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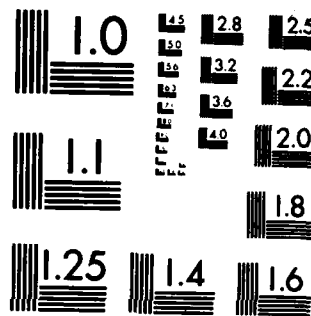
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A REVIEW OF RESEARCH IN
CHEMICAL-ACOUSTIC COUPLING

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Introduction

In this review we have summarized previous research in chemical-acoustic interactions and extracted from it those elements which may be most important to reactive flows of exothermic materials. The basic idea of chemical-acoustic coupling is that there can be feedback between sound waves occurring naturally in a chamber and the energy released in a chemically reacting region. Exothermic reactions can both generate sound waves and amplify the natural ones. Conversely, sound waves may substantially decrease the chemical reaction times. Thus what might start out as a small amplitude wave can, through various feedback mechanisms, be reinforced and grow substantially. As we shall outline below, the extent of such coupling depends on the exothermicity, the activation energy, and the ratio of a chemical to an acoustic time.

In the material presented below we are stressing what has been learned about chemical-acoustic interactions in a premixed, laminar medium. We are not including those types of instabilities that are generated by fluid motions coupling with sound waves, e.g. vortex generation or Tollmien-Schlichting waves. We are also not immediately concerned with noise which is generated by random fluctuations but which does not feed back to the chemistry through resonant coupling. However, although they are hydrodynamic in origin, we will briefly mention and give references to work in acoustic-flame oscillations.

We have separated the research in chemical-acoustic interactions into three topics. The first deals with the characteristics of pressure waves generated by energy release. The second addresses the question of how ambient sound waves are altered by a nonequilibrium reactive mixture. The final topic considers how the presence of sound waves effects the ongoing chemical kinetics. It is important to note that most of the work referred to below is analytical and therefore rather idealized compared to even the simplest physical environment in which these effects occur. Recently there have been some experiments to verify the analyses and some numerical simulations which allow certain complications to be examined. We conclude this review with a summary of the chemical-acoustic issues that require further research.

Pressure Waves Generated by Heat Release

When heat is added to a small volume of the gas in a chamber, the density in that volume is reduced, the volume increases, and sound waves are generated which propagate into the surrounding medium. According to the physical and chemical properties of the background medium and the amount and time history of the energy deposition, these waves will either attenuate, amplify, or form into shocks.

Until recently, all of the theoretical studies of the properties and effects of such waves used either linearization or perturbation methods which restricted their validity to small amplitude sound waves or weakly dispersive background media. For example, the early work of Chu (1955) used a linearization technique to address two aspects of this local heat addition problem: how strong are the pressure waves generated and when do they transition to shocks. By relating the problem of a moderate heat release in a tube to that of two pistons, he calculated pressure, density and temperature fields. Some extensions of these ideas to large energy release were possible, but requied an exact solution and so were only obtainable in rather idealized geometries. Other authors have analyzed the propagation of one-dimensional waves in various media. A review of pressure wave propagation through real gasses has been given by Lick (1967). Later Blythe (1969) described propagation of low amplitude waves in a weakly dispersive medium, and Chu (1970) examined weak nonlinear waves in nonequilibrium mixtures.

In general, as pressure waves propagate through a medium, they can decay due to expansion, damp due to frictional forces, or disperse due to non-equilibrium chemistry. The pressure produced by adding heat depends on the

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rate of heat release, and the time rate of change of the heat release is responsible for generating sound waves. As pressure waves propagate through a nonequilibrium medium, their shape changes in time, producing frequency dispersion. When the amplitude of the wave is large, there are nonlinear effects leading to mode-coupling, which we can call amplitude dispersion. Pressure waves generated from heat release are altered by a competition between both effects.

Perhaps the most uncomplicated observation of the direct effects of pressure waves generated by energy release have been in both incident and reflected shock tube experiments. (See, for example, Meyer and Oppenheim (1970), Strehlow and Cohen (1962), or Edwards et al. (1981). Here ignition starts at or near the contact discontinuity or the reflecting wall. Pressure waves which move forward into unignited material produce fluctuations in the material not yet ignited. These waves have negligible effect in some mixtures, but in others they can accelerate the shock, initiate flames, and generate turbulence. Such behavior has been studied by Oran et al. (1982) and Kailasanath and Oran (1983). By numerically solving a full set of conservation equations coupled to a detailed representation of the chemical kinetics, they were able to consider large amplitude waves in highly dispersive, very reactive materials and thus carry the solution into the nonlinear regime.

In general the problems in exothermic materials are highly nonlinear, dispersive in both amplitude and frequency, and occur in nonidealized environments. Thus we feel that because of the theoretical restrictions, most of the analytical work to date has qualitative importance but it certainly cannot tell the whole story. When a highly reactive and exothermic mixture

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burns it generates a spectrum of pressure waves moving in all directions. First, there is the possibility that the waves generated could couple directly with and thus amplify the natural frequencies of the chamber. However, those waves moving towards previously reacted material probably decay, whereas those moving towards the unreacted material could heat or perturb unreacted material enough to reduce the ignition time. The tendency of the flame zone to expand might be halted as expansion fans catch up with the front. The coupling mechanism whereby pressure waves enhance reaction rates is discussed below in more detail.

The Influence of Energy Release on Sound Waves

Given a prescribed energy change, we ask how the propagation of sound waves is altered. In particular, the question we are asking is under what conditions will the sound waves be amplified, attenuated or be influenced by dispersive effects. This question was first addressed by Lord Rayleigh (1941) who considered how oscillations are driven by a periodic heat release. He concluded that it is important when the energy is released in the cycle of the sound wave. For example, the amplitude will increase when heat is added in phase with the pressure, and decrease when it is added out of phase. Also, the frequency is raised if heat is added at a quarter period before the pressure peak, and lowered if heat is added a quarter period after the pressure peak. A description of the Rayleigh criterion is given by Putnam (1964a).

The studies by Einstein (1920), Chu (1958), and Clarke and McChesney (1964) showed that sound waves propagating in a mixture which is slightly

perturbed from equilibrium will always be attenuated. However, the more interesting and relevant problem concerns the changes in sound waves as they propagate through a nonequilibrium, chemically reacting mixture. Studies of the acoustic properties of such media have been done primarily by Toong and associates and by J. F. Clarke. In addition, there is the complementary work by Gilbert and associates and Blythe.

The earliest work by Clark (1973, 1974) and Srinivasan and Vincenti (1975) considered the propagation of sound waves through systems which initially were not in equilibrium. Clarke (1973, 1974) primarily studied the response of diffusion flames to sound waves. Srinivasan and Vincenti (1975) were interested in the acoustic effects due to disequilibrium in the vibrational and rotational states of the ambient gas. The result of these studies is that under proper conditions, due to the disequilibrium in the background, a disturbance is amplified.

Later work by Clarke (1977, 1978a and b, 1979, 1981) and Blythe (1979) considered propagation of sound waves through spatially uniform but explosive mixtures. The amplifying and accelerating effects of energy release were noted. Another issue which is less relevant to our problem is whether the sound waves would steepen into a shock before thermal explosion of the whole mixture.

Toong and associates (Toong (1972), Toong et al. (1974), Garris et al. (1975), Patureau et al. (1977), Abouseif et al. (1979)) showed that when sound waves propagate through nonequilibrium, chemically reacting mixtures, they can be amplified or attenuated depending on the details of the coupling between them and the chemistry. They showed there were two kinds of effects:

one type in which there are mean changes in the averaged background state and the other type due to fluctuations in reaction rates. The relative importance of these effects depends on the order and enthalpy of the reaction, the activation energy, and on the ratio of a characteristic chemical to acoustic time, Ω. For example, for a zeroeth order reaction and a large value of Ω, i.e., relatively slow chemistry, the main effects are from fluctuations in the chemical rates due to variations in external conditions imposed by the sound waves. Whereas for very low frequency waves or constant reaction rates, the mean changes due to chemistry are most important. The conditions leading to sound wave amplification in exothermic materials are reaction rates increasing with temperature, a reaction order greater than zero, and a large activation energy. Conversely, a constant chemical rate, order equal to zero, or small activation energy promotes attenuation. When the amplification becomes large, nonlinear coupling effects become important and much of the classification of possible interactions breaks down.

The studies by Patureau et al. (1977) and Abouseif et al. (1979) are particularly interesting because the authors have set up an experiment to test a number of the conclusions of the theoretical analyses mentioned above. These experiments used a premixed H_2-Cl_2-Ar mixture in which reactions were initiated at a set time by ultraviolet radiation. The test of the large Ω limit validated the theories. They also observed very large amplification when Ω become small, indicating that nonlinear effects were becoming important and the theory was breaking down.

Gilbert et al. (1972, 1973) used linearization techniques and WKB solutions to study acoustic effects in transient chemical reactions. They

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focussed on the case of large Ω for a system far from equilibrium. Their conclusions were generally in agreement with Toong. Since their emphasis was primarily on the effects on chemistry, discussion of this work is in the next section.

The most important conclusion from the work of Toong et al., Clarke, Gilbert et al., Blythe, and Srivanison and Vincenti is that for large values of Ω, i.e. for long chemical times compared to acoustic times, there can be an amplification of the sound waves due to coupling to the energy release process. When Ω becomes small, that is, the characeristic times become more comparable, the coupling becomes very strong and nonlinear processes dominate.

The Influence of Sound Waves on Chemical Reactions

The presence of sound waves can stimulate chemical reactions, oscillations, and instabilities by creating fluctuations in the temperature and pressure and thus locally altering the reaction rates. This effect has been examined theoretically by Gilbert et al. (1972, 1973) and Toong and associates (Toong (1972), Toong et al. (1974), Garris et al. (1975)) using analytic perturbation and linearization methods and by Oran and Boris (1982) using numerical simulations.

Gilbert et al. (1972, 1973) examined the frequency dispersion and amplitude changes for a single-step unimolecular decomposition or conversion, $A \rightarrow B + C$ or $A \rightarrow B$. They then suggested that it would be possible to determine the pressure and temperature dependence of chemical reaction rates by measuring the rate of change of the amplitude and frequency of sound waves

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during the period of energy release. This is similar to the idea proposed by Oran and Boris (1982) in which sound wave perturbations of chosen frequencies would be used to calibrate the sensitivity of reactive mixtures to fluctuations. They also suggested using sound waves to enhance and then measure radical concentrations which would ordinarily be below instrumental sensitivity. Then unperturbed concentrations could be inferred.

Most of the work mentioned up until now has looked at rather idealized problems which involved a one-step Arrhenius reaction rate and rather simplified fluid dynamics in order to obtain either analytical solutions or otherwise simple forms to solve. Oran and Boris (1982) performed numerical simulations which solved the set of coupled partial differential equations coupled to a complete set of ordinary differential equations describing the chemistry of H_2-O_2 reactions. Two important results came from this work. The first is that the presence of sound waves can significantly alter the chemical kinetics even before there is any significant energy release, i.e, in the induction period. The mechanism here is to produce variations in temperature and pressure consistent with a constant entropy condition. The second conclusion is that when chemical induction times and acoustic periods are close, there can be a significant decrease in the induction time itself. Mixtures containing H_2 will in fact exhibit a range of pressure and temperature in which the chemical induction time is extremely sensitive to sound wave perturbations. In this regime sound waves can decrease the induction time by orders of magnitude. If an analogous effect occurred in the ramjet fuel, it could provide a mechanism for strong coupling of energy release to acoustic modes.

The Interaction of Sound Waves and Flames

It is well known that sound waves interacting with flames can trigger oscillations in flames. The interaction of pressure waves and a flame can create a system of transmitted and reflected waves which reflect from each other and various walls or obstacles in the system. These waves interact again with the flame and, when conditions are favorable, feedback effects can lead to oscillations of considerable amplitude. Gaydon and Wolfhard (1979), in their review of the early work on premixed flames, have said:

> "It is important to realize that the energy imparted to the flame by the sound is very small compared with the energy release in the combustion, and unless the sound can trigger some instability, such as the vortices in the sensitive flames, we should not expect sound to produce any marked change. However, although the energy contribution is small, the amplitude of the movement due to sound is not so small, and there are some interesting changes in flame shape."

They also note that the presence of sound waves narrows the region of conditions in which a flame is stable. However, the mechanisms primarily responsible for these effects are not directly connected to the chemical release; they are interactions between physical properties such as the flow field and flame area. The heat release provides the source of energy and defines the flame front. Acoustic-flame interactions in premixed flames have been studied by Putnam and Dennis (1953 a,b), Kaskan (1953), Blackshear (1953), Bailey (1957) and Chu (1956). Good reviews of these and other work are given by Gaydon and Wolfhard (1979) and Putnam (1964b).

There have also been studies of acoustic interactions with diffusion flames by Toong et al. (1965) and Clarke (1973). Clarke has shown that, given the right combination of activation energy and heat of reaction, instability can occur. Toong and associates have attributed the oscillations

they see to Tollmien-Schlichting waves. What must be resolved is whether these oscillations can occur at frequencies important to ramjet combustion.

Conclusion and Discussion

Considering the complexity of the inter? ion between fluid dynamics and chemistry it is important to establish the level at which research is needed in chemical-acoustic coupling. Below we list some of the questions important to chemical-acoustic interactions and make several recommendations for further work.

Pinpointing the important time and space scales for a particular system is required for deciding what and when effects are important. The question of which are the relevant time scales to use when comparing chemical and acoustic effects is not resolved. A characteristic acoustic time is usually defined as the sound speed divided by a characteristic length. However, in a ramjet the sound speed varies theroughout the chamber due to temperatures and composition variations. The choice of length scale is also important: the transverse or radial length of the material or combustor or some combination for unusual geometries. While in general these timescales can be calculated, they will vary for each system configuration. Methods of determining these modes need to be found.

There are at least two chemical times to consider: the chemical induction time of the fuel and the time for energy release. We know that for H_2-O_2 mixtures, sound waves can drastically alter both of these times. Changes in induction time are subtler and not as expected. The effects of sound waves on both of these time scales should be evaluated for other materials.

This should involve experiments, perhaps along the lines of Abouseif et al. (1979), as well as simulations, perhaps similar to those of Oran and Boris (1982).

Also, if the mixture contains enough droplets which have not yet fully evaporated, we need to consider evaporation and perhaps diffusion times. In this case we might also have to consider acoustic-flame interactions for diffusion as opposed to premixed flames.

It is important to carry the coupling analyses into the nonlinear regime, consider chemical mechanisms more complicated than one Arrhenius rate, and to consider multi-dimensional effects not present in planar geometries. A part of this requires numerial computations. However, the analytic theories provide insights which are crucial for defining important sales and understanding the physics in the simplest terms. They must also be used for benchmarking numerical calculations which can represent more complicated physical and chemical processes (see, for example, Kailasanath et al. (1982) and Abouseif et al. (1979)).

More experimental work looking specifically at the mechanism of chemical-acoustic coupling is important. The only such work we have been able to find is by Patureau et al. (1977) and Abouseif et al. (1979). It would be particularly enlightening to perform more studies of the behavior of a sound wave of given initial amplitude and frequency in a mixture whose stoichiometry is varied within the flammability limits. It would also be instructive to study how sound waves effect both propagating flames as well as mixtures which are uniformly ready to react.

Finally, there have been two types of technological breakthroughs in the last twenty years that we should exploit in this problem. The first is the advance in diagnostic techniques: CARS, LIF, LDV, etc., which will allow us to examine both the chemistry and flow fields in more detail. The second is the advance in computational power which will allow is to model more complicated, nonlinear problems that may be intractable analytically.

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