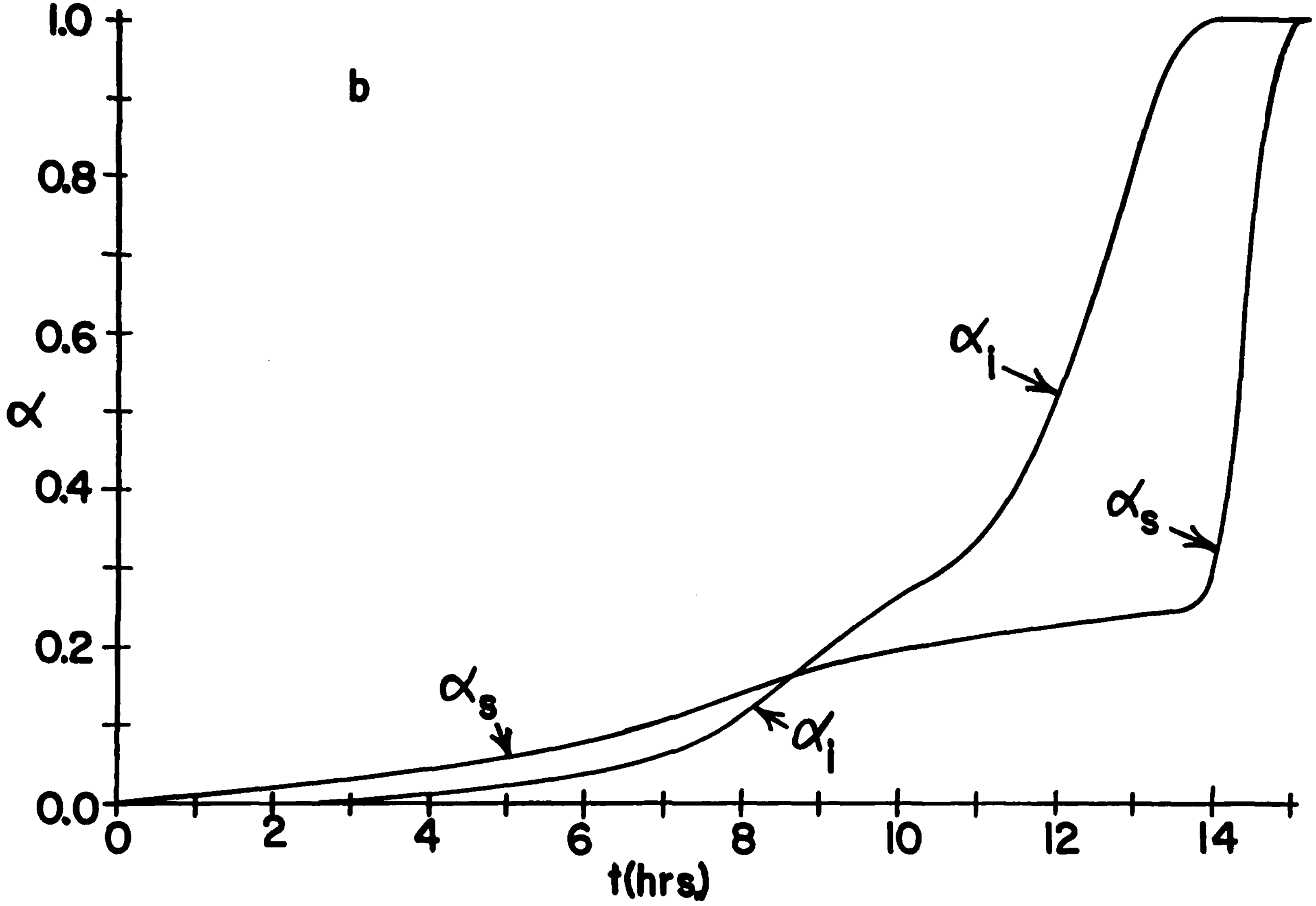


Figure 3a


b
α
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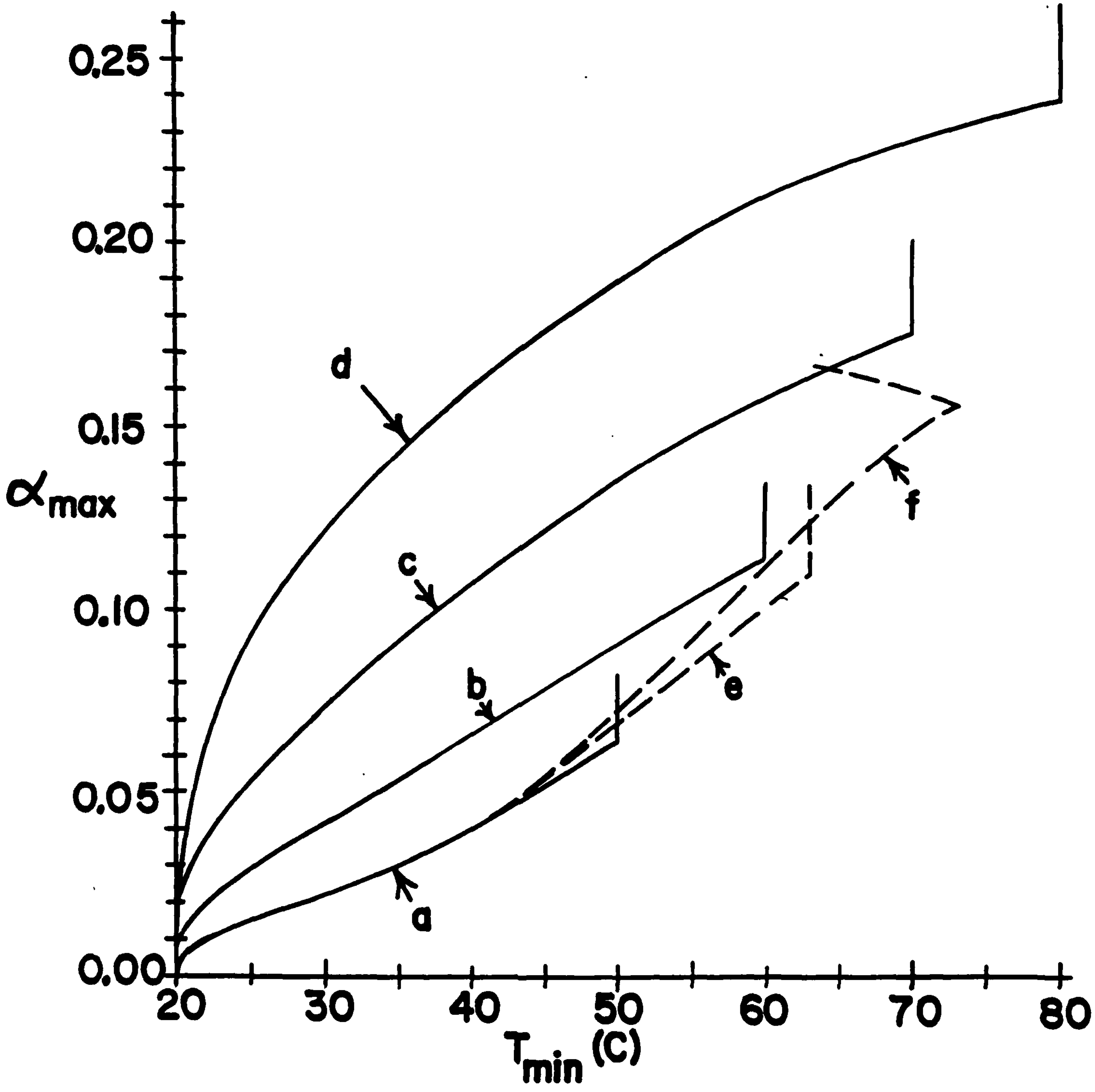
0.8
0.6
0.4
0.2
0.0
0
2
4
6
8
10
12
14
t(hrs)
α_l
α_s
α_s
α_l

Figure 3b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0.20
0.15
0.10
0.05
0.00
α_{max}
20
30
40
50
60
70
80
T_{min} (C)
a
b
c
d
e
f

Figure 4

[illegible]

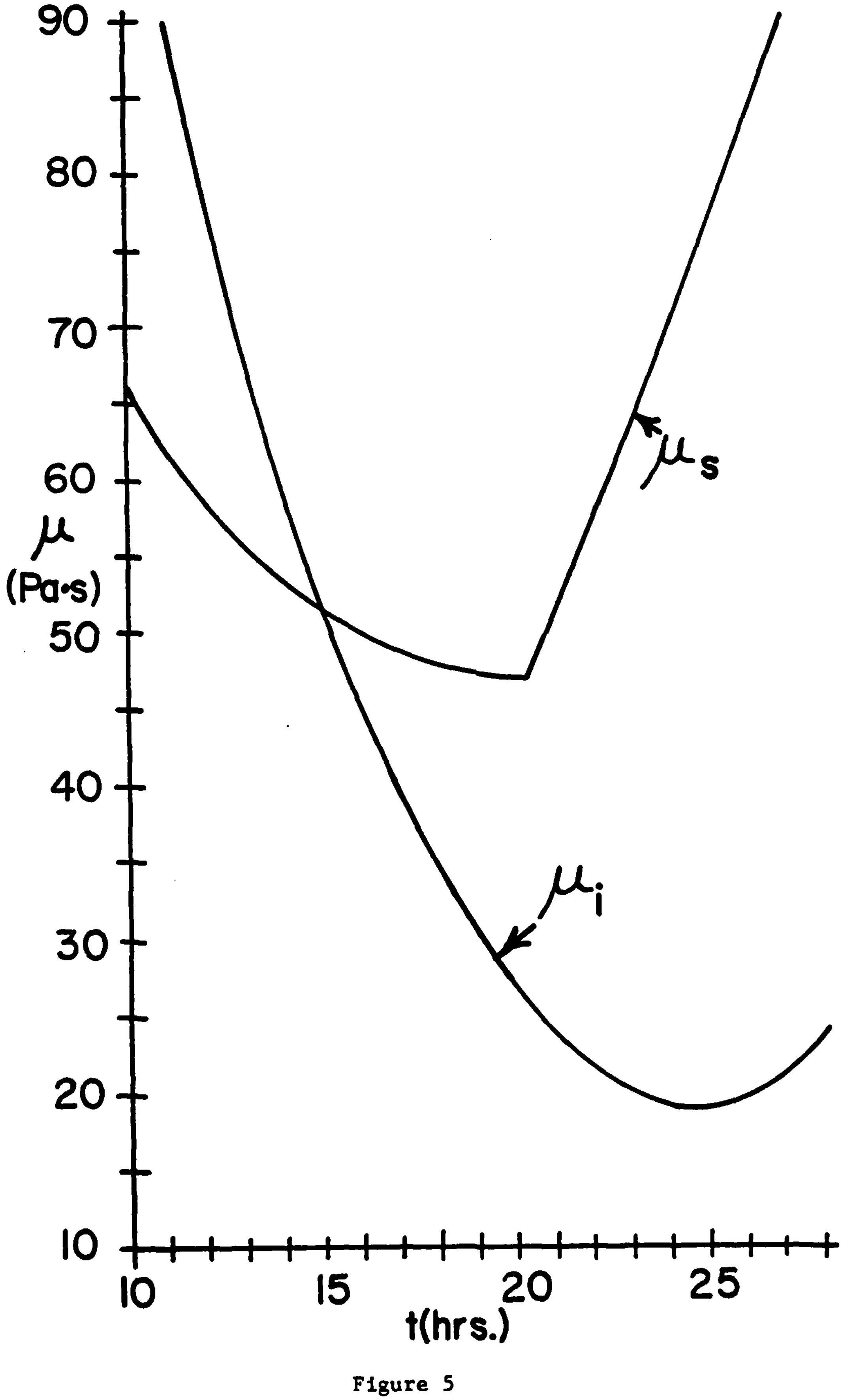

Figure 5

FIG 5 — UTELLMING & WALKER

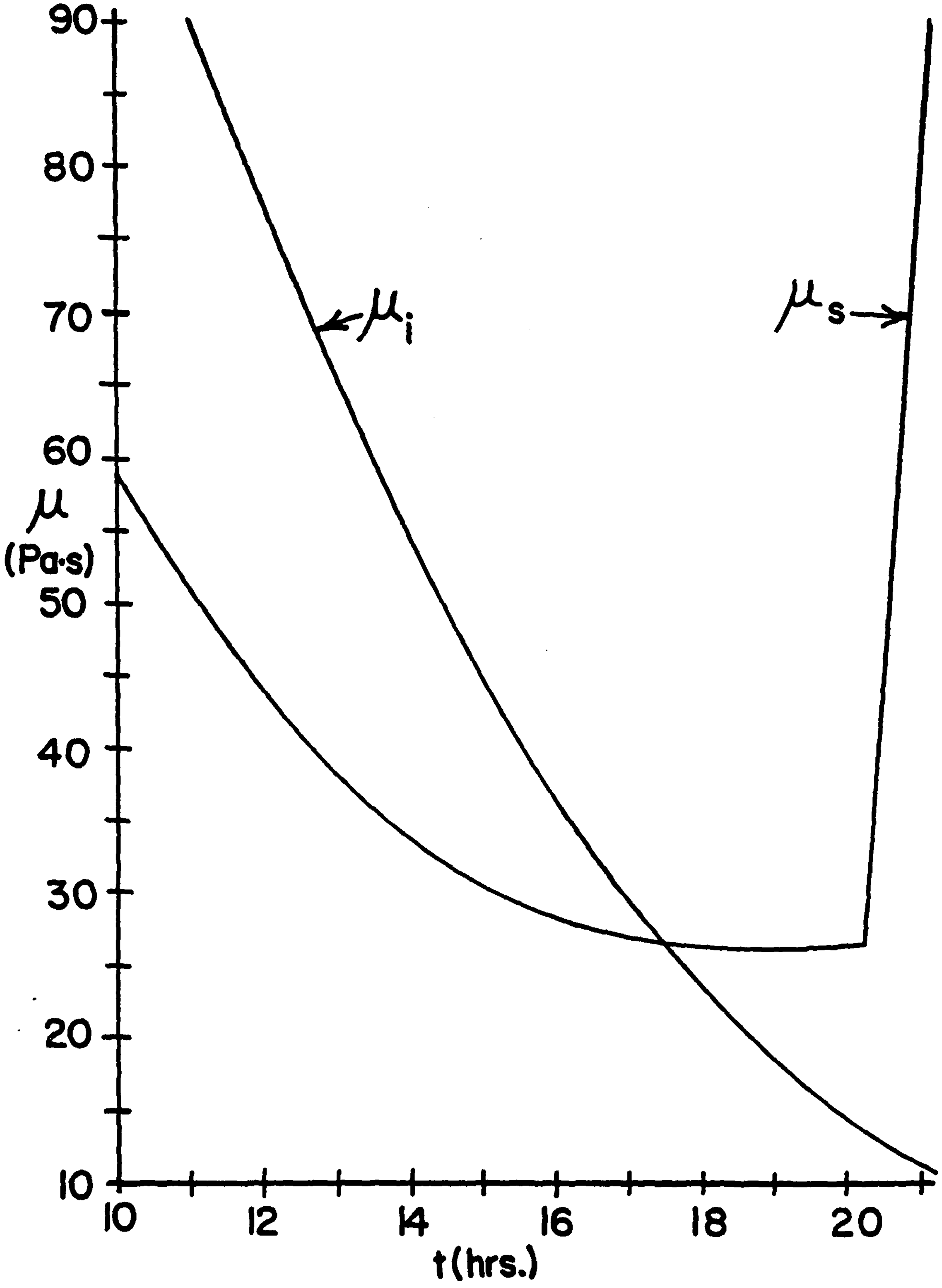

Figure 6

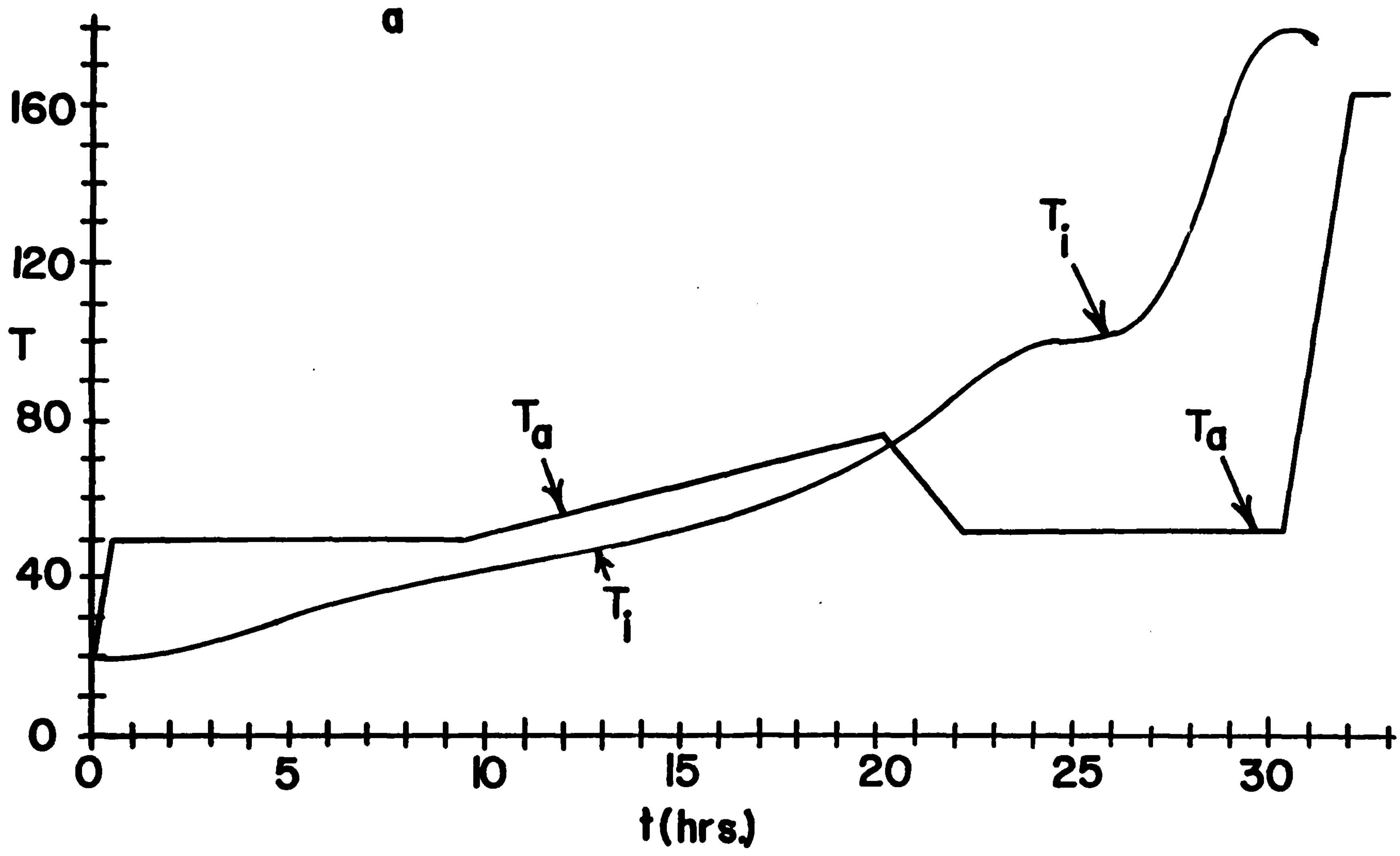

Figure 7a

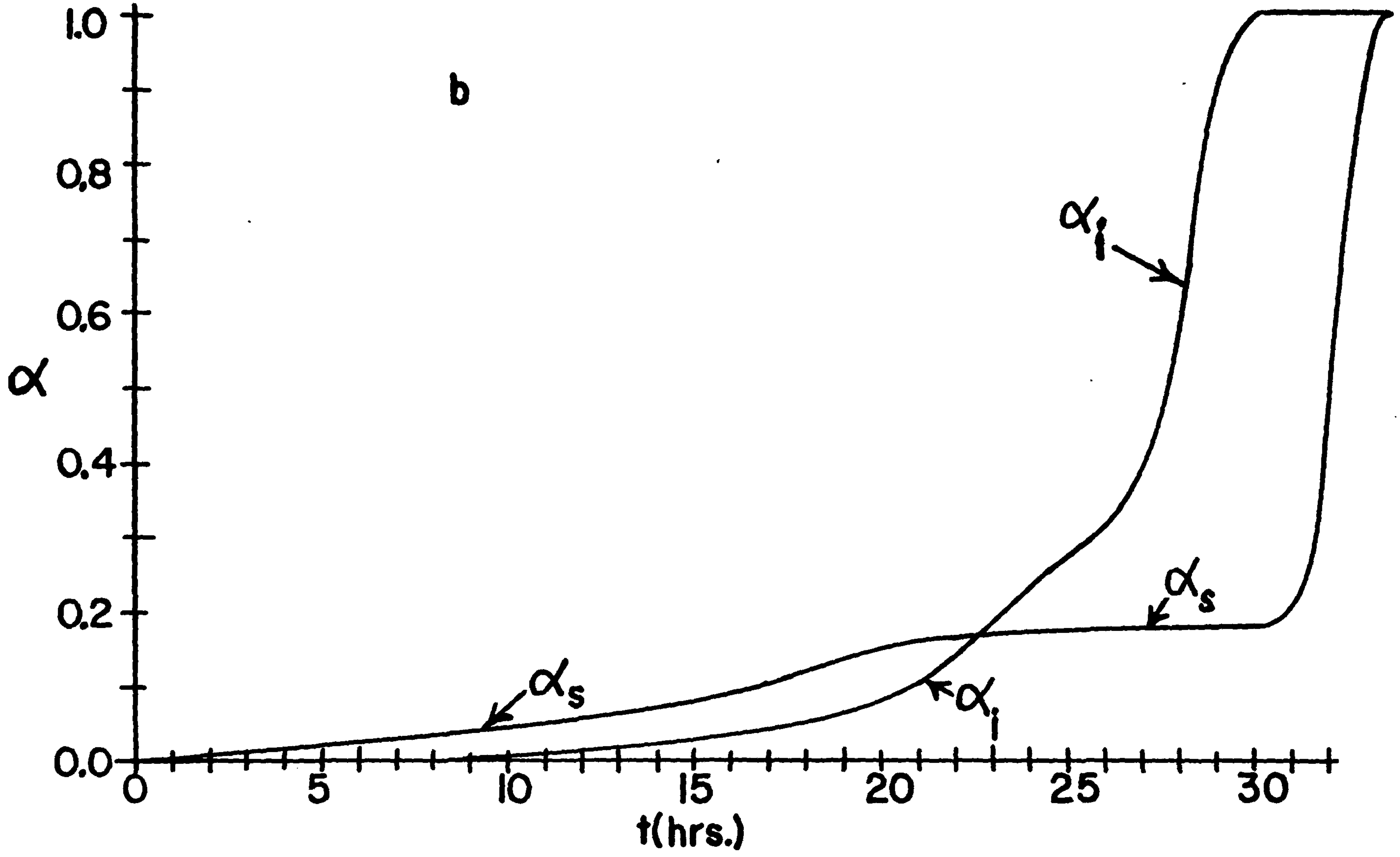
b
α
1.0
0.8
0.6
0.4
0.2
0.0
0
5
10
15
20
25
30
t(hrs.)
α_i
α_s
α_s
α_i

Figure 7b