

# Growth of Droplet in Radiation-Induced Plasma of Own Steam

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**Abstract**—The theoretical approach is developed to describe the change of drops in the atmosphere of own steam and buffer gas under irradiation. It is shown that the irradiation influences on size of stable droplet and on the conditions under which the droplet exists. Under irradiation the change of drop becomes more complex: the not monotone and periodical change of size of drop becomes possible. All possible solutions are represented by means of phase portrait. It is found all qualitatively different phase portraits as function of critical parameters: rate generation of clusters and substance density.

**Keywords**—Irradiation, steam, plasma, cluster formation, liquid droplets, evolution.

## I. INTRODUCTION

THE essential property changes in structural materials are induced by irradiation [1].

It was established experimentally that irradiation can significantly change the conditions of the phase-structural transformations and technological conditions for the coexistence of substances that are in different phases. Investigation of radiation influence on the conditions for the coexistence of disordered substances, including gases and liquids is of particular interest in the design of nuclear equipment and new-generation reactors, for instance molten-salt reactors.

Processes in gases and liquids under irradiation are actively studied [2], [3]. However, the kinetics of phase changes in gases and liquids under irradiation analyzed insufficient, and many aspects are still not been investigated.

Irradiation changes the properties of the gas and the liquid and affects the processes near their interface [4]. Molecules are excited, ionized and destructed under irradiations. The energy of internal degrees of freedom of the molecules is redistributed. The equilibrium velocity distribution and the equilibrium structure of short-range ordering of the liquid are disturbed. The ionized and excited molecules often react with neutral molecules and form stable clusters [5], [6].

The substance (primarily the liquid phase, where the relaxation passes faster) is heated due to relaxation of the radiation-induced excitations. A liquid surface is "spattered" by high-energy particles and small droplets can be destroyed by irradiation. Generation of surface disturbances, changes in

the structure of the liquid surface lead to a change in evaporation and absorption of molecules.

The peculiarity of radiation effects is, first of all, in the combination of various mechanisms operating at very different time scales. Radiation-induced excitation and ionization of molecules lasts about  $10^{-13}$  seconds. An excited molecule can transfer energy to an unexcited molecule. Various restructuring of excited (ionized) molecules occur within  $10^{-10}$  sec. As a rule, restructuring leads to primary damage of molecule and radical formation. Active products, such as excited molecules, ions and free radicals interact with each other. New radicals are generated; molecules are changed; and clusters are formed very active. These processes last about  $10^{-6}$  s.

Characteristic time of variation of the droplet size is considerably greater than the characteristic times of each of these processes.

## II. STATEMENT OF THE PROBLEM

Let us consider droplets in atmosphere of own steam under irradiation. Let us consider stage of growth of droplet when nucleation of new droplets can be neglected, their concentration is constant, and they all have approximately the same size. Due to irradiation weakly-ionized plasma is generated. The clusters of molecules are formed as result of the excitation and ionization of molecules of the steam. The droplet grows by absorbing both molecules and their clusters. Reducing the size of the droplet is due to thermal emission its individual molecules. Speed of emission depends on droplet radius and temperature. Since cluster formation is much faster the change of droplet size, at studying growth of the droplet we assume in our model that the formation of the cluster is instantaneous. It means that kinetics of cluster formation is not studied and we use merely a given number of clusters that are generated per unit time under irradiation. Cluster size is assumed to be equal and lower than threshold of formation of liquid droplets. Life-time of cluster is function of temperature. Clusters can diffuse and their diffusion coefficient can be distinguished from the one of molecules.

Let  $V$  is volume per one drop. The total number of molecules in this volume  $n_0V$  is constant. It consists of molecules of steam ( $n_gV$ ), molecules included in clusters ( $mnV$ ) and in droplets ( $4\pi R^3/3v_0$ ). Thus

$$n_0 = n_g + mn + (4\pi NR^3)/3v_0 \quad (1)$$

Rate of change of the average density of the complexes ( $n$ ) and the droplet radius ( $R$ ) are described by the equations

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This work was made possible by support from NRF of South Africa.

$$\frac{dn}{dt} = Kn_g - n/\tau - 4\pi NR D_c n \quad (2)$$

$$\frac{dR}{dt} = \frac{v_0}{R} (D_g n_g + m D_c n - D_g n_g^{eR}) \quad (3)$$

Here  $n_g^{eR} = n_g^e \exp(2\sigma v_0 / RkT)$  and  $n_g^e$  are equilibrium densities of molecules near droplet and far away from it.  $N = I/V$  is number of droplet per volume;  $m$  is number of molecules in the cluster.  $K$  is part of molecules that ionizes (excites) per time.  $\tau$  is life-time of cluster.  $v_0$  is volume per molecule in droplet,  $D_g$  and  $D_c$  are diffusion coefficients of stream molecules and clusters.

A theoretical approach to evolution of complex density and the droplet radius is developed via formalism of Poincare. Our goal is to identify all possible quality different solutions of system (1), (2) for different conditions and to build corresponding phase portraits.

Due to conservation of the total number of molecules the variables are limited and satisfy the following inequalities:  $n_0/m \geq n \geq 0$ . The equalities are reached when all molecules are included in clusters and when there are no clusters.  $(3v_0 n_0 / 4\pi N)^{1/3} \geq R \geq 0$ . The equalities are reached when all molecules are included in droplets and when there are no droplets.

Let us introduce new variables:  $x = m D_c n / D_g n_g^e$ ,  $y = R / r_0$ ,  $t' = t / \tau$  and new parameters:  $r_0 = (4\pi N / 3)^{-1/3}$ ,  $y_0 = 2\sigma v_0 / r_0 kT$ ,  $x_0 = n_0 / n_g^e$ ,  $\mu = D_g / D_c$ ,  $\xi = 1 / v_0 n_g^e$ ,  $\alpha = 3\tau D_c / r_0^2$ ,  $\beta = mK\tau / \mu$ ,  $\gamma = \tau D_g / \xi r_0^2 = \alpha\mu / 3\xi$ .

Then system of (2), (3) transform into

$$\frac{dx}{dt'} = f_1(x, y) \equiv \beta n_0 - (1 + \beta\mu)x - \alpha\gamma y - \beta\xi y^3 \quad (4)$$

$$\frac{dy}{dt'} = f_2(x, y) \equiv \frac{\gamma}{y} (x_0 + (1 - \mu)x - \xi y^3 - \exp(y_0/y)) \quad (5)$$

and variables must satisfy inequalities

$$x_0 - \xi y^3 \geq \mu x \geq 0 \quad (6)$$

The changes of the droplet radius ( $y$ ) and density of clusters ( $x$ ) are completely determined by (4), (5) and by the values of the droplet size and the density of clusters in the initial time. A set of initial conditions defines a family of solutions, one for each choice of the initial condition. This family can be divided into classes of qualitatively different solutions. Every class

includes qualitatively similar solutions. Solutions belonging to different classes are qualitatively different. A clear and adequate representation of such a partition into classes is given by the phase portrait. The purpose of this paper is not to find explicit solutions but to give an exhaustive description of all solution classes and the change of the partition into classes with the change of control parameters. We use as control parameters  $\beta$  and  $x_0$ . The first control parameter is related to the rate of radiation-induced generation of clusters, the second one is related with the total amount of molecules in the considered volume.

### III. ISOCLINES AND STATIONARY SOLUTIONS

Structure of the phase portrait is determined by critical points (stationary solutions) of systems (4), (5) and by their topological type. The critical points are intersection of  $dx=0$  and  $dy=0$  isoclines. The first isocline is

$$x = X_I(y) \equiv \frac{\beta(x_0 - \xi y^3)}{1 + \beta\mu + \alpha\gamma} \quad (7)$$

At each point of this isocline the tangent to the phase trajectories are parallel to the  $y$ -axis ( $dx = 0$ ).

The second isocline is

$$x = X_{II}(y) \equiv \frac{x_0 - \xi y^3 - \exp(y_0/y)}{\mu - 1} \quad (8)$$

At each point of this isocline the tangent to the phase trajectories are parallel to the  $x$ -axis ( $dy = 0$ ).

Function (7) decreases monotonically from  $x = \beta x_0 / (1 + \beta\mu)$  at  $y=0$  to zero at  $y = (x_0 / \xi)^{1/3}$ . The function (8) has a single extremum and vertical asymptote  $x=0$ . The form of the isoclines shows that the isoclines intersect either one or two times, or isoclines do not intersect.

In order to determine the region of parameters for which there are stationary solutions, find the bifurcation point and the topological type of the critical points, we exclude  $x$  from (7) and (8) and represent one of the control parameters as a function of  $y$

$$\beta = B(y) \equiv \frac{-(1 + \alpha\gamma)Z(y; x_0)}{D(y; x_0)}$$

where

$$D(y; x_0) = x_0 - \xi y^3 - \mu \exp(y_0/y),$$

$$Z(y; x_0) = x_0 - \xi y^3 - \exp(y_0/y).$$

Zeros of the function  $B(y)$  satisfy equation  $Z(y; x_0) = 0$ .

Asymptotes of  $B(y)$  are defined by equation  $D(y; x_0) = 0$ .

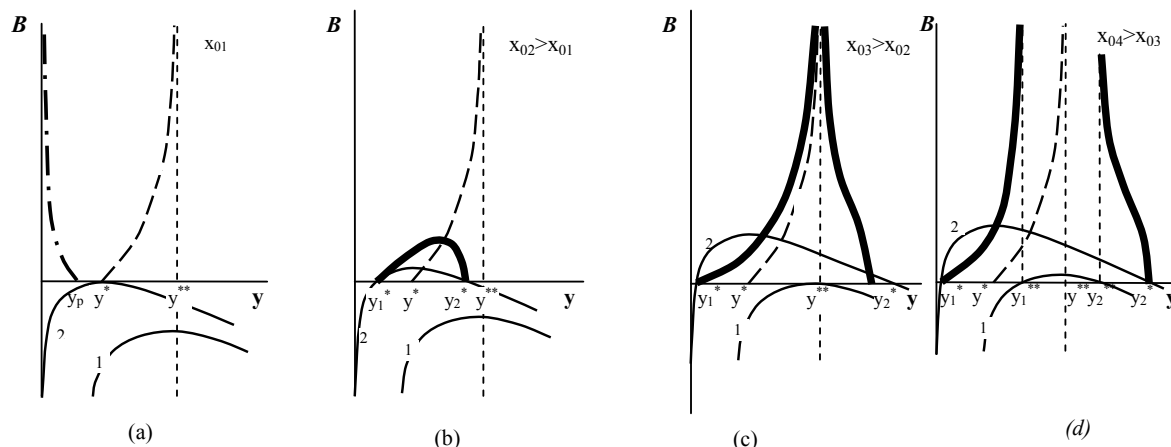


Fig. 1. Positive branch of function  $B(y)$  (for different values of  $x_0$  (The thick solid line —: 1 -  $D(y)$ ; 2 -  $Z(y)$ );  $\mu > 1$ ).

For  $\mu > 1$  the function  $B(y)$  has the only maximum and for  $\mu < 1$  it has the only minimum. Let us consider a family of functions  $B(y)$ , which is born by change of the second critical parameter  $x_0$ . By  $y_1^*$  and  $y_2^* \geq y_1^*$  denote the roots of equation  $Z(y; x_0) = 0$ ; by  $y_1^{**}$  and  $y_2^{**} \geq y_1^{**}$  denote the roots of equation  $D(y; x_0) = 0$ . By definition, put  $x_0^* = \xi y^{*3} + \exp(y_0/y^*)$  and  $x_0^{**} = \xi y^{**3} + \exp(y_0/y^{**})$  where  $y^*$  is position of maximum of function  $Z(y; x_0)$  and  $y^{**}$  is position of maximum of function  $D(y; x_0)$ . The values  $y^*$  and  $y^{**}$  satisfy equations  $3\xi y^{*2} = \frac{y_0}{y^{*2}} \exp(y_0/y^*)$  and  $3\xi y^{**2} = \mu \frac{y_0}{y^{**2}} \exp(y_0/y^{**})$ ; thus they do not depend on  $x_0$ .

Let us consider cases  $\mu > 1$  and  $\mu < 1$  separately:

If  $\mu > 1$ , then  $D(y; x_0) < Z(y; x_0)$ ,  $y^* > y^{**}$ ,  $x_0^* < x_0^{**}$ . For  $x_0 < x_0^{**}$  the function  $B(y)$  is positive hence there exist no stationary solutions of the system for any  $\beta$ . A unique root of the function  $B(y)$  arises at  $x_0 = x_0^*$ . It is equal to  $y^*$  if  $\beta = 0$  (Fig. 1 (a)). With growing  $x_0$  the root falls into two roots, which are  $y_1^*$  and  $y_2^* \geq y_1^*$ . The function  $B(y)$  is positive when  $y_2^* > y > y_1^*$  (Fig. 1 (b)). Now, when  $x_0^* < x_0 < x_0^{**}$  and  $\beta < \beta_{\max}$ , there exist two stationary solution of system (4), (5):  $y_1$  and  $y_2 \geq y_1$ ,  $y_2^* > y_2 > y_1 > y_1^*$ , which coincide at  $\beta = \beta_{\max}$ . And there are no stationary solution if  $\beta > \beta_{\max}$ . Here  $\beta_{\max}$  is maximum value of positive branch of function  $B(y)$ . Here and further we find the stationary value of  $x$  using (7) or (8).

When value of  $x_0$  grows, maximum of function shifts to right and up. At  $x_0 = x_0^{**}$  vertical asymptote of function  $B(y)$  arises at point  $y^{**}$  thus two stationary solutions,  $y_1$  and  $y_2$  take

place for any value of  $\beta \geq 0$  and  $y_1^* \leq y_1 \leq y^{**} \leq y_2 \leq y_2^*$  (Fig. 1 (c)). If  $x_0 > x_0^{**}$ , the asymptote divides into two vertical asymptote at points  $y_1^{**}$  and  $y_2^{**}$ . Thus two stationary solutions,  $y_1$  and  $y_2$  take place for any value of  $\beta \geq 0$  and  $y_1^* \leq y_1 \leq y_1^{**} \leq y^{**} \leq y_2^{**} \leq y_2 \leq y_2^*$  (Fig. 1 (d)).

If  $\mu < 1$ , then  $D(y; x_0) > Z(y; x_0)$ ,  $y^* < y^{**}$ ,  $x_0^* > x_0^{**}$  (value of  $x_0^*$  does not change, value of  $x_0^{**}$  decreases). For  $x_0 < x_0^{**}$  function  $B(y)$  is negative thus there are no stationary solution of system (4), (5) for any  $\beta$ . At  $x_0 = x_0^{**}$  vertical asymptote of function  $B(y)$  arises at point  $y^{**}$  and two stationary coincident solutions,  $y_1 = y^{**} = y_2$  arise as  $\beta$  tends to infinity ( $\beta \rightarrow \infty$ ). With increasing  $x_0$  ( $x_0 > x_0^{**}$ ) the asymptote divides into two vertical asymptote at points  $y_1^{**}$  and  $y_2^{**}$ . The function  $B(y)$  is positive between the asymptotes. Thus the system (4), (5) has two stationary solutions,  $y_1$  and  $y_2$  ( $y_1^{**} \leq y_1 \leq y_2 \leq y_2^{**}$ ) for  $\beta > \beta_{\max}$ . They coincide if  $\beta = \beta_{\max}$ . If  $\beta < \beta_{\max}$ , there is no stationary solution. Here  $\beta_{\max}$  is minimum of the positive branch of function  $B(y)$ . With increasing  $x_0$  the minimum of function  $B(y)$  shifts to right and down. At  $x_0 = x_0^*$  the minimum of function  $B(y)$  touches x-axis at point  $y^*$  thus two stationary solutions,  $y_1$  and  $y_2$  take place for any  $\beta \geq 0$ , and  $y_1^{**} \leq y_1 \leq y^* \leq y_2 \leq y_2^{**}$ . With increasing  $x_0 > x_0^*$  the region in which function  $B(y)$  is positive divides into two ones. The only stationary solution takes place in each region if  $\beta \geq 0$ ,  $y_1^{**} \leq y_1 \leq y_1^* \leq y^* \leq y_2^* \leq y_2 \leq y_2^{**}$ .

The stationary solution  $y_1$  is unstable saddle point. The stationary solution  $y_2$  is stable. It can be stable node or spiral.

## IV. RESULTS AND DISCUSSION

Obtained results allow to describe conditions existence of stable liquid droplets in atmosphere of own stream under irradiation and to find dependence of droplet size on parameters  $\beta$ ,  $x_0$ , and  $\mu$ , namely on substance density, intensity of irradiation, and on the ratio of the diffusion coefficients of free molecules and clusters.

Without irradiation clusters are not form ( $\beta=0$ ) and their concentration is equals to zero ( $n=0$ ). State of the system is determined by density of substance (given temperature is constant). If the value of  $x_0$  is less critical one, the droplets evaporate. If the value of  $x_0$  is more critical one, there exist two stationary droplets: stable (large) and unstable (small). If initial droplets are smaller than small stationary ones, they evaporate. If initial droplets are more than small stationary one, their size tends to large stationary size. If the value of  $x_0$  is equal to critical one, there exists bifurcation of stationary solutions: they coincide and vanish with decreasing density of substance,  $x_0$ .

A state of the system under irradiation is determined by the rate of cluster generation and substantially depends on the ratio of the diffusion coefficients of free molecules and clusters. It can be different [6].

When diffusion coefficient of clusters is less than the one of free molecules, then region of parameters, under which stable droplets exist, shrink. At a low density of substance but sufficient for the existence of a stationary droplets, stationary droplet size decreases to "critical", coincides with it and disappears with increasing the cluster generation rate. If density substance grows, the "critical" and the stationary radii converge to a certain limiting value, which increasing with growth of substance density. This is happen because during formation of the clusters with low mobility the substance flux to the droplet decreases.

If diffusion coefficient of clusters is more than the one of free molecules, then region of parameters, under which stable droplets exist, expand. And there can be oscillations of size droplet near the stationary value.

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## V. CONCLUSION

Irradiation has effect on existence conditions of stable droplets and on their size. Droplets can exist only if the density of substance exceeds the "critical" value. If the clusters are less mobile than the free molecules of steam, the region of existence of droplets shrinks. Otherwise, the region of existence of droplets expands under irradiation.

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