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THE SAFETY OF HOT SELF-HEATING MATERIALS IN COOL SURROUNDINGS - A METHOD OF ANALYSIS

by

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SUMMARY

If a piece or pile of self-heating material is heated, for example during a manufacturing process and then exposed to cool surroundings, it may cool down to a stable safe condition or if the cooling is insufficient, heat to ignition. This paper describes the theory of a new, relatively simple, method of discussing such a problem and so finding criteria for specifying the safe initial conditions, i.e. the necessary degree of cooling. Practical applications will be discussed elsewhere.

In the special case of a very low surface heat loss one can neglect internal temperature differences and the theoretical treatment is then trivial. When the surface losses are very high the surface temperature is close to the ambient and one can readily adapt published computer solutions to a similar problem arising in explosion theory. The general case has hitherto not been dealt with by any method which is both as simple and, as judged by the two extreme cases, as accurate.

The criterion developed relates the critical explosion parameter δ to the excess temperature θ_o and the cooling characteristic of the equivalent inert material.

KEY WORDS: Self-heating, safety, analysis, heat transfer.

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INTRODUCTION

The determination of the long term steady condition to which a piece of self-heating material can be safely exposed makes use of the steady state theory of thermal explosion or ignition[1, 2, 3].

The condition is derived from equating the rate of heat generation to the rate of heat loss. In its simplest form the heat balance is

$$VAe^{aT_m} = HS(T_m - T_o)$$

where
- A depends on the reaction properties
- V is the volume of material at temperature T_m
- S is the surface area
- H is the heat transfer coefficient
- T_o is the temperature of the surroundings

and a is a constant in a term e^{aT} representing the dependence of the heat generation rate on temperature.

It follows that

$$\frac{aVAe^{aT_o}}{SH} = a(T_m - T_o)e^{-a(T_m - T_o)} \qquad \ldots\ldots (1)$$

$$\leqslant {}^1/_e$$

from which the relation in Fig. 1 follows. Equation (1) is a steady state relation, but if the initial temperature of a piece of material is T_i ($\neq T_m$) one can see that cooling will take place if the initial condition lies within the shaded area of Fig. 1 and self-heating if it does not. The upper branch BC may be regarded as giving the critical initial condition and the arrows show the direction of the temperature change. Steady solutions allowing for conduction are known for slab, cylindrical, and spherical geometry, but only for a unique initial temperature distribution within the material which is not uniform. However, for the simple model described above we have assumed that the temperature is uniform and this can only correspond to a very low ratio of external thermal conductance to internal thermal conductances, i.e. a small Biot No. (= Hr/K) where K is the conductivity of the material and r a characteristic dimension (e.g. V/S).

If, say, a slab at uniform temperature is placed in cool surroundings and Hr/K is not small, the edges will cool more quickly than the centre. The assumptions underlying Fig. 1 do not then hold. The assumption of an initial uniform temperature cannot correspond to the assumption of any steady state solution and we are forced to seek a solution to our problem from non-steady state theory in order to find the safe size and temperature.

APPROXIMATE THEORY OF CRITICAL INITIAL CONDITIONS

Derivation of equations

The heat balance of unit volume of material is written as

$$\rho c \frac{\partial T}{\partial t} = K\left\{\frac{\partial^2 T}{\partial x^2} + \frac{j}{x}\frac{\partial T}{\partial x}\right\} + Ae^{aT} \qquad \ldots\ldots (2)$$

where
- t is time
- ρ is density
- c is specific heat
- x is distance from centre
- j is zero for a slab, 1 for a cylinder and 2 for a sphere

At the surface $x = \pm r$ and we shall assume the Newtonian cooling condition

$$K\frac{\partial T}{\partial x} = -H(T - T_o) \qquad \ldots\ldots (3)$$

We shall discuss the case where at $t = 0$

$$T = T_1 \quad \text{for} \quad -r < x < r$$

This is appropriate when many small items are placed together in a pile.

The conditions are shown diagrammatically in Fig. 2. The central temperature will initially rise and will subsequently fall if cooling from the surface is strong enough.

Equation (1) can be rewritten as

$$\frac{\partial \theta}{\partial \tau} = \frac{\partial^2 \theta}{\partial y^2} + \frac{j}{y}\frac{\partial \theta}{\partial y} + \delta e^{\theta} \qquad \ldots\ldots (4)$$

where

$$\theta = a(T - T_1)$$

$$y = x/r$$

$$\delta = A r^2 e^{aT_1} a/K$$

$$\tau = Kt/\rho c r^2$$

At the surface

$$\frac{\partial \theta}{\partial y} + \alpha(\theta + \theta_0) = 0 \qquad \ldots\ldots (5)$$

where

$$\alpha = Hr/K$$

At $\quad t = 0, \quad \theta = 0 \qquad \ldots\ldots (6)$

and we define

$$\theta_0 = a(T_1 - T_0)$$

δ and θ_0 are defined with respect to T_1 though with a modification of the boundary condition both could be defined with respect to T_0. T_1 is a conventional choice since reactions which obey laws other than e^{aT} e.g. the Arrhenius Law, can be approximated by e^{aT} over a limited temperature range, and the matching is best made at high temperature. We seek the critical values of δ i.e. δ_c.

No general analytic solution is known for equation (2) but Merzhanov et al[4] and Bazykin et al[5] have obtained computer solutions for $\alpha \to \infty$. These solutions have been reviewed by Merzhanov[6]. The case $\alpha \to 0$ for which equation (1) applies has been widely studied.

Friedman[7,8] has earlier described a method which could be used for this problem but it is more difficult to apply in some cases and relies on previously computed quantities, which are not available for finite values of α.

We shall proceed to obtain an approximate solution from which one can derive the critical condition and to show that for $\alpha \to \infty$ the results agree to within 10 per cent with computed results and that they are correct for $\alpha \to 0$.

A 10 per cent error in δ_c corresponds to a 5 per cent error in estimating the safe radius or about 1°K, in the critical value of T_1 so the method is presumably acceptable for practical purposes for all values of α (and initially uniform temperatures).

Approximate solution

The approximation we employ is to treat the conduction losses from an elementary volume as equal to those from an inert solid heat initially in the same way. The method, which could be adapted to other initial conditions, is as follows.

We consider the solution when δ is zero, i.e. when the material is inert. This gives a solution θ_i, viz. $\theta_i(\alpha, y, \tau)$ which by virtue of the linearity of the equations and boundary conditions is proportional to θ_o and we shall write this as $-\psi\theta_o$ (since θ_i is always negative), ψ is zero at $\tau = 0$. ψ is some times available as an analytic function, sometimes as a series, sometimes as graphical or tabular presentations. We use this solution to estimate the conduction cooling in equation (4).

$$\frac{\partial^2 \theta}{\partial y^2} + \frac{j}{y}\frac{\partial \theta}{\partial y} \doteq -\theta_o\left(\frac{\partial^2 \psi}{\partial y^2} + \frac{j}{y}\frac{\partial \psi}{\partial y}\right)$$

For an initially uniform temperature with cooling at the edges of the material and self-heating at the centre, this approximation underestimates the conduction cooling of the centre itself, since the curvature of the $\theta(x)$ curve is greater than that of the inert solid.

Equation (4) now becomes

$$\frac{d\theta}{d\tau} = -\theta_0 \frac{\partial\psi}{d\tau} + \delta e^{\theta} \qquad \text{...... (7)}$$

We now turn to discussing equation (7). We shall assume that from symmetry we know a priori where the temperature is highest, i.e. where instability develops. Henceforth θ and ψ refer to values at this position.

The approximate transient equation

Equation (7) will be used to find the value of δ_c, (in terms of θ_0 and α) which separates safe θ_0 where the temperature may at first rise but subsequently falls from dangerous or super-critically high θ_0 where the temperature rises to high values. From the nature of our approximation to the conduction cooling we shall underestimate δ_c so from the point of view of safety the results will be conservative.

Following Boddington[8] who dealt with $\psi \propto \tau^{1/2}$ we solve equation (7) to obtain

$$e^{\theta} = \frac{e^{-\theta_0\psi}}{1 - \delta\int_0^{\tau} e^{-\theta_0\psi}\,d\tau} \qquad \text{...... (8)}$$

Figure 3 shows some typical variations of θ with τ for gradually increasing δ.

For all δ, θ must eventually tend to ∞ because $\psi \rightarrow 1$ and $\delta\int_0^{\tau} e^{-\theta_0\psi}\,d\tau$ is unbounded and must reach unity at some value of τ for any finite δ. This, however, is a consequence of our approximation and has no physical significance; it can be removed by regarding the rate of heat generation as $\delta e^{\theta} - \delta e^{-\theta_0}$ instead of δe^{θ} and replacing ψ by $\psi + \delta e^{-\theta_0}\tau/\theta_0$.

For $\delta < \delta_1$ there is a maximum temperature followed by a fall.

The maximum θ occurs when

$$\delta e^{\theta} = \theta_0 \frac{d\psi}{d\tau} = \theta_0 \psi'$$

which combined with equation (8) gives

$$\frac{1}{\delta} = \int_0^{\tau_m} e^{-\theta_0 \psi} d\tau + \frac{e^{-\theta_0 \psi_m}}{\theta_0 \psi'_m} \qquad \ldots\ldots (9)$$

which expresses δ as a function of the time τ_m when the peak occurs. In Appendix I it is shown that this has a stationary value when ψ'' is zero and this defines a limit to the time and hence defines δ_1, the largest for which $\theta(\tau)$ exhibits this type of behaviour.

There is a relatively narrow range of δ $(\delta_1 < \delta < \delta_2)$ where the constantly ascending temperature curve has inflexions and above this $(\delta > \delta_2)$ a progressively rising temperature without inflexion. Expressions for δ in terms of the time at which an inflexion occurs can be obtained on similar lines to the above determination of δ_1, and hence δ_2 can be found. The algebraic determination of δ_1 or δ_2 is straightforward and leads to a parametric determination of $\delta(\tau_m)$ and $\theta(\tau_m)$ but is somewhat cumbersome (see Appendix I).

Clearly the definition of criticality is somewhat arbitrary for this approximate model though clearly we should expect δ_1 to be a low estimate.

A comparison of δ_2 with the computed solutions for a sphere and a slab show it also is too small (by about 15 - 25 per cent).

We week some other characteristic in our theory which will compare with the computed values and we can find one in seeking a <u>sufficient</u> condition for super-critical states.

A sufficient condition for criticality

From equation (8) it follows that

$$\frac{e^{-\psi\theta_0}}{1-\delta\tau} > e^{\theta} > \frac{e^{-\psi\theta_0}}{1-\delta e^{-\theta_0\psi}\tau}$$

so we have upper and lower limits to θ .

Consider

$$Z = \frac{e^{-\psi\theta_0}}{1-\delta e^{-\theta_0\psi}\tau}$$

Now

$$\frac{dZ}{d\tau} = \left\{\frac{e^{-\psi\theta_0}}{(1-\delta e^{-\theta_0\psi}\tau)^2}\right\}\left\{\delta e^{-\theta_0\psi} - \theta_0\psi'\right\} \qquad \ldots\ldots (10)$$

θ is always rising if $\delta e^{-\theta_0\psi} > \theta_0\psi'$ and infinite temperatures are reached as $\tau \rightarrow \frac{1}{\delta}e^{\theta_0\psi}$

ψ is an increasing function of τ so our condition can be shown in Fig. 4. The conditions for a finite rising temperature are satisfied in the shaded area and the least value of δ which gives $\frac{dZ}{d\tau}$ positive as θ approaches infinity is at the intersection X so that $\delta > \delta_3$ is a sufficient condition for super-critical states and thus this choice for $\delta_c = \delta_3$ is given by

$$\tau_c\,\theta_0\,\psi'(\tau_c) = 1 \qquad \ldots\ldots (11)$$

$$\delta_3 = e^{\theta_0\psi(\tau_c)}\big/\tau_c \qquad \ldots\ldots (12)$$

This $\delta_c(\theta_0)$ locus satisfies $\frac{d\delta_c}{d\theta_0} = \delta_c \Psi$.

Equation (11) defines a time τ_c and equation (12) defines a δ_c in terms of this τ_c and we have upper and lower bounds for this δ_c from

$$\frac{1}{\tau_c\left(1-\theta_0\psi(\tau_c)\right)} > \delta_c > \frac{1}{\tau_c}$$

Our first approximation is to take the lower bound to obtain the critical condition from

$$\frac{\theta_0}{\delta_c}\psi'\left(\frac{1}{\delta_c}\right) = 1 \qquad \ldots\ldots (13)$$

a result that relates δ_c directly and simply with the way the equivalent inert hot body behaves. $\theta_0\psi'$ is the rate of fall of the central temperature of the equivalent inert hot spot. $\frac{1}{\delta_c}$ is the adiabatic induction time, so we can state an approximate criticality criterion for symmetrical hot spots in words.

The numerical value of δ_c is the reciprocal of the dimensionless adiabatic induction time and is equal to the rate of fall of the central temperature at this adiabatic induction time.

It is readily shown from conventional conduction theory that for $\alpha \to 0$

$$\psi'(\tau) \simeq e^{-1/4\tau}\Big/\sqrt{\pi}\,\tau^{3/2} \quad (\tau \ll 1) \text{ for a slab}$$

$$\psi'(\tau) \simeq e^{-1/4\tau}\Big/2\sqrt{\pi}\,\tau^{5/2} \quad (\tau \ll 1) \text{ for a sphere}$$

We find that δ_c calculated from equation (13) and these expressions for ψ' give better agreement with the computed results than does δ_1 or δ_2, and moreover is simpler, so for these pragmatic reasons we choose it as our criterion of criticality.

The approximation $\delta_c \tau_c = 1$ raises θ_o for a given δ_c and reduces our estimate of δ_c for a given θ_o, so equation (13) is not itself the sufficient condition for criticality – only an approximation to that given by equations (11) and (12).

δ_c calculated from equation (13) is within 10 per cent of the computed results for the slab and sphere ($\alpha \to \infty$). A more detailed discussion of the mathematical aspects of the above is given in Appendix I. The comparison shown in Fig. 5 is with results computed for a slightly different boundary condition, since only these are available. This different condition is the transient diffusion across a stolid/solid boundary instead of a quasi steady constant relation between θ and $d\theta/dy$ (equation (5)). It gives a different value of ψ but for $\tau \ll 1$ the asymptotic forms given above for ψ' are valid apart from a factor of 2 in the actual temperature difference as between the surface and the centre, so for $\tau_c \ll 1$ (i.e. $\delta \gg 1$) the two problems become identical and we can, as we have done, use the one to obtain results for the other.

The special case $\alpha \to 0$

When $\alpha \to 0$, ψ is readily shown to be

$$\psi = \left(1 - e^{-(HS/V\rho c)t}\right) = 1 - e^{-\frac{\alpha S r}{V}\tau}$$

For slab, cylinder and spheres

$$\frac{Sr}{V} = 1 + j$$

Thus

$$\theta_o = \frac{\delta_c}{\alpha(1+j)} e^{\alpha(1+j)/\delta_c}$$

and for large θ_o

$$\delta_c = \alpha(1+j)\theta_o$$

i.e.

$$A e^{aT_i} = \frac{HS}{V}(T_c - T_o)$$

which is in effect a repetition of equation (1).

Solution for various α

To find δ_c for α other than very small or very large we make use of conduction theory to find ψ . With equation (13) this gives the required values of $\delta_c(\alpha, \theta_o)$ which are shown in Fig. 6 for the sphere. The errors are likely to be greatest when θ is smallest and the region $\theta < 2$ requires special study which is beyond the scope of this report.

DISCUSSION

For any size of pile and any initial temperature we can calculate θ_o and δ . Provided this δ is less than δ_c for infinite α we can find the minimum cooling necessary for safety.

This approximate theory has been justified only by comparison with computed results. We note that $\chi\left(= \frac{1}{\delta_c} \theta_o \psi'(\frac{1}{\delta_c}\right)$ is strictly less than 1 and this can be regarded as a change in the scale of θ_o . But for initial temperature distributions which are uniform the presence of the term $e^{-1/4\tau}$ in the term for ψ' makes δ_c vary only weakly with θ_o so errors in the θ_o scale correspond to much smaller errors in δ_c .

This would not be so for an initial distribution of temperature which is peaked in the centre and errors in δ_c are then greater. In practice this may happen if storage piles a r e built so that the outsides are made up of items which have been exposed longer to the cool surroundings. This obviously makes for safety and, for a given initial peak central temperature, we should regard the results given here as being on the safe side.

The application of equation (13) is relatively straightforward and the above treatment can readily be applied to other geometries, e.g. cubical or rectilinear piles with little additional difficulty since solutions for $\psi(\tau)$ are available in the literature for many such shapes. It should be noted that whilst the analytic form for these is more complicated than for the simple geometries discussed here, we are usually concerned with short times and simplifications exist for these conditions. The Laplace transform method of finding ψ is especially suited for short times.

A table of $\psi(\tau)$ can readily be turned into a table of ψ' by a simple numerical procedure and hence $\frac{1}{\theta_o}(=\tau\psi'(\tau))$ readily evaluated with τ being replaced by $\frac{1}{\delta}$. A graph of ψ must be differentiated with of course the possibility of graphical errors.

It can be seen from equation (13) that it may sometimes be more appropriate to consider $1/\theta_o$ and $1/\delta_c$ as the relevant dimensionless parameters.

ψ is sometimes presented in tabular and graphical form. These may be more useful than analytic forms especially when τ_c is neither very small (high δ_c) nor very large (low δ_c). ψ is often obtained as a series for the immediate values of τ_c that arise if the Biot No. α is small (small piles or low cooling coefficients). This still gives $1/\theta_o$ explicity in terms of $1/\delta$ (as a series) so allowing a graph of $\delta(\theta_o)$ to be drawn but it may be more convenient to use tables or graphs of ψ .

CONCLUSION

A theory is given for determining the safety of hot self-heating materials. It can be applied to various symmetrical shapes of material and it allows safe temperatures - or minimum cooling requirements - to be specified in terms of the self-heating and geometrical properties of the material at risk.

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APPENDIX I

We have

$$e^{\theta} = \frac{e^{-\theta_0 \psi}}{1 - \delta \int_0^{\tau} e^{-\theta_0 \psi} d\tau}$$

The stationary values (Fig. 3) satisfy

$$\theta_0 \frac{d\psi}{d\tau} = \theta_0 \psi' = \delta e^{\theta}$$

i.e.

$$\frac{1}{\delta} = \int_0^{\tau_m} e^{-\theta_0 \psi} d\tau + \frac{e^{-\theta_0 \psi_m}}{\theta_0 \psi_m'} \qquad \text{...... (1A)}$$

where ψ_m and ψ_m' are functions of τ_m the time for the stationary value.

We seek the smallest value of $1/\delta$ (highest δ) for which a solution exists.

Now $$\frac{d}{d\tau_m}\left(\frac{1}{\delta}\right) = - \frac{e^{-\theta_0 \psi}}{\theta_0 (\psi_m')^2} \psi_m''$$

so the smallest value of $1/\delta$ satisfying equation (1A) is that for which ψ_m makes ψ_m'' zero*.

Since $\psi' \to 0$ at $\tau \to 0$ for our boundary conditions and $\psi = 1$ for $\tau \to \infty$ there must be a τ_m when $\psi_m'' = 0$.

This τ_m can be found for a known ψ and then equation (1A) gives a critical $\delta_c(\theta_0)$

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* For this solution it makes no difference if ψ is redefined as

$$\psi + \delta e^{-\theta_0 \tau} / \theta_0$$

We now consider the inflexions which, from equation (7) must satisfy

$$-\theta_0\psi'' + \delta\left(\delta e^{\theta} - \theta_0\psi'\right) = 0 \qquad \text{...... (2A)}$$

$$\delta e^{\theta} = \theta_0\psi' \pm \sqrt{\theta_0^2(\psi')^2 + 4\theta_0\psi''} \qquad \text{...... (3A)}$$

At low values of τ $(\tau < \tau_m)$ ψ'' is positive and so we consider the positive sign only.

$$Z + \theta_0\psi' = \frac{2\delta e^{-\theta_0\psi}}{1 - \delta\int_0^{\tau} e^{-\theta_0\psi}\,d\lambda} \qquad \text{...... (4A)}$$

where ψ , ψ' and ψ'' are functions of τ_i the time at the inflexion and

$$Z = \sqrt{\theta_0^2(\psi')^2 + 4\theta_0\psi''}$$

$$\frac{1}{\delta} = \frac{2e^{-\theta_0\psi_i}}{\theta_0\psi'_i + Z} + \int_0^{\tau_i} e^{-\theta_0\psi}\,d\lambda \qquad \text{...... (5A)}$$

The smallest $1/\delta$ giving a root of equation (5A) is obtained after differentiation of equation (5A)

$$\frac{d}{d\tau_i}\left(\frac{1}{\delta}\right) = e^{-\theta_0\psi} - \frac{2\theta_0\psi' e^{-\theta_0\psi}}{\theta_0\psi' + Z} - $$

$$- \frac{2e^{-\theta_0\psi}}{(\theta_0\psi' + Z)^2}\left\{\theta_0\psi'' + \frac{2\theta_0^2\psi'\psi'' + 4\theta_0\psi'''}{2Z}\right\}$$

Thus $\dfrac{1}{\delta}$ has a minimum when

$$(\theta_0\psi' + Z)(Z - \theta_0\psi') =$$

$$= 2\left(\theta_0\psi'' + \frac{\theta_0^2\psi'\psi'' + 2\theta_0\psi'''}{Z}\right)$$

i.e. $Z\psi'' = \theta_0\psi'\psi'' + 2\psi'''$

both sides being positive.

Hence

$$\theta_0 = \frac{(\psi''')^2/\psi''}{(\psi'')^2 - \psi'\psi'''} \qquad \ldots\ldots \text{(6A)}$$

and so $Z = \theta_0 \left| \dfrac{1}{\psi'''}\left(2(\psi'')^2 - \psi'\psi'''\right)\right|$

From equation (6A) one can find, for a given τ_i, $\theta_0(\tau_i)$. Hence, from equation 5(A) δ_i can also be found as a function of τ_i so by choosing a series of values of τ_i, δ_i and θ_0 can be found parametrically.

Redefining ψ as $\psi + \delta e^{-\theta_0\tau}/\theta_0$ complicates the treatment by leading to

$$\theta_0 = \frac{(\psi''')^2 + (\psi'')^2\delta e^{-\theta_0}}{\psi''\left((\psi'')^2 - \psi'\psi'''\right)}$$

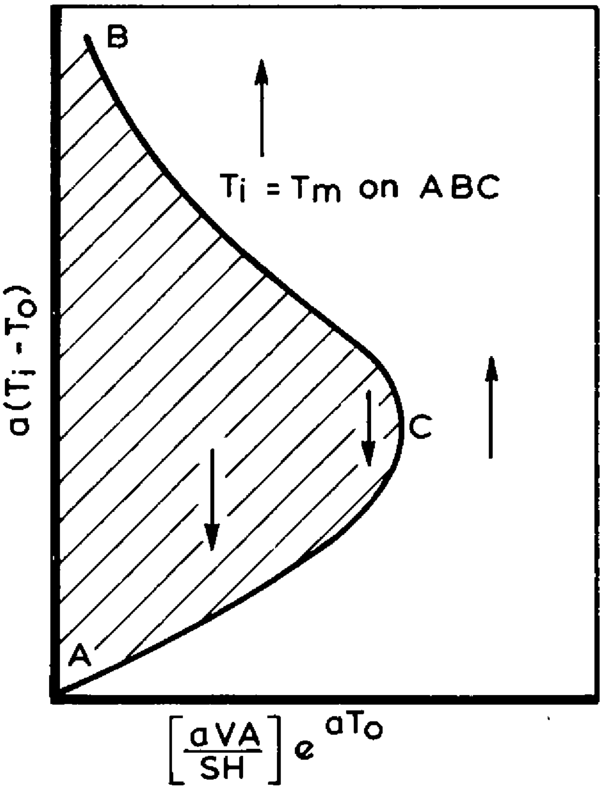
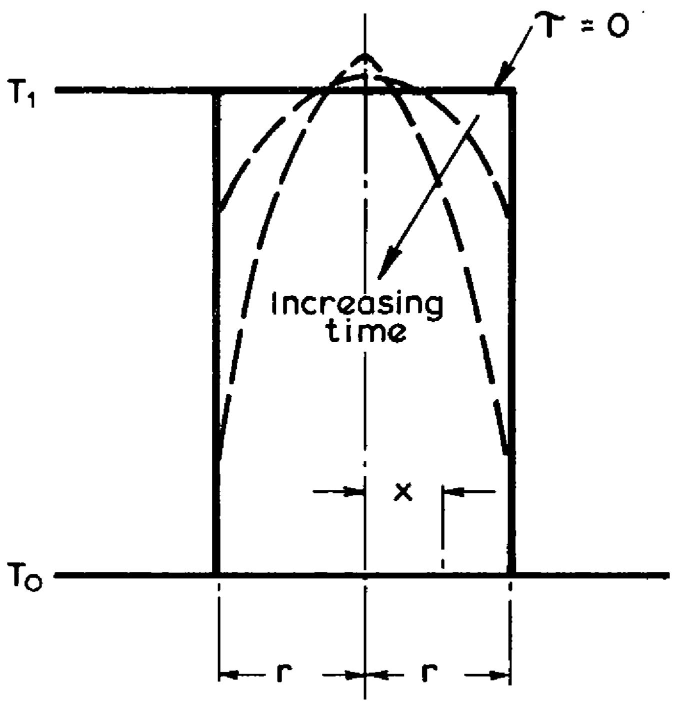


FIG. 1. CRITICAL SELF-HEATING CONDITIONS FOR $\alpha\,[=Hr/K] \rightarrow 0$



$$H(T - T_O) = -K\left[\frac{dT}{dx}\right]_r \text{ at surface}$$

FIG. 2. TEMPERATURE DISTRIBUTION IN RADIALLY SYMMETRICAL "HOT SPOT"

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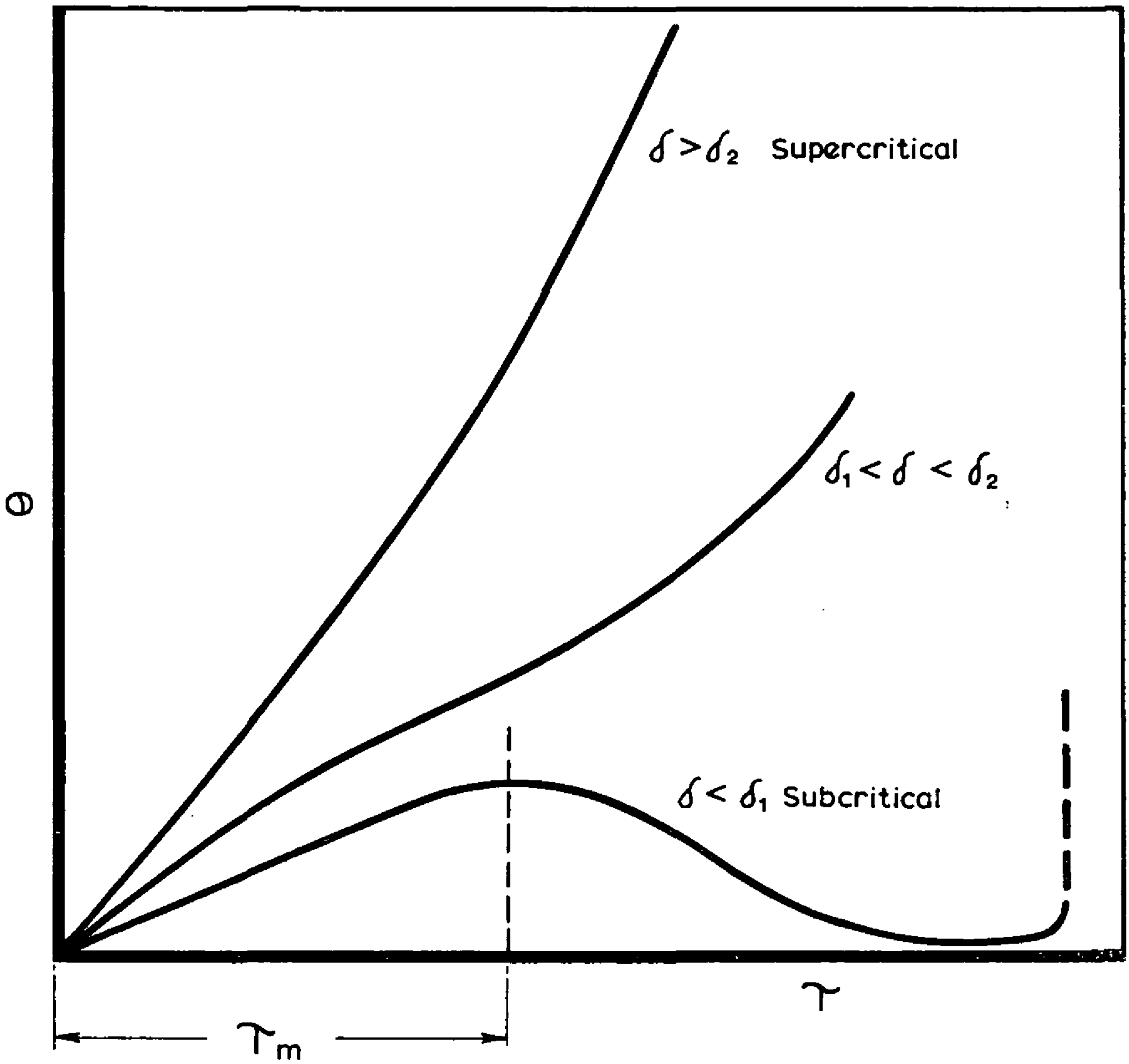


FIG. 3. TYPICAL TEMPERATURE-TIME BEHAVIOUR

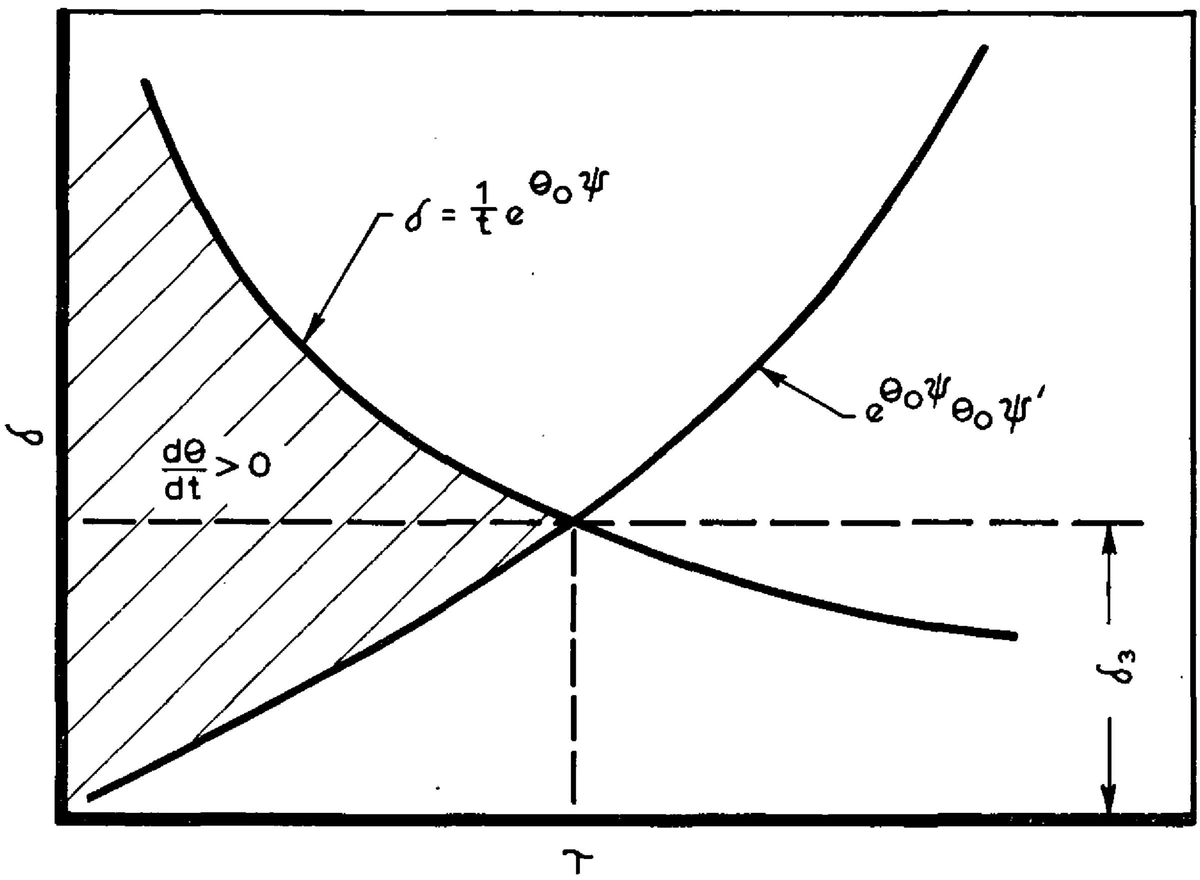


FIG. 4. THE SUFFICIENT CONDITION FOR IGNITION

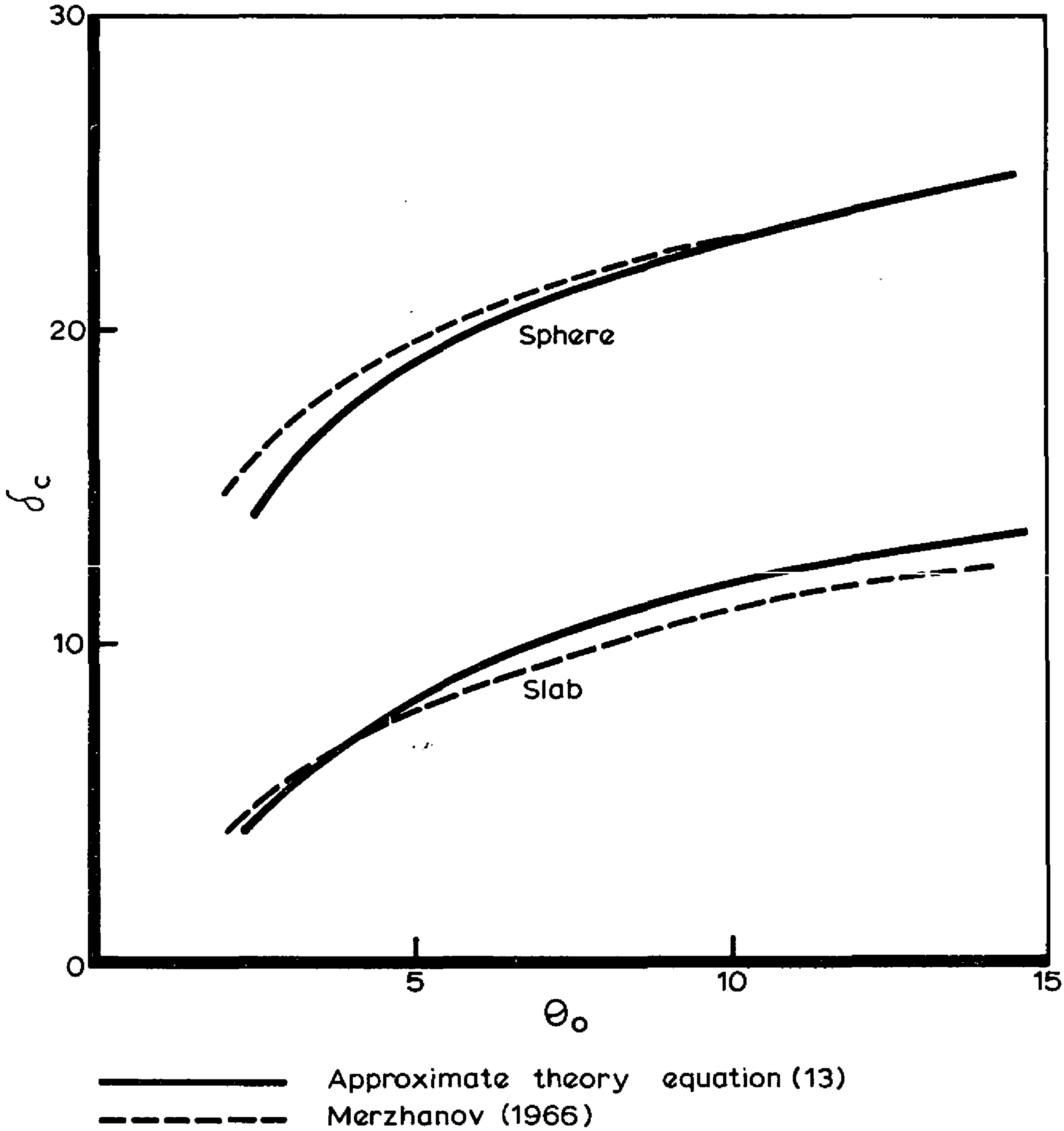
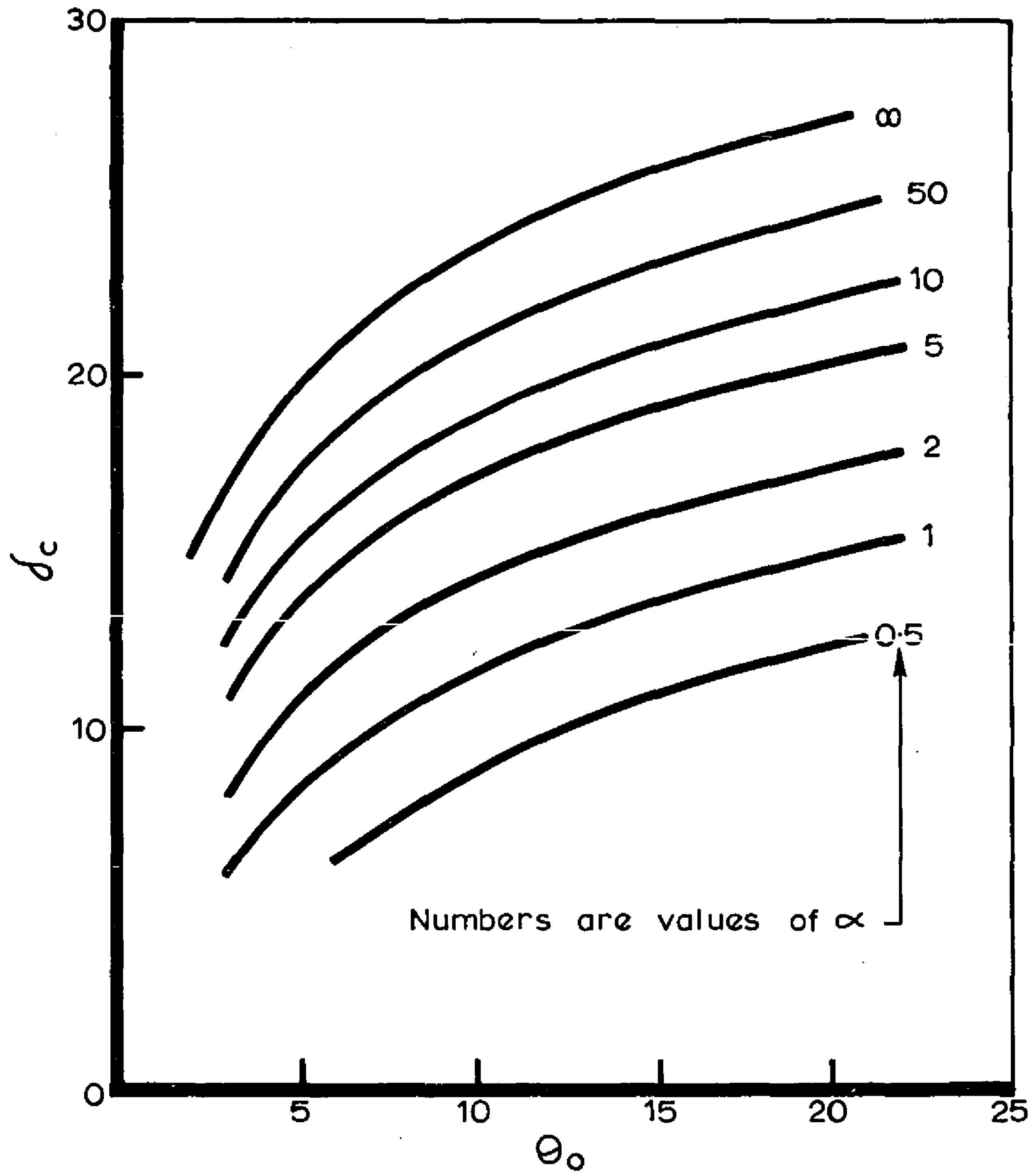


FIG. 5. CRITICAL δ_c – COMPARISON OF RESULTS



$\delta_c \longrightarrow 3hr\,\Theta_o$ for $\alpha \longrightarrow 0$

FIG. 6. CRITICAL δ_c FOR SPHERES — NEWTONIAN COOLING