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Mathematical Modeling of the Gas Bubbles in the Microwave Discharge of Boiling N-Heptane

Yu. A. Lebedev[1], A. V. Tatarinov[1], I. L. Epstein[1] and K. A. Averin [1,2]

[1]A.V. Topchiev Institute of Petrochemical Synthesis
Russian Academy of Sciences
Leninsky pr. 29, Moscow
119991, Russian Federation

[2]Moscow Institute of Physics and Technology State University
Moscow region, Dolgoprudny, Institutsky per., 9
141700, Russian Federation

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Abstract

Numerical modeling of formation of the gas bubbles in boiling n-heptane, initiated in the microwave discharge at atmospheric pressure, has been performed. The model is based on joint solution of the Maxwell equations, Navier-Stokes equation, heat equation, continuity equations for electrons and the Boltzmann equation for free electrons of the plasma. The calculations allowed to describe the dynamics of the formation of gas bubbles in the liquid and to estimate the characteristic times of various processes in the system. The size of the bubble in the experiment is comparable with the size of the bubble in our simulations. The phase field method has been successfully applied for the description of boiling of n-heptane processed in the microwave discharge.

Keywords: microwave discharge, n-heptane, 2D modeling, two-phase fluid

1 Introduction

Low temperature plasma is now widely used for solving scientific and applied problems. Finding new approaches to the plasma application requires the development of methods for its study. Typically both computational and experimental approaches are used. They mutually complement each other and allow obtaining reliable information about the plasma parameters. There are situations, when the use of experimental methods is difficult and then the main burden of research is laid upon the mathematical modeling. This fully corresponds to the intensively developing field of obtaining plasma in a volume of dielectric fluid [1-6].

This work presents a model of the microwave discharge generated in the vicinity of the metal conductor immersed in a liquid n-heptane. This work is a continuation of research for the study of physico-chemical processes in microwave discharges in liquid hydrocarbons, conducted by the authors [7-9]. The possibility of using phase-field method [10] for the simulation of plasma-chemical processes is demonstrated.

2 Mathematical formulation of the problem

A cylindrical chamber with metal walls of diameter 4 cm and height 5 cm is partially filled with liquid n-heptane. The energy from the microwave source is supplied from below via a coaxial waveguide. A part of the bottom of the cylindrical chamber, marked by the line OABC, permeable to microwave fields and not permeable for liquid and vapor (Fig.1). At the bottom of the camera there is a small cylindrical cavity OAB of diameter 0.3 cm and height 0.1 cm.

Fig. 1. The simulation area and initial distribution of liquid and vapor phases of n-heptane. A piece of border ABC is permeable to microwaves and impermeable to n-heptane. The input of the microwave power is marked by the arrow

At the initial moment, the lower part of the cavity is filled with saturated vapor. This is in fact a gas bubble nucleus of the saturated vapor which would be further involved in the process of bubble boiling. Plasma of gaseous n-heptane is ignited in the gas bubble nucleus, which leads to local heating and growth of the size of the bubble. The position of the boundary between liquid and gas phases changes with time.

For analysis of the formation, growth and detachment of the bubble the phase field method [10] has been applied. This method is one of the single-fluid approaches to modeling of the two-phase flows in which both phases are described by one system of Navier-Stokes equations with an assumption that the liquid-vapor interface has a finite thickness and is characterized by a rapid but smooth change of density, velocity and other physical parameters. For a transition between two phases the dimensionless phase field variable φ is introduced, where $-1 \leq \varphi \leq 1$, $\varphi = -1$ for the vapor phase, $\varphi = 1$ for the liquid phase. The volume fractions of the vapor phase V_{fv} and liquid phase V_{fL} are determined as follows: $V_{fv} = (1-\varphi)/2$, $V_{fL} = (1+\varphi)/2$. The variable φ is determined from the Cahn - Hilliard equation [11].

The two-dimensional axisymmetric model includes the Navier-Stokes equations for two-phase subsonic flow of incompressible fluid and compressible gas, the heat conduction equation, the Maxwell equations for the microwave field, the Boltzmann equation for the plasma electrons and the balance equation for the electron density.

For the determination of constant rate of ionization reaction the electron energy distribution function (EEDF) has been calculated. The Boltzmann equation for a stationary, isotropic part of the EEDF, recorded in two-term approximation of the expansion of the EEDF into spherical harmonics [12] has been used. Unfortunately, the self-consistent set of cross sections for n-heptane for calculation of the EEDF is unknown. In such cases, for description of electrons kinetics for heavy hydrocarbons it is common to use known sets of cross sections for lighter hydrocarbons [13]. According to [14] cross-sections of vibrational and electronic excitation of saturated hydrocarbons C_3-C_7 are close. Therefore, in the first approximation, we used a set of cross sections for propane [15]. For calculation of the ionization coefficient of n-heptane we used the total ionization cross section for n-heptane, which was measured in [16] in the range from 10 to 86 eV.

The Navier-Stokes equation

$$\rho\frac{\partial \boldsymbol{u}}{\partial t} + \rho(\boldsymbol{u}\cdot\nabla)\boldsymbol{u} = \nabla\cdot[-p\boldsymbol{I} + \eta(\nabla\boldsymbol{u} + (\nabla\boldsymbol{u})^{\boldsymbol{T}})] + \rho\boldsymbol{g} + G\nabla\varphi$$

$$\nabla\cdot\boldsymbol{u} = \dot{m}\delta\left(\frac{1}{\rho_V} - \frac{1}{\rho_L}\right)$$

Here G is chemical potential of the fluid, η is dynamic viscosity, p is pressure, ρ and $\boldsymbol{u}$ are the density and velocity of the fluid, index L relates to the liquid phase, V – to the vapor phase, $\dot{m}$ is the evaporation rate of the liquid, δ [1/cm] is a smooth-

ed representation of the interface between two phases, $\delta = 6V_{fL}V_{fV}\frac{|\nabla\varphi|}{2}$, $\boldsymbol{g}$ is free fall acceleration. The density of the fluid is determined from the relation: $\rho = V_{fL}\rho_L + V_{fv}\rho_V$, where ρ_L and ρ_V are density of the liquid and vapor, respectively. The vapor density is determined from the state equation of a perfect gas.

Along with the forces of pressure, viscosity and buoyant force the force of surface tension is introduced in the form of a volume source $G\nabla\varphi$ [17].

$$G = \frac{\lambda}{\varepsilon^2}(-\nabla\varepsilon^2\nabla\varphi + (\varphi^2 - 1)\varphi))$$

Here the quantity λ is the mixing energy density, ε is a capillary width that scales with the thickness of the interface. These two parameters are related to the surface tension coefficient σ_s via: $\sigma_s = (2\sqrt{2}/3)\,\lambda/\varepsilon$.

The mass flux emanating from the interface surface $\dot{m}$ is linked with the value of the heat flux at the interface as

$$\dot{m} = -\left(\frac{M_w}{\Delta H_{vl}}\right)\boldsymbol{n}\cdot k_V\nabla T_V$$

Here M_w is the molecular weight of vapor, ΔH_{vl} is the enthalpy of evaporation, k_V is the coefficient of thermal conductivity, $\boldsymbol{n}$ is the unit vector normal to the boundary.

The Cahn - Hilliard equation for φ

$$\frac{\partial\varphi}{\partial t} + \boldsymbol{u}\cdot\nabla\varphi - \dot{m}\delta\left(\frac{V_{fL}}{\rho_L} + \frac{V_{fV}}{\rho_V}\right) = \nabla\cdot\frac{\gamma\lambda}{\varepsilon^2}\nabla\psi$$

Here ψ is the auxiliary variable, governed by the equation: $\psi = -\nabla\cdot\varepsilon^2\nabla\varphi + (\varphi^2 - 1)\varphi$. γ is the mobility which determines the time scale of the Cahn - Hilliard diffusion. It is determined empirically and must be large enough to retain a constant interfacial thickness, but small enough to allow a convective flow passing through the interface. Nonwettability of the walls was used as the boundary conditions, i.e. it was considered that

$$\boldsymbol{n}\cdot\frac{\gamma\lambda}{\varepsilon^2}\boldsymbol{\nabla}\varphi = 0$$

The heat conduction equation

$$\rho c_p\frac{\partial T}{\partial t} + \rho c_p(\boldsymbol{u}\cdot\nabla)T + \nabla\cdot(-k\nabla T) = Q_s + Q_{chem} - \frac{\dot{m}}{M_w}\delta\Delta H_{Vl}$$

Terms in the right part of this equation correspond to the heating of the gas by the microwave plasma, the thermal effect due to chemical reactions and the temperature change associated with the vaporization of the liquid. Here c_p and k are the heat capacity and the thermal conductivity of the mixture; they are calculated by taking into account the volume fraction of each phase:

$$c_p = V_{f\,L}c_{p\,L} + V_{fV}\,c_{p\,V}$$
$$k = V_{f\,L}k_L + V_{f\,V}k_V$$

Q_s is a heat source, associated with the fraction of microwave energy, which goes into plasma heating: $Q_s = \alpha\sigma\left|E_m^2\right|$, σ is the electric conductivity of the plasma: $\sigma = \upsilon_{col}\varepsilon_0\,n_e/n_c$, υ_{col} is the collision frequency of electrons with neutrals, ε_0 is the dielectric permeability of vacuum, $n_c \approx 7\times10^{10}$ cm^{-3}is a critical value of plasma

density for a given frequency of the microwave field f = 2.45 GHz. α is a fraction of the microwave energy, which goes into heat. Q_{chem} is the thermal effect of thermal decomposition of n-heptane: $Q_{chem} = -H_h r_h$, H_h, r_h is the enthalpy and reaction rate of decomposition of n-heptane. When calculating the value of α, it was assumed that the heat Q_s is the energy that goes into the excitation of vibrational degrees of freedom of molecules of n-heptane. It was determined from the solution of the Boltzmann equation.

For modeling of thermal decomposition of n-heptane we used a simplified approach, based on our simulations with full kinetic scheme of its pyrolisis [18]. The calculated time dependence of the n-heptane concentration for different temperatures allowed us to obtain an approximate expression for the characteristic time of n-heptane decay τ_x as a function of gas temperature T: $\tau_x = 1.46 \times \exp(-14461/(T-200))$.

The balance equation for the electron density

$$\frac{\partial n_e}{\partial t} + \nabla \cdot (-D_{am}\nabla n_e + n_e \boldsymbol{u}) = k_i \left(\frac{E}{N}\right) * [C_7H_{16}] * n_e - k_{rec} * n_e^2$$

Here D_{am} is coefficient of ambipolar diffusion, $D_{am} \approx \mu_i \varepsilon_x$, k_i, k_{rec} are coefficients of ionization and volume recombination of the main ion of n-heptane respectively, μ_i is mobility of the main ion, ε_x is characteristic energy of the main ion.

For calculation of the ionization constant of n-heptane we used the total ionization cross section, measured in [16]. Measured partial ionization cross sections of n-heptane are also presented in [16]. It is shown that near the ionization threshold (~10 eV) the main ion is the molecular ion $C_7H_{16}^+$ and above 16 eV the main ion is ion $C_3H_7^+$. The analysis made with our previously calculated EEDF showed that in the range of our interest, i.e. at values E/N <400 Td, the ion $C_7H_{16}^+$ can be considered as the main ion with accuracy ~20%.

The modeling has been carried out numerically, using finite element method implemented in the commercial package Comsol 3.5a [19]. The system usually reached stationary or periodic temporal distribution of the spatial variables at t ~ 0.1-1 sec after about 2-3 hours of computation on 12-core 2.3GHz Xeon of 64 bit system with 32 GiB of RAM.

3 Results and discussion

The simulations allow tracing the evolution of the boiling process. Initially a small bubble of saturated gas is placed inside of the cavern. Scenario of further process strongly depends on the input microwave power P_0. When the power of the microwave signal supplied to the system exceeds 100 W, the periodic separation of bubbles with their further ascension begins. At a power greater than 500 W the first bubble is formed and rises from the cavern to the surface, after which a pillar of saturated gas follows the bubble and also lifts to the surface.

The diameter of the resulting bubbles is comparable to the diameter of the cavern. In our case it is a few millimeters. The fluid velocity is about a centimeter

per second. The plasma exists only inside the cavern, in close proximity to the central electrode. When the bubble separates from the cavern and leaves the electrode, the plasma disappears inside it. This is because the microwave field is concentrated near the end of the central electrode and falls very sharply outside.

Note that the simulations allowed determining the characteristic times of the processes of the bubble formation, its isolation and further floating up to the surface. The snapshots of the boiling process at time t=0.1 s at various values of the microwave power are shown in figure 2. The higher the level of input microwave power, the greater the speed of ascent and diameter of the bubble.

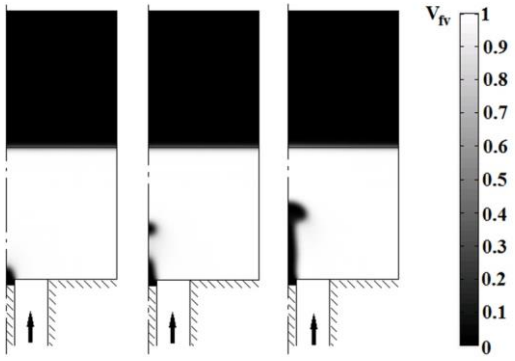

Fig. 2. Snapshots of the volume fraction of the vapor phase V_{fv} at time t=0.1s for different levels of input microwave power. From left to right: P_{inc} = 300, 500, 700 W. Simulations

For comparison, the gas bubbles in boiling n-heptane obtained in simulations and in the experiment are shown in figure 3. In experiments the microwave discharge in boiling n-heptane is sustained at the end of the microwave antenna immersed in a liquid hydrocarbon. The detailed description of the experimental setup can be found in [7-9]. In the simulations the microwave antenna was located just below the bottom of the tank with n-heptane, and the initial bubble was placed inside the cavern at the end of the antenna. Since the geometries of the microwave antenna are different, we note only the qualitative proximity of the emerging bubble.

Conclusion

The present work is the first self-consistent two-dimensional model of the microwave discharge in liquid n-heptane, which takes into account a number of key physical and chemical processes in the boiling liquid and plasma: evaporation, heat transfer, forming and ascent of bubbles, microwave heating, cooling due to the thermal dissociation of n-heptane, its ionization and recombination. The first results have been achieved. The model allows looking after the evolution of the boiling process and estimating the characteristic times of the process of formation of bubbles.

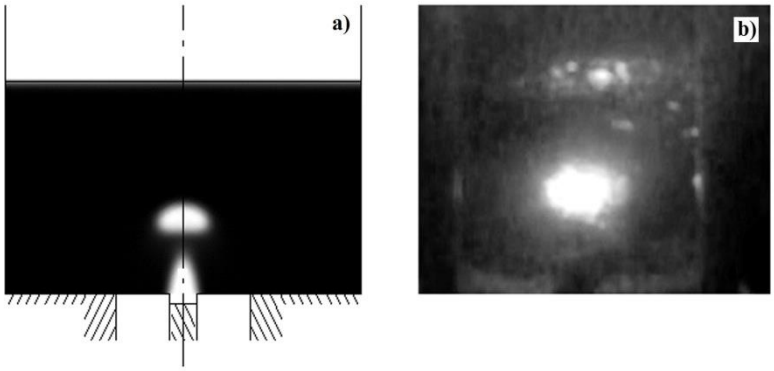

Fig. 3. a) Snapshots of the volume fraction of the vapor phase V_{fv} at time t=7×10^{-2} s at P_{inc} = 1 kW. Simulation. b) Photograph of the discharge at time t=1.2×10^{-2} s after the ignition. P_{inc} = 500 W. Experiment

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