Intra - Bubble - Combustion: Diffusion Limit

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Abstract

The present work considers an idealized mathematical model motivated by the application of chemically processing crude oils using submerged oxygen bubble jets [1]. In this theoretical analysis we examine a fully transient, spherically symmetric oxidizer gas bubble immersed in an unconfined, otherwise quiescent, single-component constant density viscous liquid fuel. At the bubble surface the liquid vaporizes supplying fuel vapor which then diffuses inside the bubble, reacting finally with the oxygen initially in the bubble in a spherical diffusion flame. The present paper focuses in the dynamics of this diffusion flame inside the oscillating bubble, coupled with the consequences of momentum-, mass- and heat- transport across the bubble (gas/liquid) interface. To gain insight into these complex interactions, the combustion process itself is approximated by means of the 'thin-flame' (Burke-Schumann) limit. Hence, flame ignition or extinction phenomena are not dealt with here. Three zones are considered —two inside the bubble (where the heat and mass diffusivities are taken to be equal) and one outside (the thermal boundary layer in the liquid)—using a Chebyshev-based orthogonal collocation method [2, 3].

Our theoretical analysis reveals that the dynamics of this single bubble heterogeneous combustion system depends on the square of the ratio of the characteristic liquid inertial time (proportional to the initial bubble radius and the square root of the liquid density) to the characteristic diffusion time within the bubble. This dimensionless parameter plays the role of the (dimensionless) effective inertia of the two-phase dynamical system.

As applications of the present theoretical model, the 'conversion' of such a bubble reactor (amount of fuel processed per bubble oscillation) is computed as a function of the dimensionless effective inertia, along with the total time to complete the consumption of the initial O2(g) inventory [2, 3]. In our future work this mathematical model will be broadened to incorporate the formation and survival of important reaction intermediates (such as acetylene) on the fuel-rich side of the gaseous diffusion flame.

1 Introduction

1.1 Scheme of Intra - Bubble - Combustion

The process called *Intra-Bubble-Combustion* is characterized by a vapor phase combustion chemical reaction between a fuel and a oxidizer, which takes place inside a bubble. Depending on whether fuel and oxidizer are initially mixed inside the bubble, or whether only one of them is initially present in the bubble while the other is in the liquid surrounding it there are two well defined limits.

In the *premixed* limit [4] both reactives are initially present in the bubble. In this case, once ignition takes place inside the bubble (for instance by means of a laser pulse focalized inside the bubble [5]), a *premixed flame* traverses the bubble until the reactive mixture is consumed.

On the other hand, in the *diffusion* limit only one of the reactives is initially present in the bubble (usually the oxidizer), while the other (usually the fuel) is initially present in the liquid surrounding the bubble. In this case, as the fuel vaporizes towards the interior of the bubble, the reaction takes place in a *diffusion flame*, until the initial inventory of oxidizer is consumed. Hence, the problem of Intra-Bubble-Combustion in the diffusion limit appears as the 'inverse' of well known spray vaporization and combustion.

The dynamics of premixed flame propagation inside a bubble have been considered in a previous work [4]. In the present work we will focus on the diffusion limit. From the technological point of view the diffusion limit is the most relevant case. However, the premixed limit is important because a lean premixed reactive mixture ignited by means of a laser pulse, is considered as one of the possibilities for producing ignition of the subsequent diffusion flame [5].

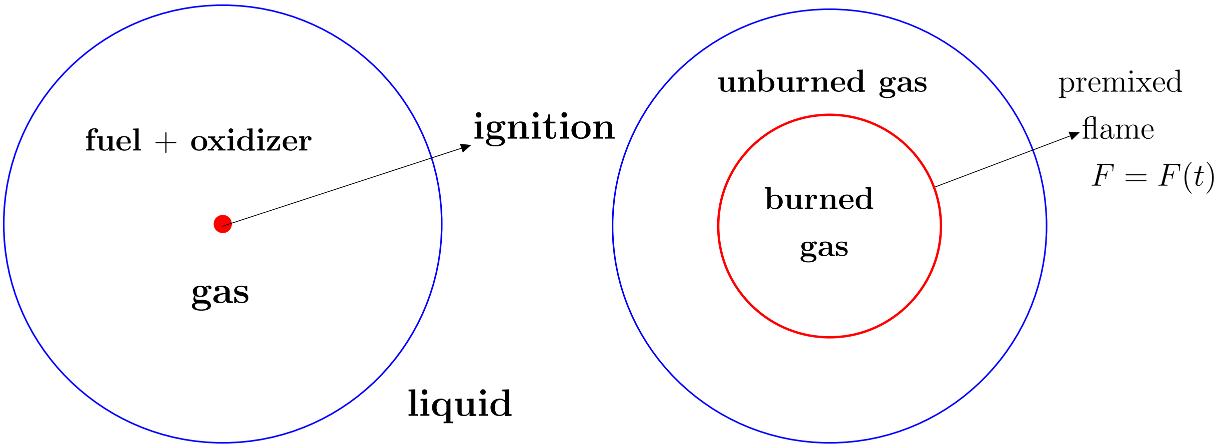

Figure 1: Scheme of Intra-Bubble-Combustion in the premixed limit.

1.2 Motivation

The technological motivation of the present theoretical work is related to the design and optimization of submerged gas jet combustors or reactors for synthesis oriented chemical industry. This kind of Intra-Bubble-Combustion reactors can be useful when the liquid fuel is difficult to 'atomize' due to its high effective viscosity, and/or chemical aggressiveness. The present work is also relevant for waste treatment using aggressive molten solvents, as, for instance, in molten salt oxidation.

2 Modelization

2.1 Assumptions

The present theoretical work focuses on the basic dynamics of a diffusion flame inside a bubble, coupled with momentum, mass, and heat transport across the bubble surface.

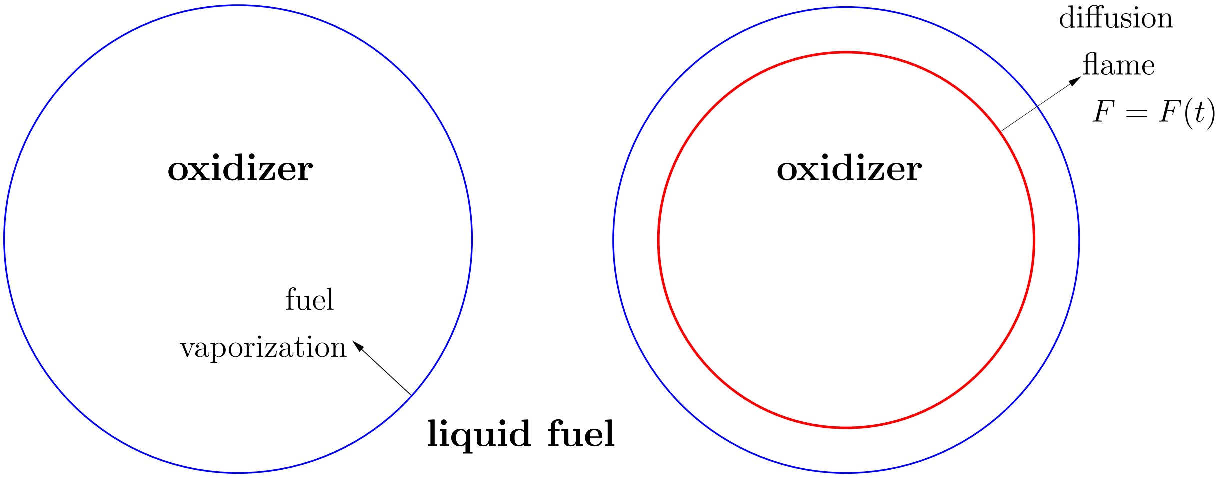

Figure 2: Scheme of Intra-Bubble-Combustion in the diffusion limit.

In the present model a spherically symmetric bubble in an unconfined, otherwise quiescent, liquid is considered. The gas inside the bubble is considered as an ideal gas with constant properties with unit Lewis number. Fick and Fourier transport are the only diffusion processes included, *ie.* cross transport processes as Soret or Dufour are neglected.

The liquid phase in which the bubble is immersed is considered as an incompressible liquid. At the bubble surface Vapor/Liquid equilibrium (VLE) conditions are assumed.

The chemical reaction that takes place in the gaseous diffusion flame is approximated by a single step reaction between the fuel and oxidizer

$$\mathrm{O} + f\ \mathrm{F} \rightarrow (1+f)\ \mathrm{P} + (\ f\, Q^{\star})$$

which is described by means of the Burke-Schumann (BS) flame-sheet limit

$$\omega_O \cdot \omega_F = 0$$

Hence, ignition and extinction phenomena are not considered in the present model.

According to the former assumptions the spacial domain divided in 3 regions

- Gas, oxidizer region: between the center of the bubble and the diffusion flame $0 \leq r < F(t)$.
- Gas, fuel region: between the diffusion flame and the bubble surface $F(t) < r < R(t)$.
- Liquid: exterior region $R(t) < r < \infty$.

2.2 Conservation equations

Under the mentioned assumptions, the conservation equations that govern the process under consideration are given by:

- Total mass conservation

$$\frac{\partial \rho}{\partial t}+\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\rho v\right)=0$$

- Oxidizer mass conservation

$$\rho\left(\frac{\partial \omega_O}{\partial t}+v\frac{\partial \omega_O}{\partial r}\right)=\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\rho D\frac{\partial \omega_O}{\partial r}\right)$$

- Fuel mass conservation

$$\rho\left(\frac{\partial \omega_F}{\partial t}+v\frac{\partial \omega_F}{\partial r}\right)=\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\rho D\frac{\partial \omega_F}{\partial r}\right)$$

- Energy conservation

$$\rho c_p\left(\frac{\partial T}{\partial t}+v\frac{\partial T}{\partial r}\right)=\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\kappa\frac{\partial T}{\partial r}\right)+\alpha T\frac{dP}{dt}$$

  where α is the thermal expansion coefficient, and v the mass-averaged radial velocity.

- Linear momentum conservation
  - In the gas phase it is assumed that the characteristic time of the process under consideration is very small as compared to the characteristic acoustic time in the bubble. Hence, linear momentum conservation in the gas phase implies that the pressure is homogeneous, although time-dependent, inside the bubble

    $$Ma \ll 1 \rightarrow p=p(t), \quad \text{in the gas}$$

  - In the liquid phase linear momentum conservation is described by the Navier-Stokes equations

2.3 Boundary conditions

The boundary conditions that complete the former set of governing equations are

- Continuity of all fields at the diffusion flame (located at $r=F(t)$)
- Burke-Schumann Condition at the diffusion flame

$$\omega_O(F)=\omega_F(F)=0$$

- Vapor-Liquid Equilibrium at the bubble surface (located at $r=R(t)$)

- Mass flow continuity for all the species at the flame

$$f\,\frac{\partial \omega_O}{\partial r}\bigg|_{r=F(t)-} = -\,\frac{\partial \omega_F}{\partial r}\bigg|_{r=F(t)+}$$

- Energy flow continuity at the flame

$$\kappa\frac{\partial T}{\partial r}\bigg|_{r=F(t)-} - \kappa\frac{\partial T}{\partial r}\bigg|_{r=F(t)+} = Q^\star\,\rho D\frac{\partial \omega_F}{\partial r}\bigg|_{r=F(t)+}$$

- Mass flow continuity at the bubble surface

$$\left(v-\dot{R}\right)\rho\Big|_{r=R(t)-} = \left(v-\dot{R}\right)\rho\Big|_{r=R(t)+} = -\frac{\dot{m}}{4\pi R^2}$$

$$\left[\left(v-\dot{R}\right)\rho\omega_F - \rho D\frac{\partial \omega_F}{\partial r}\right]_{r=R(t)-} = -\frac{\dot{m}}{4\pi R^2}$$

  (it is assumed that the fuel is the only species that can cross the vapor-liquid interface, oxidizer or product dissolution/condensation are not considered).

- Energy flow continuity at the bubble surface

$$-\,\kappa\frac{\partial T}{\partial r}\bigg|_{r=R(t)-} = L^\star\frac{\dot{m}}{4\pi R^2} - \kappa\frac{\partial T}{\partial r}\bigg|_{r=R(t)+}$$

- Momentum flow continuity at the bubble surface, described by means of the Rayleigh-Plesset equation

$$R\,\ddot{R} + \frac{3}{2}\dot{R}^2 = \frac{p(t)-p_A}{\rho_L}$$

3 Numerical method

The former system of Partial Differential Equations has been solved by means of the Orthogonal Collocation Method. To that end each spatial region has been mapped to a fixed interval, and then the Orthogonal Collocation Method has been applied using Chebyshev nodes in terms of the mapped variable (z).

The mappings and numbers of abscissae ($\hat{N}$) used on each sub-domain are

- Gas, oxidizer region:

$$0 \le r < F \qquad \rightarrow \qquad z = \frac{r}{F}$$

  This region has been solved by means of OCM in terms of z^2 with $\hat{N} = 6+1$ Chebyshev abscissae (6 interior abscissae and 1 boundary abscissa).

- Gas, fuel region:

$$F < r < R \qquad \rightarrow \qquad z = 2\frac{r-F}{R-F} - 1$$

  This region has been solved by means of OCM in terms of z with $\hat{N} = 17 + 2$ Chebyshev abscissae.

- Liquid, exterior region:

$$R < r < \infty \qquad \rightarrow \qquad z = \exp\left(-\frac{r-R}{2\delta_T^L}\right)$$

  where δ_T^L is the liquid thermal boundary layer thickness. This region has been solved by means of OCM in terms of z^2 with $\hat{N} = 29 + 2$ Chebyshev abscissae.

In the present calculations the liquid fuel considered is n-dodecane. As already mentioned, ignition or extinction phenomena are not considered in the present model. In this respect it is assumed that the ignition has been produced by means of the combustion of an initially lean mixture [4]. As a consequence of that process, the initial pressure in the bubble (p) is slightly higher than in the liquid far from the bubble (p_A). The initial overpressure in the bubble considered in the present numerical calculations has been $\Pi(0) \equiv p(0)/p_A = 3$.

4 Dynamics

The present model shows that the dynamics of the process under consideration is governed by a dimensionless number (M) defined as the squared ratio between the characteristic diffusion time in the bubble and the characteristic time related to the liquid inertia.

The characteristic diffusion time in the bubble is the relevant time scale t_c of the process. This characteristic time scale is given by

$$t_c = \frac{R_0^2}{\alpha_A} \cdot \frac{f}{Q} \frac{p(t=0)}{p_A}$$

where R_0 is the initial bubble radius, α_A as the characteristic value of the gas thermal diffusivity at ambient pressure $(p = p_A)$ and ambient temperature $(T = T_A)$, and where

$$Q = \frac{Q^\star}{c_p T_A} \gg 1$$

is the dimensionless heat release, ($Q^\star$ is the dimensional heat release in the chemical reaction per unit fuel mass). As can be seen, as a consequence of the heating produced by the large heat released in the chemical reaction, there is a decrease in the characteristic diffusion time.

On the other hand, according to the Rayleigh-Plesset eq. the characteristic liquid time t_L is given by

$$t_L = R_0\sqrt{\frac{\rho_L}{p_A}}$$

where ρ_L is the liquid density. Hence, in terms of the dimensionless units

$$a = \frac{R(t)}{R(0)}, \quad \Pi = \frac{p(t)}{p_A}$$

the Rayleigh-Plesset eq. is given by

$$M \left(a\,\ddot{a} + \frac{3}{2}\dot{a}^2 \right) = \Pi - 1$$

where

$$M = \left(\frac{t_L}{t_c} \right)^2$$

represents the effective liquid inertia.

In terms of the dimensional quantities the parameter M is given by

$$M = \frac{\rho_L \alpha_A^2}{p_A R_0^2} \cdot \left(\frac{Q}{\Pi(0) f} \right)^2$$

and can take values in a wide range between the small inertia limit (reached for $M \ll 1$) and the large inertia limit (reached for $M \gg 1$). The dynamics of the process is qualitatively different in these two limiting cases.

On one hand, in the small inertia limit ($M \ll 1$) the liquid reacts almost instantaneously in the characteristic time scale defined by the diffusion flame. As a consequence, in this case the process takes place under conditions similar to ‘constant pressure’.

On the other hand, in the large inertia limit ($M \gg 1$) the liquid almost does not react at all in the characteristic time scale defined by the diffusion flame. As a consequence, in this case the process takes place under conditions similar to ‘constant volume’.

5 Bubble reactor optimization

From the technological point of view, the main result shown by the present simple model is the conditions that lead to a maximal bubble reactor yield. In general the reactor yield will be proportional to the total fuel vaporized mass. In this respect, the present model shows that the maximal value of the total fuel vaporized mass is reached for values of M close to 1.

The present result can be explained as a consequence of the combined effect of the large radius increase reached when M is not large, and the small pressure in the bubble during most of the bubble lifetime reached when M is not small.

Another important result from the point of view of bubble reactor optimization is the Bubble combustion time.

The results found in this respect show that the combustion time is minimum for a value of M slightly smaller than 1.

The physical explanation of that results is that in the small inertia limit the combustion time increases as a consequence of the oscillatory bubble response, while in the large inertia limit it increases as a consequence of the reduced exchange surface.

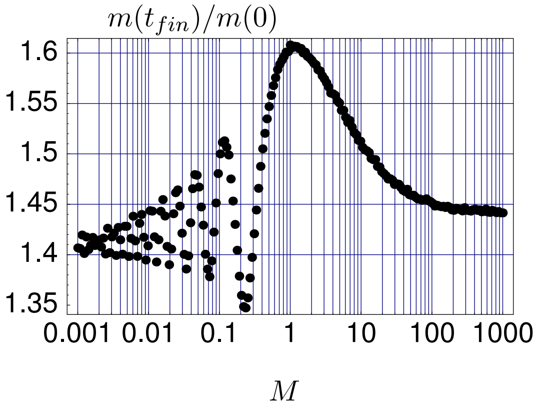

Figure 3: Bubble mass at the end of the process.

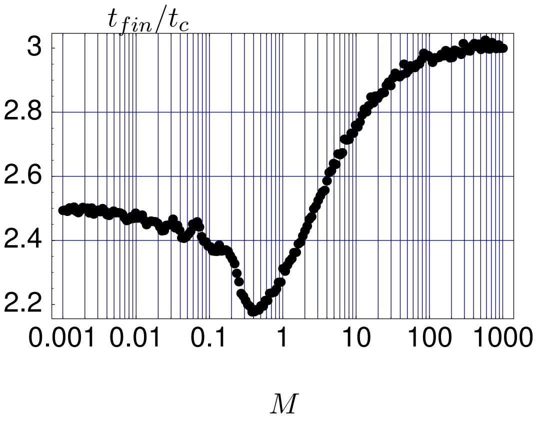

Figure 4: Bubble combustion time.

6 Conclusions

- The dynamics of the process is governed by the effective inertia parameter M, defined as the squared ratio between the characteristic time scales defined by diffusion in the bubble and by the liquid inertia
$$M = \left(\frac{t_L}{t_c}\right)^2$$
- In the small inertia limit ($M \ll 1$) the process takes place at constant pressure after a short oscillatory transient.
- In the large inertia limit ($M \gg 1$) the process takes place under conditions close to constant volume.
- The maximum bubble reactor yield has been found for values of M close to 1.

- The physical reason for this result is because in that case there is significant surface increase and bubble pressure decrease during the combustion time, which favors the fuel vaporization at the bubble surface.

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