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Utilization of High-Density Strained Hydrocarbon Fuels for Propulsion

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To increase the range of liquid-hydrocarbon-fueled ramjets and cruise missiles, a new class of high-density strained hydrocarbon fuels has been synthesized and characterized. By virtue of the increased density and additional strain energy introduced during synthesis, these fuels have increased the volumetric heat of combustion. The oxidation chemistry and short-term and long-term stability for storage of these fuels have been studied. Because of the high carbon-content-to-hydrogen ratio of the molecules, there is an increased tendency for sooting. Several methods have been identified to eliminate this problem. These include sequential timed fuel injection, retarded fuel-stream velocities, and the use of additives. It has been shown that soot generation and transport can be controlled and that these fuels microexplode rapidly, thus offering potential for utilization in propulsion systems to obtain increased range.

Introduction

INCREASED range, speed, stealth, reliability, and reduced vulnerability are required of Navy propulsion systems. For a given volume of fuel used, increasing the combustion efficiency of the propulsion system can increase the range. Research has been pursued over the past couple of decades to achieve this. The increase in efficiency obtained by further improving mixing and combustion and combustion control will, however, be marginal. Navy propulsion systems are volume limited, and a more attractive and logical way to increase the range for a given propulsion system would be to increase the energy per unit volume of the fuel taken aboard. For liquid-hydrocarbon-fueled ramjets and cruise missiles, a new class of high-density strained hydrocarbon fuels has been synthesized.[1] These fuels, by virtue of their increased density, and the additional strain energy stored during synthesis, when properly burned in the combustion chamber, can substantially increase the range.

In conventional hydrocarbon fuels, the angle between the bonds is 109.5 deg and, by the open nature of the molecules, they are not packed densely. The strained hydrocarbon molecules studied are strained to form triangles, squares, and polygons, or strained further to form tetrahedrons and prisms (Fig. 1), thereby increasing the density and adding strained energy.[1] In Fig. 2, the gravimetric heat of combustion is plotted against the density of common hydrocarbon fuels, including other fuels synthesized elsewhere.[1] As the density of a conventional hydrocarbon increases, a decline in the gravimetric heat of combustion is noted. To take full advantage of this increase in density, the gravimetric heat of combustion should remain constant. However, the present fuels exhibit an increase in the heat of combustion with an increase in density, resulting in improved volumetric and gravimetric heat release. Consequently, improvement in range of volume-limited (greater extent) and weight-limited (lesser extent) propulsion systems is achieved. The synthesis aspects of these fuels have been reviewed by Roy.[1] Because of the strained nature of the fuels, storage and long-term shelf life are to be considered. The combustion of these fuels poses several problems, the most significant being a high propensity to sooting that is due to the high carbon content (carbon-to-hydrogen ratio) of the fuels. The other issues in the combustion of these fuels are the influence of the strain energy on the decomposition[2] process and any difference in its injection/atomization characteristics and spray and vortex dynamics compared with those of conventional liquid hydrocarbon fuels, liquid-phase reaction[3] vaporization, and microexplosion[4] characteristics. These issues are illustrated in Fig. 3.[5]

Early fuel-synthesis efforts focused on strained hydrocarbon fuels containing azido groups.[6] It has been shown that certain diazido compounds are effective in controlling the droplet burning rate and microexplosion characteristics and thus moderate combustion instability.[6] The idea behind this approach is that the combination of the inherent strained energy in the substrate fuel molecule coupled with the attachment of azido functionality could yield a superior fuel. The concept is called synergetic microexplosion, in the sense that microexplosion is coupled with exothermic strain release in the fragmentation of the strained hydrocarbon.[7] The most strained hydrocarbon is cubane (C_8H_8, Fig. 1), in which the strain energy is the result of cyclobutane rings fused together in the cubane molecule. The strain energy of cubane is 161 kcal/mole (Ref. 8). The other molecules considered, namely, dihydrobenzvalene (DHBV) (C_6H_8) and quadricycline (C_7H_8) have strain energies of 72.8 and 78.7 kcal/mole, respectively (Santoro, R., Private Communication, 1999).[9]

Storage Stability

Although these fuels were chosen for their higher density and strain energy, other chemical and physical properties, as well as short-term and long-term stability for storage, should also be considered for propulsion applications. The DHBV prepared by Moriarty[7] contained 20% benzene as the major impurity and other trace impurities. The results of a study on the effect of long-term storage[10] of DHBV are shown in Fig. 4. Here the lowest trace (original mixture) is a portion of the in infrared (IR) spectrum of a typical DHBV/argon matrix sample prepared immediately after the sample was received. The main peaks that are due to DHBV are labeled D, peaks that are due to benzene are labeled B, and the unlabeled peaks are either contaminants or, in some cases, a combination band from DHBV and benzene. The second trace (old mixture) shows the IR spectrum of a matrix grown from the same DHBV/argon mixture after storage for 3 months at room temperature in the dark. It can be seen that the DHBV has almost decomposed during storage. A mass spectrum of this decomposed mixture shows a significant intensity of mass 80, 79, and 77 as well as lower mass cracks, therefore suggesting that the decomposition was simply an isomerization, as this cracking pattern is very similar to that of DHBV. Another sample prepared by Moriarty was stored for a year and used for Fourier transform IR (FTIR) experiments. Although this sample also contained the same amount of benzene as impurity, no decomposition was noted.[11] The only observable difference between these samples is that the first was slightly colored, and the second was clear. It appears that the trace contaminants could possibly accelerate or even catalyze the

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decomposition of DHBV. It becomes necessary to identify such contaminants that can cause rapid decomposition of the fuels, and attention needs to be focused on synthesizing these fuels in a pure form.

Because these fuels are expensive to synthesize, it is necessary to characterize these fuels by use of very small quantities, and the not-so-promising candidate fuels can be eliminated. A unique instrument, a micro-flow-tube reactor/mass spectrometer (micro-FTRMS) has been designed and built to meet the requirement of analyzing small samples.[12] This instrument will allow the determination of those fuels that indicate promise for follow-up.

Droplet Combustion Studies

Because of the small quantities of fuels synthesized, spray combustion studies were initially carried out with surrogate fuels that have similar properties, whereas droplet combustion studies were carried out with fuels synthesized at different university laboratories.

To study the droplet burning characteristics of these fuels, a single monodisperse stream of droplets was axially injected downward into the postcombustion region of a premixed flat flame burner.[13] The droplet size could be varied from approximately 150 to 250 μm, and the chamber could be pressurized. The composition of the gaseous mixture fed to the burner was controlled to yield combustion temperatures of approximately 1000–1200 K. The amount of soot emitted per drop is shown in Fig. 5 as a function of a normalized time, defined as $(1 - d^2/d_0^2)$, where d is the diameter of the droplet and d_0 is the initial diameter, for JP-10, benzene, and a mixture of 20% benzene and 80% DHBV.[4] As the droplet burns, the amount of soot first increases and then decreases. As can be seen, the sooting propensity of DHBV mixture is approximately the same as that of benzene and approximately five times higher than that of JP-10.

Only very small quantities of methyl cubane were synthesized, and hence single suspended droplet experiments were conducted. The flame was characterized by intense sooting, and imaging of the droplet regression was not possible. Therefore additional experiments were performed[14,15] with mixtures of methyl cubane and n-nonane under reduced pressures (the boiling points of these two components are quite close so that preferential gasification effects can be eliminated). The burning rate constant K (the time rate of change of the square of the droplet diameter) for combustion at 0.25 and 0.13 atm is shown in Fig. 6. When the values of K for pure nonane and methyl cubane at 0.13 atm are compared, it is seen that methyl cubane burns at least twice as fast as nonane, which is a decisive advantage.

In these fuels, the liquid-phase reaction is most likely to occur in the relatively warmer portion of the droplet near the surface rather

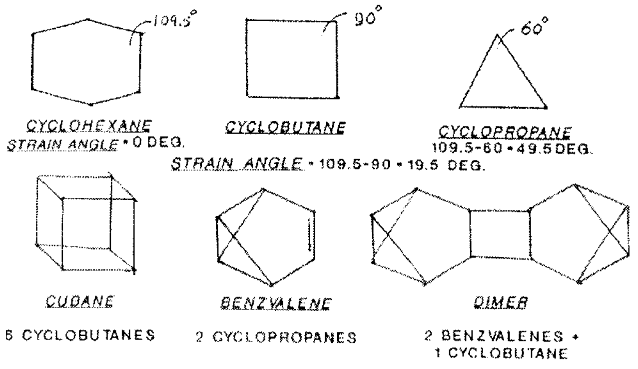

Fig. 1 Strained molecular hydrocarbons.

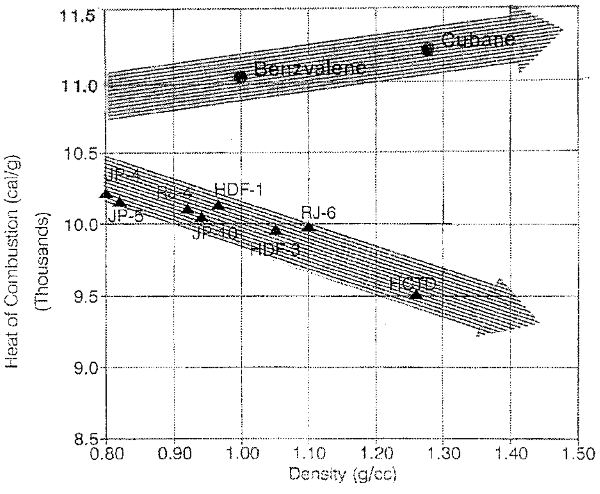

Fig. 2 Comparison of heat of combustion of various fuels.

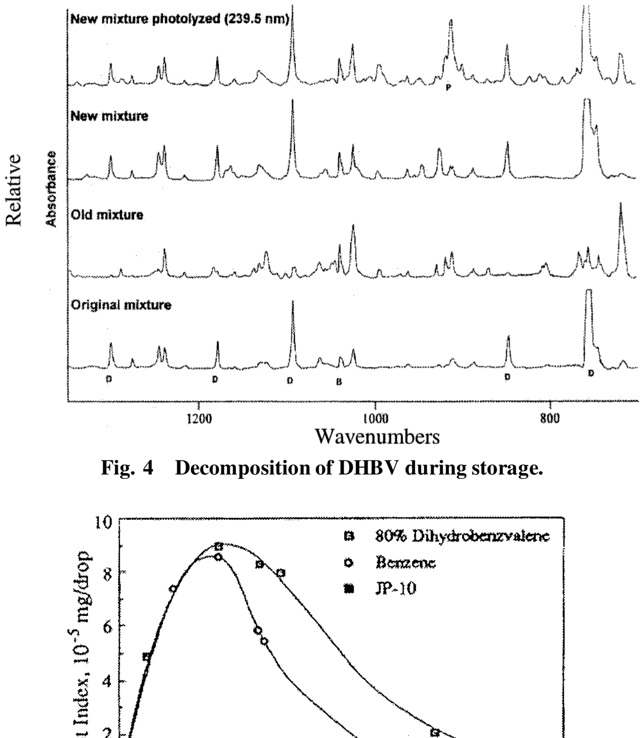

Fig. 4 Decomposition of DHBV during storage.

Fig. 5 Soot emission from high-energy fuels.

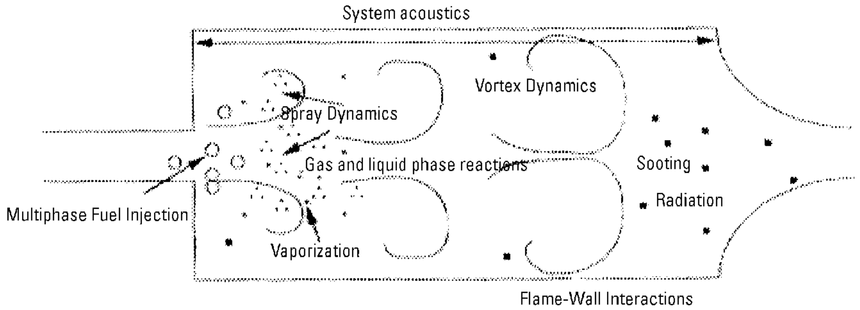

Fig. 3 Combustion issues.

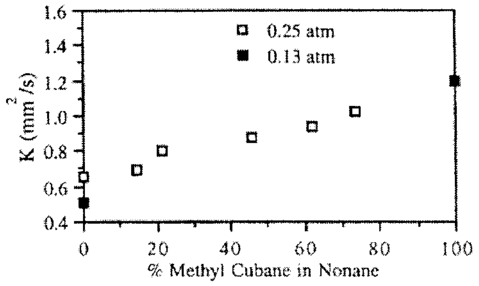

Fig. 6 Droplet burning rate constant *K* for mixtures of methyl cubane and *n*-nonane at reduced pressures.

than the center of the droplet. It is not likely that the liquid-phase reaction occurs in a very thin spherical shell around the edge of the droplet, which would require very fast reactions at relatively low liquid temperatures. Hence it is likely that this reaction occurs over a finite domain where liquid and gas exist. With a large content of gas, all of the gas cannot remain in solution, but rather bubbles could exist, and a bubbly region forms.[16] With this hypothesis, numerical analysis was performed. The analysis indicated that the bubble radius grows with radial position and time, causing the droplet to swell initially. A model incorporating decomposition of the droplet surface predicted that the droplet radius continuously increases with time until surface decomposition becomes appreciable. Afterward, the droplet mass and radius decrease with time.[16] However, experiments with certain diazidoalkanes did not show any bubbly foam formation at the droplet surface.[13] Although foaming combustion is an essential feature of solid propellant combustion, it is not clear that foaming occurs in liquid propellant combustion because of the much lower surface temperatures and smaller molecules compared with those of solid propellants. However, it could be due to the possibility that the resolution obtained in the experiment is not sufficient to identify this very thin region.

Soot Studies in Diffusion Flames

Several investigations were carried out with different methodologies to minimize or eliminate soot that forms during combustion of these energetic fuels. A variety of methods have been tried in the past to achieve rapid mixing between the fuel and oxidant-rich streams, which is known to be effective to control soot. However, the way in which the mixing is carried out is also very crucial. Existing evidence from both turbulent and laminar jet flames and from empirical industrial practice indicates that soot reduction in diffusion flames can be achieved if the fuel-stream velocity is retarded. This is schematically shown in Fig. 7, in which the hydrodynamics of nonpremixed flames is sketched.[17] The soot-containing regions are sketched for conditions under which fuel-stream velocities are larger (enhanced) and smaller (retarded) compared with those of the air stream. Because of their size, the soot particles do not diffuse like gas molecules, but are convected at the local velocity of the flow field. In the case of enhanced fuel-stream velocities, entrainment caused by the higher-velocity fuel stream causes streamlines to cross the flame toward the fuel side. Consequently, soot particles nucleate mainly near the fuel-rich side of the flame sheet and are convected away from the flame surface. This results in long residence times at fuel-rich conditions, and soot particle growth is enhanced. These particles are difficult to oxidize completely when they reach the fuel-lean portions of the flame. Thus the potential for soot emissions is increased. On the other hand, with retarded fuel-stream velocity, entrainment from the higher-speed air stream causes streamlines to cross the flame toward the side of air. The soot particles then begin to nucleate in the cool core of the flow and are drawn immediately toward the oxidation zone. Because they do not have sufficient time to grow in the fuel-rich region, they are readily oxidized. Thus retarding the fuel velocity offers the potential of reducing soot emission from the high-sooting fuels under consideration.[18]

In diffusion flames, the mixing occurs in two stages. In the first stage, large amounts of the reactants are brought together through large-scale mixing, dominated by vortex dynamics, and in the second stage, molecular contact between the reactants is promoted by small-scale turbulence. Minimizing locations of unmixedness is important to reduce soot formation that is due to localized high fuel-to-air concentrations. Hence synchronized periodic injection of fuel into air vortices at a timing that ensures energizing of the vortices and homogeneous mixing was investigated in another effort.[19] For a certain time delay, islands of unmixed fuels were generated inside the core of the air vortices, leading to high soot formation, whereas for other time delays, a homogeneous mixture of air and fuel was promoted, reducing the soot level nearly to zero. Simultaneous planar laser-induced fluorescence (PLIF) of OH and Mie scattering images of soot was used to identify the relationship between the initial soot formation, the vortical structures, and the fuel-injection locations (Fig. 8). It was shown that dual fuel injection during a single air-vortex period can reduce the soot formation by several orders of magnitude relative to an unforced flame (Fig. 9). Further, significant improvement in the combustion energy release was also obtained. The first fuel injection, which is synchronized with the formation of the incipient vortex, mixes homogeneously and initiates intense combustion in the vortex as it is convected downstream. The second injection penetrates the jet and brings the fuel into the lean combustion products of the prior vortex. The process therefore simulates staged combustion without mechanical stages. This methodology has been applied to practical systems, resulting in efficient soot-free combustion, higher temperatures, and an increase in energy release.

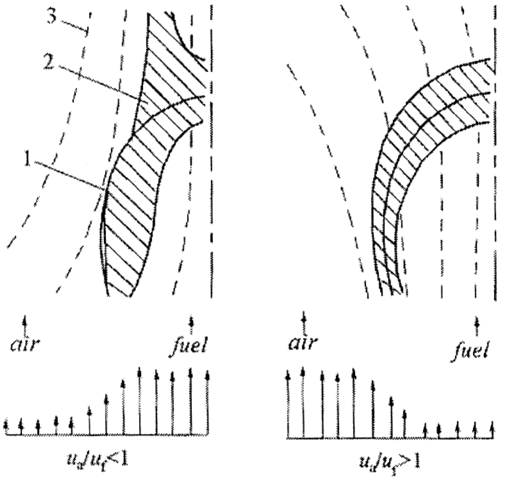

Fig. 7 Effect of reactant stream velocities on soot processes within laminar mixing layers for nonpremixed hydrocarbon-fueled flames. 1, flame sheet (typical); 2, soot layer (typical); 3, streamline (typical).

The control mechanisms developed in gaseous-fueled flames were applied to liquid-fueled flames.[20] It was found that proper atomization was critical to the success of this technique. Although the fine component of the spray follows the vortical flow and participates in the control, larger droplets, because of their inertia, travel through the vortices and burn in a sooty flame above the region of vortex combustion. To guide the droplets into the desired spatial location, the interaction between fuel droplets and large-scale temporal features in the flow (vortices) was utilized. By synchronizing the fuel-injection process with a phase-delayed pressure signal, it was possible to control the timing of fuel injection precisely with respect to the air-vortex shedding process. The droplet trajectories were significantly affected by this timing. Droplet dispersion characteristics of pulsed sprays at different timings are shown in Fig. 10.[21] The improvement of spray characteristics was attempted through custom-built atomizers.

The fuel-additive approach has long been used to control soot emissions and was investigated in the control of soot of the strained high-energy fuels.[22] The physical considerations in the choice of additive are temperature effects that are due to the increased heat capacity of the additive, dilution by a less energetic additive, differential diffusion that reduces the concentration of soot precursor species, and reduced thermal diffusivity that results in smaller residence time in the soot growth region. Experiments were conducted with benzene (C_6H_6) and quadricyclane (C_7H_8). Because of the heavy sooting characteristics of these fuels, a fuel mixture approach

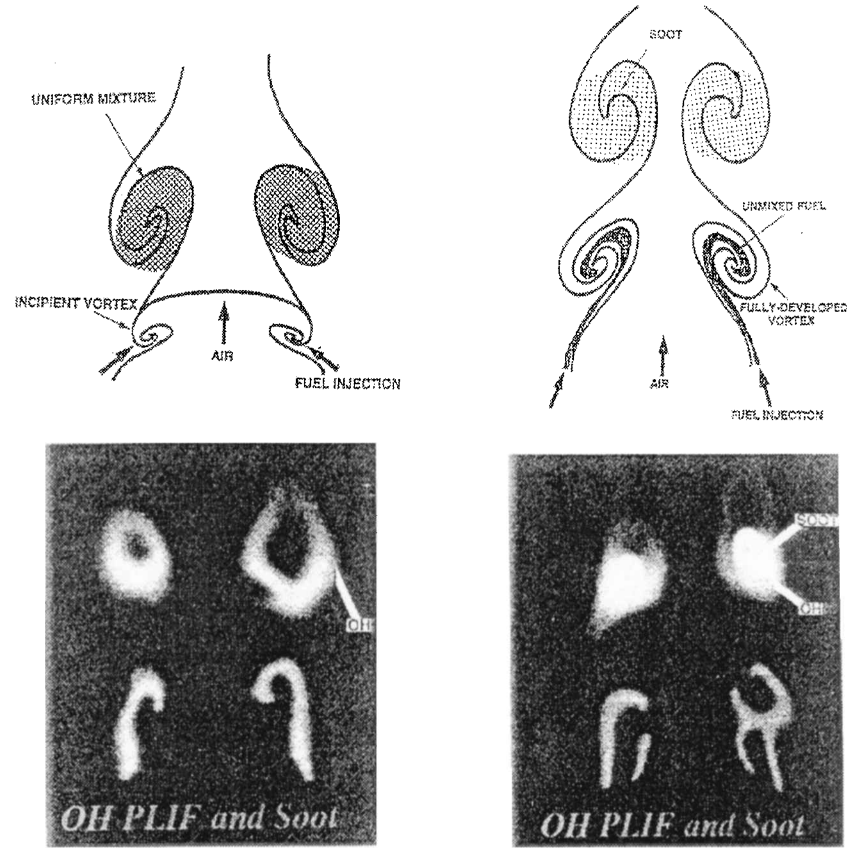

Fig. 8 Soot generation with and without controlled fuel injection.

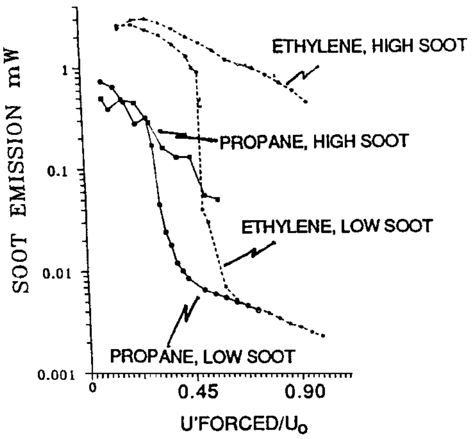

Fig. 9 Comparison of soot emission in uncontrolled and controlled ethylene and propane flames for different forcing levels.

was adopted with methane as the base flame. Figure 11 indicates that quadricyclane has a higher propensity to sooting than benzene. After methanol addition, it was observed that the soot volume fraction (f_v) is reduced significantly for heights at or below 20 mm. However, higher in the flame, soot reduction is comparatively lower. CS_2 was found to be very effective in reducing the sooting nature of these fuels. A 60% soot reduction was observed with the addition of 25% CS_2 (Fig. 12). Here, it is expected that most of the sulfur atoms in CS_2 are converted to SO_2, which subsequently form the intermediate HSO_2. This catalyzes H, OH, and O radical combination reactions, thereby reducing their concentration close to their equilibrium values. As polycyclic aromatic hydrocarbon (PAH) formation and growth are sensitive to H radical concentrations, such effects can significantly affect net soot formation.

The advances in supercomputing and parallel processing have made it possible now to perform detailed calculations of complex multiphase reacting flows.[23] As a complement to laboratory experiments, numerical simulations were performed[24,25] both to obtain further understanding of the mechanisms involved and to perform detailed parametric evaluation. In contrast to physical experiments, numerical experiments have the advantage of changing any desired parameter at a time. Simulations were made with timing the particle (which simulates fuel) injection in or out of phase with the forcing at the shedding frequency. Dispersion increased when injection was in phase with the forcing because the particles traveled downstream with the flow structures. Decreased dispersion was observed when injected out of phase. These simulations also demonstrated that, by choosing suitable forcing frequency and timing the fuel injection, one could achieve enhanced dispersion of fuel droplets at different locations in the combustor, depending on the application. Simulations were also performed to determine the effect of particle size on its distribution in the vortical structure (Fig. 13). These results can be utilized as the basis for the design of atomizers with optimum performance.

A study on electrostatic dispersion and evaporation of dense and dilute clusters of drops was undertaken to determine the effectiveness of this technique in soot control.[26] The effects of mechanical dispersion on electrostatically charged as well as uncharged polydisperse clusters of drops were numerically investigated. It has been shown that drop-induced mechanical dispersion cannot achieve the same beneficial effects as electrostatic dispersion in terms of reducing the mass fraction of the evaporated fuel and gas density (to reduce soot nucleation while promoting evaporation).

From the comparison of the above methodologies, it is evident that synchronized fuel injection seems to be the most promising. The rapid development in microelectromechanical systems, sensors, and actuators would make active control of fuel modulation in real systems possible. Research is underway to achieve this goal. Perhaps a hybrid technique involving more than one control methodology may be the solution for utilization of these fuels, in compliance with external constraints as well.

High-Energy-Density Solid Hydrocarbons

Pentacyclic alkene dimers are high-energy-density (HED) hydrocarbons that exhibit attractive features for combustion systems because of their high density (to $\rho = 1.2$–1.3 g/cm^3). They contain

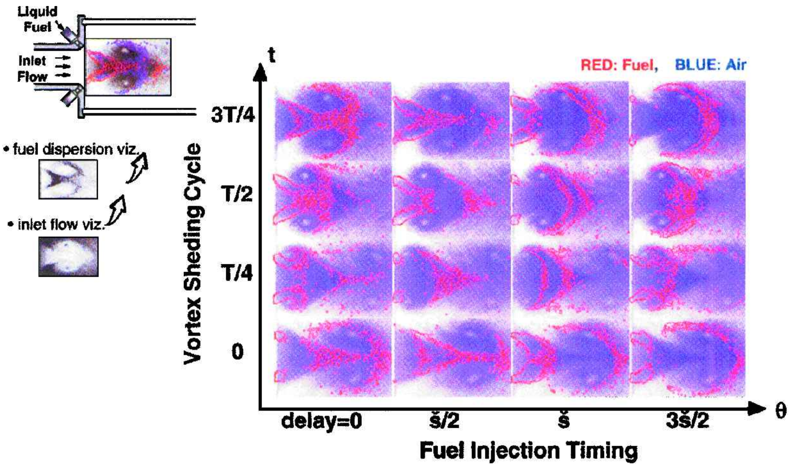

Fig. 10 Spatial distribution of ethanol droplets (points) as a result of interaction with a large air vortex (continuous lines).

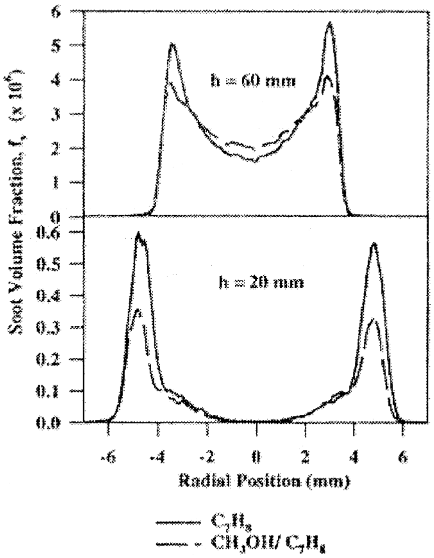

Fig. 11 Soot volume-fraction profiles in methanol-added quadricyclane flames for measurement heights *h* of 20 and 60 mm.

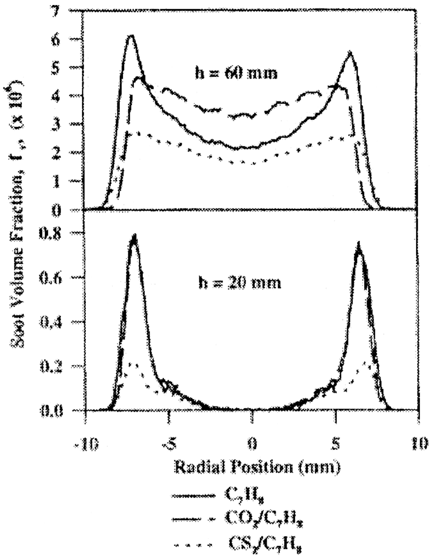

Fig. 12 Soot volume-fraction profiles in CS_2-added quadricyclane mixture flames for measurement heights *h* of 20 and 60 mm.

a moderate amount of strain energy that contributes to the energy output during combustion. In addition, they dissolve in large portions in existing liquid-hydrocarbon formulations, forming stable solutions to 18% w/w (Ref. 27). To assess their application to air-breathing propulsive devices, investigations have been focused in two areas: 1) evaluation of combustion characteristics and, 2) modeling of the solid–gas interface of HED fuels in a turbulent reacting flow. Droplet combustion experiments with binary solutions of methylated pentacycloundane (MPCU) dimer in JP-10 indicated high heat output of the binary system, doubling the pure JP-10 burning rate. Evaluations of liquid HED formulations based on pentacyclic cages, as individual compounds, exhibited microexplosive combustion and burning rates comparable with those of azido-organic compounds.[27] Although the pentacycloundane(PCU) dimer and spirocyclic alkene dimers dissolve in only a small quantity in liquid hydrocarbons, the MPCU alkene dimer dissolved in the proportion of 22% w/w in a realistic additive to existing fuel systems.[28] Products resulting from the chemical decomposition of HED compounds were analyzed with a quantitative measurement of the combustion products of MPCU by FTIR spectroscopy. Figure 14 shows the increase in the burning rate 2 > 2 of an 18% solution of a MPCU alkene dimer [$C_{22}H_{22}(CH_3)_2$] in JP-10 (Ref. 29). Further, a new synthesis path produced a mixture of MPCU isomers with increased solubility in JP-10. Solutions of 50% w/w have been obtained with stability at low temperatures verified down to 70°C. The 50% w/w mixture increased the JP-10 burning rate by a factor of 5 (Ref. 30).

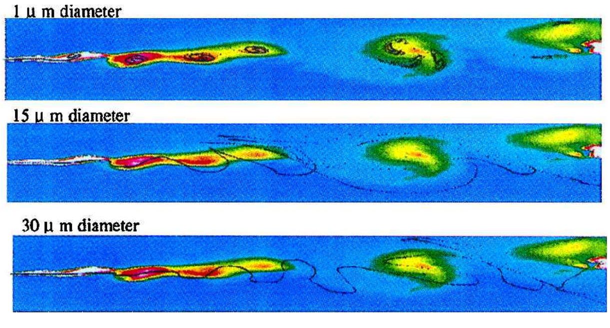

Fig. 13 Vorticity and particle distribution.

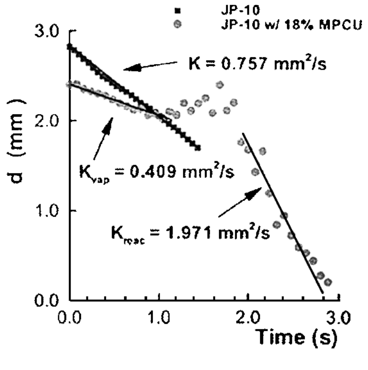

Fig. 14 18% solution of MPCU alkene dimer, $C_{22}H_{22}(CH_3)_2$, in JP-10 indicates a burning rate increase by a factor of >2.

Conclusions

A new class of high-density hydrocarbon fuels with strained molecules has been synthesized and characterized. Their combustion characteristics have been investigated to assess their suitability for utilization in the propulsion systems for ramjets, cruise missiles and possibly for aircraft. Systems studies indicate a substantial increase in the range of vehicles using these fuels for propulsion. However, there are several scientific issues that have to be resolved before these fuels can be utilized. Small quantities of various candidate fuels have been synthesized, and their properties, oxidation chemistry, and reaction pathways have been characterized. Droplet combustion studies indicate a propensity in soot formation; however, these fuels possess good microexplosion and fast burning properties. An innovative approach of sequential fuel injection with respect to the vortex generation in the mixing layer has resulted in orders-of-magnitude reduction in soot. Other methods investigated include control of velocities of fuel and oxidant and the addition of fuel modifiers. These fundamental studies indicate that this class of fuels shows promise for longer range and speed of propulsion systems for the given tank volume. Efforts have been undertaken to produce large quantities of these fuels, as well as to demonstrate the utilization of these fuels in real systems. Although the number of steps involved in the synthesis of these fuels has been reduced through research, the cost would be still far above what commercial applications would dictate because of the higher price of starting materials. However, as with most new products, continued research, alternative starting materials, improvements in the processes involved, and large-scale production will bring the fuel price down. Studies should also focus on the environmental impact in using these fuels. Each application could have specific issues to be addressed. Experience has shown that the development of new fuels takes several years. In the near term, these fuels may be added to conventional fuels to enhance performance and range. However, in the long range, the payoff from these fuels will be substantial.

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