

Detection Method for Ionic Species Based on the Electroacoustic Effect

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The development and application of a new ion detection method based on the electroacoustic (EA) effect is described. An EA signal, produced by applying a pulsed-type electric field to an electrolyte solution in an electroacoustic cell, is dependent on the electrical and thermal properties of the electrolyte and can be detected by using a conventional gas microphone system. The EA signals, generated in this fashion, are proportional to the square of the amplitude of the pulsed-type electric field and show an inverse dependence on the modulated frequency, as found in other acoustic detection systems. The results of this study demonstrate that the EA signals observed with the new system display a linear dependence on the concentration of the electrolyte over a 3 order-of-magnitude concentration range (ca. 10^{-7} – 10^{-4} M). The detection limit of this system was shown to be as low as 29.9 ppb for an aqueous solution of HCl. The results also indicate that the EA signal is proportional to the equivalent conductivity of electrolytes in aqueous solution. As a consequence, the new method has the potential of being used as a universal detector for ions in solutions. An important property of this detection system is that it can be applied to in situ ion detection, and as a result, it can be employed in kinetic studies to follow the progress of ionic chemical reactions.

Introduction

Acoustic wave technologies have been developed as unique techniques to study materials when more conventional methods are not applicable. Photoacoustic spectroscopy (PAS)¹ provides a means to directly measure the energies absorbed by materials as a result of their interaction with a photon beam. Owing to its high sensitivity and relatively simple experimental setup, this method has become a powerful tool for investigating the properties of solids and liquids. The photoacoustic technique has been employed to study liquid-phase samples containing low optically absorbing substances² that cannot be effectively probed by conventional spectroscopy. Despite its high sensitivity, the photoacoustic technique has some drawbacks, including the expense and wavelength selection limitations of lasers. Other acoustic techniques, such as thermoacoustic oscillation in cryogenics³ and the pulsed electroacoustic method for measuring charge accumulation in solid dielectrics,⁴ have also found important applications.

Studies using acoustical methods to determine the properties of electrolytes were initiated by Debye in 1933.⁵ Debye predicted that the propagation of an acoustic wave in a solution containing ions would induce an electric potential of the same frequency as that of the acoustic wave. This effect was experimentally verified,^{6–8} but the work showed that the method could not be used to determine electrolyte properties because only small and irregular potential differences were induced by acoustic waves. In 1985, Oja and co-workers⁹ demonstrated that an acoustic wave can be produced by applying an ac electric field to a charged colloidal solution. The phenomenon, termed the electrokinetic sonic amplitude (ESA) effect, was further developed^{10,11} in efforts directed at defining the dynamic mobility and the electrokinetic potential of colloid solutions.

High sound wave amplitudes in electrolyte samples could be also obtained when the fluid particles were driven directly by volume change forces by employing an ac magnetic field to induce motion in a ferromagnetic liquid.¹²

In the 1970s, Aamont and Murphy^{13,14} developed the thermophone, a device for generating an acoustic wave in metals by applying electricity, and used it to calibrate photoacoustic spectrometers. In addition, another method for electric field generation of acoustic waves, the so-called electroacoustic (EA) effect, in electrolyte solutions was uncovered by N. Tankovsky and co-workers.^{15–18} In this system, acoustic waves are produced in buffered electrolyte solutions that are in contact with a working liquid via a thin-foil diaphragm that serves as an electrode. In earlier investigations of acoustic wave generation, the thermophone technique was employed to investigate the influence of electric fields on solid materials, and the EA effect was used to describe the momentary movement of electrolytes in an ac electric field. However, these studies were not designed to quantitatively evaluate the sample concentration dependence of the acoustic signal, and as a result, they did not reveal the analytical potential of the technique.

Recently, we developed a new method for detecting ionic species that employs acoustic waves generated in solutions of electrolytes by using a periodic pulsed-type electric field. In contrast to the conventional electroacoustic method,^{9,11–14} a doubly modulated ac electric field is used as the energy source in conjunction with platinum (Pt) electrodes. As a result, the high-frequency, pulsed-type alternating current that is produced leads to exceptionally fast movement (few thousand times per second) of charged particles between the electrodes. The friction due to the interaction of the charged particles with solvent molecules causes heat to be released. Because the electric field is amplitude modulated in a low-frequency envelope, the alternating heat generated in the cell produces a low-frequency

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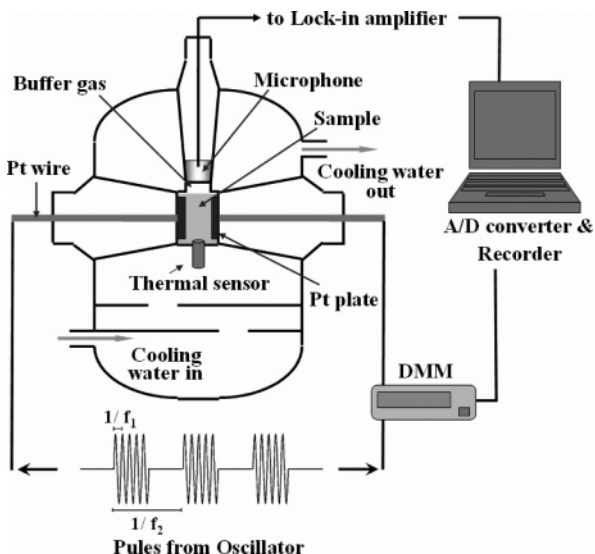


Figure 1. Schematic diagram of the EA detection system and EA cell. The shape of the applied pulsed-type electric voltage is shown.

acoustic wave in the gas above the solution. The acoustic signal generated in this manner reflects the thermal transport properties (eg., thermal conductivity and thermal diffusivity) of the electrolyte solution.

Generation of Electroacoustic Signal

In the conventional EA effect, movement of ionic particles of an electrolyte between two electrodes is induced by an application of a high-frequency ac electric field. This behavior produces an acoustic signal in the EA cell.¹⁴ However, the new electroacoustic method we have developed uses a high-frequency ac electric field as the energy source that is modulated by relatively low-frequency field. Specifically, an acoustic wave at frequency f_2 is generated in an electrolyte solution by applying a pulsed-type electric field, while a second ac electric field with high frequency f_1 is modulated within the envelope of the relatively low frequency f_2 wave. The shape of the synthesized pulsed-type electric field applied to the solution is shown in Figure 1.

When a modulated pulsed-type ac electric field is applied between two electrodes of an EA cell, ionic particles in the electrolyte move rapidly in a direction that depends on their electrical conductivity. During the alternating, short ($1/2f_2$ s) time periods, the ac current passes through the solution, and heat is periodically generated in the electrolyte, giving rise to an acoustic wave with a frequency of f_2 in the gas above the solution. Because current in an electrolyte solution results from ion motion, the current density is the sum of contributions made by both cations and anions. When u_j is defined as the mean drift velocity of ions j , each cation that is not farther from a fixed plane than the distance ($u_+ \times 1$ s) passes in one second. The same definitions apply to the anions. Thus the current density J can be written as shown in eq 1

$$J = (z_+c_+u_+ + |z_-|c_-u_-)F \quad (1)$$

where F is the Faraday constant ($96\,485\text{ C mol}^{-1}$), z_j and c_j are the respective ionic charge and concentration of the ion j .

The induced ion velocity u_j (eq 2) caused by an ac electric field, $E = E_0 \exp(i\omega t)$ applied between two electrodes, is defined by Tankovsky and Burov¹⁶ from the equation of motion for an ion having a mass m_j , an effective solvation radius a_j , a viscosity coefficient η , and an electrical charge e , taking into account

the exponential time dependence, $\exp(i\omega t)$:

$$u_j = \frac{e|z_j|E}{6\pi\eta a_j + i\omega m_j} \approx \frac{e|z_j|E}{6\pi\eta a_j} \quad (2)$$

Because $\omega m_j \ll \eta a_j$ for frequencies up to 10^{10} Hz, the upper approximation can be justified.¹⁶ For a general binary electrolyte $M_{v+}X_{v-}$ where v_j is the number of ions, j , the limiting (infinite dilution) molar conductivity, Λ_0 , can be expressed as shown in eq 3.

$$\begin{aligned} \Lambda_0 &= \lim_{c \rightarrow 0} \Lambda = \frac{J}{cE} \\ &= \frac{Fe}{6\pi\eta} \left(\frac{v_+z_+^2}{a_+} + \frac{v_-z_-^2}{a_-} \right) \end{aligned} \quad (3)$$

When the area of and distance between electrodes is A and d , respectively, the current between two electrodes can be written as

$$I = JA = c\Lambda_0 EA \quad (4)$$

During the time interval of $1/2f_2$, the accumulated electrical energy ($I \times E$) is expressed as shown in eq 5.

$$W = \frac{Ad}{2} c\Lambda_0 E^2 \frac{1}{f_2} \quad (5)$$

According to Aamodt and Murphy,¹³ the time-dependent acoustic signal associated with uniform heating of an ionic solution in a sample cell is derived by using the diffusion equation, which consists of the source (or driving) term and the cell-response term. The source (or driving) term incorporates the dependence of the acoustic signal at time t on the sample properties, and its value is dependent on the manner in which the sample is heated. The cell-response term describes the effect of the physical and thermal properties of the cell on the acoustic signal. Suppose that the thermal conductivity of the buffer gas is zero so that no energy is transferred into the buffer gas from the sample surface. Thus, the source term would be proportional to the temperature at the sample/buffer gas interface. Assuming that the electrical heating of the sample generates uniform heating in the sample, the rate of heat generated, $H(t,x)$, is independent of space, x , i.e., $H(t,x) = H(t)$.

Note that the acoustic signal in ionic solution in the presence of an ac electric fields consists of a driving function $H(t)$, modified by a series of response functions, which is the electrical power unit volume for electrically heated samples. The applied electrical energy is converted into both the kinetic energy and heat. In most cases, the kinetic energy associated with the ion motions is so small that it can be ignored compared with the potential energy or heat, which is transformed into the generated electroacoustic signal. Thus, by taking into account the accumulated electrical energy, W , in eq 5, the heat generated in electrolyte, which is uniform in space and time, can be defined as displayed in eq 6.

$$H(t) = \frac{Ad}{2} c\Lambda_0 \frac{1}{f_2} \{E_0 \exp(i2\pi f_1 t)\}^2 \quad (6)$$

It is evident from eq 6 that the heat generated, causing the electroacoustic signal, should be linear to the concentration, c , and the limiting molar conductivity, Λ_0 , of the ions. As can be also seen, the interesting feature of the electroacoustic effect is

that the application of the electrical energy that is frequency modulated in intensity results in heat variations that are also modulated and of the same frequency. The f^{-1} dependence in eq 6 arises from the fact that the amount of heat generated in the system per cycle decreases as the modulated frequency increases, giving rise to the acoustic signal amplitude that is inversely proportional to the frequency of the modulation, f_2 . Because the values in the parenthesis specify the complex envelope of the sinusoidal variation at high-frequency f_1 , the equation may be evaluated for the magnitude and phase of the acoustic wave produced by the electroacoustic effect. The actual physical heat variation is given by the real part of $H(t)$, and the phase of the acoustic signal is governed by the imaginary part of $H(t)$. At the signal processing stage, the phase-sensitive detection technique in conjunction with a low-pass or band-pass filter can eliminate the small alternating high-frequency components, so that the magnitude of the acoustic signal turns out to be independent of frequency f_1 of the electric field.

Experimental Section

A block diagram displaying the experimental arrangements used in this study is shown in Figure 1. The electric source is synthesized by using a sweep function generator (Hung-Chang, Inc., model G305), which produces a pulsed-type electric voltage via a burst mode with a high-frequency component superimposed on a low-frequency envelope. In this system, an electric chopper instead of a mechanical chopper is used to avoid temperature effects caused by constant heating of the solution. The voltage from the function generator is electronically amplified (Töllner Electronic Ins. GmbH, TOE 7608S) and applied between two Pt electrodes in the EA cell. A low-frequency (f_2) acoustic wave is then generated in the EA cell. An alternating high-frequency current of 20 kHz with a relatively low voltage is used to eliminate possible electrolytic redox reactions and direct current polarization effects.

The acoustic signals are detected by using an electret microphone with a built-in field-effect transistor preamplifier (Radio Shack model 270-092B), which is in contact with air as a buffer gas above the solution. The acoustic wave detected by the microphone is amplified and detected by using a lock-in amplifier (Stanford Research System, SR 850) tuned to the modulation frequency and then analyzed by a personal computer. Both the voltage and current at the liquid sample are separately measured by using a digital multimeter (DMM, Keithley Instruments, Inc., model 197A) in order to calculate the total power applied to the cell. The current is also recorded by a personal computer at the same time with the signal. The lock-in amplifier and the DMM are interfaced by using an IEEE-488 interface board (Keithley Instruments, Inc.). Note that the use of a lock-in amplifier allows observation of only the principal Fourier component of the sine wave caused by electric heating. The temperature of the solution is measured by using a thermocouple in the cell and monitored with a digital temperature indicator (Hanyoung, AT3-K).

The handmade Pyrex glass electroacoustic cell used in these experiments is shown in Figure 2, where the microphone is placed at the top of the cell. The total volume of the cell is ca. 7 mL (liquid sample of 5.5 mL and gas layer of 1.5 mL). The Pt plate electrodes are 5 mm in width, 10 mm in length, and 1 mm in thickness. The electrodes are separated by 10 mm and placed at the ends of the electroacoustic cell. The EA signal detected by using the instrument described above is dependent on the conductivity Λ_0 of electrolytes, which typically varies greatly with temperature. It has been reported that conductivities

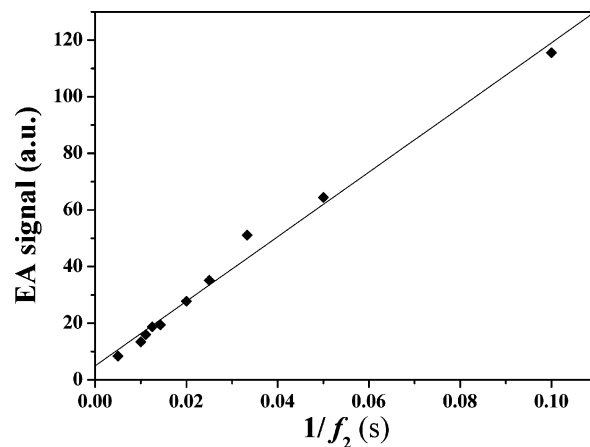


Figure 2. EA signal as a function of the chopping frequency f_2 with 0.001 M KCl at f_1 of 2 kHz and constant applied voltage of 200 V_{pp} ($R^2 = 0.9944$).

of electrolyte solutions deviate by ca. 2–3% for a 1 °C temperature change from 25 °C.^{16,17} To avoid these effects, the temperature of the EA cell is rigorously maintained at 25 ± 0.5 °C. All solutions and calibration standards containing ions are prepared with 18-M Ω water (Milli-Q academic A10 system, Millipore Corp.). A blank measurement using 18-M Ω water is made prior to all ion signal measurements. Each data point is an average of five measurements, and in each case, the data are normalized with the reference.

Results and Discussion

Four main factors are responsible for generating pressure in the vicinity of an ion between the two parallel-plane electrodes of the EA cell when an ac electric field is applied.¹⁶ The first comes from the electroacoustic effect associated with translational motion of ions that resembles a piston oscillating in a liquid. The second source arises from the nonhomogeneous electric field created by the interaction of the ions and the external electric field with solvent molecules in the vicinity of the ion. The thermoelastic force due to the local heating produced by solvent friction in the region close to the ion is the third factor. Finally, a nonhomogeneous medium mechanism, a component of the electric force density, also contributes. These factors, summed over all ions, contribute to the pressure generated in the EA cell. On the basis of the assumption that the electrolyte is a resistance medium and that the temperature change of the electrolyte is entirely and uniformly transformed into an acoustic wave, the signal intensity detected by the microphone will be dependent on several factors. These include the applied voltage, current, chopping frequency, concentration, and physical properties of ions, and the sensitivity of a microphone.

Studies were designed to characterize the dependence of the EA signal on the applied voltage and current. In one experiment, a 0.001 M KCl solution, electric field frequency f_1 of 2 kHz, modulation frequency f_2 of 20 Hz, and amplitude of a voltage range of 20–220 V_{pp} were used. The results showed that the EA signal is proportional to the square of the amplitude of the pulsed-type electric field, where the current was directly proportional to the voltage. Thus, the EA signal is proportional to the input power (W), with the predicted quadratic dependence on E . In addition, the EA signal (Figure 2) exhibits a clear dependence on the reciprocal of the chopping frequency f_2 in the range of 10–200 Hz (0.001 M KCl solution, constant voltage of 200 V_{pp} applied at fixed f_1 of 2 kHz). The signal shows good

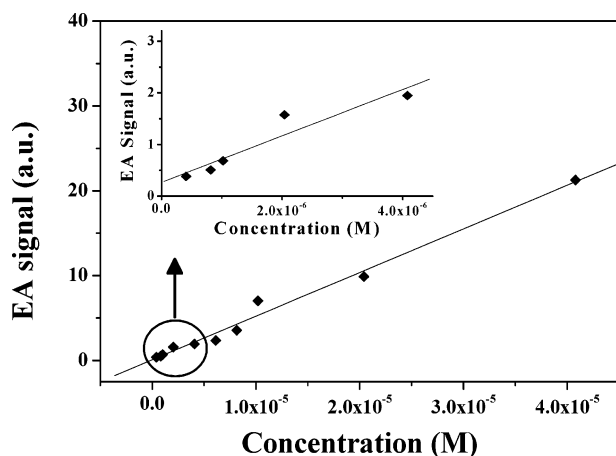


Figure 3. Plot of EA signal vs concentration of HCl. Each point is the average of five consecutive runs normalizing with 1×10^{-5} M KCl solution ($R^2 = 0.9934$). The upper graph represents the zoom in circle (4×10^{-7} – 4×10^{-6} M concentration ranges).

reproducibility, where the relative standard deviation (RSD) lies within 5% ($R^2 = 0.9944$). This result confirms that acoustic signals produced by periodic electrical or optical heating are inversely proportional to the chopping frequency at a constant power.⁹ The combined results prove that the EA signal follows exactly the pattern seen in photoacoustic experiments.

To evaluate the influence of the high-frequency input voltage, EA signals were recorded on a 0.001 M KCl solution at a chopping frequency f_2 of 20 Hz, applied voltage amplitude range of 100 V_{pp}, as a function of the frequency f_1 in the range of 400–20 kHz at the fixed electric current. The signal was found to be independent of frequency f_1 of the pulsed-type electric field. The pulsed-type ac electric voltage used in this study produced an acoustic signal, which has a frequency that is the same as the modulation frequency, f_2 . If the EA signal varying at frequency f_2 is generated only by frictional heat produced by the translational motion of the ions, the frictional heat generated during the first $1/2 f_2$ second would be dependent on the total distance over which the particles move during that time period. Because, at the same value of electric current, the distance ($v_j \times 1/2 f_2$) is not related with the frequency f_1 , the EA signal measured is independent of the applied ac voltage with the frequency f_1 .

To probe applications of this new methodology for quantitative analyses, various ion detection experiments were carried out to obtain calibration curves of the EA signal vs concentra-

TABLE 1: Comparison of Sensitivity of EA Signal for Different Salts with Their Conductivities

	conductivity (Λ_0) ^a	sensitivity ^b	R^2	detection limit (pmole) ^c
HCl	425.95	5.142	0.9934	4.1
AlCl ₃	411.93	4.621	0.9993	4.7
MgCl ₂	258.68	2.589	0.9993	9.1
KCl	149.79	1.838	0.9996	14.1
KBr	151.90	1.697	0.9999	14.5
KF	128.88	1.334	0.9998	18.3
NaCl	126.39	1.182	0.9993	20.0
LiCl	114.97	0.881	0.9999	27.9

^a Λ_0 (10^{-4} m² S mol⁻¹): limit molar conductivity referred to aqueous solution at 25 °C (refs 20–21). ^b Sensitivity: slope in plots of EA signal (V)/concentration (M). ^c Signal-to-noise ratio of 2.

tion. In Figure 3 is shown the observed dependence of the EA signal on HCl concentration in the range of 4×10^{-7} – 4×10^{-5} M (amplitude of ac electric field of 200 V_{pp}, $f_1 = 2$ kHz, and $f_2 = 20$ Hz, at 25 °C). The results clearly demonstrate that the EA signal is directly proportional to the concentration of HCl with a detection limit of 29.9 ppb for HCl (18-M Ω water as the blank) at the signal-to-noise ratio of 2. As shown in Figure 4, EA signals arising from solutions of aluminum chloride, magnesium chloride, potassium chloride, potassium bromide, potassium fluoride, sodium chloride, and lithium chloride show a linear relationship over a 3 orders-of-magnitude range of concentration (1×10^{-6} – 4×10^{-4} M). In Figures 3 and 4, although at lower concentrations some EA signals show the RSD values in the range of 5–10%, most signals exhibit a fairly good reproducibility with the RSD values of less than 5%. Thus, EA signals obtained by using the new EA technique display good linear relationships with electrolyte concentrations over fairly wide concentration ranges. The limit of detection (LOD) and R^2 values for the ionic solutions are listed in Table 1. It is also noted in Figure 4 that the slopes of the curves, which represent the sensitivity of the detection response, are changed according to the ionic solutions. This demonstrates that the signal sensitivity is closely related to the intrinsic properties of ionic samples. Consequently, these results strongly suggest that the method will find general applications as an effective technique to detect ionic species in electrolyte solutions.

The EA signals observed in the above experiments are dependent on the conductivities of the solutions. The intrinsic properties of both the positive and negative ions, including the number of valence electrons, effective solvation radius, and nuclear charge, will influence the conductivity. In light of this,

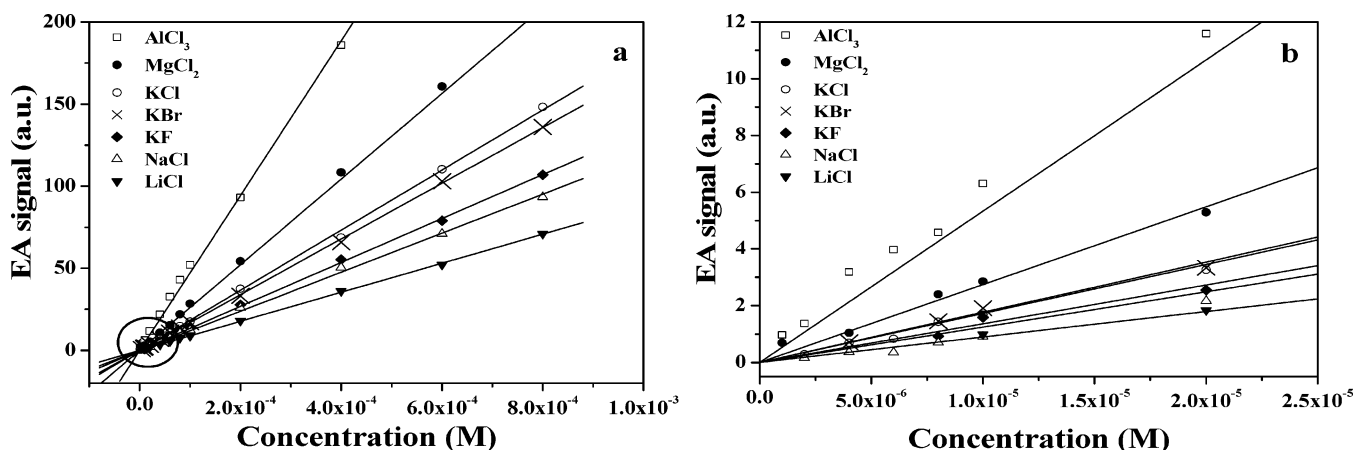


Figure 4. (a) Plots of EA signals vs concentrations of seven salts (\square : aluminum chloride; \bullet : magnesium chloride; \circ : potassium chloride; \times : potassium bromide; \blacklozenge : potassium fluoride; \triangle : sodium chloride; \blacktriangledown : lithium chloride). (b) Plots of signals vs lower concentration marked as circles in (a). Each point is the average of five consecutive runs normalizing with 1×10^{-5} M KCl solution.

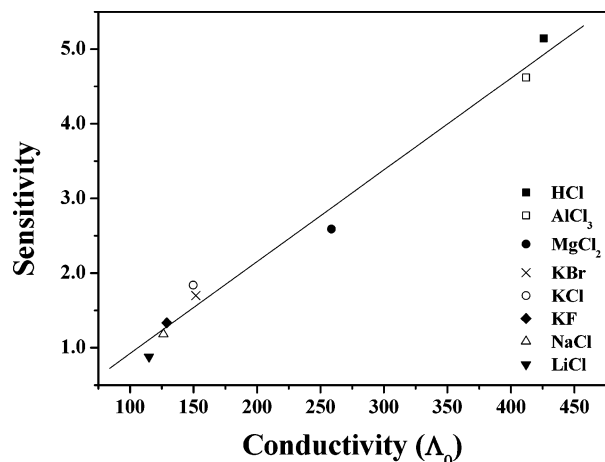


Figure 5. A plot of the sensitivity (slope in plots of EA signal vs concentration) for different salts as a function of their conductivities ($R^2 = 0.9914$).

the observation that, for alkali cations bearing the same counteranion Cl^- , the magnitude and sensitivity of the signal increase in the order of Li^+ , Na^+ , and K^+ is interesting. Note that the metal cation dependence of the signal intensity is in line with their increase in molar conductivity. The same effects are observed for cationic species with higher nuclear charges, i.e., the larger signal is produced by metal cations with higher conductivities. These conclusions arise from inspecting the correlation that exists between the slope in plots of EA signal vs concentration, expressing the sensitivity of the EA signal, and conductivity shown in Figure 5 and summarized in Table 1. The plot displays a linear dependence of the signal sensitivity on the limiting conductivities for the salts. Importantly, the EA signals from two salts with similar conductivities are similar, even though the effective solvation radii or atomic weights of the metal cations are different. Because these results are obtained from measurements made under identical conditions (including concentrations), they show that electroacoustic signals can be used as a universal measure of ion properties that are directly related to the conductivity. As a consequence, it appears that this EA method will find its important application in direct measurement of the ionic conductivity in solutions.

In this study, it is not attempted to investigate the minimum detectable volume of the solution because the present study focuses mainly on the demonstration of measurements of acoustic signals from the electrolyte solutions. For the acoustic wave generation, the volume of samples is not generally restricted, as can be seen in other acoustic detections such as the photoacoustic spectroscopy. It is thus believed that the EA method described in this work can be applicable to the detection of microliter samples of ionic solutions with the improvement in the cell design. It is also noteworthy that current changes associated with movement of ionic species are also measured and the results display features that parallel those accompanying the acoustic measurements. These observations point out the

meritorious feature of the new technique that allows electric current measurements in an EA cell by a high-precision digital multimeter simultaneous with those of acoustic signals.

Conclusions

An electroacoustic method based on simple pulse modulation of an applied ac electric field has been developed for the first time in these studies. Several limitations of conventional EA methods are eliminated by using this new technique. The new EA method is highly sensitive for the detection of most electrolytes in solutions, and it has a good linear response to electrolyte concentrations over wide ranges. As a result, the method can be used to both detect and quantitatively analyze ionic species. Its quantitative capability suggests that the new EA method can be further developed into a new strategy for highly sensitive ion detection. It is also instructive to recall that the concepts of the EA detection in this work possess strong implications for the promise of serving as a universal detector, where this method is employed in conjunction with the ion chromatography. Moreover, the method has the potential of being used as an in situ probe to investigate the kinetics of ionic chemical reactions¹⁹ and to simultaneously evaluate the electric and thermal properties of liquid samples.

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