A SEMI-ANALYTICAL MODEL FOR LASER-INDUCED ABLATION OF FIBRE-RES--ETC(U)

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A SEMI-ANALYTICAL MODEL FOR LASER-INDUCED ABLATION
OF FIBRE-RESIN COMPOSITES

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ABSTRACT

Laser-induced ablation of fibre-resin composites is considered using a steady-state, one-dimensional model. In the analysis, it is attempted to clarify the physical and chemical aspects of ablation rather than to predict ablation rates. It is shown how suitable measurements may be used in conjunction with this model to determine loss rates for combined gaseous and combined non-gaseous products of ablation, and to determine the average enthalpy of each of these two losses. The model is restricted to incident irradiances in excess of approximately 30 MW/m² and to cases in which the ablation rate varies with irradiance in a linear fashion. Because some of the required measurements may be difficult to make accurately, this analysis should be regarded as a preliminary to more detailed work.

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described here. This model is restricted to linear variations of ablation rate with incident laser irradiances, and to irradiances above approximately 30 MW m$^{-2}$, for which some simplifying assumptions, not otherwise valid, can be made. In addition, only steady-state ablation is considered. Shortcomings in the model and in its application are discussed.

2. PHYSICAL MODEL

Figure 1 shows the model adopted here for the steady-state ablation of a fibre-resin composite. Physically, it is based on standard concepts [1-4]. The model is one-dimensional in the $x$ direction with $x = 0$ fixed on the ablating surface so that material appears to move in the positive $x$ direction and alters composition chemically as it moves. Three zones of composition exist which have fixed boundaries for a given ablation rate $v$.

In zone A, virgin material extends effectively to $x = -\infty$. At an unspecified value of $x$, zone B begins in which some decomposition of the resin occurs at a rate that increases with the temperature $T(x)$ according to the Arrhenius law [8]. However, the resin pyrolysis is not advanced to the stage in which a hard char is formed. The model assumes that, in this zone, no decomposition of the fibres occurs. Thus fibres like kevlar, which have similar degradation temperatures to resins, are not included in the model. Zone C is the surface char zone, and is assumed to begin at a measurable $x = x_1$. Resin pyrolysis in this zone produces a hard char, and decomposition or melting of the fibre also occurs. The fact that surface char layers usually have a reasonably well-defined thickness is well established [1-3], and is a consequence of the large difference in the temperature stability of char compared with the stability of undegraded resins. In zone C, gas produced from the resin in zone B may undergo further decomposition before emerging at $x = 0$. Much of the surface char will be lost as gas, but some of it, molten material, and undecomposed fibres may be thermomechanically removed from positions close to $x = 0$. The analysis here assumes that the thermomechanical removal rate cannot be measured directly by any means. Any reaction with environmental gases occurs only in the gas phase external to the ablating body because the evolving gases convectively block environmental gases.

A fraction $\delta$ of the incident irradiance $P_L$ penetrates the gases above the surface. All of the $\delta P_L$ is assumed to be absorbed at $x = 0$ because chars have very high absorption coefficients [9] at the laser wavelengths of interest.

The profiles of temperature $T(x)$ (above ambient temperature) in zones B and C may differ but are assumed to be linear, with $T(x_1) = T_1$, small compared with $T(0) = T_s$ at the surface. (In fact, $T(x)$ in zone C is unlikely to be linear, but the validity of this assumption is defended in Sec. 3.3). In zone A, $T(x)$ is of no importance even for conditions prior to steady-state because the line of demarcation between zones A and B may be chosen at any $x$ value for which $T \ll T_1$; that is, the enthalpy of the virgin material may be made arbitrarily small. Thus the zone of virgin material is included in the model only for the purpose of completing the physical description.
A SEMI-ANALYTICAL MODEL FOR LASER-INDUCED
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1. INTRODUCTION

For military and industrial applications, high energy lasers may be used to penetrate or cut fibre-resin composite materials. In the case of continuous-wave lasers, incident irradiances are typically between 30 MW m\(^{-2}\) and 1 GW m\(^{-2}\). Ablation of fibre-resin heat shields has been extensively studied for aerodynamic heating rates below 25 MW m\(^{-2}\) [1-4] and much of this work is relevant to laser-induced ablation, which has received less attention [5-7].

Except in some unusual circumstances, the complexity of the physical and chemical ablation processes for fibre-resin composites is such that no method currently exists which enables the ablation rate of these materials to be accurately predicted. For laser-induced ablation, three basic inadequacies in present theoretical analyses have been identified [5]. Firstly, the loss rate and enthalpy of solid or liquid material removed thermomechanically cannot be calculated, and appears to be very difficult to measure directly. Such removal may be caused by spallation arising from pressure of pyrolysis gases or by rapid release of thermal stresses in the char, and if an imposed gas flow exists, aerodynamic shear and pressure forces may assist this removal. Secondly, thermophysical constants (e.g. char thermal conductivity) and reaction rate constants are often poorly known. These constants determine in part the final gaseous ablation products and their relative concentrations. Thirdly, the laser radiation may be partially absorbed before reaching the material by pyrolysis gases or by products of reactions between these gases and environmental gases (usually air). The magnitude of this "gas blockage" effect is difficult to estimate.

This report presents a simple analysis which, in conjunction with experiments on a given material, enables quantitative information to be obtained on some aspects of the three factors mentioned above, and which attempts to clarify the physics of the ablation processes. An ability to predict ablation rates awaits more detailed analyses applied individually to each particular composite, but these analyses may be based on the model
Some assumptions are made concerning the two types of thermophysical parameters of relevance, namely mass density and thermal conductivity. The resin density $p_R$ in zone B and the density $p_C$ in zone C are assumed to vary with $x$ at rates that are not significantly greater than the corresponding profiles for $T(x)$. Many complex factors influence these density profiles, but the validity of this assumption is supported by considerable experimental evidence [1,2]. A non-critical assumption is that the thermal conductivity $k_B$ in zone B is known or is easily measured. This is a good assumption because the conductivity of the un decomposed fibres is the dominant contribution to $k_B$ [10], and this conductivity is known or easily determined. An important feature of the present model is that the conductivity in zone C is not assumed to be known. Several other analyses assume that char conductivity is a measurable quantity, but this appears to be a very poor assumption in view of the dominance of radiative transport through porous char at high temperatures [11].

3. ANALYSIS

3.1 Theoretical Approach

The model utilizes conservation of energy and continuity of mass with the main aim being to determine from measurements the rate at which material is removed thermomechanically, and the average enthalpy of this material at the time of removal. Individual knowledge of these two quantities provides a better appreciation of the ablation process than simply a knowledge of their product. The ultimate aim of the model is to acquire a capacity to predict the behaviour of composites under various conditions of irradiance.

Consider first the hypothetical case in which all material is removed in gaseous form. Conservation of energy in the steady-state gives [12]

$$\delta p_L - p_R = \rho_v H_G$$

where $p_R$ is the energy lost by radiation and convective cooling per unit area per unit time, $\rho_v$ is the density of the virgin material, $v$ is the ablation rate, and $H_G$ is the enthalpy of the gases at the time of removal, measured with respect to the virgin material. This enthalpy represents an upper limit to the average enthalpy of ablation products in the more general case in which a fraction of the material is removed thermomechanically. In this latter case, Eq. (1) becomes

$$\delta p_L - p_R = \rho_v v \{ H_G + F \rho_H \}$$

where $F$ is the mass fraction removed thermomechanically, and $H_H$ is the average enthalpy of the non-gaseous products at the time of removal.
The aim here is to determine $H_G$ from Eq. (2) after evaluating the other parameters from experiments and from theory involving known chemical details. Most of the present analysis is concerned with the evaluation of $F$ (Section 3.3). Progress towards this aim is possible if the analysis is now subjected to two main restrictions.

Firstly, the parameters $\delta$, $P_R$, $H_G$, $H_S$, and $F$ are all considered to be constants over the $P_L$ regime of interest. From Eq. (2) this means that $v$ is linearly dependent on $P_L$. As explained in Sec. 3.2 this restriction also means that, for gaseous products, only equilibrium decomposition is considered. The variation $v(P_L)$ can be determined experimentally [7,12], and if found to be linear, this restriction is likely to apply. Little possibility exists that compensating variations in $H_G$ and $H_S$ with $P_L$ will result in a linear $v(P_L)$ because $H_S$ will tend to be constant; thermomechanical removal occurs when the material has attained a particular physical condition. (If $F$ is non-constant, this is observable independently of $v(P_L)$ - see Sec. 3.3.) Linear variations for $v(P_L)$ have been observed for several ablating composites [6,7]. However, a non-linear $v(P_L)$ has been observed for kevlar-epoxy ablating in an airflow, and this was attributed to a strong dependence of $H_G$ on $P_L$ arising from nonequilibrium decomposition effects [7].

The second major restriction here is that only the regime of irradiance $P_L$ in excess of approximately 30 MW m$^{-2}$ is considered. As explained later in Secs. 3.2 and 3.5, this effectively enables $H_G$ and $P_R$ to be evaluated. This is not a serious restriction because in practice lasers are rarely used at irradiances below 30 MW m$^{-2}$ for cutting or penetrating non-metals.

### 3.2 Determination of $H_G$

For $P_L > 30$ MW m$^{-2}$, the gaseous ablation products may be assumed to be a small number of elements or simple compounds such as C, Si, CO, SiO, H, H$_2$, CH, C$_2$H and C$_4$H [5]. Relative concentrations of these product species may be predicted with sufficient accuracy by analytical or numerical methods in order to identify a small number that are dominant. This is so because uncertainties in chemical kinetic rate constants become less significant at temperatures sufficiently high to induce almost complete dissociation of hydrocarbons. Exceptions occur, for example, when the heating rates experienced by a material during ablation are faster than those rates that allow equilibrium decomposition conditions. Such nonequilibrium effects cause the gas enthalpy $H_G$ to vary with $P_L$, and thus $v(P_L)$ is non-linear [7]. The present model accommodates only those nonequilibrium events which lead to non-gaseous ablation products, i.e. thermomechanically-removed products.

If attention is restricted to equilibrium decomposition for gaseous products (i.e. high $P_L$ and linear $v(P_L)$), $H_G$ may be obtained in part from the heat of formation of the material considered to have been formed from the few dominant gaseous ablation products. Since these gases are simple compounds or elements, their own heats of formation are low and do not strongly influence the overall heat of formation. Thus considerable uncertainty in the gas composition may be tolerated. In order to obtain $H_G$, the sensible
heat content $C_p T_s$ of the gas must be added to the overall heat of formation (where $C_p$ is the average specific heat of the gases).

3.3 Determination of $F$

This is the only parameter which requires for its evaluation a consideration of the zones in Fig. 1. For zone C, the mass flowing in at $x = x_1$ equals that flowing out at $x = 0$; i.e.

$$v_{PF} + v_{PR1} + M_{GR} = fM_{GR} + M_{GC} + M_S \quad (3)$$

where $p_F$ is the mass density of undecomposed fibres, $p_{R1}$ is the mass density of non-gaseous resin remaining at $x_1$, $M_{GR}$ is the mass of gas evolved through $x_1$ from the resin per unit area per unit time, $f$ is the mass fraction of this gas that emerges at $x = 0$, $M_{GC}$ is the mass of gas evolved through $x = 0$ from char decomposition per unit area per unit time, and $M_S$ is the rate at which mass is lost thermomechanically per unit area. For zone B, continuity of mass gives

$$v_{PW} = fM_{GR} + M_{GC} + M_S \quad (4)$$

Equations (3) and (4) give

$$v_{PW} = fM_{GR} + M_{GC} + M_S$$

$$= fM_{GR} + M_{GC} + F v_{PW} \quad (5)$$

It is now assumed that $M_{GR}$ can be determined from a simple measurement using a sample of the resin. When a resin is heated, a mass fraction of it forms a relatively stable char while the remaining fraction $r$ is evolved as gas. Measurements of $r$ are not uncommon, and a typical value for epoxy is 0.8 [13]. The heating rates employed in these measurements are slower than those induced by a laser because observation of resin weight loss with temperature is necessary in the former case. Since the pyrolysis behaviour at low heating rates may not be the same as those at high heating rates, it is assumed here that the fraction $r$ is the same in both cases, but that the temperature for stable char formation ($T_i$ in the case of laser heating) may be different. (Determination of $T_i$ is discussed later in this section). Thus $M_{GR}$ in Eq. (5) can be replaced by

$$M_{GR} = r p_{R} \quad (6)$$

where $p_{R}$ is the known density of virgin resin.
The additional assumption is now made that \( f = 1 \), i.e. that although further dissociation of the gas represented by \( \mathbf{M} \) may occur in zone C, none of this gas is deposited there. Reactions that may occur between this gas and char in zone C are viewed as exchange reactions (mainly of carbon) without loss to \( \mathbf{M} \). This non-interactive nature of \( \mathbf{M} \) and zone C may be expected because for the very high temperatures in zone C (about 3,500 K at the surface), decomposition processes are dominant over recombination processes [4,5].

In order to determine \( \mathbf{M} \) for use in Eq. (5), the Arrhenius law is assumed for the decomposition of the char [8]:

\[
\mathbf{M} = \int_0^\infty \rho_C B_C \exp(-E_C/(RT)) \, dx
\]  

(7)

where \( \rho_C = \rho(x) \) is the density of solid plus liquid in the char, \( B_C \) is the frequency factor, \( E_C \) is the activation energy, and \( R \) is the gas constant. Since the chemical composition of zone C is very complex, the use of a single expression of this sort represents a gross simplification. However, use of Eq. (7) is somewhat justified if it is viewed not in terms of describing a set of chemical processes but as an expression for which an empirical fit is to be obtained based mainly on physical considerations.

An approximate solution for the integral term in Eq. (7) is possible because invariably \( E_C/(RT) >> 1 \). Thus the exponential term increases very rapidly with \( T \), while \( \rho_C \) varies slowly; i.e. most of the contribution to \( \mathbf{M} \) comes from the part of the char near the hot surface. Therefore, \( \rho_C(x) \) may be approximated by the density \( \rho_S \) at the surface, and \( T \) may be expanded near \( T_S \) as

\[
T = T_S + bx
\]  

(8a)

where

\[
b = (T_1 - T_S)/x_1
\]  

(8b)

thus

\[
T^{-1} = (T_S - bx)/[(T_S - bx)(T_S + bx)]
\]

\[
= (T_S - bx)/(T_S^2 - b^2 x^2)
\]

\[
= T_S^{-1} - bx/T_S^2 \quad \text{(since } b^2 x^2 << T_S^2 \text{)}
\]  

(9)
Hence

\[ x = (\frac{T_S^2}{b})(T_S^{-1} - T^{-1}) \]  

(10)

and

\[ dx = -(\frac{T_S^2}{b})d(T^{-1}) \]  

(11)

Using Eq.(11) to integrate Eq.(7) and putting \( \exp(-E_C/(RT_S)) >> \exp(-E_C/(RT_1)) \) gives

\[
M_{GC} = \frac{\rho B C R T^2}{S C - S_1} \exp(-E_C/(RT_S)) \\
= \frac{\rho B C R T^2 x_1}{E_C(T_1 - T_S)} \exp(-E_C/(RT_S))
\]  

(12)

As a brief aside it is interesting to note that, if a constant char temperature \( T_S \) is assumed instead of Eqs. (8), the less-accurate result equivalent to Eq. (12) is

\[ M_{GC} = - \rho C B C x_1 \exp(-E_C/(RT_S)) \]  

(13)

Equations (5), (6) and (12) give

\[
v_p = \frac{\rho B C R T^2 x_1 \exp(-E_C/(RT_S))}{E_C(1 - F - r (\rho R/\rho_c))(T_1 - T_S)}
\]  

(14)

Equation (14) may be expressed as

\[
\ln(v_p) = \ln \left( \frac{\rho B C R T^2 x_1}{E_C(1 - F - r (\rho R/\rho_c))(T_1 - T_S)} \right) - \frac{E_C}{RT_S}
\]  

(15)

The first term on the right hand side of Eq. (15) is likely to vary with \( T_S \) much more slowly than the second term, and some experimental evidence supports this expectation [14]. Here, it will be assumed that the first term is constant with \( T_S \), at least for a limited regime of interest. Thus, if the variation of \( T_S \) with \( v \) is measured (this would involve varying \( P_L \)), a plot of \( \ln(v_p) \) against \( T_S^{-1} \) should give a straight line with a slope equal to \(-E_C/R\). This procedure is similar to a thermogravimetric analysis [15].
The parameter $F$ may then be obtained from the intercept of the straight line with the axis $T_S^{-1} = 0$ if it is assumed that $\rho_S$, $B_C$, $x_1$ and $T_1$ are measurable. This assumption is probably the poorest of all those in this report, and it will be further discussed here and in Section 4.

The three quantities $\rho_S$, $B_C$ and $x_1$ may be obtained from post-ablation analyses. This approach is commonly used for ascertaining heat shield performance [16]. Although measurements of $\rho_S$, the char density at the surface, and $x_1$, the char layer thickness, are quite simple measurements for cold char, it is necessary to assume that the post-ablation values are indicative of the values existing during ablation. In this respect, high speed photography of the ablation at the time that laser irradiation ceases may be used to indicate if any char loss continues after irradiation. Fortunately, the thermal conductivity of char is usually higher than that of the uncharred material in zone B, and thus the char layer thickness and density may be expected to "freeze" during the rapid cooling that would exist in most environments of interest immediately following cessation of irradiation. The measurement of $B_C$ involves thermogravimetric analysis of the resulting char, and if performed properly the analysis should yield a value for $E_R$ in agreement with that obtained previously. Such an analysis should utilise temperatures and heating rates as high as possible, and use an inert atmosphere in order to simulate the convective blockage effect of pyrolysis gases that exist at the high laser-heating rates. However, the accuracy that may be expected from this technique is poor [15].

The quantity $T_1$ may be determined by slowly heating a sample of the resin in order to find the lowest temperature at which it is relatively stable. Typically, this temperature is only about 700 K [13], and thus $T_1 \ll T_S$. Hence errors in deducing $T_1$ are not of major concern. As mentioned earlier in this section, the behaviour at low heating rates may not accurately represent that at high heating rates, so some check on the accuracy of the above approach may be warranted. This may be achieved by utilising the frequency factor $B_R$ and the activation energy $E_R$ for decomposition of the resin. If unavailable, $B_R$ and $E_R$ may be obtained by a standard thermogravimetric analysis. Then, in analogy with the analysis leading to Eq. (12), mass continuity of resin for zone B (a modification of Eq. (4)) gives

$$v_{\rho_R} = M_{GR} + v_{\rho_R1}$$

$$= M_{GR} + (1 - r)v_{\rho_R}$$

$$\frac{\rho_{R1} B_R T_1^2}{aR_r} \exp \left(- \frac{E_R}{RT_1} \right)$$

(16)
where $\rho_R$ is the known density of the virgin resin, $\rho_{R1}$ is the measurable density of the pyrolysed resin in zone B near the interface at $x_1$, and $a = \partial T/\partial x$ in zone B (compare Eqs. (8)). The quantity $a$ is readily available from the heat conduction equation and the requirement for energy conservation across the interface, i.e.

$$k_B a = \nu P_R H_R$$

(17)

where $k_B$ is the known thermal conductivity in zone B as discussed in Sec. 2, and $k_B a$ is equal to the rate of heat flow into zone B; $H_R$ is the enthalpy required to char the resin, and is known or easily measured. Thus $T_1$ is available from Eqs. (16) and (17). This completes the determination of the four quantities $\rho_S$, $R_C$, $x_1$ and $T_1$ shown by Eq. (15) to be necessary to evaluate $F$.

3.4 Determination of $\delta$

In most cases with cw lasers, $\delta$ will not be small compared with unity [17,18]. The simple gaseous ablation products are unlikely to cause absorption, but absorption may be significant in gases such as CO$_2$ and H$_2$O formed from reaction of the ablation products with the cold environmental gas in front of the ablator [19]. Experimentally, these absorbing gases may be precluded by using an inert gas such as argon in place of the environmental gas of interest, thus making $\delta = 1$. By measuring the ablation rate $\nu$ in both types of atmosphere, $\delta$ for the reacting atmosphere may then be deduced. Such a procedure has been performed in the case of ablating samples of glass-epoxy and kevlar-epoxy composites [7].

Alternatively, $\delta$ can be obtained from Eq. (2) and a measure of $\nu(PL)$ provided that $P_R$ is known (see Sec. 3.5). If the results for $\nu(PL)$ are extrapolated back to $\nu = 0$, then Eq. (2) shows that $\delta = P_R/PL$ when $\nu = 0$.

3.5 Determination of $P_R$

The quantity $P_R$ is the remaining parameter required in Eq. (2) in order to find $H_S$. It is limited by the melting (or sublimation) temperatures of ablating elements. For $P_s > 30$ MW m$^{-2}$, the surface temperature $T_S$ is usually in excess of 3000 K [5,7], and convective cooling rates are much smaller than radiative cooling rates. Thus present theoretical approximations adequately determine the former rate despite the large uncertainties [20]. The radiative cooling contribution is given by $\epsilon \sigma T_S^4$ where $\epsilon$ is the total surface emissivity, and $\sigma$ is the Stefan-Boltzman constant. At 4400 K, $\epsilon T_S^4 = 20$ MW m$^{-2}$. For ablating fibre-resin composites, $\epsilon > 0.8$, and thus if $T_S$ is measured to within 200 K, the absolute uncertainties in $P_R$ are only around 3 MW m$^{-2}$, which can be ignored compared with $P_L$ for $P_L > 30$ MW m$^{-2}$. 
4. DISCUSSION AND CONCLUSIONS

At present no analytical or numerical technique, including the one described here, can quantitatively predict the significance of thermomechanical removal effects without some measurements. The analysis described here shows how this significance may be determined from some suitable measurements, at least in principle, but because some of the assumptions in the theory are poor, the main value of the analysis is that it delineates the important physical and chemical parameters and clarifies the physical concepts. Thus the analysis should be considered to be preliminary to more detailed work.

The model is restricted to cases in which \( v \) is observed to vary with \( P_L \) in a linear fashion, and to cases in which \( P_L > 30 \text{ MW m}^{-2} \). Its main shortcomings are that it assumes that the frequency factor \( B_C \) for the char can be determined experimentally, and that the char thickness \( x_1 \) can be determined from a post-ablation measurement. The latter assumption could probably be justified in some specific cases, but since typically \( x_1 < 2 \text{ mm} \) when \( P_L > 30 \text{ MW m}^{-2} \), the accuracy of post-ablation measurement for \( x_1 \) may even then be poor. However, in theoretical treatments for the ablation of composite materials, shortcomings of this or greater magnitude are, by necessity, often tolerated.
5. REFERENCES


FIG. 1  Physical model for the ablation of a fibre-resin composite material
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