Mathematics and mechanics. Physics

UDC 544.54;544.55

NONEQUILIBRIUM PLASMACHEMICAL PROCESSES -THE BASIS OF FUTURE PLASMA TECHNOLOGIES

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Laws of molecule excitation in basic electronic condition have been considered. It was shown that nonequilibrium excitation of oscillatory degrees of molecule freedom was the most effective for initiation of chemical reactions. Plasmachemical processes proceeding in such conditions have a number of advantages allowing at their use in traditional manufactures decreasing power inputs and increasing productivity. Conditions realized at pulse excitation of gas mixes are also favorable for organizing chain chemical processes. Experimental data of chain chemical process realization in plasma of pulse electronic beam were presented. Other perspective plasma technologies pulse radiolysis of liquid phase hydrocarbons at low temperature in conditions of electronic beam influence with high current density, initiation of carbon-black formation process in nonequilibrium conditions were also considered.

Introduction

Modern large-tonnage chemical production using traditional approach – thermal activation of chemical process faces the problem of energy saving. Further development of industrial base results in increasing the volume of separate productions, unreasonable resource consumption for developing equipment, depletion of minerals, metals and fuel.

Initiation of the majority of chemical reactions requires energy consumptions for overcoming activation reaction threshold. This energy is consumed for decomposition of initial product or one of components of original mixture into reactive radicals. For this purpose various physical and chemical processes are used. Thermal heating of gas mixture is a widely used technique for overcoming activation barrier of chemical reaction. Possessing a number of advantages (simplicity of implementation, universality etc.) it has very low coefficient of energy utilization. Heating gas mixture energy should be consumed for heating other components of gas mixture, chemical reactor beside energy for initial molecule dissociation. For example, at fluorine molecule thermal dissociation gas should be heated to the temperature higher than 700 °C. In this case specific energy consumed for fluorine atom formation amounts to 54,6 eV [1] while binding energy of F₂ molecule amounts only to 1,4 eV [2]. In processes of thermal dissociation of polyatomic molecules the coefficient of directed energy use is lower as they posses higher heat capacity. Application of plasma pulsed sources is perspective due to three main reasons:

- reduction of reactor wall heating;
- implementation of chemical reactions at lower temperatures;
- selective excitation of molecule vibration level.

1. Main laws of molecule nonequilibrium excitation in plasma

Combination of reaction zone with gas-discharge one allows heating locally agents to high temperatures without heating reactor walls that reduce significantly unproductive energy loss. The given conditions are simply implemented at gas mixture excitation by continuous electron beam, in arc discharge and other. In this case reduction of reaction barrier is also achieved by participation of free radicals and atoms which are efficiently operated in gas discharges.

The next step in decreasing energy consumptions for carrying out chemical processes is the use of nonequilibrium processes characterized by significant energy excess at separate inner molecule degrees of freedom in comparison with thermodynamically equilibrium state. In this case gas temperature for a number of agents may be lower than 400 K that reduces energy consumption for heating reactor walls, initial components of gas mixture as well as simplifies hardening (stabilization) of chemical process products.

At molecule excitation (in ground electronic state) its internal energy is distributed over different degrees of freedom – to molecule translation motion, to atom oscillation in polyatomic molecules relative to each other and to molecule rotation. Molecule dissociation occurs in the case when energy at vibration levels exceeds certain value. Molecule atom oscillations relative to each other cause the increase of interatomic distance to the value at which force of intermolecular bindings becomes low that results in molecule breakup. Molecule translational motion or its rotation do not result in dissociation (without collisions with other molecules or reactor wall) therefore, energy of translational and rotational motion is unproductive one from the point of view of initiation of chemical reactions. It is obvious that the most efficient method of molecule dissociation and chemical reaction initiation is selective vibration (nonequilibrium) excitation as in this case energy consumption (to translational and rotational motion) decreases.

High efficiency of chemical reaction initiation due to selective excitation of vibration molecule degrees of freedom caused intensive researches of various ways of molecule nonequilibrium excitation. For today the following main methods may be singled out: chemical excitation, gas-dynamic gas expansion, excitation by optical radiation and electron impact in plasma. Selective excitation of vibration molecule degrees of freedom in plasma is the most universal method used for chemical reaction initiation in various gas-phase and liquid-phase compounds.

As experimental investigations showed at collision of electron having kinetic energy in the range of 0,5...3 eV with a molecule the efficient excitation of vibration and rotational degrees of freedom of this molecule occurs [3, 4]. At energy of incoming electron of 0,5...1 eV up to 90 % of its energy is consumed to excitation of vibration levels, it is right as well for other molecules [3].

But dissociation energy of the majority of studied molecules is in the range of 4...10 eV [2]. The efficiency of transferring such energy quantum to molecule vibration levels at molecule interaction with one electron is very low. Energy loss of electron with initial energy more than 4 eV at collision with molecule usually used for its ionization or excitation of electron subsystem. Investigations [3, 4] showed high efficiency of molecule dissociation at excitation of CO_2 molecule by electronic impact.

Dissociation efficiency was calculated as ratio of energy consumed for conversion of CO₂ initial molecules (product of molecule dissociation energy and amount of decomposed molecules) to energy transferred by electronic impact to molecule vibration levels. At electron energy about 1 eV almost all energy transferred to molecules by electrons is consumed for their dissociation. And the region of specific heat input in which dissociation efficiency is the highest coincides with the region in which incoming electron energy is mainly consumed for vibration level excitation [3]. It is conditioned by the fact that relaxation process of molecule vibrationly excited molecule to equilibrium state occurs as a result of multiple collisions (Z>100) of excited molecules with unexcited ones (or with reactor walls) [5, 6]. Many molecules are known for which Z at homomolecular collisions may be equal to several tens of thousands at general relative molecule rate.

At vibration relaxation the collision of excited molecules and exchange of vibration quanta occur as well. But because of anharmonicity of atom oscillations in molecule the opposite exchange processes of vibration quanta of molecule are of asymmetric character [7]. At collision of two molecules having different energy at vibration levels the additional excitation of highly excited molecules occurs but not stabilization of energy reserve at vibration levels of collided molecules. This mechanism (Trinor mechanism) results in energy rise at vibration levels of separate molecules to the level exceeding dissociation energy and as a result their breakup.

These features of transfer process of incoming electron energy to a molecule and the process of intermolecular exchange of vibration energy as well as dissociation mechanism of vibrationly excited molecules in ground electronic state determine high efficiency of plasma-chemical process.

2. Chain plasma-chemical processes

Significant success in development of sources of high-current pulse electron beams [8] initiated investigations of plasma formation processes by high-energy electrons and channels of their energy dissipation. In comparison with volume discharges the pulse electron beams allow easily solving the problem of excitation of large volumes of reaction gas at high pressure that is important for developing high-capacity process installation.

Survey of experimental investigations [9] showed that pulse electron beam is inefficient for carrying out direct plasma-chemical processes in which energy of electron impact is the only source of chemical reaction initiation. In the case of plasma formed by electron beams in plasma chemistry only low-energy electrons (in comparison with energy of beam electrons) formed in relaxation process of primary electron energy at formation of ionization cascade of secondary electron plays role. Small part of energy of beam accelerated electrons is consumed for excitation of molecule vibration levels. Calculations made by the authors [10, 11] showed that at electron kinetic energy more than 30 eV the degradation cascade energy in nitrogen is generally consumed for ionization and excitation of molecule electron level. Electron energy loss for excitation of rotational, vibration and translational degree of freedom of molecule ground state at low energy does not exceed 17 % and decreases to 10 % at electron energy increase. As a result of this electron beam energy consumption for decomposition of one molecule of initial gas mixture exceeds significantly (in 10...100 times) its dissociation energy.

Estimating efficiency of carrying out plasma-chemical processes in the case of gas mixture excitation by pulse electron beam it should be also taken into account that accelerator efficiency (ratio of electron beam energy to total energy consumed by accelerator) does not exceed, as a rule, 50 %. Energy consumption for decomposition of initial molecule exceeds respectively 20...40 eV.

But forming low-temperature plasma by pulse electron beam in comparison with many other methods significant decrease of energy consumption for gas-phase compound conversion is revealed in some cases. The analysis of experimental works devoted to decomposition of various compounds impurities (NO, NO₂, SO₂, CO, CS₂ and others) in the air by pulse electron beam showed that its energy consumption for decomposition of one gas molecule is even lower than its dissociation energy [9]. It is conditioned by the fact that at beam influence the conditions favorable for chain processes are formed. At sufficient chain length the electrophysical device supports insignificant part of total energy consumption for chemical process. The main source in this case is thermal energy of initial gas or energy of exothermic chemical reactions of chain process (for example, oxidation or polymerization reactions). It is important to mention that carrying out chemical process at temperature lower than equilibrium one allows synthesizing compounds unstable at higher temperatures or selectivity of synthesis of which is low at high temperatures. Temperature decrease at chain chemical process is similar to catalytic effect. Chain process may occur in the whole gas volume that increases significantly reaction velocity in comparison with heterophase catalytic process. Reaction high velocity required for industrial technologies is supported by branched chain processes. But their considerable disadvantage is connected with process explosive behavior that increases significantly production danger. This disadvantage is removed by chain process initiation out of self-ignition region at external influence.

In paper [12] it is shown that at certain values of current density of pulse electron beam sulfur dioxide oxidation may occur at low energy consumption (lower than 1 eV/molecule SO₂) as a result of chain process with ion-molecule reactions of chain continuation. Further development of experimental investigations of chain processes in plasma created by electron pulse beams was attempted by a group of Yu.N. Novoselova in the Institute of electrophysics RAS UD [9]. The results of investigations of electron pulse beam influence on mixtures simulating combustion gases were presented as well. In particular, the influence of electron beam parameters, external electric field and composition of irradiated gas on the process of its purification from SO₂ was examined. Existence of optimal values of current density of electron beam, its pulse duration and strength of external electric field at which consumptions for removing one molecule of SO₂ are minimal (0,7 eV/molecule) was shown. The boundary of separation of chain and radical mechanisms of purification was determined by SO₂ concentration.

Similar investigations were carried out in removing molecules of NO and NO₂ from modeling mixture radiated by pulse electron beam of microsecond duration [9]. It was obtained that at low concentrations of NO the value of energy consumption is significant and amounts to tens of eV/molecule. The increase of [NO]₀ results in energy consumption decrease. At [NO]₀=0.55 % its value decreases to 2 eV/molecule (NO dissociation energy equals 6,5 eV, O_2 –5,12 eV). It indicates the implementation of chain mechanism of NO decomposition in these conditions. The increase of pulse duration or electron beam current density (that is increase of specific heat input) causes the decrease of purification efficiency and increase of energy consumption for removing one toxic molecule. Minimal values of energy consumption for decomposition in these tests amounted to the value 3...4 eV/molecule and corresponded to minimal values of pulse duration and current density of electron beam which might be implemented at the device used in the test. Similarly to experiments on decomposition of SO₂ admixture in air the decrease of purification degree and growth of energy consumption at increase of absorbed dose of electron pulse beam (at increase of pulse duration or current density) indicate the implementation of new mechanism of admixture decomposition.

In [13] the results of studying the process of influence of pulse electron beams of nano- and microsecond duration on nitrogen-oxygen mixture of atmospheric pressure with impurity of carbon disulfide CS_2 are given. Two modes of impurity conversion depending on parameters of used beam and differ both in mechanism and end products were revealed. At irradiation of mixture with nanosecond beam of electrons it was obtained that minimal value of energy consumption was equal $0.8 \, \text{eV/molecule}$ that is considerably lower than dissociation energy of carbon disulfide molecule equal $7.6 \, \text{eV}$. The process of CS_2 conversion in non-self-maintained volume discharge initiated by nanosecond beam was studied as well. It was obtained that in the range of

strength of electric field E from 0 to 1,5 kV/cm the value of energy consumption changes insignificantly and is in the range of 0,6...0,9 eV/molecule. The increase of field strength results in significant growth of energy consumption the value of which at E>10 kV/cm exceeds 35 eV/molecule.

In the mode of mixture irradiation with the beam of microsecond duration the linear decrease of CS_2 concentration at growth of radiation pulse amount was stated as well. In the range of $[CS_2]_0$ =0,3...1% energy consumption for carbon disulfide conversion amounted to 1,8...1,2 eV/molecule respectively. Low values of energy consumption indicate the fulfillment of chain process. The model of plasma-chemical processes initiated by electron beams in nitrogen-oxygen mixture with carbon disulfide impurity was developed for analyzing mechanisms of CS_2 conversion.

SF₆ decomposition was experimentally studied at special-purpose pulse electron accelerator TEU-500 (500 kV, 200 kA/cm², 60 ns) [14]. It was obtained that in conditions of plasma formed by pulse electron beam SF₆ decomposition in the mixture with N₂ occurs efficiently [15]. For the mixture SF_6+N_2+Ar (6:6:1) electron beam energy consumption for dissociation of one molecule of SF₆ amounted to 5,0 eV that is lower than standard formation enthalpy equal 12,4 eV/molecule. It indicates the implementation of chain process initiated by pulse electron beam. The results of experimental investigation of SF₆ decomposition in the mixture with H₂ under the influence of pulse electron beam are given in the work [16]. Electron beam energy consumption for SF₆ decomposition was lower than 2 eV/molecule. It confirms that SF₆ conversion in the mixture with H₂ occurs as well by chain mechanism.

The results of experimental investigation of WF_6 decomposition in the mixture with nitrogen under the influence of pulse electron beam at 300 K are given in works [16, 17]. Electron beam energy consumption for decomposition of one molecule of WF_6 did not exceed 0,24 eV. The reason of abnormally low energy consumption (rather low than formation enthalpy of WF_6 –17,8 eV [2]) for recovering tungsten from WF_6 under the influence of pulse electron beam is chain reaction occurrence in plasma-chemical reactor.

Under the influence of electron beam on mixture SiCl₄+H₂+Ar at increase of pulse amount the diminution of SiCl₄ and HCl life were recorded [18]. Powder of dark color which obtained white color after letting-toair into reactor was formed in the reactor volume [19]. In this case electron beam energy consumption for dissociation of SiCl₄ molecule amounted to 1,9 eV (uppers estimate without taking into account electron beam energy loss for ionization and excitation of Ar and H₂). It is considerably lower than dissociation energy of SiCl₄ equal 6,6 eV [2]. The process of SiCl₄ decomposition at pulse electron beam influence had a number of peculiarities. The products of plasma-chemical process (controlled by pressure change in close reactor) were heated to the temperature exceeding significantly heating by electron beam only. The reason of low energy consumption of a beam for SiCl₄ decomposition and significant gas heating is chain reaction occurrence in plasma-chemical process at energy output.

3. Perspective plasma technology

Studying decomposition of liquid-phase hydrocarbons at heating showed that the process is implemented as a chain one (thermal cracking) [20]. At low temperatures (~400 K) when reaction thermal initiation does not occur without radiation the active centers – free radicals which may start chain reaction of hydrocarbon cracking occur at irradiation. Interaction of radicals containing small number of carbon atoms with initial hydrocarbon is the pronounced chain process which is initiated by radicals.

But at interaction of ionizing radiation with substance in liquid phase the main part of secondary electrons formed as a result of medium ionization has rather low path owing to its comparatively low energy and therefore, generates ionization and excitation in immediate vicinity to the place of its formation. This effect observed at liquid media radiolysis obtained its name «effect of a cell» of Frank-Rabinovich. The influence of the effect of a cell comes to maintenance of high concentration of radicals and long-living excited products formed in dikes already after termination of primary excitation acts during some time. The influence of this effect is especially evident at general yield of radiolysis products which are formed less in liquid phase than in gas phase. It results in the fact that radiation chemical yield does not increase molecule unit per 100 eV at temperature lower than 600 K.

Pulse radiolysis of liquid phase hydrocarbons at low temperature in the conditions of influence of electron beam with high current density (more than 200 A/cm²) and sum kinetic electron energy in a pulse up to 250 J is of great interest. In this case the conditions favorable for multiple collisions of particles with excess energy on internal degrees of freedom are formed. These conditions are similar to nonequilibrium excitation of gas phase medium and promote chain reactions. High power of absorbed radiation dose is obligatory as well for developing such conditions of radiolysis. Lifetime of active radicals formed in track at radiolysis does not exceed 10...100 ns therefore duration of radiation exposure should be less than 100 ns. Radiolysis of liquid hydrocarbons in these conditions has not studied and it is of scientific and practical interest for studying mechanism of radiation cracking of liquid phase hydrocarbons at low temperature.

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Another perspective direction of applying high-current pulse electron beams is initiation of the process of sooting in nonequilibrium conditions. It is known that conversion of hydrocarbon fuels at combustion pass the stage of formation of polycyclic aromatic hydrocarbons. They are centers of formation of carbon-black particles [21]. Radical-precursors are required for sooting therefore the process in equilibrium conditions does not occur at low temperatures. Besides, carbon-black precursors are pyrolyzed and oxidized at higher temperature so that sooting in equilibrium conditions (1000...2000 K) finishes by pyrolytic carbon synthesis. Radical formation at influence of pulse electron beam on hydrocarbon gas mixture with oxygen may initiate the process of formation of polycyclic aromatic hydrocarbons at temperature lower than equilibrium sooting. In these conditions the process may be finished at the stage of aromatic hydrocarbon synthesis if duration of external influence does not exceed duration of this stage. It allows developing new technology of synthesis of synthetic liquid fuel or complex hydrocarbons of isomerous structure of natural gas.

Conclusion

The results of investigations indicating the decrease of specific energy consumption at implementation of some chemical reactions in conditions of pulse plasmaformation are given. Laws of molecule excitation in ground electronic state are examined. It is shown that nonequilibrium excitation of vibration degrees of freedom of molecules is the most efficient for initiation of chemical reactions. Conditions implemented at pulse excitation of gas mixtures are favorable for organizing chain chemical processes. Experimental data of implementation of chain chemical processes in plasma of pulse electron beam are given.

Use of pulse electron beam supports chain reaction occurrence in gas phase at comparatively high pressure (up to 1 atm) in volume more than 1 litre. Other perspective plasma technologies — pulse radiolysis of liquid phase hydrocarbons at low temperature in conditions of influence of electron beam with high current density, initiation of sooting in nonequilibrium conditions are considered. It is shown that peculiarities of gas phase chemical processes in conditions of pulse plasma influence indicate the possibility of their application in large-tonnage chemical production.

Work is fulfilled at support of RFBR, grant 06-08-00147.

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Received on 7.12.2006

UDC 537.9

FEATURES OF HIGH-FREQUENCY CAPACITOR DISCHARGE COMBUSTION PROCESS IN MEDIA WITH DISPERSE PHASE

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Measurements of current, voltage and amplitude of electric field of dusty high-frequency capacitor discharge have been carried out. Increase in the current proceeding in discharge at its dustiness by substances with potential ionization less than 7 V was established. Electromagnetic wave parameters extending in dusty high-frequency capacitor discharge were calculated. The absence of characteristics change in discharge electromagnetic field at degrees of its dustiness $v < 10^2$ was shown.

Gas discharges in plasma technology are usually used only as a source of plasma jet. However, substance processing in gas discharge directly has a number of advantages in comparison with substance processing in plasma jet. First of all, higher temperature of plasma and lower axial temperature gradient are typical for gas discharge. Presence of electric fields and higher concentration of electrons in discharge area make plasma catalytic properties more defined.

It is more optimal to use high-frequency discharges of capacitive type to carry out processes in discharge area. Discharges of this type have large volume of discharge plasma at low level of power supplied to discharge. Walls of plasma-chemical reactor and intensification of processes occurring in plasma may be refined by amplitude modulation.

In this work electro-physical characteristics of high-frequency capacitive discharge dusty by dielectric and conducting materials were measured. The diagram of experimental set-ap is given in Fig. 1. High-frequency ca-

pacitive discharge with a ring electrode with the diameter of 48 mm was used. Presence of the second grounded electrode, as experience has shown, does not substantially influence [1] the characteristics and modes of capacitive discharge combustion. The discharge was excited in quartz tube with the diameter of 36 mm. Discharge combustion was carried out in the air at atmospheric pressure. Discharge power varied from 1 to 3 kW. Electromagnetic field frequency amounted to 40 MHz. Dispersion of materials which were used for discharge dust amounted to 20...60 mkm. Powder was supplied to central discharge area by pneumatic feeder. Consumption of plasma forming gas amounted to 0,6 m³/h.

Voltage-current characteristics of discharge in free consumption mode and in the case of discharge plasma dustiness by dielectric and conducting particles were measured. The degree of discharge plasma dustiness determined as a ratio of spray substance volume to total volume of discharge chamber changed from 0 to 10⁻⁴. Discharge dustiness degree was measured by determine