

Modulation of Impedance in DNA Solutions by Ions and Molecules:

2. Effect of Tetraalkyl Ammonium Ions

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To understand natural biological events, it is important to understand interactions involving DNA-electrolytes-biomolecules – water. This prompted a critical look at the behavior of electrolytes(1), DNA(2,3), and molecules that interact with and chemically modify DNA(4). We had studied the frequency response of the concentration dependence of DNA, the influence of alkali metal ions, and peroxide. At present the impedance technique is used mostly to understand corrosion behavior of metals and batteries. The comprehensive list of ions and molecules included in our studies is intended to further develop impedance technique to provide information on solute-solute, solute-solvent, and solvent-solvent interactions near the electrode surface. We have observed that the impedance behavior of DNA in the absence of any background electrolyte is highly sensitive to frequency changes. The admittance data at frequencies less than 100Hz indicate solvent structural changes. DNA is sensitive to electrostatic interactions as exhibited by our results with alkali metal ions(2). The present study is intended to further explore these electrostatic interactions with the added attraction of introducing some hydrophobic groups in the ions.

Recent summary of the electrochemical analysis of nucleic acids(5) reveals that very low concentrations of DNA were studied at negative potentials of the mercury electrode. To achieve packing of DNA, we have used fairly large concentrations. We have also explored the region of both positive and negative potentials of the mercury electrode in order to simulate biological surfaces, and dopant effects.

The admittance data of 1 mg/mL calf thymus DNA in the presence of 0.01M solutions of ammonium, tetramethyl, tetraethyl, tetrapropyl, and tetrabutyl ammonium chlorides at pH 6.0-7.0 at 1000 and 100 Hz shown in Figures 1 and 2 indicate that with increasing length of the alkyl chain the admittance is less. Also the data clearly distinguish the influence of the alkyl chain especially at higher frequencies. The differences are much less significant at low frequencies. Also the nature of admittance is

different at high and low frequencies. The impedance data are shown at 0.3 V in Figure 3. Even though the impedance data are not different enough to characterize each tetraalkyl ammonium ion, it clearly shows a trend to reverse the direction of impedance at low frequencies. We have observed this earlier in systems such as palladium lipoic acid(6) and DNA-peroxide(4). We have attributed this in part to the close packing of molecules near the double layer which facilitates semiconduction. DNA by itself does not exhibit this tendency. The tetraalkyl ammonium ions also do not seem to exhibit such tendencies. The observed reversal behavior can be explained in terms of contributions from ion-solvent interactions and the interaction of the ions with the phosphate in DNA.

Figure 1: Admittance of 1mg/mL DNA with 0.01M Tetraalkylammonium Chlorides at 1000Hz

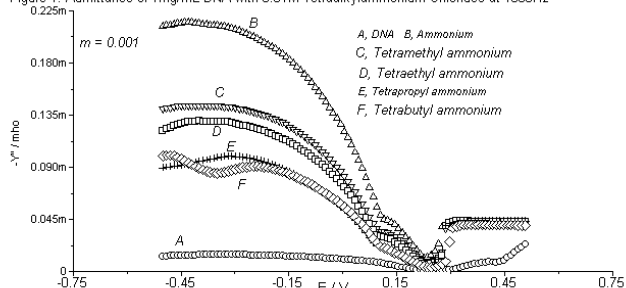


Figure 2: Admittance of 1 mg/mL DNA with 0.01M Tetraalkylammonium Chlorides at 100 Hz

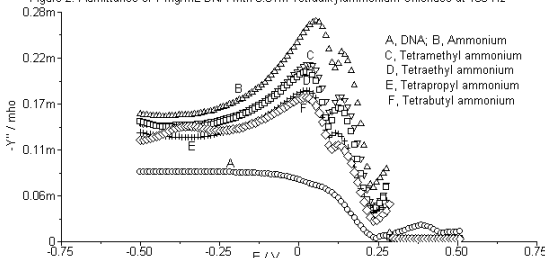
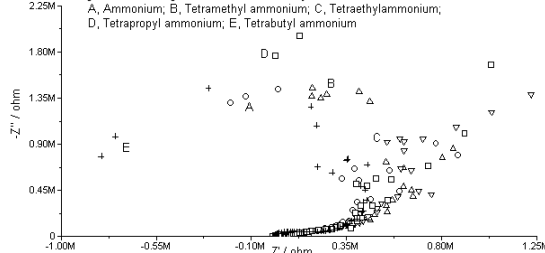


Figure 3: DNA 1mg/mL with 0.01M Tetraalkyl ammonium Chlorides at 0.3V



References:

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