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Lichtverstrooiing door oplossingen van enkele colloid-electrolyten

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S U M M A R Y

The aim of the present work is to test the applicability of the light scattering method to dilute solutions of charged macromolecular substances such as polyelectrolytes and ionic micelles, and to investigate to what extent light scattering data can give information about the behaviour of these systems under various conditions, e.g. in the presence of varying amounts of extraneous electrolytes, at several temperatures etc.

The first part consists of three theoretical chapters in which the main problems encountered in the experimental sections are dealt with.

In chapter I a survey is given of the general theories of light scattering. A separate paragraph deals with the significance of the second virial-coefficient and the light scattering by multi-component systems. Particular attention has been paid to the various ways of handling data and interpretation of results (§ 6).

Chapter II deals with the most important current theories regarding the behaviour of dilute solutions of polyelectrolytes. The basic assumptions and essential features of the theories of Katchalsky c.s. and Hermans-Overbeek are reviewed and compared.

Chapter III gives a survey of the various current conceptions and energetic considerations concerning the formation of micelles in soap-solutions. In particular Debye's theory is amply discussed.

In the first part of chapter IV a detailed description of the apparatus used is given. It also contains diagrams of the optical and electronic parts of the turbidimeter. In view of the corrections to be applied, special attention has been paid to the optics of the secondary (scattered) beam. The refractive index increment is measured by means of a differential refractometer of which a description is given. It is pointed out that special precautions are required in order to keep the solutions optically as clean as possible. This condition forms the main drawback of the light scattering technique. Our solutions were prepared by diluting with ultrafiltered water and were submitted to centrifugation immediately before use. An apparatus for dust-free cleaning of glassware such as cuvettes or pipettes is described.

The turbidimeter is calibrated by means of a stable silica-solution "Ludox", the turbidity of which is determined by measuring the extinctions at various wavelengths in a simple visual photometer. The calculated turbidities of the common standard

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solvents such as benzene or toluene agree fairly well with those obtained by Carr and Zimm. The results are checked by the turbidimetric determination of the molecular weights of sucrose-octa-acetate and an international polystyrene sample. As a first application the molecular weights of commercial samples of colloidal iron oxide solutions are determined. The colloidal particles in these solutions prove to be spherical.

Chapter V contains a discussion of the results obtained by light scattering measurements on solutions of two polyelectrolytes, viz. polymethacrylic acid (PMA) and the sodium salt of a carboxymethylcellulose (NaCMC). A conductometric titration shows that the PMA molecules possess a much higher charge density, which fact seems to be responsible for the different light scattering properties observed: The dyssymmetric of the light scattered by solutions of PMA in water appears to pass a minimum value on dilution. On dilution of NaCMC no such phenomenon could be observed with certainty. A qualitative explanation for this different behaviour is given. In both cases the K_c/R_{90} -values drop sharply at high dilutions, so that extrapolation to zero concentration is impossible.

A more thorough investigation of the properties of the NaCMC molecule was carried out with five sets of mixtures of NaCMC and NaCl of constant total ionic strength. No irregular phenomena were observed in this case, and Zimm-plots could be given. A comparison of these Zimm-plots showed a decrease of the second virial coefficient at increasing ionic strength. A linear relation between the second virial coefficient and the reciprocal ionic strength proved to be valid. Although the so-called Donnan term seems to be responsible for this behaviour, the calculated values do not agree with the experimental ones.

The size of the macro-ion undergoes some changes when the total ionic strength is altered by addition of electrolytes. It was possible to follow these changes quantitatively by means of dyssymmetry measurements. At high ionic strength the polyelectrolyte molecule shrinks to a random coil, and expands on dilution because of an increased dissociation of the ionic groups. The limiting sizes for these two extreme cases agree fairly well with those calculated by Pals on account of viscosimetric data. The theory of Hermans and Overbeek could be checked in two ways: one of them gives good results, but the other being more sensitive to experimental errors leads to values which can not be reconciled with theory.

Light scattering measurements on solutions of a number of soaps (alkyltrimethylammoniumbromides and alkylpyridiniumbromides) are discussed in chapter VI. After an introduction in which the