COMMENTS ON EXPERIMENTS ON THE SELF-HEATING OF FIBRE INSULATING BOARD

by

P. H. Thomas

Summary

This note discusses the results given by Mitchell[1] on the
ignition and self-heating of fibre insulating board in the light of
simple theory. Some values are derived for the apparent activation
energies of fibre insulating board and the rates of reaction
at various temperatures. These appear to be somewhat larger than
expected.

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1. Introduction

Mitchell[1] reports two kinds of experiment; the first, the measurement of the self-heating in terms of the temperature rise, when various volumes of fibre insulating board are held in different ambient temperatures and the second, the minimum ambient temperature at which this self-heating proceeds to ignition. The times after which this ignition occurs are also given. Three types of board were used.

The differential equation for a solid in which heat is generated is

$$\nabla^2 T = \frac{1}{k}\frac{dT}{dt} - \frac{q}{K}\rho \quad \dots\dots\dots\dots\dots\dots\dots(1)$$

where ∇^2 is the Laplacian operator
T is the temperature (absolute)
k is the thermal diffusivity
'q' is the rate of generation of heat per unit mass
ρ is the density
and K is the thermal conductivity.

At the surface the boundary condition is

$$H(T - T_A) = -K\frac{dT}{dx} \quad \dots\dots\dots\dots\dots\dots\dots(1i)$$

where H is the heat transfer coefficient at the surface, and the suffix 'A' denotes ambient conditions.

For simplicity we assume that the reaction rate is independent of time (see Appendix I) and that the Arrhenius Law is obeyed so that

$$q = Q.f.e^{-E/RT} \quad \dots\dots\dots\dots\dots\dots\dots(2)$$

where Q is the heat of reaction/unit mass
f is a constant
E is the apparent activation energy cal/gm/mole
R is the universal gas constant.

It is implicit in this assumption that the reaction is temperature not air rate controlled.

Also for simplicity only one dimensional heat flow (linear or radial) is considered, i.e.

$$\nabla^2 = \frac{d^2}{dx^2} + \frac{n}{x}\frac{d}{dx}$$

where n = 0 for a slab
= 1 for radial flow in a cylinder
= 2 for radial flow in a sphere.

It will simplify the analysis if we employ three dimensionless parameters

$$\delta = \frac{Q\rho f E r^2 e^{-E/RT_A}}{K R T_A^2} \quad \dots\dots\dots\dots\dots\dots\dots(3i)$$

$$\theta = \frac{E(T-T_A)}{RT_A^2} \quad \dots\dots\dots\dots\dots\dots(3ii)$$

$$\chi = Hr/K$$

where 'r' is the radius of the sphere or cylinder or the half width of the slab.

It is interesting to note that $\frac{\delta}{\theta}$ can be written

$$\frac{\delta}{\theta} = \frac{Q \rho f \, e^{-E/RT_A} \, r^3}{K \frac{(T - T_A)}{r} r^2}$$

which is of the form

heat generated chemically
heat lost by conduction

Provided $\frac{T - T_A}{T_A} \ll 1$, equation (1) may be written

$$\nabla^2 \theta = \frac{r^2}{h} \frac{d\theta}{dt} - \delta e^{\theta} \quad \ldots\ldots\ldots (4)$$

A simple self-heating equation which considers parabolic temperature distribution in the material for the steady state can be written

$$\theta \propto \delta e^{\theta}$$
$$i.e. \quad \delta \propto \theta e^{-\theta}$$

This equation shows the essential feature of self heating namely that there is a maximum value of δ for which a steady state is possible [2] [3]. This critical value δ_c is a function of α [4], though in these experiments δ_c for all but the smallest samples is in effect constant.

From the definition of δ it follows that $\log \frac{\delta_c T_A^2}{r^2}$ should vary linearly with $1/T_A$, the slope determining the value of E. The values δ_c appropriate to spheres of diameter equal to the sizes of the piles of material used experimentally are listed in Table (1) of Appendix II. From these it becomes possible to plot Mitchells ignition data, as shown in Fig. (1). The data do in fact appear as straight lines and give values of 21,000 - 30,000 cals/gm mole for E. This is greater than the estimate of about 20,000 or less made by Bowes [5].

2. The use of self-heating data

Provided δ does not exceed δ_c a steady state can theoretically exist. In practice, after a long time the reaction rate is not independent of time, so that it is not strictly permissible to consider steady state conditions. This is however neglected in these considerations (see Appendix I). For the steady state it is possible to calculate the relation between θ_0 - (the suffix o denoting conditions at the centre $x = 0$) and the value of δ. This computation has been performed for certain values of α (Appendix III) and the results are shown in Fig.2. The only measurements made in the experiments were the actual temperature rise $T_0 - T_A$, the value T_A and the size of specimen 'r'. In order to use equations (3) and the relation between $\delta \propto \theta_0$ in Fig.2 to determine E, a somewhat cumbersome procedure would have to be adopted, if the whole range of values of $T_0 - T_A$ were to be included. If, however, only small values of self-heating are considered a simple procedure is practical. It may be shown that if on the right hand side the steady state version of equation (4) θ is approximated to θ_0 - a constant - the solution for a sphere is

$$\theta_0 = \frac{\delta}{6} \left(\frac{2 + \alpha}{\alpha} \right) e^{\theta_0} \quad \ldots\ldots\ldots (5)$$

It may be seen from Fig.2 that for $\theta_0 < 0.4$ this is not an unreasonable approximation.

If $\theta_0 < 0.4$ then $T_0 - T_A < 0.4 \dfrac{T_A^2 R}{E}$ which for T_A equal to 550°K, $R = 2$ cals/gm mole/°C and E equal to 20,000 cals/gm mole gives $T_0 - T_A < 12$°C

If $T_0 - T_A \ll T_A$, δe^{θ_0} is approximately equal to $\dfrac{Q \rho f E \tau^2 e^{-\frac{E}{RT_0}}}{K R T_A^2}$

It then follows from equation (5) and the definitions of θ and δ that

$$\frac{\alpha}{2+\alpha}\left(\frac{T_0 - T_A}{\tau^2}\right) = \frac{Q \rho f}{6 K} e^{-\frac{E}{RT_0}} \qquad \ldots\ldots\ldots(6)$$

i.e. a plot of $\log \dfrac{\alpha}{2+\alpha} \dfrac{(T_0 - T_A)}{r^2}$ should vary linearly with $\dfrac{1}{T_0}$.

Since equation (6) over-estimates θ it follows that the plot of the experimental results according to equation (6) would be expected to be convex upwards.

The results which are shown in Fig. (3) do in fact suggest such a curvature. The value of E obtained from the slope at small values of $T_0 - T_A$ is 22,000 cals/gm mole which is in reasonable agreement with the other estimate of 24,000 for the wood fibre insulating board.

There is a maximum value of θ_0 which for a sphere is 1.69[6] whereas, using the values for E given above a value of θ_0 of 2.9 was obtained without ignition. A possible explanation of this discrepancy is that the theoretical values of θ_0 are less reliable than those of δ. For example, it may be shown (see Appendix IV) that if e^θ is replaced by $1 + \theta$ in the equation for a slab, a value of 5.1 is obtained for the critical δ. This is 1.6 times the correct value. On the other hand there is no upper limit to the value of θ_0. A second possible reason for obtaining values of θ_0 larger than theoretically possible is that the rate of heat generation may decrease with time as reacting material becomes exhausted. Thus self-heating which would cause ignition were the heating to be constant, would be insufficient if the rate of heat output fell with time. The extent of this effect could not be found without a more detailed analysis than is attempted here.

3. Rate of generation of heat

It follows from equations (2) and (31) that

$$q(T_A) = \frac{K \delta R T_A^2}{\rho E r^2} \qquad \ldots\ldots\ldots(7)$$

which means that the scale of $\delta T_A^2/r^2$ in Fig.(1) can be related directly to q which is plotted in cals/gm/min. The value assumed for K was 10^{-4} c.g.s. units. In the data obtained from the rise in temperature on heating we have from equation (6)

$$\frac{\alpha}{2+\alpha} \frac{T_0 - T_A}{\tau^2} = \frac{\rho}{6K} q(T_0)$$

Here again the scale $\dfrac{K}{2+\alpha} \dfrac{T_0 - T_A}{r^2}$ can be directly related to q and this is done in Fig.3.

These values of q are much larger than those obtained from ignition data (Fig.1.) which are themselves about 10 times those estimated by Bowes[5] and Raskin & Robertson[7]. A rate of heating of 10 cal./gm/min. at 250°C is equivalent to self heating at $\frac{1}{2}$ °C s⁻¹ which also suggests that the figure is too large. Although an error in measuring T of t per cent produces an error in q of $\dfrac{E}{RT} t$ per cent i.e. about 25 times t this cannot account for

the discrepancy which, moreover is not a result of the physical
assumptions made (Bowes, for example, found that a law of the
Arrhenius kind was obeyed) nor of this particular mathematical
treatment.

Consider the following simple model of a sphere large enough
for the surface temperature to be considered equal to the ambient
oven temperature (i.e. $\alpha \to \infty$). If T_0 is the central
temperature then the heat loss from the sphere is _less_ than
$q(T_0) \frac{4\pi}{3} r^3 \rho$. On the other hand the heat loss is _greater_
than $4\pi r^2 \frac{K(T_0 - T_a)}{r}$.
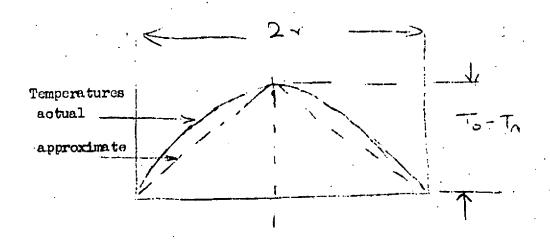

Fig.4. Real and approximate temperature
distribution sphere.

Hence $q(T_0) > \frac{3K}{r^2}(T_0 - T_a)$ (The full theoretical model gives the
result $q = \frac{6K}{\rho r^2}(T_0 - T_a)/r$ see equation (16) (Appendix III) for small
values of $(T_0 - T_A)$. Clearly even an approximate treatment shows
that the estimated rate of heat generation is greater than 5 times
that measured by Bowes[5] and Raskin & Robertson[6].

4. Time for ignition to occur

Equation (4) must be solved by direct computation if the term
$\frac{dT}{dt}$ is retained. Some computations have in fact been made by Copple et al[8]
but these do not cover a sufficiently large number of values of
δ near the critical for the time to ignite under conditions just
above the critical to be determined.

Despite this, it can be seen from equation (4) that for any
given value of δ , including that just in excess of the critical
value, the ignition-or mathematically, the point at which $\frac{d\theta}{dt}$ becomes
infinite - corresponds to a certain value of $\frac{kt_1}{r^2}$ t_1 being the time
to ignite. Theoretically the value of $\frac{E}{R}(T_A - T_1)$ where T_1 is the
initial temperature of the solid will have some effect, but this is
probably not large: in simple conduction heating the time for the
centre to reach, say 95 per cent of the improved surface temperature
is also given completely by a certain value of $\frac{kt}{r^2}$ i.e. 0.4.

The larger the value of δ the smaller, relative to the other
terms in equation (4) is the conduction term.

Thus for $\delta \gg 1$

$$\frac{r^2}{K}\frac{d\theta}{dt} = \delta e^{\theta} \quad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots(8)$$

i.e. $\frac{kt_1}{r^2} = \frac{1}{\delta}$

From equation (8) it follows that

$$t_i = \frac{c}{qf} \frac{RT_A^2}{E} e^{\frac{E}{RT_A}} = \frac{cRT_A^2}{E q(T_A)} \quad \ldots\ldots\ldots (9)$$

where c is the thermal capacity.

This in the more general form

$$t_i = A\, c^{\frac{E}{RT_A}}$$

is known as the induction equation and is often the means of determining E.

For critical conditions

$$t_i \propto r^2$$

and Mitchell's data for the ignition times of different sized specimens are shown in Fig.(5) to follow this relationship very closely.(For the wood board $t_i/r^2 = 0.75$).

However, the three materials do not give comparable values for t_i - and as the three materials appear to have similar values for q it would seem that any difference between materials in the values of t_i/r^2 would be due to differences in 'k' - the thermal diffusivity. This is unlikely to vary by as much as a factor of 60 which is that apparent in the results between wood and cotton felted fibre insulating board and it is thus clear that the preceding analysis is inadequate. Some other factor, such as the diffusion of air must be relevant. So long as it is a diffusion factor there will be a square law between the time to ignite and the linear size.

5. Discussion and conclusions

An attempt has been made to analyse data on the self-heating and self-ignition of fibre insulating boards according to a simple thermal theory. The values of activation energy are somethwat larger than evaluated by Bowes but the materials although similar were not identical and they may well have slightly different activation energies. Though there does appear to be a real difference between the data from the experiments of Mitchell on the one hand and of Bowes and Raskin & Robertson on the other, Mitchell's data gives much higher rates of self-heating. The rates of heating calculated from the ignition and self-heating data of Mitchell are not in agreement. This suggests that the simple theoretical model is inadequate.

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Time dependent reactions

The heat output calculated from Mitchell's self-heating experiments is larger than that calculated from his ignition experiments (compare Figs. 1 and 3) and both sets give values larger than obtained by Bowes and from the results of Raskin and Robertson.

We consider here to what extent this can be explained by allowing for the dependence of the rate of heat output on time.

If the loss of reactant with time is negligible for the conditions of Bowes' experiments then equation (2) may - as assumed by Bowes - be used to calculate E. This was determined as 16,000 cal/gm mole. At 250°C q was measured as $1/60$ cal g^{-1} s^{-1} and these, together with an assumed value for Q of order 100 cal/gm (9) gives f as 800 s^{-1}, a value which, like E, is much smaller than would be expected for a cellulosic product (10). Typical values are in fact, for E, 25-35,000 cal/gm mole and for f $10^8 - 10^{12}$ s^{-1}.

The effect of loss of reactant is to reduce the variation of heat output with temperature, which appears to reduce E. While, qualitatively this is a feature of Bowes' results, quantitatively the explanation is inadequate as shown below.

We write "w" the concentration of primary reactant and we have for a first order reaction

$$dw = -w f e^{-E/RT} dt \qquad \text{....... (10i)}$$

and

$$q = -Q \frac{dw}{dt} \qquad \text{....... (10ii)}$$

For isothermal heating we have from equation (10)

$$q = Qf e^{-\frac{E}{RT}} e^{-f e^{-E/RT} t} \qquad \text{.......(11)}$$

and we can write

$$q = q_0 e^{-\frac{t}{f} q_{13}} \qquad \text{......(12)}$$

where

$$q_0 = Qf e^{-E/RT} \qquad \text{.......(13)}$$

If the heating is not isothermal but, say, a progressive rise in temperature, the exponent of t in equation (12) is less. If the heating is according to

$$\frac{1}{T} = \frac{1}{T_0} - \beta t \qquad \text{........(14)}$$

this reduction factor can be readily evaluated. Thus from (10i) and (14)

$$\int_1^w \frac{dw}{w} = -f \int_0^t e^{-\frac{E}{R}(\frac{1}{T_0} - \beta t)} dt$$

$$\log_e \frac{1}{w} = f \left[\frac{e^{-\frac{E}{RT}} - e^{-E/RT_0}}{E\beta/R} \right] \qquad \text{......... (15)}$$

For any appreciable rise in temperature the second exponential term is negligible c.f.c $^{-E/RT}$ and from (15) (10ii) and (13) we have

$$q = q_0 e^{-\frac{f e^{-E/RT}}{E\beta/R}}$$

$$= q_0 e^{-\frac{q_0}{Q E\beta/R}}$$

Now whatever the value of $\frac{QEB}{R}$ the value of $\frac{q}{q_0}$ cannot be less than $\frac{1}{e}$ (i.e. $\frac{1}{2.7}$,) if q is increasing with rise in temperature. It therefore follows that the difference between Bowes' values of q and those derived from Mitchell's which is of order 5 - 10 cannot be explained by the loss of primary reactant.

If there were loss of reactant in Mitchell's experiments, the actual estimation of the rates of heat output would be affected in a way not readily calculated. They would correspond to values for heating in a time less than the actual time of the measurement, but from the preceding arguments it is not possible to explain the difference, which is of order 10, between the results of the two kinds of data by loss of primary reactant because the observed difference is greater than a factor of 2.7.

Bowes has suggested tjat loss of secondary volatile reactant may be involved and this is being considered.

The calculation of α and δ

We calculate the heat transfer coefficient H_c by convection for a sphere from the formula

$$N_{N_A} = 2 + 0.55 \left(N_{N_Y} . N_{P_A} \right)^{\frac{1}{4}}$$

where N_{N_A} = Nusselt No. = $\dfrac{H_c D}{K_A}$

N_{G_r} = Grashof No. $= \dfrac{g D^3 \Delta T}{T_A \nu^2}$

N_{P_r} = Prandtl No. $= \nu / \kappa_A$

where D is the diameter of the sphere

K is the thermal conductivity of air

g is the gravitational constant

ν is the kinematic viscosity of air

κ is the thermal diffusivity of air

ΔT is the difference in temperature between the surface and ambient temperature

T_A is the ambient, i.e. oven temperature (absolute).

The radiation coefficient H_R for a black body and a small difference in temperature is

$$H_R = 4 T_A^3 \sigma$$

where σ is the Stefan-Boltzman constant.

We have $\alpha = \dfrac{(H_R + H_c) D}{K}$

Now $\Delta T = \dfrac{R \theta_s}{E} T_A^2$

where θ_s is the dimensionless surface temperature which is known as a function of α for critical conditions. It is always less than 1 and may be shown that the term in α involving θ_s is much smaller ($<$10 per cent) than the radiation. It is sufficient therefore to take $\theta^{1/4}$ as unity and an approximate value of E of 25,000 cal/gm/mole.

A Table of the values of T_A measured by Mitchell for the various values of 'r' appears below with the computed values of α and the corresponding values of δ_c. It appears that the variation of δ with 'r' is not of great significance for these experiments.

TABLE I

'r' ins.	T_A °F abs.	α	δ
1/16	1060	2.5	1.7
1/8	946	3.0	1.9
1/4	874	4.4	2.2
1/2	856	7.2	2.6
1	816	11.6	2.8
2	762	17.3	2.9
4	726	30	3.1
6	712	41	3.2
11	688	67	3.2

Relation between θ_0 & δ for non-critical condition

The steady state equation for a sphere is from equation (4)

$$\frac{1}{x^2} \frac{d}{dx} \left(x^2 \frac{d\theta}{dx} \right) = - \delta e^{\theta} \quad \ldots\ldots\ldots (16)$$

By the substitution in equation (16) of
$$\zeta = x (\delta e^{\theta_0})^{1/2}$$

and
$$\psi = \theta_0 - \theta$$

we obtain

$$\frac{1}{\zeta^2} \frac{d}{d\zeta} \left(\zeta^2 \frac{d\psi}{d\zeta} \right) = e^{-\psi} \quad \ldots\ldots\ldots (17)$$

At the centre, $x = 0$, $\theta = \theta_0$ and $\frac{d\psi}{dx} = 0$

$\therefore$ ζ at $= 0$, $\psi = 0$, $\frac{d\psi}{d\zeta} = 0$ $\quad\ldots\ldots\ldots (18)$

And at the boundary $x = 1$

$$\alpha \theta = - \frac{d\theta}{dx}$$

$\therefore$ at $\zeta = (\delta e^{\theta_0})^{1/2} = \zeta_s$

$$\alpha (\theta_0 - \psi_s) = \zeta_s \left(\frac{d\psi}{d\zeta} \right)_s$$

Tables of ψ as a function ζ for equations (17 and 18) are given by Chandrasekar and Wares [11]. The values of ψ_s and ζ_s satisfying equation (14) for given δ and θ_0 can then be obtained.

Hence δ can be obtained as a function of θ_0 and α. The results are shown in Fig. (2).

Although there is a large variation of 'δ' with α for a given θ_0, a curve of δ/δ_c against θ/θ_0 is practically independent of α. (Fig.6).

Small values of θ_0

For small values of θ_0 we put θ equal to θ_0 on the right hand side of equation (16).

$$\therefore \frac{d}{dx} \left(x^2 \frac{d\theta}{dx} \right) = - \delta e^{\theta_0} x^2$$

Hence
$$\frac{d\theta}{dx} = - \delta e^{\theta_0} \frac{x}{3}$$

i.e.
$$\theta = A - \frac{\delta x^2 e^{\theta_0}}{6}$$

so that from (14)
$$\alpha \left(A - \frac{\delta}{6} e^{\theta_0} \right) = \frac{\delta}{3} e^{\theta_0}$$

and at the centre $x = 0$

$$\theta_0 = A = \frac{\delta(2 + \alpha) e^{\theta_0}}{6\alpha} \quad \ldots\ldots\ldots (19)$$

continuation

This relationship is also shown in Fig.(2) and it is seen that it is a very good approximation up to $\theta_o \approx 0.4$.

Since $\dfrac{E}{R}\left[\dfrac{1}{T_A} - \dfrac{T_o - T_A}{T_A^2}\right]$ is approximately equal to $\dfrac{E}{RT_o}$ so long as $T_o - T_A \ll T_A$.

$$\theta_o \doteqdot \frac{Q\rho f E \gamma^2}{K R T_A^2} e^{-E/RT_o}$$

Equation (19) may then be written

i.e.
$$\frac{\alpha}{2+\alpha}\left(\frac{T_o - T_A}{\gamma^2}\right) = \frac{Q\rho f}{6K} e^{-\frac{E}{RT_o}}$$

We consider the effect on the critical parameters δ_c and θ_o of changing the form of the heat generation function. This effect is greatest when the variation in temperature is greatest, i.e. when $\alpha \to \infty$

Let e^θ be replaced in the basic equation (4) by $1 + \theta$. Then equation (4) for the steady state becomes

$$\frac{d^2\theta}{dx^2} + \frac{n}{x}\frac{d\theta}{dx} = -\delta(1+\theta) \quad \ldots\ldots\ldots (20)$$

The boundary conditions are as before

$$x = 0 \quad \frac{d\theta}{dx} = 0 \quad \theta = \theta_o \quad \ldots\ldots\ldots (21i)$$

$$x = 1 \quad \theta = 0 \quad \ldots\ldots\ldots (21ii)$$

The solution to equation (10) and (21i) is

$$1 + \theta = (1 + \theta_o)\left[\frac{m_- e^{m_+ x} - m_+ e^{m_+ x}}{m_- - m_+}\right] \quad \ldots\ldots\ldots (22)$$

where $m_{+,-} = \dfrac{-n \pm i\sqrt{4\delta - n^2}}{2}$

We have from (21ii) and (22)

$$\frac{1}{1+\theta_o} = \frac{\left(-n + i\sqrt{4\delta - n^2}\,e^{-\frac{n}{2} - \frac{1}{2}\sqrt{4\delta - n^2}}\right)}{\left(+ (n + i\sqrt{4\delta - n^2})\,e^{-\frac{n}{2} + \frac{1}{2}\sqrt{4\delta - n^2}}\right)}{2i\sqrt{4\delta - n^2}}$$

i.e. $\dfrac{e^{n/2}}{1 + \theta_o} = \cos\sqrt{\delta - \frac{n^2}{4}} + \dfrac{n}{\sqrt{4\delta - n^2}}\sin\sqrt{\delta - \frac{n^2}{4}}$

There is no limiting value of θ_o, δ increasing to a maximum value as θ_o tends to infinity. The maximum value of δ is given by

$$\tan\sqrt{\delta - \frac{n^2}{4}} = \frac{-2\sqrt{\delta - \frac{n^2}{4}}}{n}$$

i.e. for the slab when $n = 0$ $\quad \delta = \dfrac{\pi^2}{4} = 2.47$
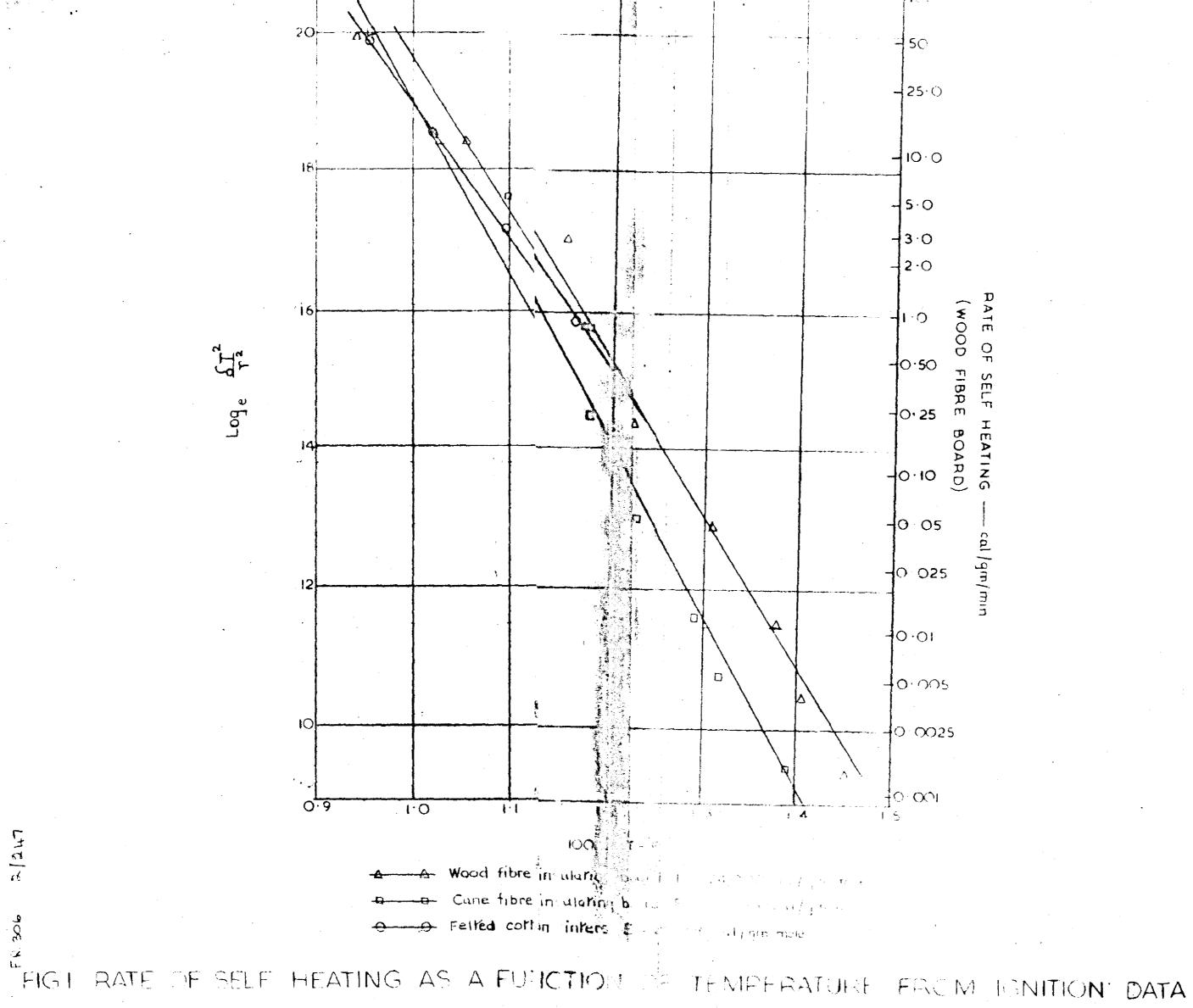
cylinder when $n = 1$ $\quad \delta = 3.65$

sphere when $n = 2$ $\quad \delta = 5.08$

The critical value of 'δ' for the sphere is only altered by 60 per cent but the critical value of θ_o is eliminated.

DOTTED LINES REPRESENT $\delta = 6\theta.e^{-\theta}\dfrac{\alpha}{2+\alpha}$

FIG. 2. THE RELATION BETWEEN δ AND θ_o FOR VARIOUS COOLING CONDITIONS FOR A SPHERE

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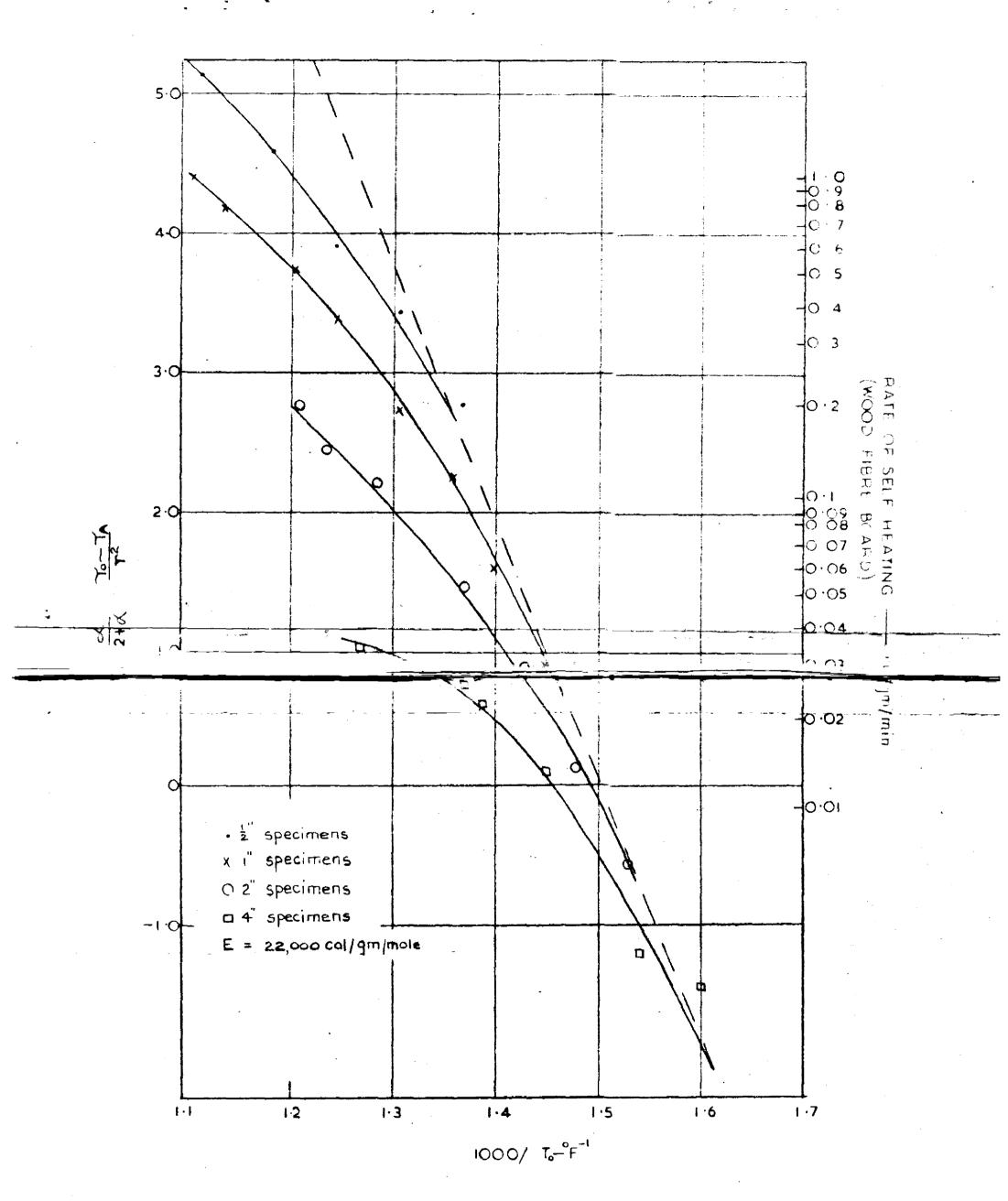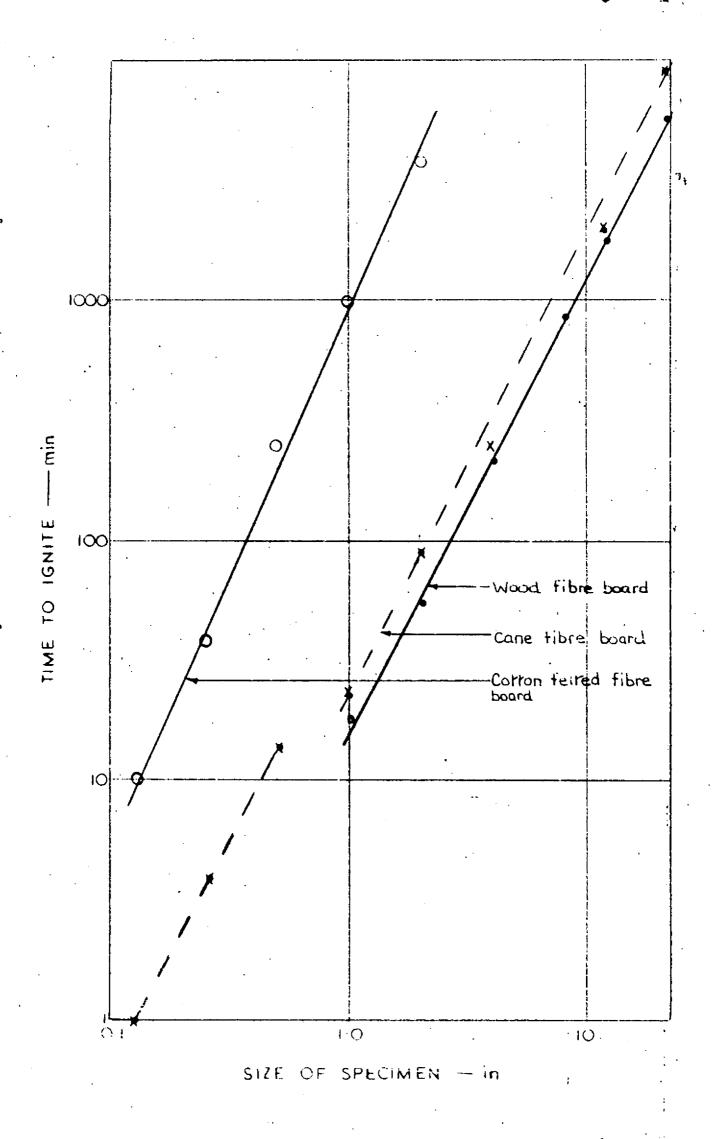FIG 1 RATE OF SELF HEATING AS A FUNCTION OF TEMPERATURE FROM IGNITION DATA

FIG. 3. RATE OF SELF HEATING AS A FUNCTION OF TEMPERATURE FROM SELF HEATING DATA
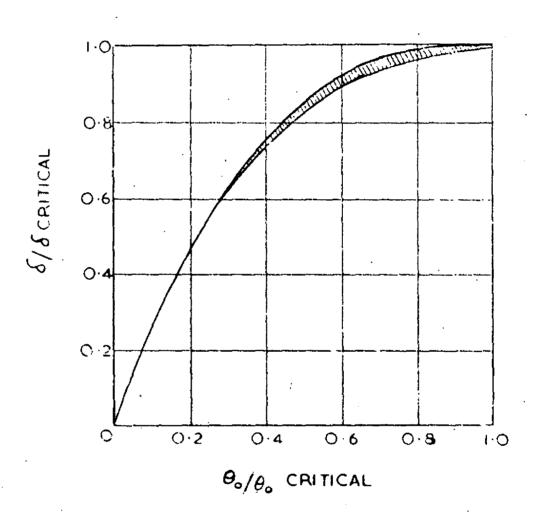
FIG.5 TIME TO IGNITE AT CRITICAL CONDITIONS

FIG. 6. THE RELATION BETWEEN δ/δ critical AND θ_0/θ_0 critical FOR ALL $\propto$