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ON THE DYNAMICS OF THE IGNITION OF PAPER AND CATASTROPHE THEORY

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Abstract

The purpose of this note is to study the phenomenon of ignition of paper considered by Shivadev and Emmons [1] from the viewpoint of stability theory and catastrophe theory [2]. It is shown that ignition results from a sudden or catastrophic change of the kinetics governing temperature from a locally stable to a locally unstable equation. Using the model of Shivadev and Emmons [1] and the above criterion, equations for the ignition temperature and the corresponding heat flux are derived. These equations are shown to provide a good match to the experimental data of Reference [1]. Further extensions of this work to combustion and the appearance of cusp catastrophes are also discussed.

1. Introduction

Nonlinear physical, sociological and engineering systems may exhibit large sudden changes in their behavior with relatively small changes in their parameters. In the theory of nonlinear differential equations, this phenomenon has been studied under Structural Stability and Bifurcation Theory [2]. More recently, Thom [2] has developed a general theory of

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elementary catastrophes (in the sense of discontinuities or bifurcations) for such systems.

In this note, we consider the phenomenon of ignition of paper studied by Shivadev and Emmons [1] and show that ignition has the qualitative properties of an elementary catastrophe. Based on the local stability properties of the nonlinear differential equations of the reaction energetics, we develop a criterion for ignition and show its usefulness in explaining the experimental data. Some proposals for the extension of these concepts to the combustion phase and for the design of new experiments to validate further theoretical results are also discussed.

2. Stability Properties of Chemical Kinetics Equations

Shivadev and Emmons [1] have given the following equations for the second phase reactions occurring during the thermal degradation and spontaneous ignition of paper sheets in air by irradiation.

$$\frac{dm}{dt} = -a_2 m \exp(-e_2/RT) \quad , \qquad m(o) = m^o \qquad (1)$$

$$mc^o \frac{dT}{dt} = q - h_o(T - T_a)^{4/3} - KT^4 + r_2 a_2 m \exp(-e_2/RT) \quad ,$$

$$T(o) = T_a \quad , \qquad (2)$$

where $h = h_o(T - T_a)^{1/3}$, $K = 2\sigma\varepsilon f$ and all the other quantities are as defined in Reference [1]*. (For easy reference, we have included a nomenclature section with numerical values used at the end of the paper.) The experiments give the critical temperature and heat flux values at ignition to be $\hat{T} = 680 \pm 15^{\circ}K$ and $\hat{q} = 0.58 \pm 0.03$ cal/cm^{2}sec respectively.

It is easily seen by numerical calculations that up to the critical temperature, the reaction rates are fairly small, but are increasing rapidly around the critical temperature. Thus the change in mass m up to ignition is quite small ($\frac{m}{m^o} \simeq .8$), and for stability analysis of equation (2), one may regard m to be a constant.

Now, let us consider equation (2) for temperature. When a particular heat flux q is applied, the corresponding equilibrium temperature is obtained by setting $\frac{dT}{dt} = 0$ in equation (2). Notice that the equilibrium in temperature is achieved very rapidly compared to the changes in mass m since below critical temperatures, the time constant of equation (2) (approximately $\frac{mc^o}{h}$) is much smaller than the time constant of equation (1). (The ratio is typically of the order 10^3.)

Let the equilibrium temperature be T_e. Then setting

$\left.\frac{dT}{dt}\right|_{T_e} = 0$ in equation (2), we obtain

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*In equation (2), we have neglected the term $[f\alpha(T_a)\sigma T_a^4]$ since it is of the order of .00563, but have retained the radiation term KT^4 since at $T = 680^{\circ}K$ (experimental ignition temperature), this term is .49 compared to $h(T - T_a)$, which is .258 cal/cm^2 sec.

$$q - h_o(T_e - T_a)^{4/3} - KT_e^4 + r_2a_2 m\exp(-e_2/RT_e) = 0 \quad . (3)$$

Equation (3) can be solved for T_e as a function of q using known values of all the other parameters*. (m may be assumed to be m^o or equation (3) may be solved for different values of the ratio $m = m/m^o$.) Figure 1 shows a plot of T_e versus q which is found to contain a sharp bend or fold at $q = q_c$.

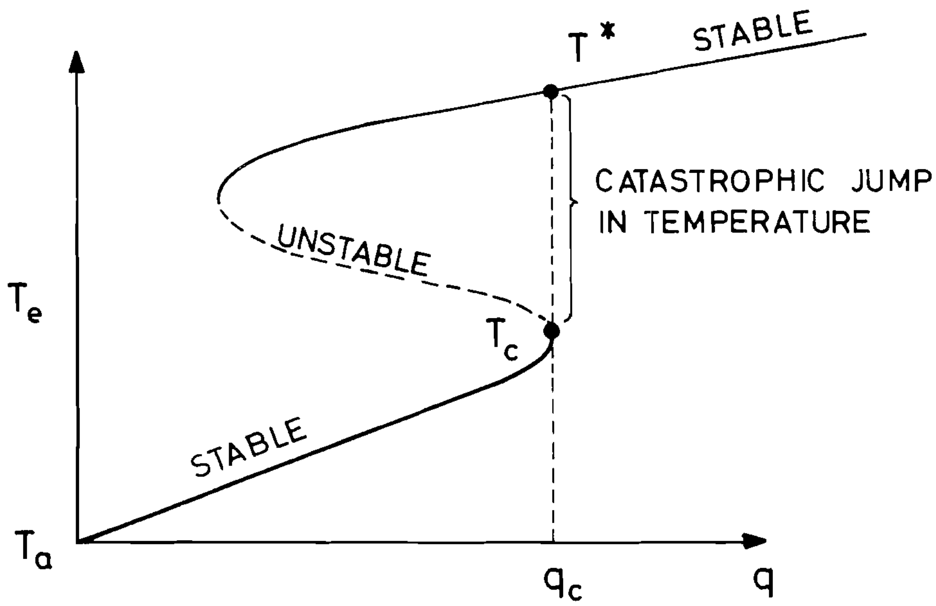

Figure 1. Plot of Equilibrium Temperature T_e versus Input Heat Flux q.

Clearly at this point $\frac{\partial q}{\partial T_e} = 0$ and immediately thereafter, the slope changes sign. In other words, the rate of increase

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*Numerically, it is much easier to assume T_e and calculate the corresponding value of q.

of the heat of reaction with temperature overtakes the rate of increase of heat loss with temperature. This also implies that the temperature equation (2) is changing from a locally stable to a locally unstable equation, since for $q < q_c$ (or $T_e < T_c$), any small increase in temperature results in a net heat loss and any small decrease in temperature results in a heat gain so that the temperature returns to the equilibrium point. But for $q > q_c$ (or $T > T_c$), any deviation results in a movement away from the equilibrium. In practice, one would observe a sudden increase in temperature (Figure 1) as the heat flux crosses the critical value q_c. The temperature essentially jumps to a new equilibrium point determined by the properties of the combustion phase. In Figure 1, the locus of combustion phase equilibria is shown by a thin solid line. Notice that this locus cannot be computed from equations (1) and (2) since the equations for the combustion phase must involve other variables such as oxygen feed rate, volume of combustion products, etc. The dotted line in Figure 1 is the locus of unstable equilibria. The S-shaped curve of Figure 1 is one of the simplest catastrophes and has been called a fold catastrophe by Thom [2]. We will discuss further properties of this curve later on after deriving the equations for ignition temperature and for critical heat flux.

3. Criteria for Ignition

Let us linearize equations (1) and (2) around the point (m, T_e) and denote the deviations by δm and δT. Also let

$$k(T) = a_2 e^{-e_2/RT} .$$

$$\frac{d}{dt}\delta m = -k(T_e)\,\delta m - \frac{e_2 m}{RT_e^2} k(T_e)\,\delta T \tag{4}$$

$$\frac{d}{dt}(\delta T) = -\frac{1}{m^2 c^o}\left[q - h_o(T_e - T_a)^{4/3} - KT_e^4\right]\delta m + \left[\frac{1}{mc^o}\left(-\frac{4}{3}h(T_e) - 4K\,T_e^3\right) + \frac{r_2 e_2}{c^o RT_e^2} k\,(T_e)\right]\delta T . \tag{5}$$

The eigenvalues of the linearized system (4)-(5) are given by the λ-roots of the determinental equation

$$\begin{vmatrix} -k(T_e) - \lambda & , & -\frac{e_2 m}{RT_e^2} k\,(T_e) \\ -\frac{1}{m^2 c^o}\left[q - h_o(T_e - T_a)^{4/3} - KT_e^4\right] & , & \frac{1}{mc^o}\left(-\frac{4}{3}h\,'T_e) - 4KT_e^3\right) + \frac{r_2 e_2}{c^o RT_e^2} k\,(T_e) - \lambda \end{vmatrix} = 0 . \tag{6}$$

Equation (6) may be written as

$$\lambda^2 + \delta\lambda + \gamma = 0 \tag{7}$$

where

$$\delta = k(T_e) + \frac{1}{mc^o}\left(\frac{4}{3}h\,(T_e) + 4KT_e^3\right) - \frac{r_2 e_2}{c^o RT_e^2} k\,(T_e) \tag{8}$$

$$\gamma = -\,k(T_e)\left\{-\frac{1}{mc^o}\left(\frac{4}{3}h\,(T_e) + 4KT_e^3\right) + \frac{r_2 e_2}{c^o RT_e^2} k(T_e) + \frac{e_2}{mc^o RT_e^2}\left(q - h_o(T_e - T_a)^{4/3} - KT_e^4\right)\right\}$$

or

$$\gamma = \frac{k(T_e)}{mc^o} \left(\frac{4}{3} h(T_e) + 4KT_e^3 \right) , \tag{9}$$

since the rest of the terms drop out due to equation (3).

The stability conditions may be expressed directly in terms of δ and γ since they are respectively the sum and the product of the roots of equation (7). When equations (4)-(5) become unstable, at least one root moves from the left half plane to the right half plane and its real part goes through zero. If the roots of equation (7) were real, this would imply that γ would go through zero, but this is impossible since from equation (9), $\gamma > 0$. Thus the roots are complex and at the critical point, $\delta = 0$. This gives us the following condition for the ignition temperature T_c:

$$k(T_c) \left[1 - \frac{r_2 e_2}{c^o R T_c^2} \right] + \frac{1}{mc^o} \left(\frac{4}{3} h\ (T_c) + 4KT_c^3 \right) = 0 \tag{10a}$$

or

$$T_c = \frac{e_2}{R} \Bigg/ \log \left\{ \frac{\left(r_2 e_2 / c^o R T_c^2 \right) - 1}{\frac{1}{mc^o a_2} \left(\frac{4}{3} h\ (T_c) + 4KT_c^3 \right)} \right\} . \tag{10b}$$

Equation (10b) is a transcendental equation in T_c and may be solved by trial and error. However, certain simplifications are possible by neglecting smaller terms and by using dimensionless variables. Let $y' = T_c/T_a$. From equation (10b)

$$y' = E_2 \Bigg/ \log \left(\frac{\frac{R_2 E_2}{y'^2} - 1}{\frac{4}{3A_2} + 4B_2 y'^3} \right) , \tag{11}$$

where $E_2 = \frac{e_2}{RT_a}$, $R_2 = \frac{r_2}{c_o T_a}$, $A_2 = \frac{m c_o a_2}{h}$ and $B_2 = \frac{K T_a^3}{m^o c_o a_2 M}$ are

all dimensionless variables.

Since $R_2 E_2 / y'^2 >> 1$, and $A_2 B_2 y'^3 >> 1$, Equation (11) may be written as

$$y' = \frac{E_2}{\log A_2 + \log \frac{3}{4} \frac{R_2 E_2}{y'^2} - \log 3 A_2 B_2 y'^3}$$

or

$$y' = \frac{E_2}{\log \frac{3}{4} R_2 E_2 - \log 3B_2 - 5 \log y'} \quad . \qquad (12)$$

In Equation (12), log y' term is much smaller than the other terms in the denominator so that a first guess for y' may be obtained by neglecting this term. Using the numbers given by Shivadev and Emmons [1] and assuming M = 0.8, a trial and error hand calculation with equation (10) gives $T_c \approx 657^o K$. The corresponding heat flux obtained from Equation (3) is $q_c = .54$ cal/cm^2 sec.

These results compare favorably with experimental values of 680 $\pm$ 15^{o}K and 0.58 $\pm$ 0.03 cal/cm^2 sec. It should be noticed, however, that T_c or y' is very sensitive to e_2 and a change of e_2 from 54 to 56 would give $T_c \approx 670^o K$ and $q_c = .61$ cal/cm^{2}sec. A slightly higher value of e_2 would also give a better fit to the transient data of Reference [1], Figure 3. It is possible to determine e_2 using nonlinear least squares or maximum likelihood techniques to obtain in some sense a "best" match to the data [3].

Remark:

1. As shown in Figure 1, $\left.\frac{\partial q}{\partial T_e}\right|_{T_e = T_c} = 0$ if the rate of change of mass m is neglected around T_c. Using Equation (3), we get an equation for T_c,

$$k(T_c)\,\frac{r_2 e_2 m}{RT_c^2} - \frac{4}{3}h\,(T_c) - 4KT_c^3 = 0 \quad . \tag{13}$$

Equation (13) is identical to Equation (10a) except for the term $k(T_c)$ which is negligible in comparison with $k(T_c)\,\frac{r_2 e_2 m}{RT_c^2}$. Thus in the present case, $\left.\frac{\partial q}{\partial T_e}\right|_{T_e = T_c} = 0$ provides a simple criterion of ignition. This method will be elaborated further in Section 4 under discussion of Catastrophe Theory.

2. In the paper by Shivadev and Emmons [1], the criterion for ignition is given as $\frac{d^2T}{dt^2} = 0$, while $\frac{dT}{dt} > 0$. Since $\frac{dT}{dt}$ is a function of both T and t,

$$\frac{d^2T}{dt^2} = \frac{\partial}{\partial T}\left(\frac{dT}{dt}\right)\frac{dT}{dt} + \frac{\partial}{\partial t}\left(\frac{dT}{dt}\right) \quad . \tag{14}$$

Now if it is assumed that $\frac{\partial}{\partial t}\left(\frac{dT}{dt}\right) = 0$ and $\frac{dT}{dt} \neq 0$, then

Equation (14) will give the same result as Equation (13). However the above assumptions regarding $\frac{dT}{dt}$ may not hold in every case since, by definition, at an equilibrium point $\frac{dT}{dt}$ and its higher time derivatives are zero. It is also very difficult to give any physical interpretation to the vanishing of $\frac{d^2T}{dt^2}$ at the ignition point. The differences in numerical values of T_c and q_c reported in [1] (715^oK and $0.68\ cal/cm^2sec$) are primarily due to neglecting the radiation and a few other terms which, as is shown here, cannot really be neglected.

4. Catastrophe Theory

The Catastrophe Theory of René Thom [2] is basically a study of the structural stability properties of dissipative systems whose state trajectories or flow fields locally minimize a potential function. Let $f(x,c)$ be such a potential function where x denotes the state of the system and c is the vector of control parameters. For a fixed value of c, the state x flows along negative gradient trajectories, viz.

$$\frac{dx}{dt} = -f_x(x,c) \quad , \tag{15}$$

where $f_x = \frac{\partial f}{\partial x}$ is the gradient function and is zero at equilibrium points. Thom [2] studies the properties of Equation (15) as c is varied slowly and shows that sudden changes in the local stability properties of Equation (15) can occur as c crosses certain boundaries in the control space.

These sudden changes or discontinuities are called catastrophes and the corresponding surfaces in the (x,c) space are called catastrophe surfaces.

The truly remarkable result that Thom [2] derives from topological considerations is that for c of dimension less than 6 and x of any dimension whatsoever, there are only a finite number of catastrophes that can occur. For example, if c is a scalar, only the fold catastrophe of Figure 1 can occur. Other catastrophes are listed in Table 1 and for each catastrophe, a generic potential function f(x,c) is also given. This potential function has the property that it is the simplest potential function that exhibits all the catastrophic properties of more complicated potential functions related to it by a diffeomorphism (i.e. differentiable, one-to-one and inverse differentiable transformation of (x,c)). An exact statement of Thom's Theorem [1,4] can be given as follows:

Let $x \varepsilon R^n$ and $c \varepsilon R^m$. Then $f_x(x,c) = 0$ is an m-dimensional manifold M in R^{n+m}, corresponding to a sheet of equilibrium points (see Figure 2 for the case n = 1, m = 2)*. The equation $f_x(x,c) = 0$ can have multiple roots for a given c and this is what gives rise to singularities of the projection map $\mathscr{X}$: M → C where C is the space of control variables.

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*In Figure 2, x corresponds to T and C corresponds to P_{o2} and Q.

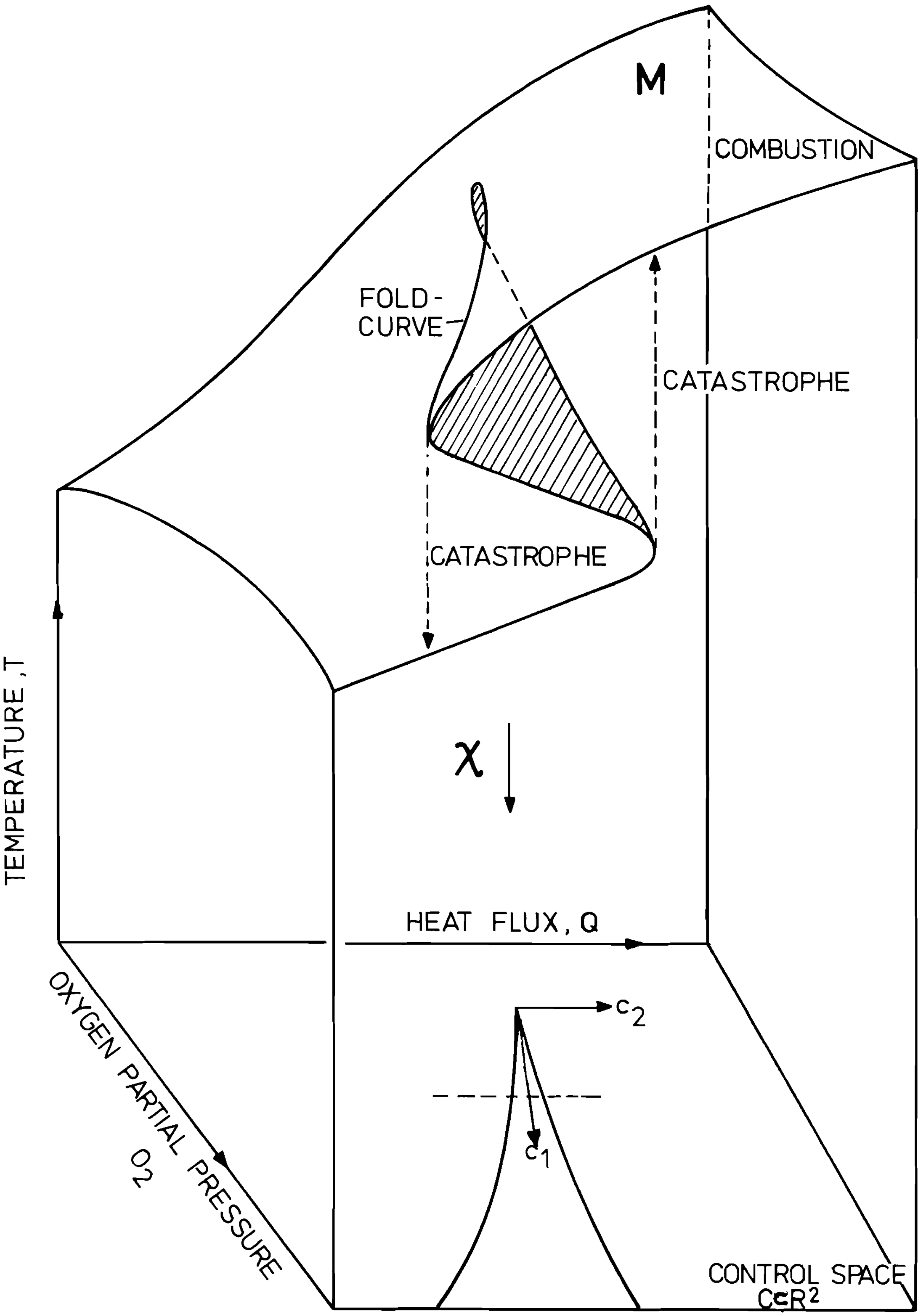

FIGURE 2. QUALITATIVE REPRESENTATION OF COMBUSTION CATASTROPHE.

Table 1. Table of Ordinary Catastrophes on Four-Dimensional Space-Time (from Thom [7]).

Control Space Dimension	Name	Organizing Center	Universal Unfolding (Generic Potential Functions)	Spatial Interpretation	Temporal Interpretation
0	Simple Minimum	$V = x^2$	$V = x^2$	A being An object	To be To last
1	The Fold	$V = x^3/3$	$V = x^3/3 + ux$	The boundary The end	To end To start
2	The Cusp (Riemann-Hugoniot catastrophe)	$V = x^4/4$	$V = x^4/4 + ux^2/2 + vx$	A pleat A fault	To separate To unite To capture To generate To change
3	The Swallow's Tail	$V = x^5/5$	$V = x^5/5 + ux^3/3 + vx^2/2 + wx$	A split A furrow	To split To tear To saw
4	The Butterfly	$V = x^6/6$	$V = x^6/6 + tx^4/4 + ux^3/3 + vx^2/2 + wx$	A flake A pocket A scale (of a fish)	To fill } (a pocket) To empty } (a pocket) To give To receive

For illustration, consider the case $n = 1$, $m = 2$ for which the generic potential function is

$$f(x,c) = \frac{1}{4}x^4 + \frac{1}{2}c_1 x^2 + c_2 x \tag{16}$$

or

$$f_x(x,c) = x^3 + c_1 x + c_2 \quad . \tag{17}$$

The equation $f_x(x,c) = 0$ can, in general, possess three real roots* and the location of these roots will change as c_1 and c_2 are varied. Figure 3 shows the variation of the stationary points of $f(x,c)$ with c_2 for a fixed $c_1 = -3$. It is seen clearly that for $c_2 < -2$, there is only one stationary point, for $-2 \leq c_2 < 2$, there are three stationary points and for $c_2 > 2$, there is again one stationary point, but corresponding to a different minimum than for $c_2 < -2$. Thus in going from $c_2 = -3$ to $c_2 = 3$, there will be a sudden jump in the equilibrium point at $c_2 = 2$ where a maximum and a minimum coalesce resulting in an inflection point. If c_2 is varied in the opposite direction, the jump will occur at $c_2 = -2$, causing hysteresis.

The singularity surfaces are characterized by the inflection point $f_{xx} = 0$ or

$$3x^2 + c_1 = 0 \quad . \tag{18}$$

Equations $fx(x,c) = 0$ and (18) give equations $c_1 = -3x^2$, $c_2 = 2x^3$ for the singularity boundaries in the control space.

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*Since x is real, we are only interested in real roots of $f_x(x,c) = 0$.

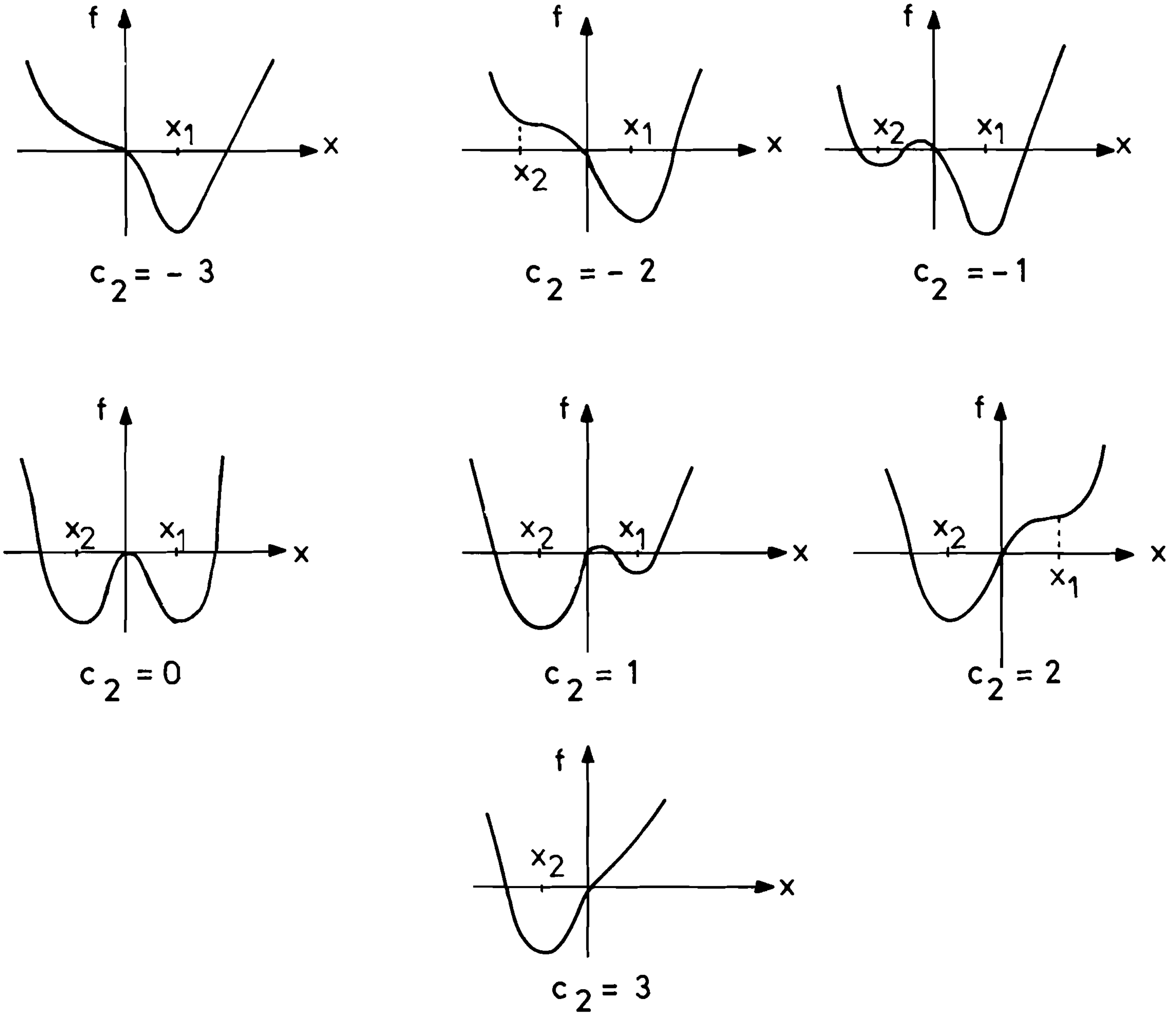


FIGURE 3. POTENTIAL FUNCTION $f(x,c)$ FOR DIFFERENT VALUES OF c_2 AND $c_1 = -3$.

The equation of the singularity or catastrophe curve is $\frac{1}{27} c_1^3 + \frac{1}{4} c_2^2 = 0$ which is a cusp as shown in Figure 2.

Now, following Zeeman [4] we can state the more general theorem of Thom [2].

Let $\mathscr{F}$ denote the space of c^{∞}-functions on R^{m+n} with the Whitney c^{∞}-topology ($\mathscr{F}$ may be regarded as the space of potential functions).

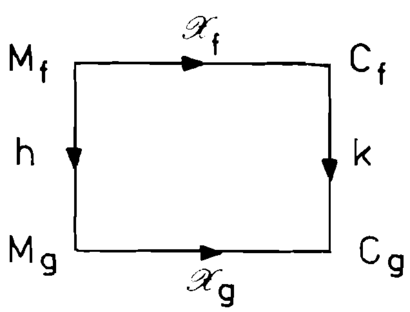
Theorem: If $m \leq 5$, there is an open dense set $\mathscr{F}_0 \subset \mathscr{F}$ which is the set of generic functions. If f is generic then

1) The manifold M_f is an m-manifold,
2) Any singularity of the projection map $\mathscr{X}_f$ is equivalent to one of a finite number of types called elementary catastrophes,
3) $\mathscr{X}_f$ is stable under small perturbations of f.

The number of elementary catastrophes depends only upon m, the dimension of control space, as follows:

m	1	2	3	4	5	6
Elementary Catastrophes	1	2	5	7	11	∞

Here equivalence implies: two maps $\mathscr{X}_f : M_f \to C_f$ and $\mathscr{X}_g : M_g \to C_g$ are equivalent if there exist diffeomorphisms h and k such that the following diagram is commutative.



If $\mathscr{X}_f$ and $\mathscr{X}_g$ have singularities at $x_f \varepsilon M$ and $x_g \varepsilon M_g$ respectively, then the singularities are equivalent if the above definition holds locally with $hx_f = x_g$. Stable means that $\mathscr{X}_f$ is equivalent to $\mathscr{X}_g$ for all g in a neighborhood of f in $\mathscr{F}$.

5. Applications of Catastrophe Theory to Fire Modelling

In Sections 2 and 3, we analyzed pyrolysis and ignition of paper with heat flux as the control variable. In actual fire modelling including combustion, there are many more control variables,e.g. oxygen partial pressure, fuel feed rate, external cooling, fuel feed temperature,etc. Not all of these control variables can be manipulated so that from an operational viewpoint, probably oxygen partial pressure and net external heat flux (which may be negative due to cooling) are the two important control variables. We are currently analyzing stability properties of some simple models of combustion based on chemical reactor analogies and the detailed results will be reported in the near future [5]. Here, based on catastrophe theory, we describe qualitatively the behavior of temperature with oxygen partial pressure and external cooling during the combustion phase. The behavior is shown pictorially in Figure 2 and the effects of changing oxygen partial pressure, p_{O2} and net external heat flux Q are easily observed. The basic hypothesis used in constructing Figure 2 is that P_{O2} is the splitting factor [4]; i.e. for extremely small values of p_{O2} (e.g. in vacuum), the effect of increasing Q is simply a temperature rise without ignition and for large values of p_{O2}, the effect of Q is ignition as shown in Figure 1. In the case where the above hypothesis is correct, the behavior shown in Figure 2 follows from Thom's Theorem.

The right hand arm of the cusp represents the ignition boundary and the left hand arm the "quenching" boundary. A hysteresis effect is seen in that the "quenching" occurs at a lower net heat flux input compared with ignition. This is a general feature of the cusp catastrophe and it will be interesting to verify it experimentally. Another general feature is divergence or extreme sensitivity which was observed by Shivadev and Emmons [1] in ignition with respect to heat flux.

In more general situations where spatial effects are also present and there are more control variables, one may observe catastrophes in time and in space, resulting in 'hot points' similar to those in chemical tubular reactors [6]. Multiphase reactions may give rise to more cusps and to more than three sheets of equilibrium points, and jumps between these points may occur as in Butterfly Catastrophes [2,4]. One of the philosophical implications of catastrophe theory is that catastrophes occur more as a rule than as exceptions in most physical, biological and social systems. Therefore, it is important in the design and operation of engineering systems to map out the catastrophe surfaces over the set of achievable parameter values.

6. Conclusions

It is shown how the general results of catastrophe theory may be applied to the phenomenon of the ignition of paper due to thermal irradiation. Based on stability considerations, equations for ignition temperature and critical heat flux are derived. These equations are shown to provide a good match to the experimental data of Shivadev and Emmons [1]. Further implications of catastrophe theory are discussed for the combustion phase when oxygen partial pressure and external cooling are used as control variables.

Nomenclature and Experimental Quantities

a_2	preexponential factor, $1.9\times10^{16}sec^{-1}$
A_2	dimensionless preexponential factor $m^oc^oa_2/h$
B_2	dimensional radiation factor, $\frac{KT_a^3}{mc_oa_2}$
c	specific heat, 0.32 cal/g^{o}c (T > 500^{o}K)
e_2	activation energy, 54 kcal/g-mole
E_2	dimensionless activation energy, $\frac{e_2}{RT_a}$
f	opacity of the paper sheet [-]
h	sum of heat transfer coefficients at top and bottom, $h_o(T - T_a)^{1/3}$ $\frac{cal}{cm^2sec^oc}$
h_o	temperature independent term in h, .0000941
k	reaction rate constant $[sec^{-1}]$
K	radiation factor $(2\sigma\varepsilon f)$, 2.06×10^{-12} cal/cm^{2}sec $(^oK)^4$
m	surface density, .0085 g/cm^2
M	normalized surface-density, (m/m^o)
q	heat rate [cal/cm^2 sec]
q_c	critical heat rate [cal/cm^{2}sec]
Q	dimensionless heat rate, $\frac{q}{hT_a}$
r_2	heat of reaction, 444 cal/g
R_2	dimensionless heat of reaction, $\frac{r_2}{c^oT_a}$

R	universal gas constant, 1.987×10^{-3} kcal/g-mole ^{o}c
t	time [sec]
T	temperature [^{o}K]
T_a	ambient temperature, 300^{o}K
T_e	equilibrium temperature, ^{o}K
T_c	critical temperature, ^{o}K
y'	dimensionless temperature, $\frac{T}{T_a}$
ε	radiative emissivity [-]
σ	Stefan Boltzmann Constant, 1.3545×10^{-12} cal/cm^{2}sec^{o}K^{4}

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