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Ultrasonic Imaging of the Electroacoustic Effect in Macromolecular Gels

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Abstract

The electroacoustic effect occurs in electrolytes and colloidal suspensions. It describes the phenomenon in which a voltage applied to the sample produces an acoustic signal or vice versa. The basic mechanism is that charged particles in the sample have various mobilities due to different masses and viscosities. Under an external voltage they respond differently to the electrical force. This results in an overall acoustic vibration. The electroacoustic effect has been the basis for many measurement tools of solutions and other materials. In this note a method to image macromolecular gel samples using the electroacoustic effect at ultrasound frequencies is presented. Radio-frequency electrical excitation produces ultrasonic signal due to spatial changes in the electroacoustic sonic amplitude of the sample, which is used to construct an image similar to ultrasonography. This method is demonstrated in agar gel and eggwhite protein phantoms. The image contrast mechanism is also discussed.

Keywords

Biological tissue; colloid; electroacoustic; electrolyte; macromolecular gel; ultrasonic imaging

Introduction

The electroacoustic effect was predicted to occur in electrolytes by P. Debye¹ many decades ago. He reasoned that ions in an electrolytic solution have different friction constants relative to the solvent and different masses, so that when acoustic waves are transmitted into the solution, the oscillatory movements of the ions lag behind the bulk solvent to various degrees. The non-uniformity in motion results in charge surplus in certain areas and deficit in other areas, leading to detectable potential differences. Debye related the voltage to parameters including the ion friction constants, ion masses, and the overall conductivity and dielectric constant of the solution. He proposed to use this phenomenon to study ion solvation. The electroacoustic effect can also be reversed²: an electrical voltage applied to a sample induces varied kinetic responses among the charged particles. The net force exerted by these particles on the surrounding medium causes acoustic vibrations, which can be detected with hydrophones. A reciprocity relation exists between the forward and reverse effects³.

The electroacoustic effect in colloidal suspensions has been studied extensively in the literature. O'Brien⁴ developed a theoretical treatment based on the double-layer model of

colloids⁵. It models the particles in the suspension as comprising of a rigid core covered by a thin fluid layer. The thin layer is polarized, i.e., the interior and exterior surfaces carry opposite charges. When one applies a high frequency acoustic wave to the suspension, the thin fluid layer slides and compresses differentially around the core, redistributing the surface charge^{4, 6}. This creates an electric dipole moment. The sum of the dipole moments of all the particles gives rise to an electric field, the colloid vibration potential (CVP). Conversely, one can apply an electric field and measure the resultant acoustic signal, called the electronic sonic amplitude (ESA). O'Brien⁴ and Dukhin and Goetz⁶ showed using this model that the CVP and ESA are determined by the size of the particle, and the characteristics of its surface layer including shear viscosity, compressibility and the zeta potential (the potential drop across the surface layer of the particle). The double layer theory has been used successfully for a wide variety of suspensions, from milk⁷ to silica emulsion⁸.

In contrast the theory for ionic solutions has not gone far beyond Debye's description. In Debye's theory the mechanisms by which momentum is transferred between the ions and the surrounding solvent is summed into the phenomenological friction constant between them. The CVP or ESA is expressed as

$$ESA = \frac{\sum_j n_j q_j m_j / \eta_j}{1 - i \frac{2\sigma}{\epsilon f}}, \quad (1)$$

where the summation is over all ion species, n_j , q_j , m_j and η_j are the number density, the charge, the mass and the friction constant of each ion species, σ and ϵ are the conductivity and the dielectric constant of the solution, and f is the frequency of the applied electric field. Tankovsky and Burov⁹ attempted detailed analysis of the electrodynamic interactions between an ion and the surrounding dipolar medium in order to derive an explicit expression for the ESA without resorting to the phenomenological friction constant, but could not reach a conclusive theory¹⁰. Nonetheless eq. (1) provides some insight into the electroacoustic effect in organic solutions or biological samples. The term nqm/η physically means the ratio between mass and friction weighted by the charge of the ions. The mass-to-friction ratio determines how readily the ions follow the movement of the solvent, i.e., the dynamic mobility of the ions. Large organic ions such as proteins may have large mass to surface area ratios and therefore large mass-to-friction ratios. For this reason their contribution tend to dominate the summation in eq. (1). The ESA or CVP in biological samples may be primarily a measure of the concentration and dynamic mobility of the charged macromolecular content.

In general there is a lack of quantitative models of the electroacoustic effect for systems other than the basic suspensions of spherical, electrically neutral particles. Nonetheless, the purpose of this paper is to demonstrate that this effect can be used to form images of samples that contain regions of different electroacoustic properties. Spatial resolutions similar to ultrasonography may be achieved if the effect is induced with radio-frequency (rf) electrical pulses in the MHz range. Consider the experiment shown in Fig.1, where an electrolytic sample is sandwiched between two metal sheet electrodes. The sample is consisted of two contiguous compartments (labeled I and II) of different materials, such as saline and agar gel. The electrodes are connected to a rf pulse generator, which produces a pulsed electric field $\mathbf{E}(\mathbf{r}, t)$ through the sample, $\mathbf{r}$ is the spatial location. An ultrasound transducer is placed above the ground electrode to receive the electroacoustic signal.

Under the electric field $\mathbf{E}$, the sample medium experiences a mechanical force $\mathbf{F}(\mathbf{r}, t)$ from the electroacoustic effect, which is related to the electric field by the electrokinetic sonic amplitude (ESA) $A(\mathbf{r}, t)$:

$$\vec{F}(\vec{r}, t) = A(\vec{r}, t) \vec{E}(\vec{r}, t). \quad (2)$$

If the sample contains both colloidal particles and ions, the ESA parameter should include the colloidal contribution and the ionic contribution in eq. (1). As explained above, the colloidal contribution is determined by the size and ζ -potential of the particles, while the ionic contribution is determined by the ratio nqm/η of the ions.

The force field $\mathbf{F}$ produces an acoustic oscillation field $\mathbf{x}(\mathbf{r}, t)$ in the sample, which should satisfy the following equation of motion assuming that acoustic attenuation is negligible:

$$\rho \frac{d^2 \vec{x}}{dt^2} - Y \nabla^2 \vec{x} = \vec{F}(\vec{r}, t), \quad (3)$$

where Y is the Young's modulus of the sample medium. In electrolytic solutions and dilute colloidal suspensions, Y and ρ are approximately uniform in the sample. We denote the acoustic pressure wave as $p(\mathbf{r}, t)$. By taking the spatial divergence of both sides of eq. (3)¹¹, and using

$$-Y(\nabla \cdot \vec{x}) = P, \quad (4)$$

eq. (3) becomes

$$\frac{d^2 P}{dt^2} - \frac{Y}{\rho} \nabla^2 P = -\frac{1}{\rho} \cdot \vec{F}(\vec{r}, t). \quad (5)$$

Here second order terms are omitted. Substituting eq. (2) into eq. (5) gives

$$\begin{aligned} \frac{d^2 P}{dt^2} - \frac{Y}{\rho} \nabla^2 P &= -\frac{1}{\rho} \cdot (A \vec{E}) \\ &= -\frac{1}{\rho} (\vec{E} \cdot \nabla A + A \nabla \cdot \vec{E}) \\ &= -\frac{\vec{E}}{\rho} \cdot \nabla A + \frac{A}{\rho} 4\pi q, \end{aligned} \quad (6)$$

where q is the charge density in the sample induced by the electric field. It is non-zero when there are spatial changes in dielectric constant or conductivity in the sample. The physical meaning of eq. (6) is that acoustic waves are emitted from areas where there are spatial variations in the electrical constants or the ESA parameter.

In the experiment shown in Fig.1, acoustic waves are emitted from the interface between compartments I and II, and propagate to the surface of the sample where an ultrasonic transducer receives them. By measuring the acoustic signal p at the surface and back-projecting its sources, one obtains a map of these gradients. The back-projection is possible if the electroacoustic signal is induced by single rf pulses. For example, if a rf pulse of 0.5 μ s duration is applied to the sample at time zero, and an ultrasound pulse is received by the

transducer at time t , then the distance between the source of the pulse and the transducer is ct , where c is the speed of sound, and the direction of the source is defined by the beam direction of the transducer. This image construction process is similar to echo ultrasonography, where time of flight is converted into spatial location. The longitudinal resolution of the image is approximately the product of the pulse width and c . In water based samples, a $0.5\mu\text{s}$ pulse gives approximately 0.7mm resolution. The transverse resolution is approximately the size of the transducer's focal point, which is usually poorer than the longitudinal resolution. The mathematics that describes the spatial resolutions is similar to echo-based sonography and passive sonar.

It should be noted that eq. (6) predicts the timing of the electroacoustic signal, but does not provide a quantitative estimate of the signal amplitude or phase. This is because it is based on the phenomenological ESA parameter. In the existing theories of the electroacoustic effect, quantitative estimates of the ESA are possible for only well controlled artificial suspensions of neutral spherical colloids. The theories for ionic solutions and gels are qualitative, and the ESA is expressed in terms of other phenomenological parameters such as ion friction and effective mass (eq. (1)). Therefore confirmation of the electroacoustic signal relies on time of flight measurements. Equations (1) and (6) do show that the electroacoustic signal reverses sign when the direction of the applied field is reversed. This can be checked experimentally. In the following section, imaging experiments and results on two different samples are described.

Methods and Results

The basic experimental setup is shown in Fig.1. The sample is contained in a plastic container of cross-section 11.5cm (width) $\times$ 5.3cm (height). The bottom electrode is an adhesive copper sheet (3M Corp) covering the bottom of the container. The top electrode is made of $15\mu\text{m}$ thick aluminum foil, which allows the ultrasound waves to propagate through it. The rf pulser (Panametric Inc.) produces exponentially decaying pulses of 400V peak voltage and $0.3\mu\text{s}$ half height width. An ultrasound sensor is placed above the aluminum foil to receive the electroacoustic signal. It is a single-element broadband 2.25MHz focused transducer of 1.9cm diameter and approximately 3.8cm focal distance (KrautKramer-Branson). In general there exists a small electromagnetic cross-talk coupling between the transducer and the pulser. If a large pulse is applied to the sample, the cross-talk induces a voltage in the transducer which in turn emits an ultrasonic pulse into the sample. The echoes of this pulse appear as coherent noise, and can overwhelm the weak electroacoustic signal. To suppress the cross-talk echoes, the transducer is shielded with a 13cm oil-filled waveguide stand-off. This shielding method is described in ref. 12. It effectively suppresses the cross-talk echoes by two orders of magnitude or more. A layer of water is placed between the stand-off and the aluminum foil as the acoustic coupling medium. Figure 2 shows a photograph of the sample container with the aluminum foil open.

Two different samples were tested. The first sample consisted of 0.2% saline solution in compartment I and cooked eggwhite in compartment II. Figure 3a shows the ultrasound signal received by the transducer, averaged over 1000 data collections for random noise suppression. The transducer was placed above the center of the sample, and the aluminum foil was the ground electrode. To trace the origins of the signal peaks, their times of flight were compared with the depths of the interfaces measured prior to the experiment. This process shows that peaks (a), (b) and (c) were produced at the aluminum sheet, the saline-eggwhite interface, and the copper sheet at the bottom of the box, at the instant of the excitation pulse; peak (d) was a reflection of peak (b) from the bottom of the box; from its time of flight, peak (e) traveled twice the distance between the transducer and the bottom of the box, therefore it was a residual cross-talk echo from the bottom of the box. Thus peaks

(a) to (d) originated from the electroacoustic effect, while peak (e) was due to the cross-talk between the transducer and the pulser.

Figure 3b shows the result with the polarity of the electrode pair reversed. Again by matching times of flight with depths of the interfaces, peaks (a) to (c) were assigned to electroacoustic signal from the aluminum sheet, the saline-eggwhite interface and the bottom copper sheet. Peak (d) was the reflection of peak (b) from the bottom of the box. Peaks (e) and (f) were cross-talk echoes of the aluminum sheet and the bottom of the box. Note that the signal from the saline-eggwhite interface reversed its phase, consistent with the phenomenological models¹.

Cross-sectional images of the sample were collected by scanning the transducer across the container surface at 1.06mm (1/24") intervals. At each position a trace similar to Fig.3 was taken. Figure 4 is the magnitude image of the sample. The saline-eggwhite interface was clearly depicted except at the two places where it was almost perpendicular to the transducer surface. In these areas the electroacoustic signal propagates horizontally, and can not be detected with a single-element focused transducer. The two electrode surfaces can also be seen. The bright line immediately below the copper sheet was the echo of the signal from the copper sheet reflected within the bottom layer of the container. The traces beyond the bottom of the box were assigned by their times of flight as follows: traces labeled (a) were reflections of the electroacoustic signal of the saline-eggwhite interfaces, reflected by the two surfaces of the bottom of the box; trace (b) was the residual cross-talk echo from the saline-eggwhite interface; the two adjacent traces labeled (c) were the reflections of the electroacoustic signal from the aluminum sheet, reflected by the two surfaces of the bottom of the box; the two traces labeled (d) were cross-talk echoes of the two surfaces of the bottom layer of the box.

In a different sample consisted of saline for compartment I and 2% agarose for compartment II, the same experiment was repeated. Figure 5 shows the cross-sectional scan of this phantom, with the only half the gray scale range as Fig. 4. The saline-agar interface is visible, but the signal amplitude is approximately one-third of the saline-eggwhite interface, taking into account the gray scale change. Upon reversing the polarity of the electrodes, the signal phase also reversed (not shown). Again the traces beyond the electroacoustic signal were assigned by their times of flight: traces labeled (a) were reflections of the electroacoustic signal of the saline-agarose interface from the bottom of the box; the group of traces labeled (b) were from the electroacoustic signal of the copper sheet, after multiple reflections between the two surfaces of the bottom layer and the saline-agarose interface; the two traces (c) were reflections of the electroacoustic signal of the aluminum sheet, reflected by the surfaces of the bottom layer; the two traces (d) were cross-talk echoes of the surfaces of the bottom layer.

The difference in signal level between the eggwhite phantom and the agarose phantom means that the ESA of cooked eggwhite is larger than agar. This may be explained the macromolecular structure of the substances. As discussed in the introduction section, the ESA parameter of ionic samples reflects the density and dynamic mobility of charged molecules according to Debye's theory (eq. (1)). In cooked (de-natured) eggwhite the proteins are extensively cross-linked. Therefore the effective mass of a protein complex can be very large, resulting in low dynamic mobility. In comparison, the agar gel contains only 2% carbohydrates, which is less than the macromolecular content in eggwhite, and the carbohydrates may be less extensively cross-linked judging from the lower rigidity of agar. Thus the macromolecular contribution to the ESA parameter in agar is lower than the eggwhite. This explanation is speculative, since there is a lack of conclusive theories for the

ESA parameter. Further experiments and theoretical modeling are needed to clearly understand the information in this parameter.

Conclusion and Discussion

The experiments described above are attempts to form images of macromolecular gels using the electroacoustic effect. They showed that combining the principle of ultrasonography with this effect, one could form images of samples that are relatively uniform in their acoustic properties, but are heterogeneous in their electroacoustic or dielectric properties. The instrumentation and the scanning method used were simple, and the resulting images had significantly lower lateral resolution than standard ultrasonography. Nonetheless they demonstrate the possibility of the method.

Definite interpretation of the image contrast is difficult due to the complexity of the electroacoustic effect itself. As shown in eq. (6), both the electrical parameters and the ESA parameter are involved. The ESA parameter contains contributions from ions as well as polarization of suspended neutral particles (colloids). Currently there is a lack of theoretical understanding of the ionic contribution, and the colloidal contribution can be estimated only for simple suspensions. To better understand the information contained in the image, one may have to rely on empirical correlation between the amplitude and phase of the signal and the various relevant factors such as macromolecular content and ion concentration. It is possible that in biological samples such as tissue, more than one mechanism contribute to the electroacoustic signal, making it difficult to relate the image to specific biological or physiological parameters. This limits the specificity of the technique.

On the instrumentation of the method, several issues are significant in improving the image quality. The first is the adaptation of array transducers and associated data-processing techniques. This will improve the image resolution, and more importantly it will allow simultaneous data collection from all elements of the array, so that in principle each image requires only one rf excitation. The benefit of using less electrical pulses is many fold. It puts less duty cycle requirement on the pulse generators, and deposits less heat in the sample. For experiments on biological subjects it reduces the likelihood of nerve stimulation¹³. Another significant issue is that this imaging method employs strong electrical pulses, thus the ultrasound transducer needs to be effectively shielded against electromagnetic cross-talk. A detailed discussion of transducer shielding methods in this context can be found in ref. 14.

The results with eggwhite are encouraging since certain tissues such as skeletal muscle have similar protein concentrations and are highly cross-linked, suggesting that they may also show large electroacoustic effects. This imaging method is non-invasive, and may provide new information that is otherwise difficult to obtain with existing medical and industrial imaging techniques.

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Fig.1. A cross-sectional diagram of the device used to demonstrate ultrasonic electroacoustic imaging.

Fig.2.

A photograph of the sample-electrode assembly. The aluminum foil is opened to the side to show the agar gel in the sample container.

Fig.3.

Traces of the ultrasonic electroacoustic signal from the cooked eggwhite phantom, collected with the setup shown in Fig.1. Fig.3a is the signal with the aluminum foil as the ground electrode. The peaks are identified by their times of flight as electroacoustic signal and cross-talk echoes. Specifically, peaks (a), (b) and (c) were electroacoustic signal from the aluminum sheet, the saline-eggwhite interface, and the copper sheet at the bottom of the box, respectively. Peak (d) was a reflection of peak (b) from the bottom of the box. Peak (e) was a cross-talk echo from the bottom of the box. Fig.3b is the signal with the copper sheet as the ground electrode, note that the polarity of the saline-eggwhite peak is inverted from Fig. 3a. The assignments of peaks (a) to (d) were the same as Fig.3a. Peaks (e) and (f) are cross-talk echoes of the aluminum sheet and the bottom of the box, respectively.

Fig.4.

A cross-sectional image of the eggwhite phantom. The electrodes and the boundary between the saline and the eggwhite compartment can be seen. The bright line immediately below the copper sheet is the its reflection within the bottom layer of the sample container. Beyond that, traces of cross-talk echoes and multiple reflections of the electroacoustic signal can be seen. The traces labeled (a) were reflections of the electroacoustic signal of the saline-eggwhite interfaces, reflected by the two surfaces of the bottom of the box. Trace (b) is the cross-talk echo from the saline-eggwhite interface. The two adjacent traces labeled (c) are the reflections of the electroacoustic signal from the aluminum sheet, reflected by the two surfaces of the bottom of the box. The two traces labeled (d) are cross-talk echoes of the two surfaces of the bottom layer of the box.

Fig.5.

A cross-sectional image of the agar phantom. The gray scale range is half of Fig.4. Similar to Fig.4, the various interfaces can be seen. However the signal level at the saline-agar interface is approximately one-third of the saline-eggwhite interface. The traces labeled (a) were reflections of the electroacoustic signal of the saline-agarose interface from the bottom of the box. The group of traces labeled (b) were from the electroacoustic signal of the copper sheet, after multiple reflections between the two surfaces of the bottom layer and the saline-agarose interface. The two traces labeled (c) were reflections of the electroacoustic signal of the aluminum sheet, reflected by the surfaces of the bottom layer. The two traces labeled (d) were cross-talk echoes of the surfaces of the bottom layer.