

## Universal Concept of Self-Synchronization of the Micro/Nano- Structures of the Energetic Materials Reactionary Zones and Micro-Scale Combustion Mechanisms

Research Article

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### Abstract

Presented research is devoted to further development of ideas of famous Russian scientist Ya.B. Zeldovich in the new area of physics - the physics of self-organizing systems in the active medium. According our hypothesis the macro-scale phenomena at the energetic materials combustion is result of self - synchronization of the micro/nano - structures of the reactionary zones. Motivated excitation source of the micro/nano - physical structures in the reactionary zones are the micro/nano - structures of the electro-magnetic fields. Our hypothesis is supported by the recent data, obtained in the model experimental systems. Micro/nano- structures in the reactionary zones is the platform for effective control by combustion processes of the energetic materials at macro-level and also programming of them. From suggested concept follows that each energetic material has a unique frequency electro - magnetic code of the reactionary zone which is the synergetic oscillatory system. The suggested concept opens new possibilities for development and micro-scale synthesis of the advanced propulsion materials through programming by the electro-magnetic field micro/nano - structures in the reactionary zones.

### Introduction

Yakov Borisovich Zeldovich worked successfully in many fields of science, from chemistry to the theory of elementary particles. He was successful everywhere. Ya.B. Zeldovich have made a definitive contribution in a new area of physics - the physics of self-organizing systems in active medium. Presented research is devoted to further development of his ideas at the new qualitative level. In the last decades, researchers have observed the excitation of micro/nano - structures and the presence of micro-torches over the burning surface of the energetic materials (EM), [1-7]. It can be stated now that such local unstable behavior is typical for the EM with well pronounced exothermic reactions in the condensed phase and evaporation on the burning surface. Both experiments and theory confirm that the micro/nano - structures excitation is a rather universal phenomenon [8, 9]. More than 50 years ago, by the numerous experiments begun by Ya.B. Zeldovich, had been established the excitation of regular spatial & time structure at stationary propagation of real processes of combustion and detonation explosion [10]. These micro/nano - structures play a role of oscillatory micro-systems in the reactionary zones. The micro/nano - structures arising in the

reactionary zones has a significant influence on controllability of ignition and combustion processes in the micro-combustion chambers. The reactionary processes of the EM can be classified as the synergetic objects.

### The Oscillatory Micro-Systems in the Reactionary Zones

As noted by Novozhilov, B.V., the solid propellant burning surface represents the oscillatory system with infinite number of freedom degrees [11]. However, such composite systems cannot be understood, analyzing their parts separately. In 1951 Zhukov, B.P. has shown, that at the EM burning on the boundary between of solid and gas phases there is a liquid-viscous layer (LVL) [12]. These data were received by the experimental way. Namely from that time has appeared opportunity for analyze the EM combustion instability and abnormal physics-chemical processes at the EM burning at a new qualitative level. Later, the existence of a melt layer was proposed by several researchers (Beckstead & Hightower, 1967 [13] Tanaka & Beckstead, 1996 [14]), (Jeppson, Beckstead and Jing, 1998, [15]); they made reference to the melt layer and estimated its thickness.

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In solid rocket-propellant combustion, the dynamic phase change from condensed to gas occurs across the melt layer (Figure 1). The melt layer maintains uniform thickness as it propagates after the initiation and growth steps [16].

The melt-layer initiation occurs upon ignition of the solid-state propellant. Across the melt-layer front, density experiences a sharp gradient, going from the condensed to the gaseous side. Following the initiation, the thickness of the hot melt-layer front grows until reaching a uniform thickness. Because the melt-layer thickness depends on pressure, the thickness during the growth step is much thicker than that observed during the propagation step. As the surface regression progresses, the pressure also rises as the flame temperature increases. When the ambient pressure is high, the melt-layer thickness is thin; when the ambient pressure is low, the melt-layer thickness is thick.

### Excitation of the Electric Charges in the Reactionary Zones

In accordance with recent experimental results [18, 19], the electro-magnetic phenomena play a key role both in the LVL and in the flame zone of the burning EM. Stability of the micro-torch structures over the burning surface is provided by the electro-magnetic field cellular structures in the LVL. The micro-structures arising in the LVL enter into interaction with flame zone and induce local changes of the area of the flame surface and intensity of heat release.

### Liquid - viscous layer

At heating from above, in the EM thin LVL the thermo-electric convection excitation occur, which induces cellular movement and formation of the synergetic micro/nano - structures. From the qualitative point of view, it can be understood that the fluctuation of temperature, due to non-uniform heating of the LVL, introduces the thermo-electric field fluctuation and it also causes fluctuation of the volumetric charge which is connected with this field. Influence of stationary, "external" thermo-electric field caused by non-uniformity of heating on the fluctuation

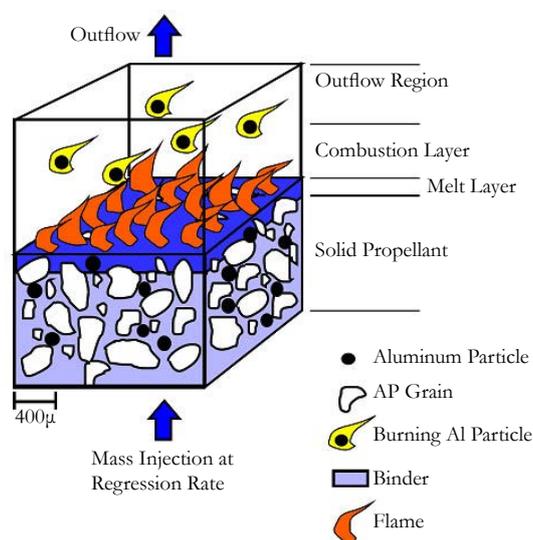
electric charge, creates the volumetric thermo-electric force. In case this force exceeds the dissipation forces, it can induce instability and movement in the LVL. In the LVL, the electric field cellular structures arise in addition to the velocity cells [8, 9]. Where the excitation of different forces is not possible the thermo-electric force can generate cellular motion in a layer of liquid semiconductor not only when the heating is from below, but also when the heating is from above. The action of thermoelectricity is most intense in thin layers.

The experimental data shows that the boundaries of the electric field micro-structures coincide with the boundaries of the convection cells ("the structures of velocity"). And on the LVL surface under influence of the thermo-electric field the electric charge is excited. In accordance with this concept, the local cells of the physical field are considered the initiators of excitation of the micro-torch structures. The electric field micro/nano-structures in the LVL gives the program for formation of the cellular-pulsating micro/nano- structures in the heated-up LVL, on the burning surface and for excitation of periodic toroidal vortex micro-structures over the burning surface. In conditions of the burning wave, where the temperature in the condensed phase increased by exponential law, the thin reactionary LVL can be considered as the molten mass with ionic properties. Concentration of the ions increases with the burning rate and determines electric conductivity of the burning surface.

### Flame zone

The most familiar description of a flame is "a local region of high temperature generated by rapid, exothermic chemical reactions". A flame is, however, also a plasma. In particular, the combustion of hydrocarbon fuels generates sufficient densities of charged species that even small flames (the flame of a humble candle) can be considered as chemically driven, non-equilibrium plasma. Flames have ions, positive and negative charges. However, due to reasons relating to mobility, flames have positive properties. In the paper [20] some experimental data on distribution of temperature and electrical potentials in the laminar diffusion flames of coaxial jets of propane and air depending on the components flow rate are presented. In accordance with experimental researches

Figure 1. The liquid-viscous layer (melt layer) in the EM reactionary zone [17].



connected with studying of electrical structure of the flame [19, 21], the flame of composite solid propellants and hybrid rocket propellants possesses own electric field of a complex structure with localization of zones of positive and negative electric charges. The own electric field of a flame is created as a result of separation of electric charges because of distinction in diffusion coefficients of carriers of charges of both signs and mobility of charged particles in the flame. Presence of distributed electric charge in the flame allows to control by shape of the flame by means of external electric field, and, hence, by distribution of the heat sources, by the heat flow into the condensed phase and, at last, by burning rate.

## Electric and Magnetic Interactions in the Reactionary Zones

The idea of using electric fields to manipulate propellant burning rates is not new. In fact, this concept was proposed at least as far back as the 1960's. In general, the application of electric fields to a combusting solid propellant has shown to increase the burning rate of the propellant, and even under certain conditions can decrease the burning rate as well [22]. In particular, application of an electric field changes the burning surface roughness [22], i.e. modifies the process of excitation of the micro/nano-structures in the reactionary zones. The priority in development of technique of control by the EM reactionary zone (LVL), by its heating at the electric current passing belongs to the Swedish inventors [23]. The simple version of the electrode system for control by heat release in the reactionary zone also is presented in the Russian patent [24], completely repeating the invention of the Swedish scientists [23]. The further development of the technique suggested in the patent [24] is the design of the composite solid propellant charge which includes the metal electrode system used as a combustible propellant component [25].

The effects, which an electric field exerts on flames, have been observed and reported in the literature for a long time. Burning velocity, flame stability, flame shape, flame luminosity, extinction limit, and soot formation, are among the effects that have been observed. Most of the studies in this field were experimental observations. In particular, application of an electric field changes the burning surface roughness, i.e. modifies the process of excitation of the micro/nano- structures in the reactionary zones. Flames interact with external electric fields, which can be used to monitor, manipulate, and enhance the processes that make up combustion through a variety of physical and chemical mechanisms. Sufficiently strong electric fields (greater than  $\sim 106$  V/m at atmospheric pressure) influence the chemistry of combustion by accelerating electrons to energies capable of exciting, dissociating, or ionizing neutral species upon impact. Even fields that are too weak to influence combustion directly can cause significant hydrodynamic flows - so-called electric or ionic winds - through the collisional transfer of momentum from accelerated charged species to the neutral gas. The electric field interacts with the charged particles in the flame - the electrons, ions and soot particles - and this collective motion of the charges in the electric field can lead to movement of the gas within the flame. The experimental studying of thermal and electrical structure of the diffusive flames shows possibility of control of the burning rate of propellant by change of heat flux to the burning surface by superimposition of a dilatational electric field

on the flame [21].

## Model Experimental Systems for Understanding of the Micro-Scale Combustion Mechanisms

Study of simple experimental systems that simulate the EM reactionary zones opens possibilities for fundamental understanding of the micro-scale combustion mechanisms. Among such experimental systems we considered the following:

### Study the characteristics of the oscillation of candle flames

The simple model nonlinear oscillatory experimental system containing a set of paraffin candles can be used for experimental studying of the phenomenon of synchronization of the torch micro-structures over the EM burning surface [28]. It was found that the two oscillators composed of a set of candles can couple with each other in a synchronous manner. Both in- and anti-phase synchronization can be observed depending on the distance between the two oscillators. All experiments were performed at room temperature under the condition without external air flow. A cylindrical candles made of paraffin (Cando Ltd., Japan) with a diameter and height of 6.0 and 50 mm, respectively, were used. The stable combustion has been noticed when a single candle burned, while oscillatory combustion has been observed when three candles burn together. When a burning candle was placed alone, the shape of the flame was stable. In contrast, when three candles were placed close together, the shape of the flames exhibited oscillation. The frequency of oscillation did not change within the experimental observations, whereas the height of the candle was slightly decreased due to the consumption of paraffin by combustion. If the distance between the two oscillators (a set of three candles as a component oscillator) was small enough, they have exhibited in-phase oscillation. As an extension of the study on a single oscillator (a set of three candles), the nature of the coupling of two oscillators, that is, two sets of three candles, was observed. When two sets of three candles were placed at a certain distance  $l$ , the flames exhibited synchronization depending on  $l$ . When the distance was smaller, that is,  $l \leq 30$  mm, the two oscillators exhibited in-phase synchronization. When the distance was larger, that is,  $30 \leq l \leq 48$  mm, the two oscillators exhibited anti-phase synchronization. When the distance was even larger, that is,  $l \geq 46$  mm, oscillations proceeded independently. In the intermediate regions around  $l \sim 30$  and  $47$  mm, one of the two modes was selected, and the system sometimes fluctuated between these modes.

In the considered paper [28], the proposed model indicates that the oscillatory combustion is induced by a lack of oxygen around the burning point. When one candle burns, sufficient oxygen is supplied. In contrast, when three candles are located close to each other, the supply of oxygen is insufficient to maintain a constant combustion compared with the supply rate of fuel, which results in the oscillatory behavior in combustion. They also suggested that thermal radiation can be an essential factor of the synchronization of candle flames. In accordance with this concept, for two sets of three candles, the main factor of interaction appears to be radiation [28]. However, they did not discuss a possible contribution of the electro-magnetic fields interaction in the reactionary zones. In connection with model experiments, provided by Japanese research team [28], the model

experiment on oxygen/paraffin combustion in e-field opposed flow burner will be considered in the following section.

### Oxygen/paraffin combustion in e-field opposed flow burner

An experimental study was conducted to determine the effects of an external DC electric field on the combustion behavior of solid fuels and solid propellants [22]. In an opposed flow configuration, a paraffin wax fuel was observed to extinguish when an electric field was applied over a wide range of conditions. The fuel was allowed to regress while combusting with gaseous oxygen. It was found that at a particular applied voltage, the combustion process was terminated.

The burning distance varied approximately linearly with voltage flux, and using electrodes with higher capacitance resulted in a more sensitive system, which provides two particular elements of control for the combustion extinguishment process. As the field strength increases, the distance the paraffin burns decreases, which was independent of polarity in either direction. Researchers have assumed that the results of the paraffin fuel study can be attributed most likely to effects of an ionic wind driving the flame front away from the fuel surface, thus not allowing sufficient heat feedback for continuous fuel pyrolyzation. The burning rates of two composite solid rocket propellants were also studied under the influence of an external DC electric field [22]. Both propellants were based on hydroxyl terminated polybutadiene and ammonium perchlorate where one has a specified amount of aluminum. Both propellants demonstrated decreases in linear burning rate when electric fields were applied regardless of polarity. The decrease was approximately linear with increasing applied voltage. Examination of the collected video showed that the aluminized propellant showed increasing luminosity when an electric field was applied and that the flame shape was altered as the propellant regressed through the electrodes. The flame shape transitioned from a long and skinny profile near the top electrode, to a short and fat profile near the bottom electrode. The decrease in average regression rates for these propellants would seem to rule out Joule heating as a mechanism for burning rate alteration. During the experiments with the aluminized formulations another interesting observation was made. As the field level was increased, there was a noticeable increase in the luminosity of the flame.

For explanation of possible reasons of reduction of the EM burning rate in the electric field a number of concepts were suggested. Currently researchers had identified some potential mechanisms for burning rate alteration due to electric fields: Joule heating, ionic wind, and an alteration in chemical kinetics.

### Kinetic singing flames

The kinetic singing flames can be considered as a simplified model of the torch micro-structures observed in the experiments over the EM burning surface [26]. Optical techniques have shown that the excitation of thermo-acoustic oscillations in singing flames is associated with periodic vortex formation in the boundary layer and the interactions of these vortices with the flame front, [26]. It has been shown experimentally that when a constant current discharge is applied to the flame zone, an internal negative feedback suppresses vortex formation in the thermal boundary layer and the pressure oscillations at all harmonics, simultaneously.

On the other hand, when a constant voltage discharge is applied, excitation and amplification of unstable combustion are observed. This method can be used for controlling unstable combustion in thermally stressed combustion chambers at higher discharge currents. The estimates show that the electrical energy required to suppress the unstable burning of a singing flame by this method is an order of magnitude lower than the chemical energy release. In addition, preliminary experiments on controlling combustion stability in more thermally stressed combustion chambers, for example, in a model of a ramjet engine, have shown that the required electrical energy is two or more orders lower. The diffusive flames, that models a flame of the composite solid propellant, are used for experimental studying of thermal and electrical structure of the reactionary zones [19-21].

Study of the simple experimental system comprising a laminar methane-air flame positioned between two parallel-plate electrodes [27] provide a basis for the electrode-free manipulation and control of combustion processes, e.g., by electromagnetic waves. A small methane flame was positioned symmetrically between two parallel aluminum electrodes ( $20 \times 20 \text{ cm}^2$ ), separated by a distance  $L = 10 \text{ cm}$ , [27]. Study [27] shows that time-oscillating electric fields applied to plasmas present in flames create steady flows of gas. AC fields induce forces localized at the surface of methane flames to create steady electric winds. Interestingly, above a critical frequency, AC fields can be used to manipulate flames at a distance without the need for proximal electrodes.

## Universal Phenomena of Self-Synchronization of the Micro/Nano - Structures of the Reactionary Zones

Self-organization of the micro/nano- structures in the EM reactionary zones is a fundamental property of the matter. Many natural and human-made nonlinear oscillators exhibit the ability to adjust their rhythms due to weak interaction. These phenomena are universal. The existence of self-synchronization in nature may seem almost miraculous. However, the main "secret" behind this phenomenon is that there exists a communication channel, called coupling, such that the entities/systems can influence each other. This coupling can be, for instance, in the form of a physical interconnection or a certain chemical process. The history of synchronization goes back to the 17<sup>th</sup> century when the famous Dutch scientist Christian Huygens reported on his observation of synchronization of two pendulum clocks which he had invented shortly before. Many interesting synchronization phenomena have been observed and reported in the literature. More importantly, it gradually became clear that diverse effects which at first sight have nothing in common, obey some universal laws, [29]. Emergent synchronization of real (non-identical) oscillators was a serious paradox in physics for a long period of time. Oscillators with similar frequencies can obviously synchronize when phase-minimizing coupling acts between them. In nature however it is practically impossible to find two "exactly" similar oscillators. Synchronization of oscillators with different frequencies in the absence of an external periodic driving is a phenomenon that is highly nontrivial. It is not obvious at all if the system will spontaneously find a common frequency and all elements will abandon their natural rhythm and synchronize following this common frequency.

Being, probably, the oldest scientifically studied nonlinear effect, synchronization was understood only in the 1920s when E. V. Appleton and B. Van der Pol systematically - theoretically and experimentally - studied synchronization of triode generators. This new stage in the investigation of synchronization was related to the development of electrical and radio physics [19]. Recently many new results were reported on complex systems capable of spontaneous synchronization. The micro/nano-structures of the EM reactionary zones can be classified as the synergetic objects. Motivated excitation source of the micro/nano - physical structures in the reactionary zones are the micro/nano- structures of the electro-magnetic fields. One of examples of collective interaction and self-synchronization of the micro/nano- structures of the EM reactionary zones are the process of excitation of the burning cells in the reactionary zones of strobes, [30]. Strobes are pyrotechnic heterogeneous compositions which show an oscillatory combustion. They have fascinated many scientists since their discovery at the beginning of the 20<sup>th</sup> century. Strobe compositions belong to the class of solid combustions. They are mixtures of powdered ingredients. All strobe compositions mentioned in the literature were discovered by trial and error methods and the mechanisms involved remain unclear. A few theories have been proposed to explain this phenomenon but the current knowledge of these mechanisms is only based on empirical methods and the physical and chemical processes that trigger the occurrence of flashes is still unclear. Explanation of such phenomenon just as a chemical oscillator is not confirmed by experimental data and detailed chemical mechanisms remain unknown. In the EM reactionary zones exists necessary conditions for realization of the phenomenon of self-synchronization. First of all this is a set of similar micro/nano- structures, which have an identical information-algorithmic condition and being in the conditions supposing fast information exchange between them. Fluctuating micro/nano- structures also are generators of electromagnetic radiation.

Taking into account earlier considered the simple experimental systems we propose hypothesis that the macro-scale phenomena at the EM combustion are result of self-synchronization of the micro/nano-  $\leftrightarrow$  structures of the reactionary zones. Self-organization of the micro/nano-structures in the EM reactionary zones is a fundamental property of the matter. The key to understanding of self-organization of the micro/nano-structures is the electro-magnetic  $\leftrightarrow$  phenomena  $\leftrightarrow$  in the EM reactionary zones. Such interaction of the electric charges and electro-magnetic fields changes the space localization of the reactionary zones and the sources of heat release. The feedback mechanism between micro/nano - structures is provided through an electromagnetic interaction. In the greatest degree, the effects of self-synchronization will be observed in the area of low pressure, when the micro-structures in the reactionary zone have the maximum size. In the area of reduced pressures the majority of anomalies of ignition and combustion are observed.

In accordance with our hypothesis, the simple model nonlinear oscillatory experimental system containing a set of paraffin candles, which has been investigated by Japanese research team [28], can be considered as the example of realization of the phenomenon of self-synchronization of the flame micro-structures. We recognize that the electro-magnetic fields interaction can be a key mechanism to realize the synchronization between two candle flame oscillators. This simple experimental example shows a role

of the phenomenon of self-synchronization in the excitation of oscillatory burning. Actually, self-synchronization of the micro/nano- structures provides the mechanism of interactions between micro-  $\leftrightarrow$  and macro-  $\leftrightarrow$  scale levels in the EM reactionary zones. Self-organizing of the EM reactionary zones is essentially new level of self-organizing which is determined by achievement of critical spacial concentration of the micro/nano- structures - by a bifurcation point.

In particular, application of an electric field, changes the burning surface roughness, i.e. modifies the process of excitation of the micro/nano-  $\leftrightarrow$  structures in the reactionary zones. Self-synchronization of the micro/nano-  $\leftrightarrow$  structures are capable to induce changes of distribution of heat flows and to induce combustion instabilities in the solid micro-propulsion systems. The new possibilities for effective control by ignition and combustion processes opens in connection with possibility of initiation of self-organizing of the reactionary zone, for instance, by use of the electric fields and the electric discharges.

## Conclusion

Presented research is devoted to further development of ideas of famous Russian scientist Ya.B. Zeldovich in the area of physics of self-organizing systems in the active medium. According our hypothesis the macro-scale phenomena at the EM combustion are result of self-synchronization of the micro/nano- structures of the reactionary zones. From suggested concept follows that each energetic material has a unique frequency electro-magnetic code of the reactionary zone which are the synergetic oscillatory system. Micro/nano- structures in the reactionary zones are the platform for effective control by combustion processes of the EM at macro-level and also programming of them. The suggested concept opens new possibilities for development and synthesis of the advanced propulsion materials through programming by electro-magnetic field micro/nano- structures in the reactionary zones.

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