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A Discussion of The Mobility and Structure of Ions in Liquid Hydrocarbons

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The mobility of ions in liquid hydrocarbons is discussed within the framework of Stokes' formula. In the Stokes model, the ions are considered as submicroscopic spheres, consisting of the ionic core (the ionized parent molecule) surrounded by a more or less solid shell of neutral molecules, which are bound by the polarization force to the ionic core. Under the influence of an applied electric field these charged particles (clusters) move with a velocity given by the Stokes' mobility formula and the applied electric field strength. The retarding effect of the liquid is assumed to be friction and the motion proceeds in a laminar regime. One result is Walden's rule. Sometimes Walden's rule is not fulfilled. Taking into account the temperature dependence of the cluster radius in Stokes' formula allows modification of Walden's rule.

A discussion of the ionic drift velocity at the high field strengths on the basis of Reynold's number or Newtons law of friction leads to the conclusion that the mobility of these ionic clusters at temperatures near the melting point will be independent of the applied electric field strength up to very high values. The measured data reported in the literature support this finding.

In another set of experiments, the radius of positive ions was increased by pre-irradiation with γ - rays. Formation of dimers led to ions with larger radii.

Key words: ion mobility, liquid, liquid hydrocarbon, Stokes formula, Walden's rule,

炭化水素液体中のイオンの移動度と構造に関する考察

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ストークスの法則の観点から炭化水素液体中のイオン移動度について考察している。ストークスモデルでは、イオンは電離した母体分子が多少とも中性分子からなる固体シェルで囲まれたイオンコアからなるサブマイクロピク的な球体と考えられる。中性分子はイオンコアに分極の力で束縛されている。電界印加下ではこれらの電荷を帯びた粒子（クラスター）はストークス移動度の法則と印加電界強度によって決まる速度で動く。抵抗力効果は液体の摩擦と層流の中での動きで決まる。その結果ワルデン則が成立する。しかし、ときにはワルデン則に従わない場合がある。ストークスの法則の中でクラスター径の温度依存性を考慮に入れるとワルデン則の変歪を示すことができる。レイノルズ数、摩擦のニュートン則を元に高電界強度でのイオンドリフト速度を考察すると、融点付近のこれらイオンクラスターの移動度は非常に高い電界まで電界依存性がないことが導かれる。文献に見られる測定結果はこの考え方を支持するものである。他の一連の実験で、正イオンの径が前もってガンマ線を照射することによって大きくなることを見出された。これはダイマーが形成された結果、イオン径が大きくなったためである。

キーワード：イオン移動度、液体、炭化水素液体、ストークス則、ワルデン則

1. Introduction

The drift mobility of simple ions in non-polar dielectric liquids has been studied experimentally for many decades but theoretical studies are few. Results for hydrocarbons and related compounds before 1970 have been summarized by Adamczewski [1]. In all these former studies, the nature of the ions remained unknown. An exception constitutes liquid helium and some other liquefied rare gases, where due to the high purity achieved, well-defined ions could be studied. Some more recent publications are reviewed in [2].

The investigation of the transport properties of ions in liquid hydrocarbons is important from various points of view. These liquids are comprised of molecules with zero dipole moment making them suitable for fundamental studies of ion-molecule reactions in a non-polar environment and of charge polarization processes occurring in a dielectric continuum. From a more practical point of view, the application of non-polar liquids in devices for ion drag pumping, their use in transformers and switches as insulating media and last but not least as detection media in liquid ionization chambers warrant more detailed studies of their charge transport properties.

In the following scheme the relations of the various topics in which ions play a major role are summarized.

An important physical property of ions moving in a medium under the influence of an electric field is their drift mobility. Its magnitude is determined by the interaction of the excess charge with the surrounding medium.

In this paper, we shall discuss the validity of Stokes' model for the explanation of the mobility of positive and negative ions in liquid hydrocarbons, especially at high electric field strengths.

The structure of the compounds discussed in the text are shown in Figs. 2a-b.

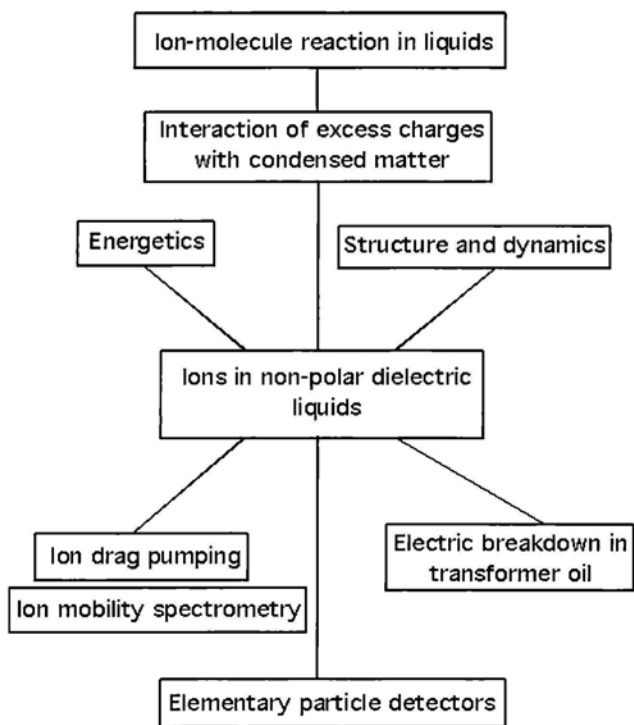


Fig. 1: Ions in liquid hydrocarbons and their relationship to various scientific and engineering topics

図1 炭化水素液体中のイオンと種々の科学技術トピックスの関係

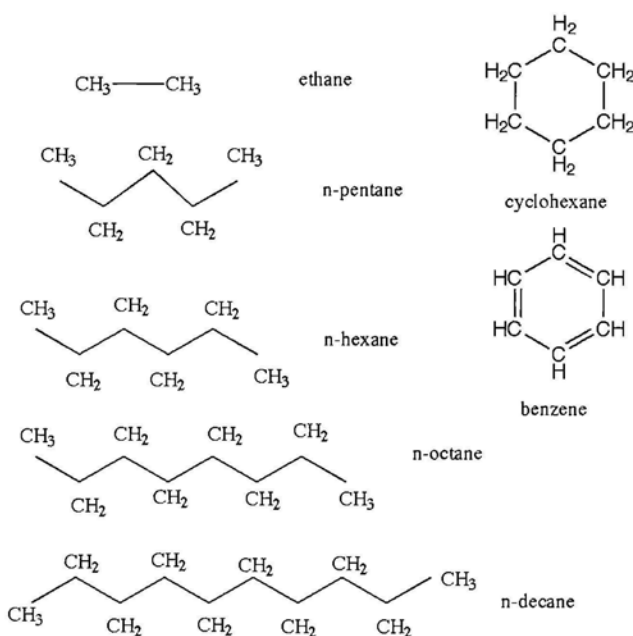


Fig. 2a: Structure formulae of some hydrocarbons

図2a 炭化水素の分子構造の例

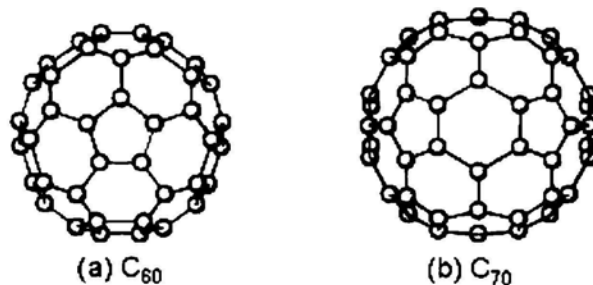


Fig. 2b: Fullerenes

図2b フラーレン

2. The Stokes' model of mobility

The most basic property studied is the drift mobility μ_{ion} . Theoretically, only simple models exist for the explanation of the measured values. One approach is to correlate the drift mobility with the diffusion coefficient D_{ion} . The Stokes-Einstein equation relates the diffusion coefficient, D , of a particle of radius R to the viscosity η by

$$D = \frac{k_B T}{C \pi \eta R}, \quad (1)$$

where k_B denotes the Boltzmann constant, T is the temperature while C denotes a constant which depends on the type of interaction between the particle and the atoms or molecules of the liquid. In the case of a "sticking" condition, in which a few layers of the liquid are firmly bound to the particle (clusters), the constant is $C = 6\pi$. In the case of a "slipping" condition, in which the interaction is minimal (bubbles), the constant is $C = 4\pi$. For ions, the diffusion coefficient is proportional to the mobility given as

$$D_{ion} = \frac{k_B T \mu_{ion}}{e}, \quad (2)$$

where e denotes the electron charge. Combining (1) and (2) yields for the mobility,

$$\mu_{ion} = \frac{e}{C \pi \eta R} \quad (3)$$

which is also known as the Stokes formula of the ion mobility. From Eq. 3 it follows that the product of mobility and viscosity is constant,

$$\mu_{ion} \times \eta = const. \quad (4)$$

This formula is known as Walden's rule. From the measured mobility, an ionic radius R_0 can be obtained. The R_0 thus determined is greater than the radius of the bare ion because of the polarization induced in the surrounding liquid. R_0 depends only slightly on temperature. From an analysis of the polarization interaction the temperature dependence of R_0 is obtained as [3],

$$R_0^4 = 2\alpha \left(\frac{e}{4\pi\epsilon_0\epsilon_r} \right) \frac{1}{k_B T} \quad (5)$$

ϵ_0 and ϵ_r are the dielectric constants of vacuum and liquid, respectively. α denotes the polarizability of the molecules comprising the liquid. The dependence on temperature is weak, but it may show up if measurements over a wider temperature range are made.

Walden's rule is fulfilled in many cases for a certain temperature range; however, deviations from Walden's rule also have been observed. For negative ions, in addition to the polarization, the repulsive interaction of the lone electron of the negative ion and the electronic shells of the surrounding atoms or liquids has to be taken into account.

3. Walden's rule

Gzowski found that for positive ions in n-hexane Walden's rule is broken and the connection between μ_{ion} and η can only be saved by

$$\mu_{ion} \times \eta^x = const \quad (6)$$

with $x = 3/2$ [4]. The measurements were made by the thin-layer method. With a pulsed x-ray tube a thin slab of x-rays is used to ionize a region near one electrode in the liquid. In this layer, positive and negative ions are present. If the applied potential at this electrode is positive, cations are injected into the bulk of the liquid, if it is negative, anions are injected. A constant current is observed until the layer of ions reaches the counter electrode (see Figure 3).

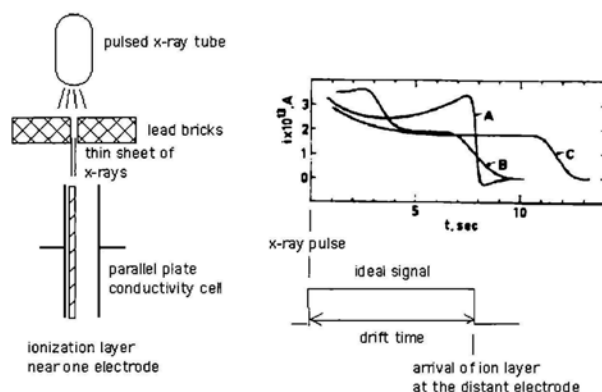


Fig. 3: Thin layer method of Gzowski [4]

図3 グゾウスキーによる薄層法 (文献4)

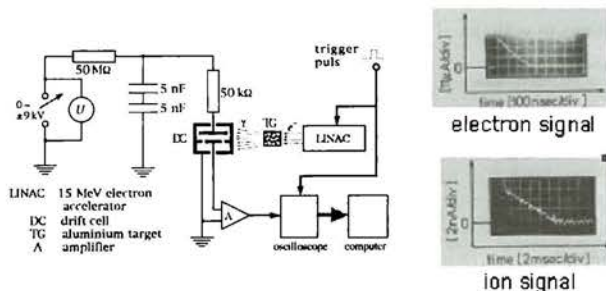


Fig. 4: Thick layer method [5]

図 4 厚層法 (文献 5)

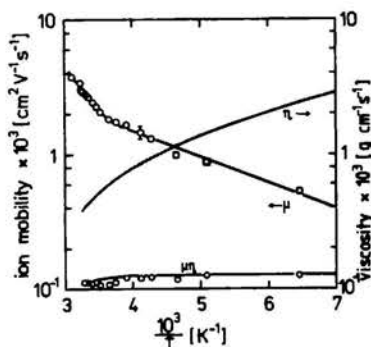


Fig. 5a: Test of Walden's rule for positive ions in liquid ethane, adapted from [5]

図 5a 液体エタンにおける正イオンに対するワルデン則のテスト (文献 5 から)

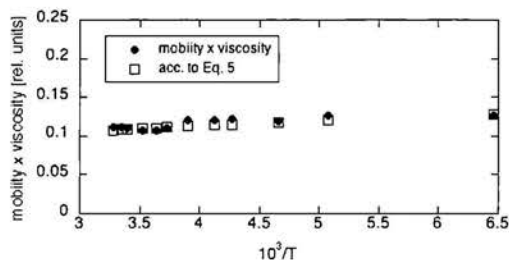


Fig. 5b: Comparison of the measured data of Fig. 5a with corrected data according to Eq. 5.

図 5b 図 5a の測定データと式 5 により補正されたデータの比較

In another method, the space between the electrodes is irradiated by a pulse of high energy x-rays. By tailoring the conditions for the generation of defined ions, mobilities can be measured (see [5]). In pure ethane, free electrons as negative charge carriers and positive ionized ethane molecules were observed [5]. For the positive ions Walden's rule was obeyed except for a small decrease of the product at higher temperatures (see Fig. 5a).

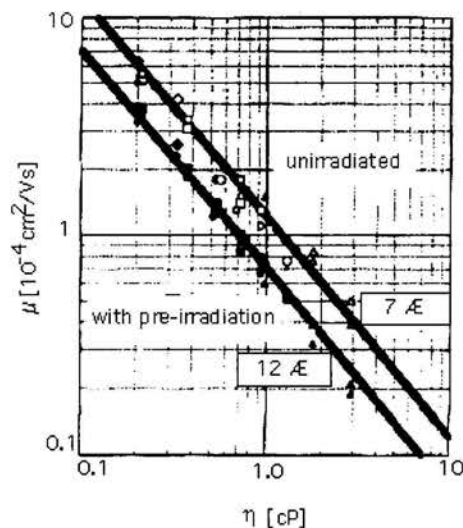
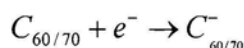
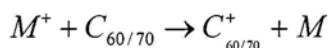


Fig. 6: Mobility of C_{60} and C_{70} -ions in liquid hydrocarbons as a function of viscosity, upper trace without pre-irradiation, lower trace with pre-irradiation; the data points refer to cyclohexane, neopentane, n-pentane, and tridecane at different temperatures, in this log-log representation the points for C_{60} and C_{70} fall on the same trace (adapted from [6])

図 6 炭化水素液体中の C_{60} と C_{70} イオンの移動度の粘度依存性
 上のトレース 前照射なし、下のトレース 前照射あり、データポイントはシクロヘキサン、ネオペンタン、n-ペンタン、トリデカンの異なる温度での値 (文献 6 から)

The slight decrease of the product at higher temperatures (lower $1/T$ values) can be rationalized with a decrease of the radius R , as given by Eq. 5. In Fig. 5b the data of Fig. 5a are compared to estimates according to Eq. 5.

If both ions are large and of similar size, then the influence of polarization becomes smaller, as was shown for positive and negative fullerene ions [6]. The mobilities of positive and negative ions of C_{60} and C_{70} were measured in various hydrocarbons and CS_2 . Positive ions are formed by charge transfer from ionized solvent molecules, while negative ions are produced by electron attachment, respectively. The ionization energy of the hydrocarbons in the liquid phase is approximately 10 eV. The ionization energy of C_{60} is about 7.6 eV. The electron affinity is 2.67 eV. Charge transfer from positive ions to the fullerenes and electron attachment to the fullerenes produces the fullerene ions,



Compared to other ions, these ions are rather big. It can be conjectured that the overall influence of the polarization on the mobility is small. The mobility data within the precision of the measurements showed no difference in mobility of the positive and negative ions. Measurements in various hydrocarbons (of different viscosities) and temperatures showed Walden's rule to be fulfilled (see Fig. 6). The data obtained in unirradiated solutions were fit with ionic radii of approximately 7 Å for C_{60} and C_{70} . Pre-irradiation with 25 krad of γ -rays produced fullerene dimers and the ionic radii increased to about 12 Å (in Angstrom units Å). The existence of these dimers was later verified by small angle x-ray scattering [13].

Generally, we notice that there exists a lack of mobility measurements in hydrocarbons with other defined ions.

In principle another mobility mechanism consisting in charge transfer between ions and neutrals of a certain solutes in liquid hydrocarbons may be possible. In a recent paper Borovkov [7] described the measurement of the diffusion coefficient of defined radical ions (positive and negative) in n-hexane and n-hexadecane. He found a higher diffusion coefficient for radical cations of triphenylmethane and triptycene compared to other hydrocarbon radical ions of similar size indicating some sort of charge transfer mechanisms.

4. Mobility at higher electric field strength

Further support for the Stokes' model comes from measurements on negative ions in various hydrocarbons. Measurements of the mobility of unknown negative ions in n-hexane as a function of the applied field strength have been reported by Chong *et al.* [8], who found no dependence of the mobility on the applied electric field up to 150 kV/cm. In n-decane, Terlecki [9] extended the measurements up to 320 kV/cm and found that the mobility remained independent of the applied field. Terlecki studied negative ions, which were produced by attachment of photo-electrons to impurities (probably oxygen) present in the liquids. These ions were studied in n-hexane, n-octane and n-decane up to electric field strengths ≥ 300 kV/cm. No variation of the mobility with increase of field strength was found. The results are summarized

in Table 1.

liquid	Mobility $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$	Field kV/cm
n-decane	3.0×10^{-4}	0 to 330
n-octane	7.0×10^{-4}	0 to 190
n-hexane	9.75×10^{-4}	0 to 100

Table 1: Terlecki data, T = 296 K (from [9])

表1 テレッキのデータ 296 K (文献9から)

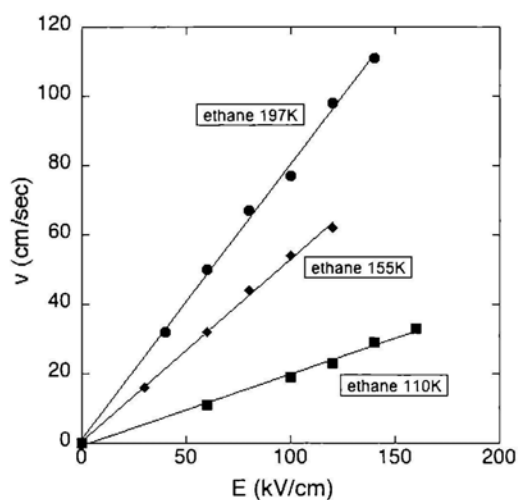


Fig. 7: Drift velocity of positive ions in liquid ethane as a function of applied electric field strength [5]

図7 液体エタン中の正イオンのドリフト速度の電界依存性 (文献5)

In our previous measurements of the positive ion mobility in liquid ethane we extended the electric field strength up to 160 kV/cm. The data showed strict proportionality of drift velocity and electric field strength (Fig. 7)

These findings are further proof that the motion of these ions proceeds in a laminar fashion. The transition between laminar and turbulent motion is determined by the Reynolds number, R_e given as

$$R_e = \frac{vl}{\kappa} \quad (7)$$

Here, v denotes the velocity, l is a characteristic length, and κ denotes the kinematic viscosity. For n-hexane the kinematic viscosity is at room temperature approximately 0.5 cSt or

$0.5 \cdot 10^{-6} \text{ m}^2 \text{ s}^{-1}$. The ionic radii are of the order of several Å up to a few nm. The mobilities of ions of unknown identity in n-hexane have been measured and values in the range of 10^{-4} to $10^{-3} \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ have been found. If we take for $l = 1 \text{ nm}$, and an applied field strength of 10^5 V cm^{-1} , we obtain for ions in n-hexane a Reynolds number of the order of $R_e \sim 10^{-3} \ll 1$. We may conclude that the motion of ions in these liquids proceeds in a laminar fashion up to rather high values of the applied electric field strength.

At much higher electric field strengths, turbulence sets in and the main effect would be a slowing down of the ion. For such conditions, Newton's law of friction should be applicable. The frictional force F_N is given as,

$$F_N = \frac{1}{2} c_w \rho A v^2 \quad (8)$$

where c_w denotes a dimensionless constant which is determined by the geometric shape of the moving object (a sphere in our case), A is the cross section of the ionic sphere, ρ denotes the density of the liquid and v is the velocity of the moving object. At any moment the frictional force should be equal to the electric force,

$$\frac{1}{2} c_w \rho A v^2 = \frac{1}{2} c_w \rho A (\mu E)^2 \quad (9)$$

where μ denotes the mobility of the ion and E the externally applied electric field strength. This equation yields a dependence of the mobility on the applied electric field as,

$$\mu = \sqrt{\frac{e}{\frac{1}{2} c_w \rho A E}} \quad (10)$$

or the mobility should be inversely proportional to the square root of the field. The deviation should occur at a field strength where mobility of Eq. 3 equals the mobility given by Eq. 10.

From both equations a critical field strength E_{crit} is obtained as,

$$E_{crit} = \frac{72 \pi \eta^2}{c_w \rho e} \quad (11)$$

The critical field depends on the viscosity and the density of

the liquid. Putting in values for hydrocarbons yields extremely high field strengths, which are outside of the experimental range. This result confirms the estimates from the Reynold number, Eq. 7.

5. Effect of pre-irradiation on the mobility

No systematic study of the changes in ion-mobility were made, which occur when the liquid or solution is pre-irradiated by gamma-rays up to a few tens of krad. In our previous studies on positive ion mobility in benzene, we found that pre-irradiation leads to the formation of slower ions as compared to the un-irradiated benzene. The results are shown in Fig. 8.

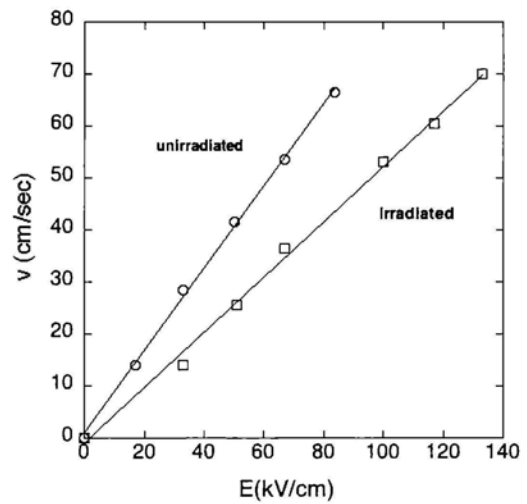
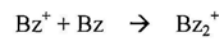


Fig. 8: Drift velocity of positive benzene ions, T=295K [10]

図 8 ベンゼンの正イオンのドリフト速度 295 K (文献 10)

Positive ions in liquid benzene produced by field ionization were found to form oligomers (see e.g. [11]). For this case, it is reasonable to assume that the parent positive ion reacts with a neighboring neutral to form a complex, which exhibits a larger radius and thus a lower mobility,



Formation of dimers of C_{60} in irradiated cyclohexane were found to exhibit a lower mobility than the positive C_{60} ion itself (see Fig. 9 and Fig. 6).

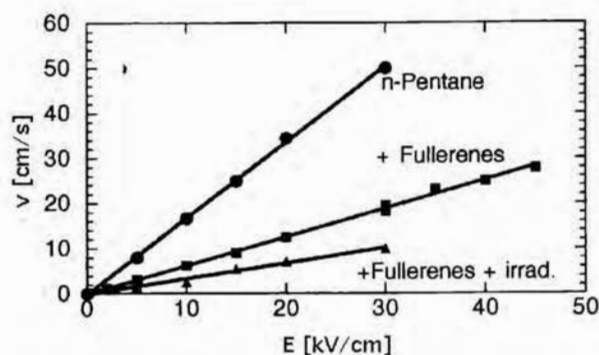


Fig. 9: Effect of pre-irradiation on the drift velocity of positive C_{60} ions; the data of n-pentane refer to the positive n-pentane molecular ion, $T=295K$ [12]

図9 C_{60} 正イオンのドリフト速度の前照射効果 n-ペンタンのデータは n-ペンタン分子正イオン 295 K (文献12)

6. Structure of ions

The structure of the ions in liquid hydrocarbons is determined by two physical processes: a) the polarization of the surrounding neutral molecules and b) by the repulsive action of the ionic shells of the ion and the neutrals.

In the case of positive ions, polarization is the predominant effect. One or several layers of induced dipoles are formed around the positive core. The radius of this cluster, R_0 is given by Eq. 5.

In ultra-pure hydrocarbon liquids, the negative carriers are electrons. In most other cases, the electrons become attached to impurity molecules (in many cases oxygen) or to voluntarily added scavengers (electro-negative molecules) present in the liquid. The lone electron exerts a repulsive action on the neutral molecules around it. Khrapak has formulated a model potential acting on the electron and solved the Schrödinger Equation for this case [14]. An important result is the fact that the radius of the orbit of the lone electron depends on the electron affinity of the scavenger molecule. Scavengers with a high electron affinity lead to the formation of negative ions, where the electron is incorporated into the electronic shell of the scavenger (for instance halogens) and the formation of a polarization cluster around the ion takes place, just like for positive ions. In the case of a low electron affinity, the electron orbit extends to a certain part outside of the scavenger molecule and repulsion of the surrounding neutrals takes place. A region of lower density is the result. Further outside, the polarization prevails. A delicate

balance between repulsion and polarization determines the radius of the ion. In the early years of mobility measurements on negative and positive ions in n-hexane a difference in mobility was found, $\mu_- > \mu_+$ [15] [16]. It is likely that the negative ions in these measurements were O_2^- ions, with a smaller radius than the positive n-hexane ions.

The influence of the competition of polarization attraction and exchange repulsion is very much apparent in liquid helium, due to its weak polarizability. A detailed study of the structure of various ions has been made recently [17].

In the case of non-polar liquids with a dielectric constant of approximately 2, in addition to the simple electrostatic argument of polarization, electrostriction and subsequently changes of the viscosity around the ions have to be taken into account. For ions in liquid xenon this has been attempted [18].

7. Concluding remarks

The interpretation of the ionic mobility in liquid hydrocarbons within the framework of Stokes' theory yields satisfactory results. Especially, it predicts that the mobility remains constant up to the highest values of the applied electric field strength. The scarce data available in the literature confirm this prediction. Near the boiling point and in supercritical conditions, the polarization shells melt away and movement of the bare ionic core has to be considered. In addition the strong fluctuations in density may lead to a different transport mechanism.

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