White Paper for *Frontiers of Plasma Science Panel*

Date of Submission:	***06/16/2015***

Indicate the primary area this white paper addresses by placing "P" in right column.
Indicate secondary area or areas by placing "S" in right column

	"P", "S"
• Plasma Atomic physics and the interface with chemistry and biology	P
• Turbulence and transport	S
• Interactions of plasmas and waves	
• Plasma self-organization	
• Statistical mechanics of plasmas	

Indicate type of presentation desired at Town Hall Meeting.

	"X"
Oral	
Poster	
Either Oral or Poster	X

Title:	Electric Discharge in Supercritical Fluids
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**(Limit text to 3-pages including this form. Font Times Roman size 11.
1 page of references and 1 page of figures may also be included. *Submit in PDF format*.)**

- ***Describe the research frontier* and importance of the scientific challenge.**

Electric discharge in supercritical fluids is a poorly studied phenomenon mostly due to the unusually high pressure and temperature needed for transition to the supercritical state of typical fluids (see Table 1 in section "Figures"). There is a very limited bibliography that discusses the electrical discharge appearance and properties in supercritical fluids. Most experiments are performed in CO2 [1-5] and explore small-scale discharges of a mm or less inter-electrode gap. From the other side, the thermodynamic and mechanical properties of supercritical fluids are of greatly increasing interest due to practical applications, such as supercritical fuel injection [6-10]. That technique is promising in terms of providing a better fuel nucleation, deeper penetration into the combustor chamber, and enhanced mixing. A wide application of supercritical injection systems is limited by the necessity of a high pressure and high temperature apparatus to provide proper conditions for typical fuels. The high-voltage pulsed electric discharge in fluids of high density (liquid) can stimulate a fast transition of fluid to supercritical state [11-13], as it is shown in Fig.1. The technical benefits of such a scheme include robust dynamic operation with supercritical fluids, deeper penetration of fluid into the cross-flow, faster evaporation, and pre-activation of the fuel by the electrical discharge. One more practical application is the protection against deadly breakdowns in electrically-insulating liquids (transformer oil).

The mechanism of the electric breakdown and the discharge development in a supercritical fluid are basically unknown. The problem is of high complexity considering the kinetic, electrodynamic and

hydrodynamic processes in multiphase media. Study of electric discharges in supercritical fluids is challenging in a fundamental aspect and promises well-recognized practical benefits.

- ***Describe the approach to advancing the frontier and indicate if new research tools or capabilities are required.***

The study of electric discharge properties and dynamics in supercritical fluid and at phase transformations is of fundamental importance. Two basic approaches are considered:

- The characterization of electric breakdown and steady-state high-voltage electric discharge in a supercritical fluid. The advantage of this approach is in conserving the supercritical state of matter during fluid heating by the discharge. The best option is to use noble fluids, like Ar, to diminish the effect of chemical reactions.
- The characterization of the discharge dynamics at transient supercritical state in commonly used liquids. This approach supposes a convenient test arrangement at ambient initial conditions and excellent access for diagnostic tools. The liquid fluid is rapidly heated and pressurized in a small volume by pulsed high-voltage electrical energy and concomitant plasma. In this phase the liquid exhibits a phase change to a supercritical fluid, and then escapes to lower pressure media via fast vaporization with subsequent partial condensation. The process of phase transformation for a model test-cell is illustrated in Figs. 1 and 2, where T is what is known as the triple point, and C is the critical point. The exact trajectory of liquid state to supercritical fluid depends on the geometry of the test cell, fluid thermal properties, and electrical power density. In a locked cell the trajectory is linear with a slope $\partial P/_{\partial T} = \beta/_{k}$, where P – pressure, T – temperature, β – thermal expansion factor, and k – compressibility coefficient. In the presence of the injection orifice, the pressure becomes lower than the estimation above; but some combination of parameters keeps it higher than the critical value. The reverse process of condensation may be realized by a variety of trajectories, all of which result in two-phase composition.

The last approach has been experimentally tested with liquid methanol (MeOH) in a cylindrical cell with diameter*length=d*L=3.5*(10-22)mm opened from one end. The power supply has a maximum voltage of U=16kV. The typical dynamics of the electrical parameters are shown in Fig.3. In this configuration, the temperature first achieves a value where the saturated vapor pressure becomes equal to the static pressure at a location near one of the electrodes. An initial non-transparent bubble is generated at this point with sequential electrical breakdown along the entire cell, as it is shown in Fig.4. The plasma formed pushes the fluid up, increasing the pressure even more due to the electric discharge power dissipation. The next stage is a dense plasma jet escaping the cell, with sequential condensation of the fluid in the ambient atmosphere.

Calculations based on the electrical measurements and on an incompressible fluid model indicate that, for some combination of test parameters, the pressure (p> 80Bar) and the temperature (T>600K) in the test cell exceed the characteristic values for supercritical condition. The electron density $Ne>2.5\times10^{19}cm^{3}$ in plasma was measured by Stark broadening of the hydrogen line Hα (656nm) of the Balmer series. To reconcile this data with the electrical conductivity measured in the test cell, the molecular number density must be higher than $N_m=10^{22}cm^{-3}$. This and other results suggest the conclusion that the technique applied allows a transition of liquid fuel to the state of supercritical fluid, with a subsequent formation of non-ideal plasma in the supercritical fluid.

Further study of phenomena of transient discharges in supercritical fluids require a significant improvement in instrumentation and data processing algorithms to be applied. Use of contact probes such as pressure/temperature transducers has a rather limited opportunity. Some conventional diagnostic tools require an extension in tabulated (simulated) data. The verified dataset for Stark broadening, for example, is available for electron density $Ne<10^{19}cm^{3}$ [14-15]. The measurement of parameters of a dense multiphase flow is of high complexity and requires use of advanced tools like the SAXS technique [10] available in Argonne National Laboratory.

- ***Describe the impact of this research on plasma science and related disciplines and any potential for societal benefit***.

This research has the potential to open a ***new domain*** of study dealing with ***non-ideal plasmas in laboratory*** conditions. A fundamental goal of this investigation is to formulate the mechanisms of electrical breakdown and electric current maintenance in a high-density media. Exploring this field could lead to some advancements and a potential breakthrough in the field of plasma science. With knowledge gained from this research it could be possible to significantly ***extend the parameters*** of the media where the plasma is generated in addition to ***spreading the diagnostic range*** to measure the plasma parameters. All of this together could potentially lead the development of a ***new transient technique*** applicable to resolve a wide range of technical problems.

This plasma-related research has a direct link to furthering the development of advanced high-speed combustion technology. A high practical benefit is expected from the plasma-based manipulation of supercritical fluids in a field of transient fuel injection in internal combustion engines, pulse-detonation engines, and, specifically, ram- and scramjets. Some scientific and practical problems could be directly related to the generation of supercritical state of fluids, such as an electrical breakdown in liquids, arcs in insulating liquids, and the mechanism of pulse ablation.

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Figures (maximum 1 page)

Table 1.

Critical parameters for selected liquids						
	Molecule	Critical temperature, Tc, K	Critical pressure, Pc, Bar	Critical density, ρ, g/cm^3	Compressibility, k, 10^{-4} 1/Bar	Thermal expansion, β, 10^{-4} 1/K
Water	H_2O	647	218	0.322	0.46	2.1
Methanol	CH_3OH	513	80	0.272	1.2	11.9
Ethanol	C_2H_5OH	513	61	0.276	1.14	10.9
Kerosene	~C_8H_{18}	~580	25	~0.24	0.45	10

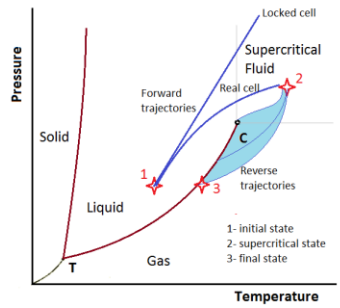


Figure 1. Schematic of phase transformations stimulated by electric discharge.

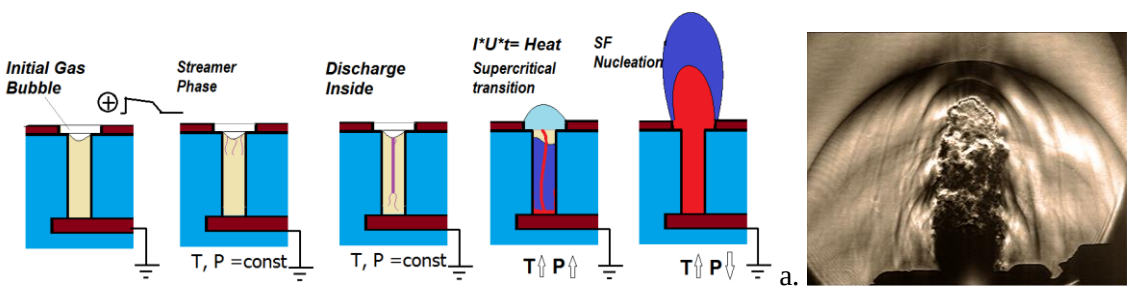


Figure 2. Dynamics of the discharge in the test cell. a – scheme; b – schlieren image of exhaust jet.

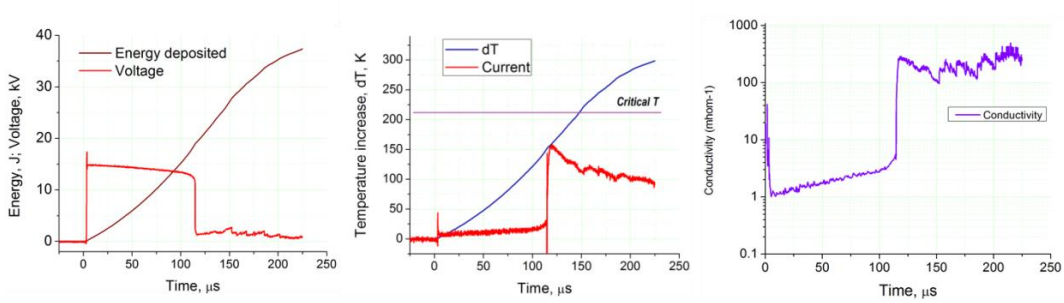
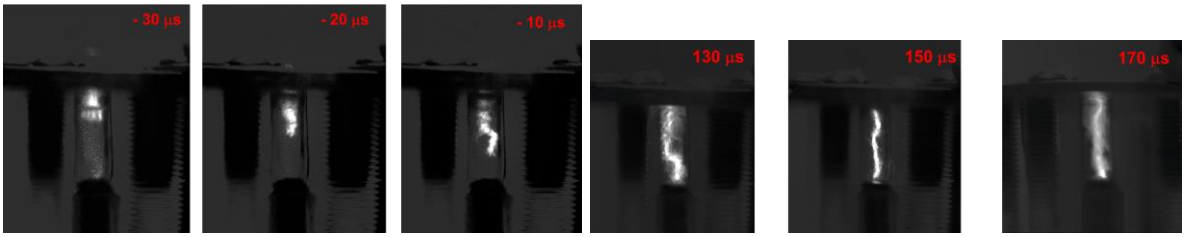


Figure 3. Basic oscillograms and calculated data at phase transformation.



a. b. c. d. e. f.

Figure 4. Discharge propagation due to streamer-to-liquid process, a-c; discharge in supercritical fluid, d-e; critical opalescence at nucleation, f.