

# TOPICS IN FILTRATION COMBUSTION

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## **Introduction**

Filtration Combustion is a subset of general combustion systems in which there is important additional upstream thermal supply or enhancement that supplements the normal upstream heat exchange of a plug-flow flame. The critical result is significantly increased flame speed, possibly or actually accompanied by superadiabatic flame temperatures. The thermal enhancement can be achieved by such means as external heat exchangers, or internal backmix flow, or by combustion in a porous matrix as in the case of filtration combustion. In filtration combustion, the thermal enhancement is provided (mostly) by porous body radiation with gas/solid convective exchange.

There are two general sub-classes of filtration combustion: (I) combustion of mixed gases *in* a porous matrix with heat release mainly in the gas phase in the pores (but not excluding catalytic reaction at the solid surface); and (II) reaction of a flowing gas *with* the porous matrix, as in solids combustion (mostly coal), with heat release mainly at the solid surface (but not excluding volatiles or CO reaction in the gas phase). The governing equations are identical, with a factor,  $\alpha$ , representing the distribution of heat release between the solid phase and the gas phase, that can be the defining differentiation between the two sub-classes. In either case, the resulting pattern of combustion phenomena can be: a combination of increased flame speed coupled with potential or actual superadiabatic flame temperatures, and possibly with singular peaking temperature profiles that can reasonably be described as "soliton like". This identifies targets for prediction, requiring modeling of the reaction system, and the procedure presented in this article is based on an integral formulation as the principal method of analysis.

## **Background**

The historical origin of filtration combustion, although not commonly identified as such, goes back more than a century; Rabinovich and Gurevich [1] identify a patent of 1867 as the scientific origin of relevant studies. The corresponding documentation of filtration combustion is quite substantial: the published collection of papers edited by Matros [2] is the most complete at this time for the Class I systems, for the Class II systems, the recognized documentation set as filtration combustion is very limited [3] although the history can again be traced back more than a century. More recently (1987), Koester [4] has about 75 citations of relevant work covering both classes. The early studies also identified evidence of incipient or actual super-adiabatic temperatures in sintering and agglomeration [5], in fluid bed combustion [6], in fixed bed combustion [3], and in pulverized coal flames [7]. This phenomenon of superadiabaticity was first discussed most completely by Weinberg [8]. For the soliton-like flames in filtration combustion, limited solution to the defining equations has been given, notably by Babkin [9], but not so far as we are aware in the more general form given here.

In this article, the focus is on the complete (integral) but simplified reaction system without initial specification of the class type, and the major basis is an unpublished approach [10] updated from earlier technical papers [11]. This procedure establishes a number of important benchmark conditions that must be satisfied in any detailed

modeling

### Model Structure and Integral Results

For combustion in or with a porous matrix, supplied by reactive gas flowing +x at velocity  $U_b$ , the matrix is stationary, and the reaction front (combustion wave) can move up or down stream at a velocity  $V_f$  (positive in the +x direction) Using co-ordinates moving with the reaction front, we define both the reactant gas mixture (sub-g) and the porous body (sub-b) moving through from left to right at mass flow rates:  $m_g = \rho u_g = \rho(U_g - V_f)$ , and  $m_b = \sigma u_b$ , where  $u_b = -V_f$ , and  $(U_g - V_f)$  is the flame velocity relative to the gas. The standard equations for the energy balance and reaction (taken for simplicity to be one-step, first-order) are:

$$\lambda_g(T_g'') - m_g c_{p,g} T_g' - h.A.e.(T_g - T_b) + \alpha.\rho_g h_f(-dc/dt) = 0 \quad (1)$$

$$\lambda_b(T_b'') - m_b c_{p,b} T_b' + h.A.e.(T_g - T_b) + (1-\alpha).\sigma_b h_f(-dc/dt) = 0 \quad (2)$$

$$-dc/dt = k.c \quad (3)$$

Transformation into dimensionless co-ordinates [4], as defined (below), these equations become:

$$\Delta_g'' - P_g \Delta_g' - B.(\Delta_g - \Delta_b) + \alpha.P_g.\eta' = 0 \quad (4)$$

$$\Delta_b'' - P_b \Delta_b' - (B/Lr).(\Delta_g - \Delta_b) + (1-\alpha).P_g.\eta' = 0 \quad (5)$$

$$\eta' = K.(1-\eta).\exp[-1/(\theta_{amb} + \omega.\Delta)] \quad (6)$$

**Integral Formulation.** Information on elements of the flame structure are obtained from the integral formulation [4], by defining:

$$\begin{aligned} B.SUM(dif) &= B \int_{-\infty}^x (\Delta_g - \Delta_b).dx \\ &= [-\Delta_g' + P_g.(\Delta_g - \alpha.\eta)]_{-\infty}^x = -\Delta_g' + P_g.(\Delta_g - \alpha.\eta) \\ &= Lr. [\Delta_b' - P_b.(\Delta_b - (1-\alpha).\eta)]_{-\infty}^x = Lr. [\Delta_b' + P_b.(\Delta_b - (1-\alpha).\eta)] \end{aligned} \quad (7)$$

**Downstream BC** At the downstream or integration upper limit, at  $x = \infty$ , the gradients are zero so that:

$$P_g.(\Delta_{g,\infty} - \alpha.\eta_{max}) = Lr. [-P_b.(\Delta_{b,\infty} - (1-\alpha).\eta_{max})] = -Hr.P_g.\Delta_{b,\infty} \quad (8)$$

$$\text{with: } Hr = \frac{[(u_b).(\sigma.h_{b,o})]}{[(u_g).(\rho.h_{g,o})]} \quad (9)$$

Since the temperatures of the gas and the porous body are the same at the downstream limit,

for  $x = +\infty$ , then

$$\Delta_{g,+\infty} = \Delta_{b,+\infty} = \alpha.\eta_{max} / (1 + Hr) \quad (10)$$

In the limit case that all heat released (or all reaction) is in the gas phase, i.e.,  $\alpha = 1$ , and for (effectively) complete combustion so that  $\eta_{max} \approx 1$ , then the exit temperature,  $\Delta_{g1+\infty} = 1/(1 + Hr)$ . This means that the exit gas temperature is only at adiabatic if  $Hr = 0$ , that is to say, that the flame is stationary. For flames moving downstream in the physical system, i.e.,  $U_g > 0$  and  $Hr > 0$ , the exhaust gas temperature is below adiabatic, but for

flames moving upstream ( $Hr < 0$ ), the reverse is true, and the exit temperatures are super-adiabatic. In addition, it can be seen that there is a lower limit of  $Hr = -1$  at which the temperatures go to infinity. This thus provides a set of important bench marks for all the flame systems.

**Flame Speed Limitations** Since, for valid physical solutions, we must have  $Hr > -1$ , this places a limit on the flame speed. Since the stoichiometric values of  $h_{i,0}$  in Eq. 9 are approximately the same for the gas and for the porous body, the difference between the gas flow velocity and the porous body "velocity" is approximately the ratio of the densities. Since these are roughly in the ratio of 1000, then we have at this limit:

$$-u_b = V_f < u_g/1000 \quad (11)$$

Thus, for a gas approach velocity of 1 m/sec, the resultant flame speed — in the +x direction (i.e., moving downstream) -- should be of the order of and less than 1 mm/sec. This estimate is in line with the findings reported by Koester [4]. This shows that the downstream flame speed is limited to a very low value that can be up to three orders of magnitude smaller than the velocity of the gas flowing through the bed. Again this represents another important benchmark value.

**Crossing Point** Temperature profiles for both the gas and the porous body are at ambient at  $x = -\infty$ ; they rise with increasing  $x$ , with the porous body exceeding that of the gas mixture, to be able to preheat it, for large negative values of  $x$ . These curves must cross if reaction is initiated. At the crossing point,  $\Delta_b = \Delta_g$ . Some bounds can be established on whether this is at the downstream exit ( $x = \infty$ ) or earlier by considering the integral solutions of Eq. 7 taken to the limit. At  $x = \infty$ , Eq. 7 for the porous body becomes:

$$\text{TotalSUM(dif)} = Lr[-P_b \Delta_b] = -Hr P_g \Delta_b, +\infty \quad (12)$$

The value of SUM(dif) at small  $x$  must initially be positive since otherwise the gas would not be preheated. Consequently, if TotalSUM(dif) is zero or negative, the two curves must have crossed. From Eq. 10, this applies at least to all values of  $Hr \geq 0$ , that is, for flames stationary or moving upstream. This is another benchmark. It will also apply to an undetermined extent for the flame moving downstream. It will be determined by the conditions at which SUM(dif) is at a maximum. This requires determination by numerical solution, however.

**Soliton Structure** A final result that can be obtained from the integral formulations is the potential for the temperatures to peak, that is, to form soliton-like structures. The criterion for this is that the gas temperature gradient is zero or negative before exit. Consider Eq. 7 written in the form:

$$\Delta_g' / P_g = -[(\alpha \eta - \Delta_g) + (Lr/P_g) \Delta_b' + (-Hr) \Delta_b] \quad (13)$$

If  $\Delta_b'$  is ever negative, then the porous body temperature will have peaked; this is impossible unless the gas temperature has also peaked, which establishes Soliton-like behavior.

Additionally, we can say that  $(1/P_g)$  is generally small, and so is  $\Delta_b'$  for very large  $x$ ; hence, as the product of two small quantities, the middle term of the RHS bracket can at the limit be neglected. Since  $\eta$  is also of order of 1 at very large  $x$ , Eq. 13 reduces to

$$\Delta_b' / P_g = -[(\alpha - \Delta_g) + (-Hr) \Delta_b] \quad (13a)$$

Clearly, if  $Hr \sim 0$ , the exit values of the temperatures are both  $\eta_{\max} \approx 1$ ; then for  $\alpha \approx 1$ , the gradient is approximately given by  $-[1 - \Delta_b] < 0$ ; hence, for  $Hr > 0$ , we can expect generally that the gradient is negative, and the gas temperature at least has peaked.

For  $Hr < 0$ , the second term in Eq. 13a is clearly positive but in this region we have from Eq. 10 that  $\Delta_g$  can exceed unity, but by an undetermined amount without numerical

computation. However, if we use Eq. 10, taking the maximum reaction efficiency as about unity, then subtracting Eq 10 from Eq. 13a, and taking  $\Delta_b'$  as zero, we find

$$\Delta_{b,\infty} - \Delta_g \approx (-Hr) \cdot (\Delta_{b,\infty} - \Delta_b) \quad (14)$$

Hence, for  $Hr < 0$ , at the presumed maximum, the RHS is positive, so that:  $\Delta_{g,\infty} > \Delta_g$ . Consequently, either the gas temperature at the maximum ( $\Delta_g'$ ) is less than the exit temperature, which is a contradiction, or the maximum is located at the exit at  $x = \infty$ , meaning thereby, that the gas temperature rises smoothly to a superadiabatic value at the exit without peaking.

Thus, the benchmark conclusion for the most probable pattern is that for  $Hr < 0$  (i.e., flames moving upstream) the gas temperatures can be superadiabatic, but they peak at the exit, and for  $Hr > 0$  (flames moving downstream) they do peak, but the exit gas temperature is below adiabatic. Consequently, this shows that there are values of parameters for which Soliton-Like flames can exist

### Numerical Solutions

The governing equations have been solved for the two cases, of (Class I) reaction in the gas phase [5,4], and of (Class II) reaction with the solid phase [2]. The solutions for the Class I conditions were simplified by assuming no solid surface reaction ( $\alpha = 1$ ), but for the Class II conditions, the gas phase reaction (primarily of CO to CO<sub>2</sub>) was found to be a major factor in the system. Details are out of place here for space, but in summary, the solutions for the Class I substantiated the benchmark expectations, with the gas temperatures rising smoothly to a superadiabatic exit maximum for flames moving upstream ( $Hr < 0$ ), and for flames moving downstream ( $Hr > 0$ ), providing soliton-type gas temperature peaks that just exceeded adiabatic, and with the temperature then declining to an exit value below adiabatic, even though the system was adiabatic.

The method of solution was to form 4th order DE's for separation of variables. The DE's formed were linear and homogeneous with constant coefficients, and could be solved analytically under the condition that the reaction rate was sufficiently small that it could be taken as zero. This was the case for the initial approach flow before reaction started, and for the exhaust flow when reaction was substantially complete. For the intervening region when the reaction rate was significant, only numerical solution was possible. The analytical solutions were used to obtain initial and final conditions for the numerical computations.

The results of the computations were as already summarized. In all cases the final reaction efficiency was so close to 100% that it could be taken at all times that  $\eta_{\max} = 1$ . Likewise, the final exit temperature for both gas and solid essentially at adiabatic for  $Hr \leq 0$ , otherwise, the exit temperature declined with increasing  $Hr > 0$ , as concluded from the benchmark integral results. The temperature curves all showed the predicted crossing, with the crossing point occurring in all cases before significant rise in the reaction efficiency,  $\eta$ . Additionally, however, the numerical calculations showed:  $\Delta_g > \eta$ ; until the reaction was essentially complete. This behavior was associated with the benchmark predictions of superadiabatic gas temperatures, in all cases, however, such superadiabaticity was not obtained for the porous body.

The earlier study of coal combustion [2] showed substantially similar results but with some significant differences. The differences derived from the more complex system involved in which the split in reaction between solid phase and gas phase was critical, additionally, the system is more complex in that there is continued filtration reaction downstream of the combustion zone, involving gasification. As a result of the gasification,

which is endothermic, the temperature profiles peaked significantly in a soliton-type of profile. The joint result of these characteristics was that the predicted upstream solids temperature was found to be highly dependent on the distribution of reaction between solid and gas phase (value of  $\alpha$ ); however, the magnitude of the temperature peak was essentially insensitive to this distribution. The values of  $\alpha$  required for acceptable experimental fit were surprisingly small, of the order of 10%, with 90% of the heat release in the gas phase.

### Conclusions

These results show the ability of the integral formulation to provide a general pattern of behavior and benchmark relative values of such parameters as the gas exit temperature, the velocity limit for the flame propagation even in substantially high speed flow. In particular, the results show the critical significance of the velocity ratio of thermal flux ratio designated by the dimensionless group  $H_r$ .

This now provides a basis for extension of the analysis to consider the modification to behavior due to variation of the locus of the reaction - in the gas phase, or at the solid surface; and the modification when stoichiometry is taken into account. These define clear targets for future developments

### List of Symbols

|   |  |
|---|--|
| (subscript) $i = g$ (gas) or $b$ (porous body) flame-                         | $u_i$ : velocity in $+x$ direction in stationary coordinates   |
| $A_e$ : internal surface area of porous body per unit mass                    | $U_g$ : gas velocity in physical coordinates   |
| $c$ : reactant concentration: mass basis (kg/kg)                              | $V_f$ : velocity of flame (in $+x$ direction)  |
| $\theta_{amb}/\omega$   | $\Delta$ : re-normalized temperature: $= (\theta_i - \theta_{amb})/\omega$   |
| $C_{p,i}$ : specific heat   | $\rho$ : gas density   |
| $E$ : activation energy   | $\sigma$ : solid density   |
| $h$ : internal heat transfer coefficient for gas to solid                     | $\omega$ : dimensionless concentration: also dimensionless temperature rise from ambient to adiabatic: $= (c \cdot h_f R / c_{p,g} E)$ |
| $h_f$ : heat of reaction  | $\lambda$ : thermal conductivity   |
| $H_r$ : enthalpy flux ratio (Eq. 10)  | $\theta$ : dimensionless temperature $= RT/E$  |
| $k$ : reaction velocity constant $= \kappa_0 \cdot \exp(-E/RT)$               | $\theta_{amb}, \theta_{ad}$ : ambient and adiabatic temperatures   |
| $K = k/u_g$   | $\eta$ : reaction efficiency   |
| $L_r$ : ratio of thermal conductivities ( $\lambda_b/\lambda_g$ )             |  |
| $m_i$ : mass flux (density x velocity)  |  |
| $P_i = u_i \cdot \rho_i \cdot c_{p,i}$ (for the porous body $\rho = \sigma$ ) |  |
| $R$ : gas constant  |  |
| $T_i$ : temperature   |  |

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