Irreversibility of Current-Voltage Characteristics in Hydrocarbon Liquids

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ABSTRACT

The I-V characteristics curves in this work have been seen irreversible, in purified saturated hydrocarbon liquids, in other words, the conduction current curves in increasing direction does not coincide with that of decreasing one. Such behavior may be attributed to the effect of space charges. It is also believed that the build up of the space charges is quicker during the increasing direction than its distraction direction in recombination processes, or due to creation of gas bubbles and liquid clusters which leads definitely to complex changes in refractive index and dielectric constant of the tested liquid.

لا انعكاسية خواص الفولتية-التيار في السوائل الهايدروكاربونية

الملخص

أستخدم في هذا البحث سوائل هايدروكاربونيـة مشبعه ووجد أن خواص منحنيات تيـار التوصيل مع المجال الكهربائي لا انعكاسية وهذا يعني أن هذه المنحنيات لا تتطابق أثناء زيادة المجـال الكهربائي وإنقاصه. أن مثل هذا التصرف يمكن أن يعزى إلى تأثير الشحنات الفضائية حيث أن سـرعة تكوين مثل هذه الشحنات تكون أكبر أثناء زيادة المجال الكهربائي مما هو علية عند عمليات أعاده التكوين ومن خلال إنقاص المجال الكهربائي، أو بسبب تكوين الفقاعات الغازية والتجمع السائلي والذي يقود حتما إلى حدوث تغيرات معقده في معامل الانكسار لهذه السوائل ومن ثم ثابت العزل الكهربـائي لمثـل هـذه السوائل.

INTRODUCTION

It has been known that irreversibility in characteristics of plasma are observed as one of the important processes in plasma diagnostics, which leads to streamer initiation and breakdown in hydrocarbon liquids. The process of charge carrier generation under the influence of a high electric field is best studied with point to plane geometry gaps, since the generation process is confined to a small volume and the influence of polarity can be analyzed.

Not too many reports have appeared in the literature, the classical work on this subject has been carried out by Halpern and Gomer (Schmidt, 1982) who investigated experimentally and theoretically the field emission and field ionization process in various liquids under non uniform electrical field conditions. Experimental results are strongly affected by the degree of liquid purity, material and the surface condition of the needle point and the tip curvature which was studied by Yamashita and Yamazawa in cyclo-hexane (Yamashita et al., 1996). Their results showed that, the structure of the primary streamer is a tiny filament for both positive and negative polarities and this tiny filament expand to form sphere within a very short time (~ 40 ns).

Previous studies (Devns et al., 1981 and Hebner et al., 1982) have shown that for electrical breakdown to occur a streamer must be initiated at one of the electrodes and travel to the opposite electrode to establish a conducting path. Of particular interest in this study are point – plane electrode geometries that produce divergent fields. That is, this work was limited to that case in which the point was the cathode and the plane was the anode.

The purity of a liquid is a very important factor in such measurements, and the convenient method in this study is the electrical one. Where the mechanism of this method has not yet been fully clarified. In general, a liquid purification by means of an electrical field may include the following processes.

1. Removal of the traces of electrolytic impurities from the effective volume of the cell.
2. Disappearance of gas bubbles accumulated in the liquid (mainly air and oxygen).
3. Disappearance of gas bubbles absorbed by the electrodes.

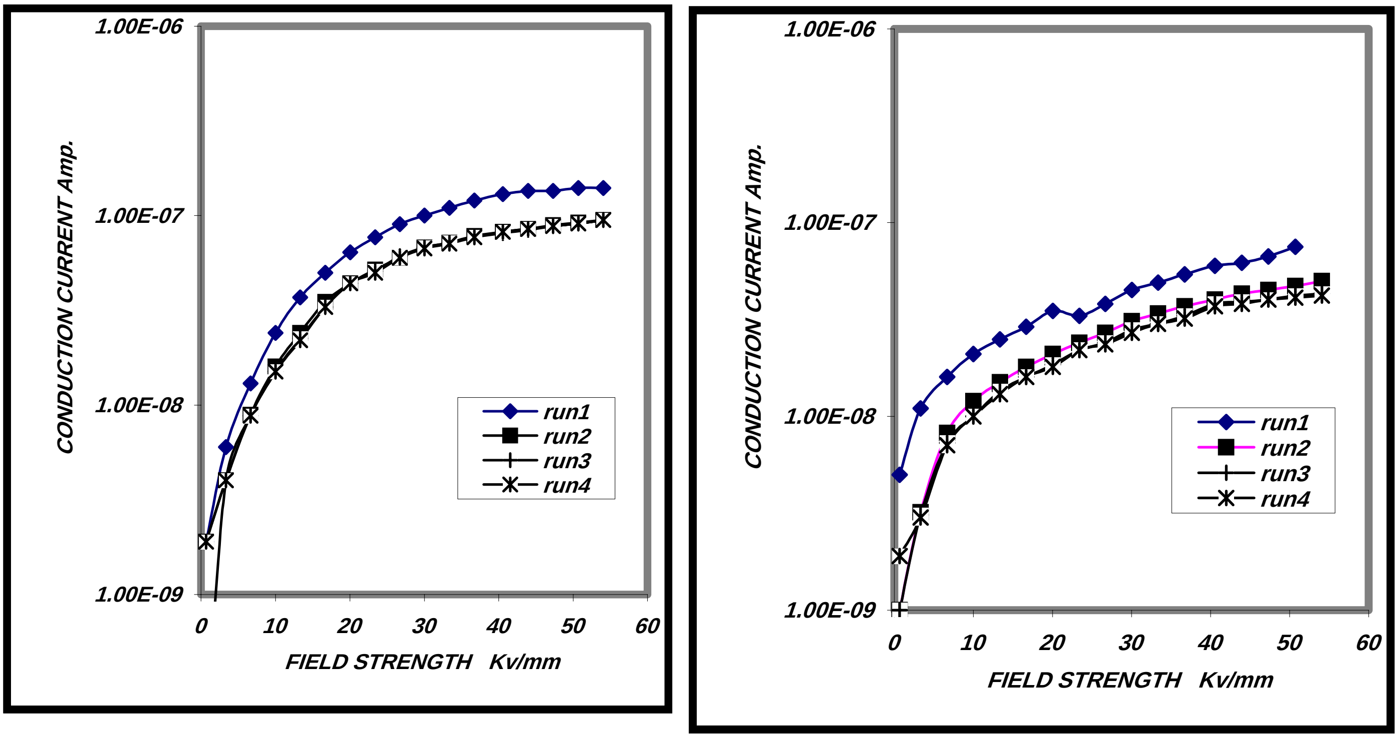
With purified liquids, the first process is generally very slow and rarely yields useful information. The processes (2) and (3) normally lead to a rapid and irreversible disappearance of conductivity immediately after switching on the voltage and then proceed to change very slowly until the final value of conductivity is reached (Adamczewski, 1969).

The current growth in a liquid under the influence of high electric field depends upon the kind of streamer that propagate along the cavity, and the creation of space charge in the bulk liquid. It is found that formation of space charge creates strong ionization layers close to the electrodes and that the whole column could deviate from electrical neutrality due to the drift of charged particles (Yan et al., 2001). However, it should be emphasized that if the assumption is made that the field at the tip of the streamer is the controlling factor, it must be further assumed that the streamer tip field is relatively insensitive to charges in the applied voltage (Hebner and Kelley, 1983).

EXPERIMENTAL

The test liquids selected for this study were n-pentane, n-hexane, n-heptane and n-octane as a series of saturated hydrocarbon liquids of form $C_n H_{2n+2}$ where, n is the number of carbon atoms in each liquid. The pure liquid was obtained by electrical purification in the test cell itself, these liquids are subjected to the action of an electric field for about 6 hours between one run and other until we get an overlapping conducting current measurements, as clearly shown in figure (1).

From this figure we can see that the conduction current has been reduced from one run to another until we reached the overlapped curves, which means that the liquid inside test cell has been purified and degassed.



Effect of electrical purification on the current growth in n-octane & n-pentane respectively.

Figure (2) shows the schematic diagram of the test cell which is used in this study and the gap between the two electrodes was fixed at 1.00 mm, by using traveling microscope.

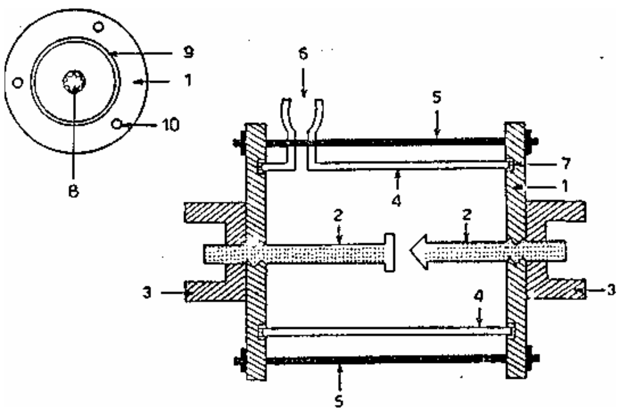


Fig. 2: Schematic diagram for the test cell

1. Obonite base. 2. Brase electrode.
3. Plexy glass. 4. Glass test cell envelope.
5. Steel fitting rods. 6. Filling port.
7. Gasget. 8. Electrode fitting hole.
9. Test cell setting grove. 10. Rod fitting hole.

The circuit diagram for setting up the voltage is shown in figure (3), and by balancing the potential across the resistance box r against the potential of standard cell μ, the applied voltage was varied until no deflection was indicated by the galvanometer.

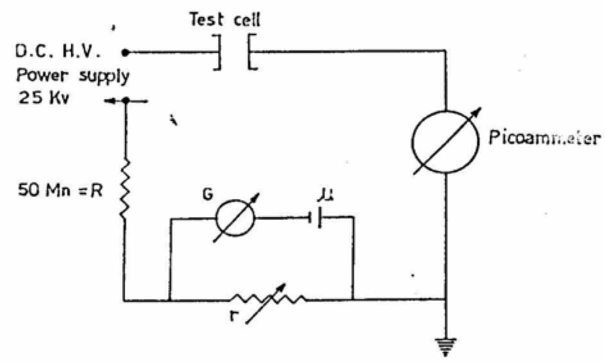
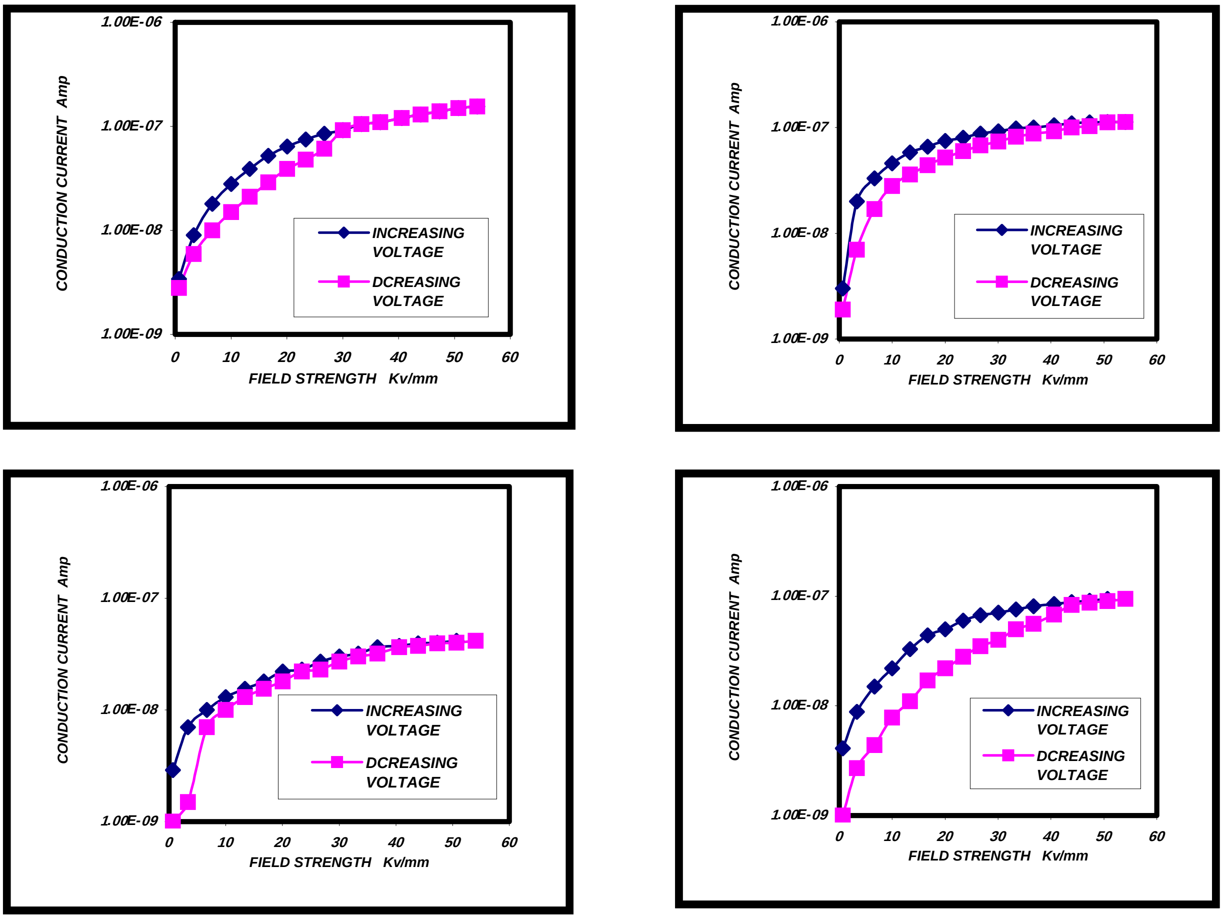


Fig. 3: the Schematic diagram of experimental circuit. G, electronic galvanometer , μ standard cell , R , standard resistor , r available resistance box .

DISCUSSION

The best study of this kind of measurements was done by using non-uniform electric field which can be produced by using point– plane electrode geometry, where the field affected in present work will be the emission one (negative point). After the purification of tested liquid in the test cell itself, the sequence of ionization processes at the time when an electric field was applied is as follows:-

When an electric field was increased, the conduction current will be increase too until we reached the saturation region as a result of secondary ionization processes in the bulk liquid by means of injected electrons from negative point to the liquid then the ionization collision will occur in small gap spacing (about 1 μm). The continuity of increasing electric field to maintain rapidly the saturation region and then to a point before the prebreakdown processes which lead to electrical spark and breakdown in the dielectric liquid. Also the creation of negative streamer from cathode to the anode and building a conducting channel could produce a high conduction current through these channels which have a negative bush type form. When we reached a point before a pre breakdown (54.1 KV/mm),the electrical field will be decreased without any irreversibility until we reached a value of applied field which is proportional with the number of carbon atoms in each tested liquid, i.e. long chain of liquid molecule. This behavior could be attributed to the high viscosity of a liquid having large number of carbon atoms which leads definitely to decreasing in ions mobility and increasing in recombination coefficient ($\mu=A\eta^{-3/2}$) where μ is the ion mobility in $cm^2\ v^{-1}\ sec^{-1}$, η is the viscosity coefficient in poise and A is a constant around 0.00053,(Adamczewski 1969), as shown in figure (4).



The irreversibility of conduction current for n-hexane, n-heptane, n-pentane and n-octane respectively.

By decreasing the field strength, the conduction current continued to flow but decreased until cutoff occurred very sharply at a such lowered field. The field-current curve has the same characteristics as that of the increasing direction field strength but lies under it, such behavior may be attributed to the effects of space charge. The space charge limited behavior occurs at sufficiently high currents, further, decreasing in a field strength leads to more decreasing in conduction current and the effect of such space charge continue to act as a field barrier for further field emission. By more decreasing in the field strength, the space charge cloud accumulation may be decreased due to the effect of recombination processes of charges. From figure(4) we can conclude that the build up of the space charge is quicker during the increasing direction of electric field strength than its distraction in recombination processes during the decreasing direction. Our tested liquid could be placed according to the classification of dielectrics in position of polar substances having optical as well as infra-red polarization (Frohlich, 1958). Such substances consist of molecules whose total dipole moment vanishes, through they contain dipolar groups of atoms.

From practical point of view the infra-red polarizability is only a small fraction of the optical polarizability, therefore, their behavior is very similar to that of non-polar substances and should satisfy the Maxwell relation $\varepsilon=n^2$ where ε and n represent the dielectric constant and the refractive index of the liquid respectively. These slightly variations in dielectric constant may be attributed to either,

1. Electrostractive density changes, due to the formation of polarized clusters within the medium throughout the ionization collisions in dielectric by electrons having sufficient energy to make these processes, or
2. The formation of bubbles in a liquid bulk itself due to the field emission and heating small amount of it which causes later local changes in refractive index of the liquid.

We have also found that the area between the two curves of increasing field direction and decreasing one can represent the power loss during recombination processes of charges in the space charge region, or may be due to heat lost in the formation of gas bubbles.

CONCLUSION

The phenomenon of irreversibility in ionization processes of purified saturated hydrocarbon liquids has been proved in this work, i.e. the curve of I-E in increasing field direction does not coincide with that of decreasing one, which may be attributed to space charge creation in the liquid bulk during increasing field which is quicker than that of the distraction of these space charges during decreasing direction in recombination processes.

These space charges were affected as a controlled factor in processes of electron injection. This phenomenon may be attributed, too, to complex changes in dielectric constant of the tested liquids due to the formation of polarized liquid clusters, and /or gas bubbles within the medium to change the refractive index of these liquids.

The area difference between the two curves(increasing direction and decreasing one) may be represented as the power loss during the recombination processes of space charges and /or due to joule energy in the formation of gas bubbles (Al-Rawachy 1992).

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