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Stokes' Law and the Mobility of Positive Ions in Mixtures of Methane and Ethane

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ABSTRACT

The mobility of positive ions was measured in mixtures of liquid methane and ethane at 110K. The applied electric field strengths extended up to 110 kV/cm. The drift velocity remained proportional to the applied field, indicating a laminar motion of the ions. Stokes law relating the mobility to the viscosity of the liquid is employed in the discussion.

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 Adamczewski¹. 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.

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 (see Fig. 1) 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 ions in liquid mixtures of methane and ethane at 110K.

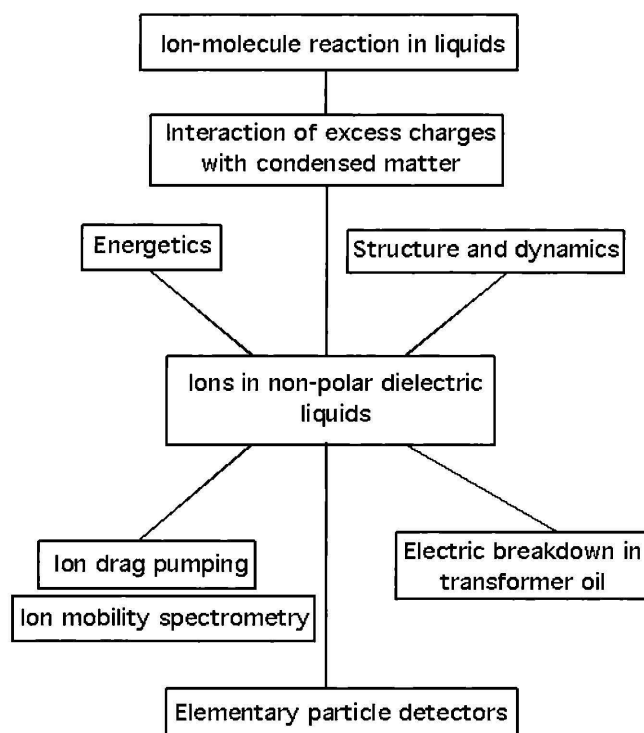


Fig. 1: Ions in liquid hydrocarbons and their relationship to various scientific and engineering topics

2. MEASUREMENT OF ION MOBILITY

The ions were produced in the liquid mixtures by ionizing the liquid with a pulse of high energy

x-rays from a linear accelerator. The experimental set-up is shown in Fig. 2.

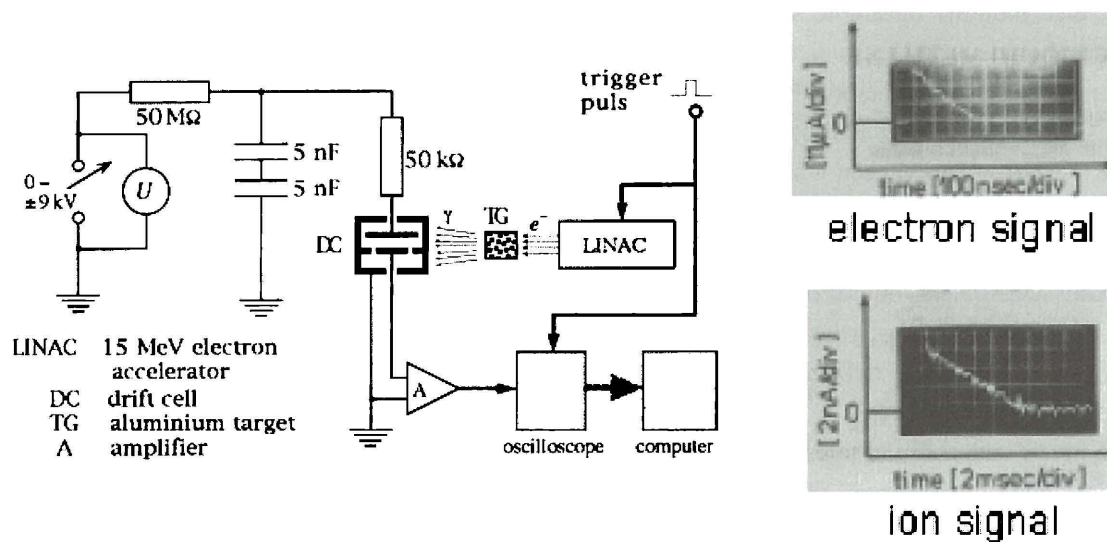
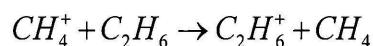


Fig. 2: Thick layer method²

Ionization leads to the formation of positive ions and electrons, which exhibit a much higher mobility as compared to the positive ions. In the mixture positive methane and ethane molecules are formed. The methane ions transfer their

charge to neutral ethane molecules, forming positive ethane ions since the ionization potential of methane (IP = 12.6 eV) is higher than that of ethane (IP = 11.5 eV),



The preparation of the liquid mixtures has been described already in the work concerning the electron mobility in these mixtures³.

3 RESULTS

The drift velocity as a function of applied field strength and the mobility of the positive ions as a function of composition is shown in Figs. 3 and 4.

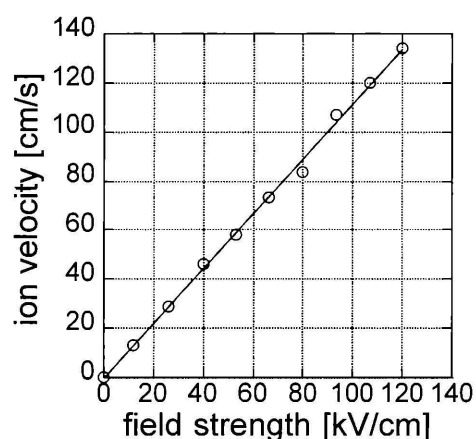


Fig. 3: Drift velocity as a function of field strength (X(eth)=0.04)

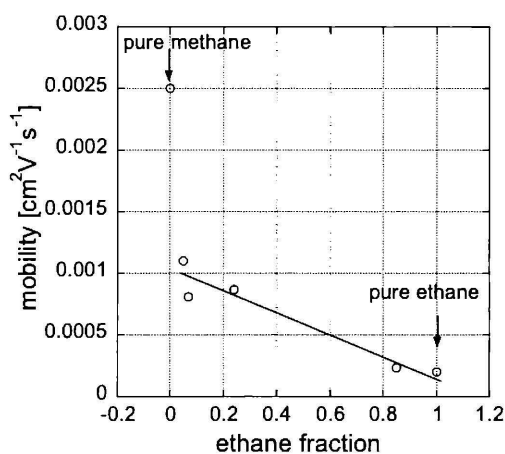


Fig. 4: Drift mobility

4 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, which can be written as

$$\eta = \frac{const}{\mu_{ion}} \quad (5)$$

In Fig. 5 the reciprocal mobility is plotted vs the ethane fraction X(ethane).

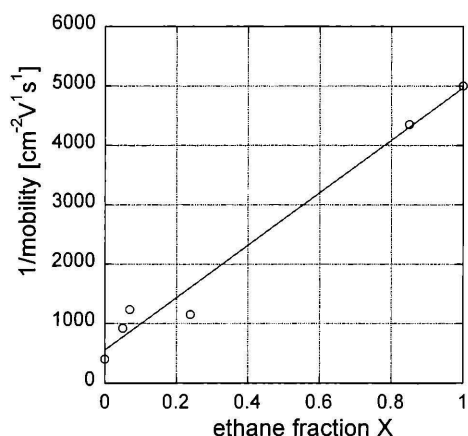


Fig. 5: Mobility as a function of ethane fraction

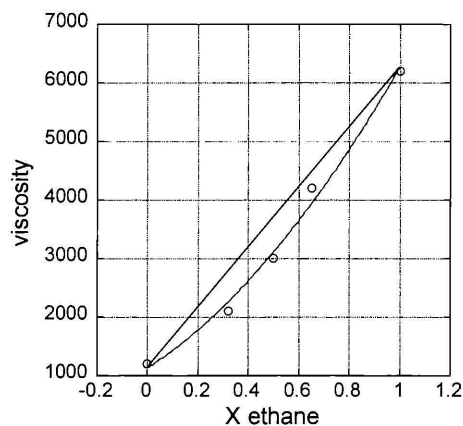


Fig. 6: Viscosity of the mixture after [4]

A reasonable straight line is obtained leading proof to the assumption of Stokes' law. Inherent in this plot is the assumption that the viscosity of the mixture is a linear function of the components, i.e. the viscosity of pure methane and pure ethane. Measurements of the viscosity of methane/ethane mixtures by Diller⁴ showed that slight deviations occur (see Fig. 6). However, the differences are small enough so that within the accuracy of the mobility measurements ($\pm 15\%$) the prediction of Eq. 5 can be taken as being fulfilled by the present data. An in depth discussion of Stokes' law and ion mobility was published recently by us⁵.

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