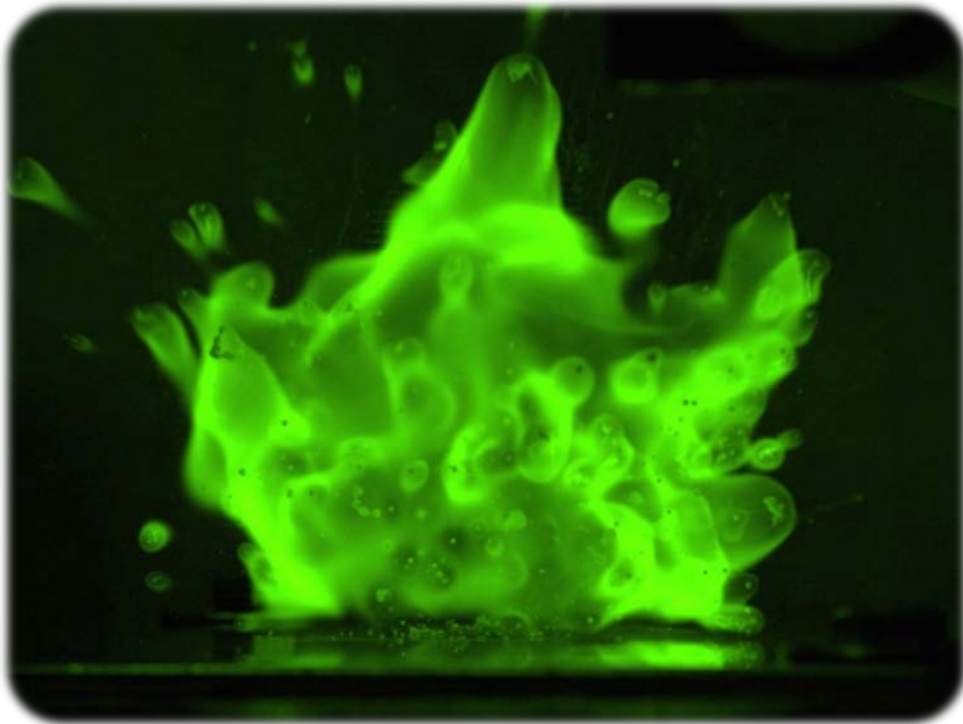# Hypergolic Ignition of Kerosene Gel with Hydrogen Peroxide for Rocket Motors

Research Project

Roni Goldin

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Nomenclature

Introduction

The present review deals with hypergolic gel propellants. Some terms should be defined [1]:

Gel is a fluid whose rheological properties were altered by gelling agents (or gellants) so that it behaves as a semi-solid: When there's no stress applied it is a solid, and under shear stress, it acts like conventional liquid. In 1990, Brinker and Scherer provided a definition of gel as "a substance that contains a continuous solid skeleton enclosing a continuous liquid phase. The continuity of the solid structure gives elasticity to the gel."

Gel propellant is a fuel or oxidizer that behaves as a solid at rest and can be atomized and combusted like conventional liquid fuel after applying shear stress.

A **hypergolic propellant combination** (usually consists of a fuel and an oxidizer) is one whose components spontaneously ignite when they encounter each other.

1: MMH-HPC (97:3)
2: Water-HPC (91:9)
3: Jet A1-Xylene-Thixatrol ST (85:7.5:7.5)
4: ISROsene-Xylene-Thixatrol ST -micro-Alumina (84:7.5:7.5:1)
5: Kerosene-Fumed Silica-PST (74-61.7:3.4-6.6:0.7-0.9)
6: Kerosene-nano-Al-Fumed Silica-PST (64.4:29.3:5.5:0.8)
7: RFNA-Fumed Silica (97:3)
8: MMH-Aerosil (94:6)

Figure 1 – Physical appearances of gel fuels, oxidizers, and simulants.

Motivation

The main reason for using gel propellant is that it combines the benefits of both solid and liquid propellants [2–4]:

- Storability and Safety - Reduces leakage and spill hazard when stored. In addition, it is insensitive to shock, impact, friction, and electrostatic discharge, preventing phenomena such as detonation, explosion, or deflagration to occur. Moreover, the volatility of gels is significantly lower than the volatility of liquids and in case of leak or spill, much less vapors will be released, thus reducing toxicity hazards.
- Packing - Flexible packing, like liquids.
- Thrust Modulation - Allows control of the mass flow rate ($\dot{m}$).
- Metallization – Suspension of energetic nano-metal additives, which increases the specific impulse (I_{sp}) and the Density impulse.

There are also great advantages for hypergolic propellants, such as simplicity and reliability because there is no need for ignition system, due to the spontaneous ignition of the combination upon contact. (The definitive property of hypergolic reactants is that they do not require a hot ignition source to initiate the chemical reaction) [5,6].

Moreover, hypergolic propellants are preferred for several rocket propulsion applications, mostly when multiple ignitions are demanded for a mission success [7].

Rheology

Rheology is the science of deformation of material. The rheological properties of a gel control its combustion, atomization, and flow characteristics. These properties depend on the chemical structure of the gel.

Non-Newtonian fluids, in general, and gels in particular, can be classified according to their rheological properties. A schematic classification can be seen in Figure 2.
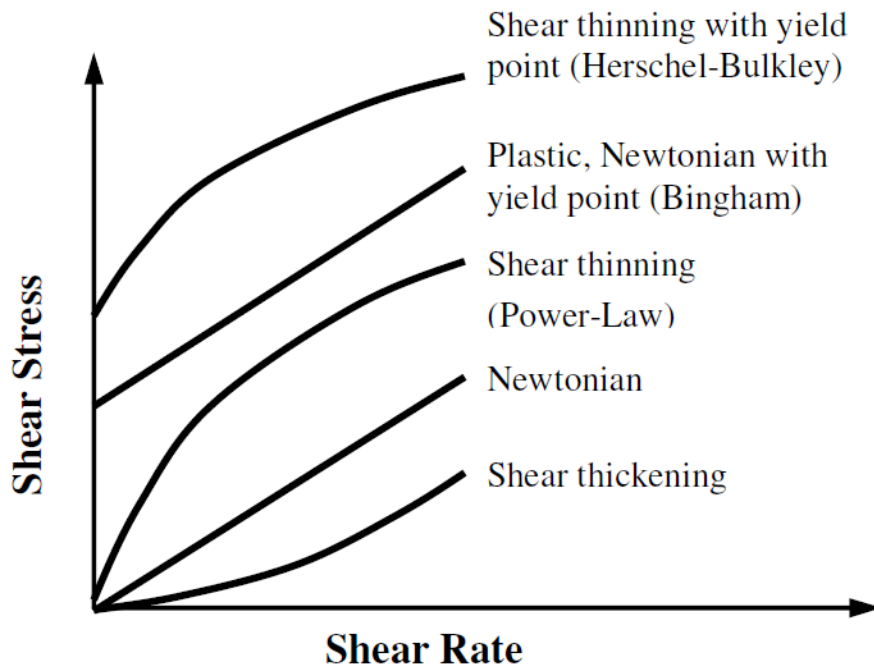
Figure 2 - Schematic classification of Newtonian and non-Newtonian fluids.

For a general viscous fluid, the stress tensor ($\boldsymbol{\tau}$) depends only on the rate of deformation tensor ($2\mathbf{D}$), and the relation is described by:

$$\boldsymbol{\tau} = \eta \cdot 2\mathbf{D} \tag{1}$$

Where η is the non-Newtonian viscosity.

A constitutive equation is a fundamental relation between force and deformation in materials, primarily liquids. Several constitutive equations are being used for gels, including:

Power law model (PL):

$$\tau = K \cdot \dot{\gamma}^n \quad or \quad \eta = K \cdot \dot{\gamma}^{n-1} \tag{2}$$

Herschel-Bulkley (HB):

$$\eta = \eta_0 + K \cdot \dot{\gamma}^{n-1} \tag{3}$$

Carreau-Yasuda (CY):

$$\tau = \eta_\infty \dot{\gamma} + \frac{(\eta_0 - \eta_\infty)\dot{\gamma}}{[1 + (\lambda\dot{\gamma})^2]^{\frac{1-n}{2}}} \quad or \quad \eta = \eta_\infty + (\eta_0 - \eta_\infty)[1 + (\lambda_{CY}\dot{\gamma})^a]^{\frac{n-1}{a}} \tag{4}$$

Herschel-Bulkley-Extended (HBE) [8]:

$$\eta = \eta_0 + K\dot{\gamma}^{n-1} + \eta_\infty \tag{5}$$

Casson:

$$\sqrt{\tau} = \sqrt{\tau_0} + \sqrt{\eta \cdot \dot{\gamma}} \tag{6}$$

Bingham:

$$\tau = \tau_0 + \eta \cdot \dot{\gamma} \tag{7}$$

The indexes $0, \infty$ represent the state at rest and at infinite shear rate respectively.

n is the flow behavior index [9]:

- In **Newtonian** fluids, viscosity is independent of the shear rate ($n = 1$).
- In **shear thinning** fluids, the viscosity decreases with increasing shear rate ($0 < n < 1$).
- In **shear thickening** fluids, the viscosity increases with increasing shear rate ($n > 1$).

Certain non-Newtonian fluids exhibit time-dependent behavior:

- **Thixotropic** - viscosity decreases under constant shear rate.
- **Rheopectic** - viscosity increases under constant shear rate.

All types of non-Newtonian fluids are summarized in the following table:

Table 1 - subcategories of non-Newtonian fluids.

Type of fluid	Special Properties	Examples
Viscoelastic	Material returns a well defined rest shape	Gels
Rheopectic	Apparent viscosity increases with duration of stress	Lubricants, whipped creams
Thixotropic	Apparent viscosity decreases with duration of stress	Paints, clays etc.
Shear thickening	Apparent viscosity increases with increased shear stress.	Sand in water
Shear thinning	Apparent viscosity decreases with increased shear stress.	Latex paint, ice
Bingham Plastic	Minimum stress needed to flow	Toothpaste, margarine

Some fluids also have yield stress. Gels for applications in rocket and air breathing propulsion systems can be designed, by choosing the appropriate gelling agent, to have a yield point in order to enable the suspension of particles. The existence of yield stress assures that particles can be added without the effect of sedimentation.

Gel propellants are usually power-law, shear-thinning, thixotropic fluids.

Atomization and Droplet Breakdown

In a liquid\gel rocket motor, atomization is a key parameter for combustion. The goal is to get droplets as small as possible, because this will enhance vaporization, mixing and combustion of the fuel with the oxidizer.

Atomization is the process whereby bulk liquid is transformed to drops. This process goes through the breakup of the jet into a number of filaments, which in turn transform into droplets. The atomization process requires applying a force to overcome the surfaces tension forces of the liquid. There are three mechanisms for it:

- Applying centrifugal forces using swirl atomizer.
- Using the surface tension between moving liquid jets and air in order to destabilize the jet and disintegrate it into filaments.
- Implementing outer supersonic acoustics or mechanical/electrostatic forces.
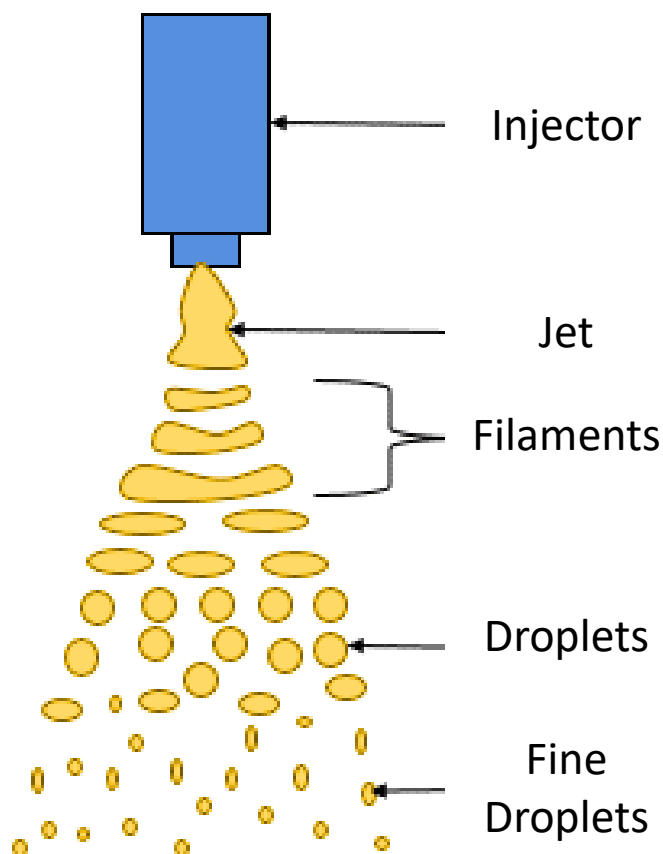
Figure 3 – Atomization process.

To describe the droplet breakup mechanisms, to the following dimensionless numbers are defined:

Reynolds number (ratio of inertial forces to viscous forces):

$$Re = \frac{\rho v d}{\mu} = \frac{v d}{\nu} \tag{8}$$

Weber number (ratio of inertia to surface tension):

$$We = \frac{\rho v^2 d}{\sigma} \tag{9}$$

Ohnesorge number (ratio of viscous forces to inertial and surface tension forces):

$$Oh = \frac{\mu}{\sqrt{\rho \sigma d}} = \frac{\sqrt{We}}{Re} \tag{10}$$

Laplace number:

$$La = \frac{\sigma \rho d}{\mu^2} = \frac{1}{Oh^2} \tag{11}$$

From the Rayleigh-Taylor or Kelvin-Helmholtz instabilities, a dimensionless time characteristic for drop breakup is obtained:

$$\mathcal{T} = \frac{v\sqrt{\epsilon}}{d} t \quad , \quad \epsilon = \frac{\rho_a}{\rho} \tag{12}$$

where ϵ is the flow field/drop density ratio.

There are 5 distinct mechanisms of drop breakup [10]:

1) Vibrational breakup , $We \leq 12$
2) Bag breakup , $12 \leq We \leq 50$
3) Bag-and stamen breakup , $50 \leq We \leq 100$
4) Sheet stripping , $100 \leq We \leq 350$
5) Catastrophic breakup , $We > 350$

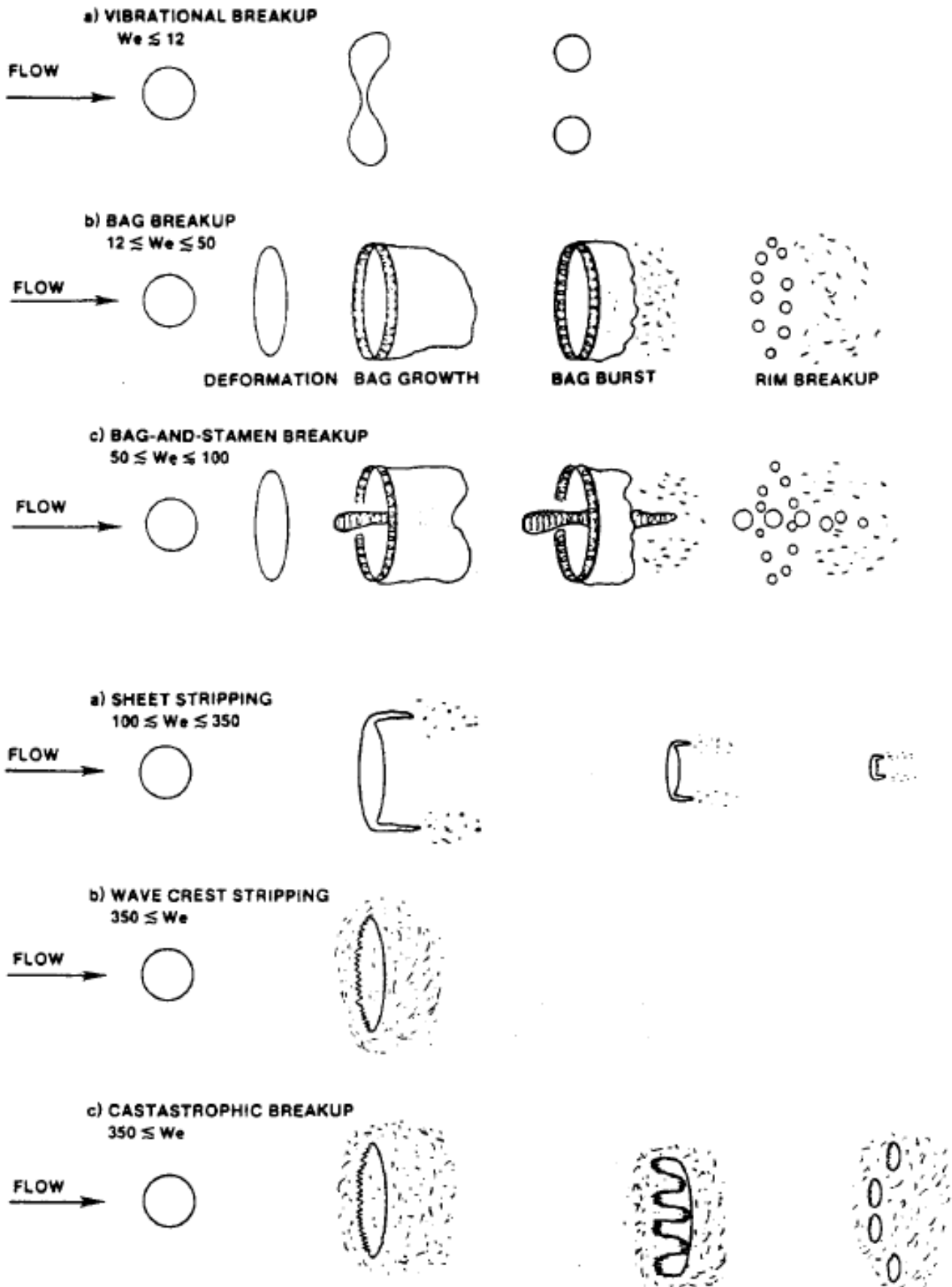All mechanisms are illustrated in Figure. 4

Figure 4 – Breakup Mechanisms.

There is a critical Weber number below which drop breakup does not occur. An empirical correlation for the critical Weber number was derived by Brodkey (1969):

$$We_c = 12(1 + 1.077 \cdot Oh^{1.6}) \qquad (13)$$

A requirement for efficient combustion in a limited combustor length is that the spray produced by the injector has a high specific surface area. Impinging jet injectors generally satisfy this requirement when used with low viscosity Newtonian fluids and sufficiently high injection velocities. The impact of the jets leads to the formation of fine droplets and thus the necessary high surface area of the droplet phase [11].

Atomization of gels is more difficult due to their non-Newtonian character [12]. The existence of yield stress and increased viscosity make gelled propellants very difficult to atomize, an obstacle to their wide use for aerospace application. In Newtonian fluids, large viscosity values produce coarse sprays, and in non-Newtonian fluids, shear and extensional viscosities can be several orders of magnitude larger. This results in reduced performance, and a longer combustion chamber is required (increased weight). The atomization of gelled propellants is significantly different from the atomization of Newtonian liquids. Hence, the injectors used to atomize the gelled propellants should be specially designed and the influence of the rheological properties, injector geometries [13] and working conditions on the spray characteristics of gelled propellants must be carefully considered [15].

For some fuels, atomization can never be achieved, and the jet get a strongly bent shape that is rarely seen in atomization of Newtonian fluids [15].

Hypergolic Ignition and Combustion Process

After the atomization, the ignition and combustion process begins. Ignition kernels form at discrete locations where fuel and oxidizer come together in the presence of the reactive particles. The fuel/oxidizer interface must be sufficiently fuel-rich for ignition to occur on millisecond time scales. When fuel and oxidizer come together in correct proportions in the presence of the reactive particles, the resulting ignition kernel produces a flame that engulfs the reactive mixture, resulting in full combustion. During the ignition process, many such kernels may form and extinguish before full-fledged ignition of the mixture occurs.

This allows two important parameters to be recorded [16]:

- "First Light" - appearance of luminous region.
- "Ignition Onset" - first kernel of light that spreads over entire spray.

Figure 5 – Images of "First Light", "Ignition Onset" and combustion of hypergolic gelled fuel with HP.

If a gel has poor atomization characteristics, it implies that larger droplets are produced, which would translate into incomplete combustion of gels inside the combustion chamber. Incomplete combustion leads to lower energy yield and hence affect the overall combustion efficiency of the system [17].

The process of the combustion of organic gellant based gel droplets is periodic. During the combustion, after the vaporization of part of the fuel, an elastic layer of gellant is formed around the droplet that prevents further fuel vaporization from the outer surface of the droplet. This causes the fuel to evaporate below the droplet surface producing bubbles, which results in droplet swelling, fuel jetting, and collapse of the remaining droplet. The process repeats itself until complete consumption of the fuel and gellant.
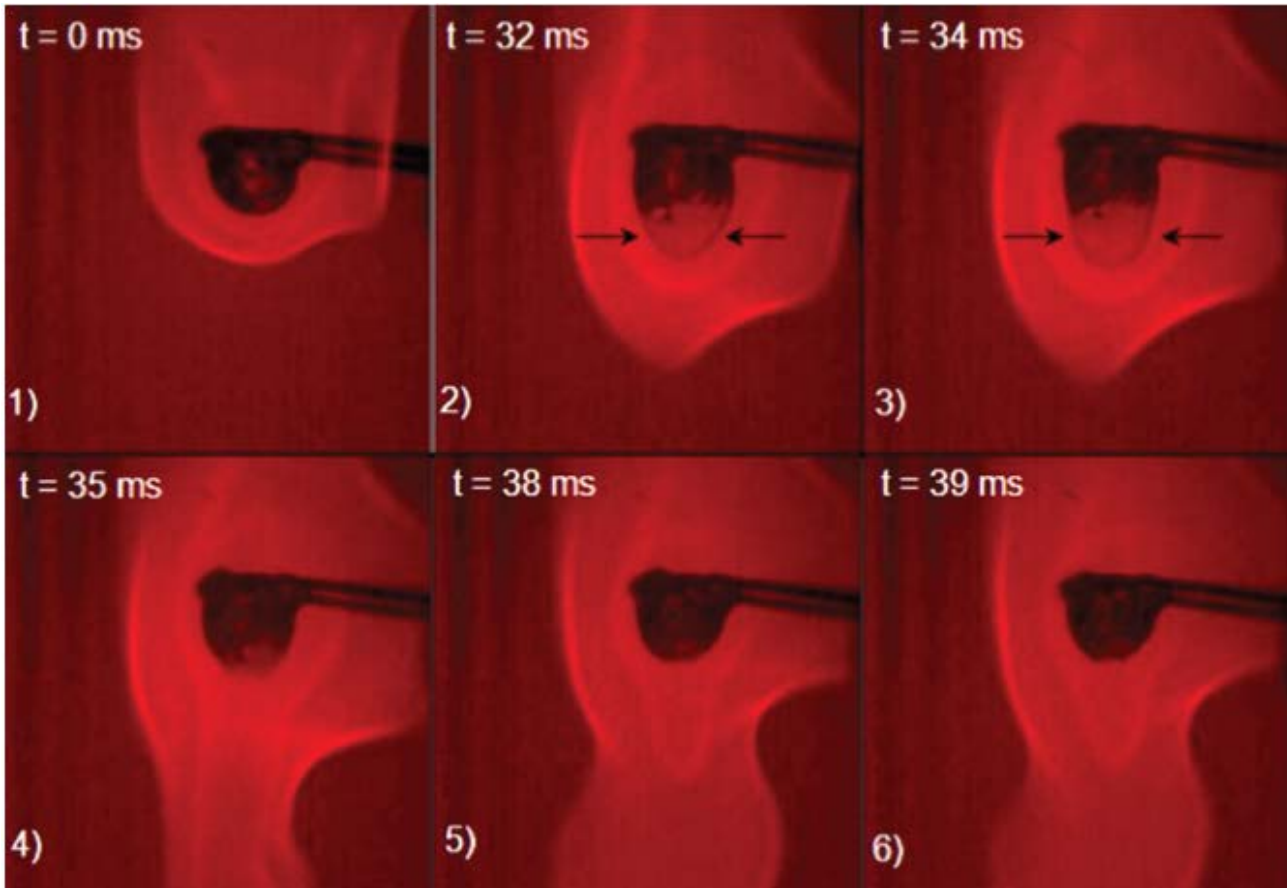
Figure 6 - Bubble formation and fuel jetting

One important measure of the hypergolic performance is the length of time between reactant contact and appearance of the flame, termed as the ignition delay time (t_{ign}). Measured in milliseconds, ignition delay time is important because longer than desired delays can lead to lower performance or cause catastrophic failure of the engine [18].

Theoretical and experimental considerations have shown that the ignition delay time values of hypergolic propellants depend on the complex interactions between many chemical and physical factors [19–23]. The chemical processes governing the ignition of reacting flows comprise many reactions between a fuel, an oxidizer, intermediate species, and products. The ignition delay time is greatly influenced by the injection technique and the physical properties of the propellants: viscosity, surface tension, and miscibility [1,25].

The main features of thermal ignition are:

- The reaction is exothermic with a significant activation energy.
- Simple kinetics are assumed.
- A significant temperature rise occurs in the control volume.
- Ignition is inevitable in adiabatic systems.
- Critical conditions for ignition exist in a non-adiabatic system and are governed by an interaction between heat release rate and heat loss rate.
- Ambient conditions and the size or shape of the reactant system are important.

The thermodynamics within the system lead to a heat production term and a heat loss term. Considering a vessel of volume V and surface area S, the rate at which heat is generated by chemical reaction is given by:

$$\dot{q}_{gen} = \frac{VQ}{N} \cdot \frac{d\xi}{dt} = \frac{VQ}{N}\left(kA^b \cdot e^{-\frac{E_a}{RT_{mix}}} \right) \tag{14}$$

It should be noted at this point that, according to Arrhenius expression, the rate of a chemical reaction never becomes zero. It falls exponentially with a decrease in temperature. Consequently, no matter how low the initial temperature of the mixture may be, the mixture must react after a sufficiently long time.

The rate at which heat is lost to the surroundings is given by:

$$\dot{q}_{loss} = hS(T_{mix} - T_0) \tag{15}$$

The heat loss term is therefore a linear function of the mixture temperature.

Considering a system in which there is simultaneous heat generation and heat loss, the overall energy conservation equation takes the form of the following equation, where the left-hand side is the rate of energy accumulation in the vessel:

$$\rho_{mix} C_{P_{mix}} \frac{dT_{mix}}{dt} = \dot{q}_{gen} - \dot{q}_{loss} \tag{16}$$

Expressing the concentrations in terms of partial pressures and the density and the heat capacity in terms of constituents' properties, we obtain an analytical expression for the ignition delay time [25]:

$$t_{ign} = \frac{R^2 T_0^3}{(P_f + P_{ox})E_a k\Delta H}\left(1 + \frac{P_f}{P_{ox}}\right)\left(C_{P_f} + \frac{P_{ox}}{P_f}C_{P_{ox}}\right)e^{\frac{E_a}{RT_0}} \tag{17}$$

The ignition performance of hypergolic bipropellants is of great importance because, for a hypergolic bipropellant system, unless the ignition delay is short enough, the thruster can experience combustion instability and/or problems associated with hard start. Once hard start occurs in the startup phase of thruster, the components of the thruster could be severely damaged due to strong pressure spikes and sometimes even destroyed [26].
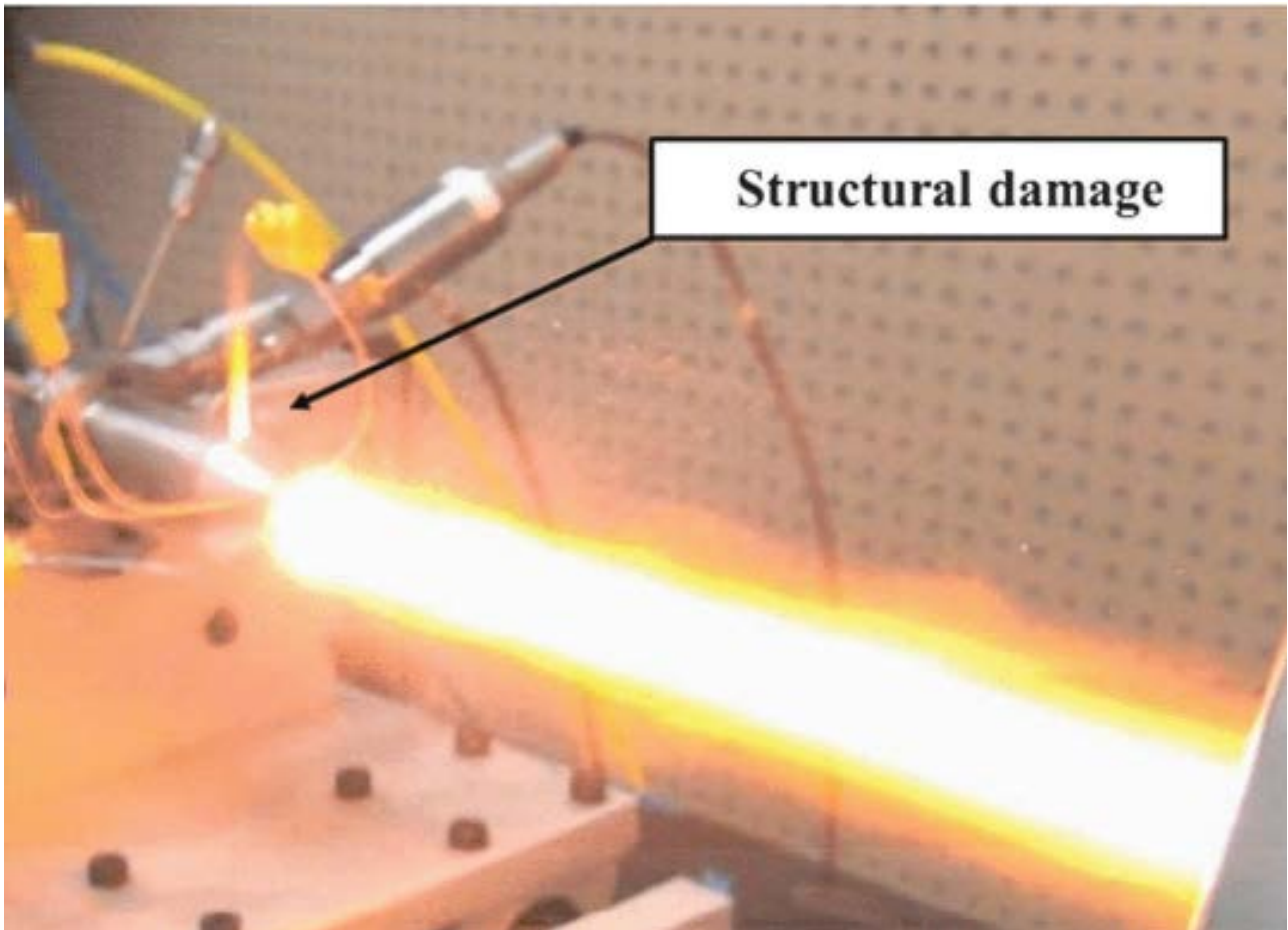
Figure 7 - snapshot of steady-state rocket thruster operation after occurring hard start.

Rocket structure

For a hypergolic bipropellant rocket, the whole combustion zone inside of the chamber can be conceptually divided (Figure 8). When the fuel and oxidizer are injected, they undergo mixing and atomization in the liquid phase before ignition. The region where liquid jets collide with substantial jet momentum is called an injection and atomization zone, thereby forming numerous mixture droplets. Rapid combustion zone is formed in between the liquid phase zone and combustion gas zone. Thus, in the rapid combustion zone, two-phase chemical intermediates react vigorously at increasingly higher temperature, and local three-dimensional turbulence and diffusion of the gas species take place via complex heat transfer processes [27].
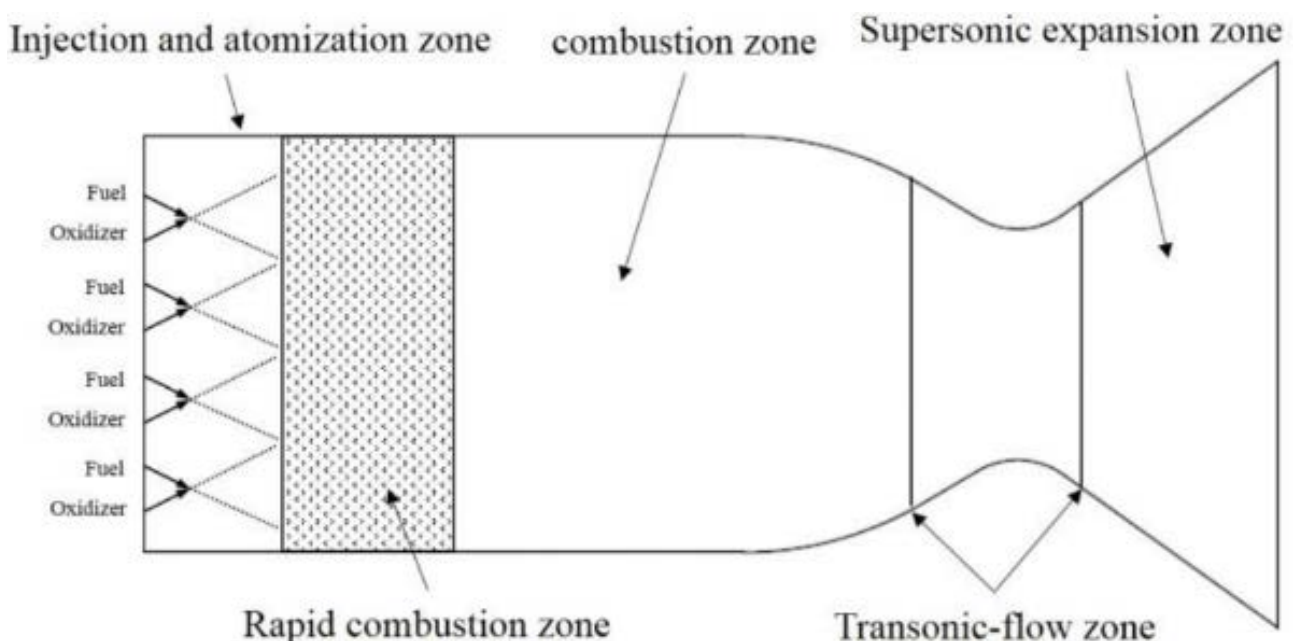
Figure 8 - Combustion zones in a combustion chamber.

Physical factors are regulated by changing the differential pressure (ΔP) on the injection nozzles: The jet mass flux and momentum grow as the differential pressure increased. Increasing the momentum of jet enhances physical interactions between the reactants regarding mixing and atomization so that hypergolic ignition can be achieved faster. However, excessive physical interactions such as oversupplying reactants with high jet velocity can trigger combustion instabilities at the startup phase.

Metal hydrides

Metal hydrides have been considered by many as a candidate for rocket propellant additives [28-29]. Their combustion with various oxidizers is highly exothermic like that of the metal atoms contained in the hydride. In addition to producing a significant amount of heat, they release hydrogen gas at temperatures far below their flame temperatures. This hydrogen gas release resulting from decomposition of these materials provides increased theoretical propellant performance in comparison to propellants that exclusively use metal fuel additives. Many metal hydrides are also hypergolic with rocket grade hydrogen peroxide and other common propellant oxidizers, simplifying motor ignition and restart.

Although metal hydrides offer improved propulsive performance over their metal constituents, their increased performance comes at a price. Many metal hydrides are either pyrophoric or appreciably degrade (dehydrogenate) in the presence of oxygen gas, excessive heat, or water vapor. This high reactivity has led to difficulty in finding fuel formulations and mixing procedures that do not result in partial or complete metal hydride deactivation.

There has been a wide interest for many years in the unusual properties and possible uses of compounds between boron and hydrogen, and a great many of them have been prepared and studied. In particular, chemical methods have been devised for preparing metallic borohydrides, which contain the monovalent group BH_4. The borohydrides of the alkali metals are much more stable than the known hydrides of boron, dissolving in water at room temperature with but slow decomposition, and resisting spontaneous decomposition in the dry state up to temperatures of several hundred ℃. Since X-ray patterns indicate a crystal lattice of metallic and borohydride ions, it appears that the relative stability of these compounds can be attributed directly to the high coulomb energy characterizing such typical ionic salts [30].

Chemicals

The hypergolic propellant combination used is composed of the following chemicals [31]:

Kerosene-based fuel (Jet-A) is a liquid mixture of carbon chains that typically contain between 6 and 16 carbon atoms per molecule and approximate molecular formula $C_{11}H_{21}$ (Jet-A). Density is 810 $\left[\frac{kg}{m^3}\right]$,flash point is 52 °C, boiling range is 165– 265 °C, auto-ignition temperature is 220°C and heat of combustion is 43.1 $\left[\frac{MJ}{kg}\right]$.

Nanosilica fumed powder (SiO_2), 0.014 [μm] has hydrophilic characteristics and can build up hydrogen bonds to create a gel when mixed with the pure liquid.

Sodium borohydride ($NaBH_4$) is a metal hydride that reacts spontaneously with HP. Density is 1074 $\left[\frac{kg}{m^3}\right]$, flash point is 70 °C, boiling point is 500 °C, and auto-ignition temperature is 220 °C.
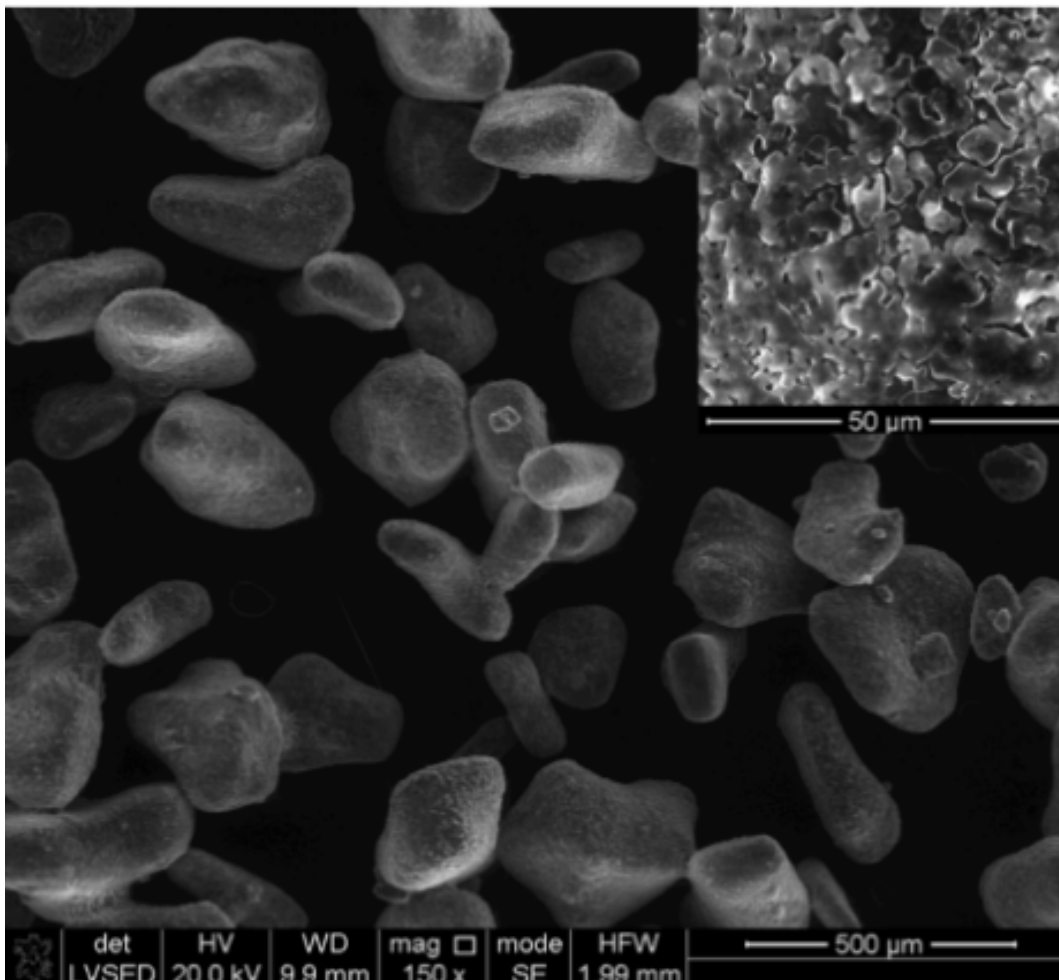

Figure 9 – SEM image of $NaBH_4$ and close-up of the particle surface showing porous appearance.

Hydrogen peroxide (H_2O_2), is in general in liquid form. Its density is 1460 $\left[\frac{kg}{m^3}\right]$, melting point is $-43\,°C$, boiling point is $150\,°C$, it decomposes exothermically into water and oxygen, and the heat of formation is $-98.2\left[\frac{MJ}{kmol}\right]$. It is a non-cryogenic, very strong, storable oxidizer/monopropellant with a high-density specific impulse. Being non-toxic, it is considered as a green propellant and its popularity is increasing due to the increase of environmental awareness since 1990 [32,33].

Hydrogen peroxide serves as an oxidizing agent in the combustion of organic fuels. The driving force behind these reactions is the conversion of oxygen in the -1 oxidation state to oxygen in the -2 oxidation state. Fuels that are strong reducing agents should facilitate this conversion and thus be very reactive with hydrogen peroxide. Under basic conditions, peroxide loses a proton and becomes much less stable. In addition to being good reducing agents, potential fuels should also be basic if possible [34].

Drop-on-Drop Tests

Hypergolic drop tests have been employed in propellant research for decades as a simple, quick, and inexpensive means of screening potential propellants and measuring ignition delay [35]. Over the years drop tests have become relatively standardized. A droplet of fuel or oxidizer is dropped from a set height into a container of oxidizer or fuel, respectively. Unfortunately, drop-on-drop tests are often conducted without rigorous control over variables known to influence ignition delay: propellant temperature, oxidizer-to-fuel ratio, ambient pressure, and ambient oxygen content. This, along with varying impact energy and mixing methods, makes it difficult to compare directly drop-on-drop experiments. Another limitation of drop-on-drop tests is that the ignition delays measured cannot be directly applied to liquid rocket injector design because the mixing times are significantly different and are often the limiting factor in hypergolic ignition.

Acknowledgments

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