How electric and ultrasonic fields assist membrane filtration

HOW ELECTRIC AND ULTRASONIC FIELDS ASSIST MEMBRANE FILTRATION

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ABSTRACT

In recent years 'assisted filtration' techniques have been receiving much attention. This paper describes how electric and ultrasound fields can be used to reduce fouling in microfiltration. Experimental data obtained over a range of operating conditions are presented that show both the effects of the individual fields and the synergistic effect of the combined fields. The parameters which control separation rate are identified and their individual contributions discussed.

INTRODUCTION

The rapid growth of high technology industries since the mid 1960s has stimulated many fields of research in filtration and separation. Although the work undertaken has often been varied and innovative the developments within separation science have largely centred on relatively few areas. Initially, research interest focussed on the modernisation of conventional separation equipment such as pressure vessel filters, filter presses and vacuum drum filters. Over the years improvements in mechanisation and instrumentation have seen the efficiency of such filtration apparatus raised to a level where some of the latest designs are now fully automated with a guaranteed discharge[1]. However, the demands of the biotechnology, pharmaceutical and electronics industries for ultra-pure solid and liquid products spawned another major field of research, namely membrane science.

The application of membrane technology to filtration aimed to provide a method for processing fine particle systems with little or no addition of contaminants (e.g. flocculants). The emergence of the reverse osmosis, ultrafiltration and microfiltration techniques has to some extent allowed colloidal dispersions with solutes in the range 0.1 nm – 10 μm to be separated efficiently[2]. While these membrane processes have frequently shown great promise their wider use at a large scale is often prevented by the problems of fouling[3,4]. The formation of a fouling layer at the septum during membrane separation can cause a rapid decline in permeate flux and result in low separation rates.

Although fouling may sometimes be reduced by mechanical means[5,6] there is currently much interest in the prevention of membrane fouling by what might be termed 'assisted filtration'. Here, electrical, sonic or other forces are utilised to modify the performance of an otherwise more conventional membrane filtration[7-12]. The potential of assisted filtration has frequently been demonstrated by laboratory scale equipment and presently these novel separation techniques must be regarded to be under exploited.

This paper considers how electric and sonic fields can influence the microfiltration of colloidal and near colloidal sized suspensions. The experimental data presented show that the imposed fields can both modify and improve filter performance over a range of operating conditions. Several parameters are identified which alter the rate a separation and their effects are discussed with respect to existing theories.

THE COLLOIDAL STATE

Most particulate substances acquire a surface electric charge when placed in contact with a polar (e.g. aqueous) medium[13]. Charging mechanisms can include ionisation, selective ion adsorption

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and unequal ion dissolution. The surface charge influences the distribution of nearby ions in solution and this together with random thermal motion results in the formation of an electrical double laver within the immediate vicinity of the particle (Figure 1). The double laver comprises a diffuse layer and a Stern laver: the latter extending approximately one hydrated ion radii from the particle surface. When the particle moves relative to the liquid, the innermost layer(s) of ions, including those within the Stern plane, move with it. The electrical potential which exists at the plane of shear is termed the zeta potential and its magnitude is dependent on factors such as surface chemistry, temperature, ion charge and solution pH.

A characteristic feature of colloidal dispersions is the large surface area to volume ratio exhibited by the particles involved. Hence, the presence of a surface charge on fine particulates in suspension can render their separation difficult and frequently requires the application of high pressures during filtration to overcome the dispersive forces generated. Such problems can become extreme with, for example, gelled materials in which the liquid phase becomes immobilised within a network of particulates[14]. The action of interparticle forces is generally compounded as particle size decreases and may also be affected by parameters such as particle shape.

THE NATURE OF ELECTROACOUSTIC SEPARATION

The electroacoustic filtration technique involves the combination of electric and ultrasound fields together with a more conventional pressure or vacuum filter. The imposed fields alter the rate of particle deposition at the filtering surface and change permeate flux levels.

The use of electric fields in filtration has shown a growing interest in the past 20 years and research workers have developed a reasonable understanding of the basic principles involvcd[15-17]. The technique relies on the application of a suitably polarised potential gradient across a filter medium to promote a reduction in the accumulation of particulates at the separating surface. The mode of filtration can be either dead-end or crossflow with the process stream usually being a dilute aqueous suspension of low electrical conductivity. It is a prerequisite that the suspension exhibits an average particle size less than 6 μm and a zeta potential greater than 20 mV in order to achieve worthwhile improvements[18]. The mechanisms involved in electrofiltration include filtration, electrophoresis, electroosmosis and several other secondary electrokinetic phenomena (see Figure 2).

Whilst the basic principles of electrofiltration are relatively well understood those of acoustic filtration/dewatering are rather less well documented. The acoustic enhancement of filtration involves the passing of sonic or ultrasonic (frequency greater than 16 kHz) sound waves through a suspension in the form of mechanical vibratory energy. It is suspected that the internal and elastic forces created by the acoustic waves cause changes to the interfacial phenomena of the solid phase[19]. This can enable an increased rate of permeate removal during filtration. The mechanisms of operation seem likely to combine mechanical, chemical and thermal effects and may promote modifications to the physical properties of a suspension at both the microscopic and macroscopic levels. Until recently the almost complete lack of understanding of the interaction between acoustics and solid-liquid mixtures had hampered previous research work to the extent that the literature which exists is largely confined to peripheral applications[20-23]. However, some recent publications have reported that acoustics can be used to assist the dewatering of fine particle suspensions[9,19]. Moreover, a synergistic effect was noted when both electric and acoustic fields were applied simultaneously and this has led to a renewed interest in the use of acoustics in filtration and separation.

EXPERIMENTAL APPARATUS

The experimental equipment used in the filtration tests is shown schematically in Figure 3. Suspension, made up to a known solids concentration and pH in twice distilled water, was pumped into a leaf filter. Here, constant pressure dead-end filtration occurred. The stainless steel (s/s) filter cell consisted of a conventional leaf filter modified by the addition of electrodes and ultrasound transducers. Two planar s/s electrodes at a separation of 4.7 cm sandwiched a 0.2 μm rated semi-permeable membrane which was approximately 17 cm in diameter. The ultrasonic horns were mounted on the upstream side of the membrane such that the sound waves transmitted through the slurry impinged on to the surface of any deposit formed at the septum. A regulated power supply with a maximum output of 400 DC at 10 A produced the constant potential, essentially uniform, electric field across the region of the filtering surface. The generator used to provide the acoustic field delivered up to 600 W of power to the ultrasonic transducer. Filtrations were carried out for between 20 and 60 mins. duration with the cumulative filtrate volume monitored throughout an experiment.

Each suspension was characterised by determining particle shape, particle size distribution and the zeta potential at various pH levels (Figures 4-6). The measurements were achieved using a scanning electron microscope and Malvern Instruments[24] light scattering devices with the pH of the suspensions altered by the addition of HCl or NaOH. When necessary, the suspensions were ground in a ball mill to bring the measured particle size to the required colloidal dimensions[25].

EXPERIMENTAL RESULTS

The following experimental results show the trends obtained by varying the filtration conditions in a systematic manner.

Figure 7 illustrates how the application of ain ultrasonic field can modify the filtration of an aqueous anatase suspension. The values on the abscissa are given by:

$$\text{\% gain volume permeate} = 100\left(\frac{V_u - V_0}{V_0}\right) \tag{1}$$

where V_0 = cumulative permeate volume with no ultrasound and V_u = cumulative permeate volume with ultrasound.

For the experimental conditions quoted the permeate rate (and hence cumulative filtrate volume) was found to be affected by both the suspension pH and the frequency of the applied sound waves. Whilst appreciable improvements were attained at certain pH levels with the acoustics, this could not be maintained over the whole pH range tested. The effect of the ultrasonics appeared to be reduced when the filter was operated with suspensions at pH = 5.3 and pH = 9. It is interesting to note that these pH values correspond approximately to the measured values for the zero and (absolute) maximum zeta potential of the dispersed phase (see Figure 6). A similar dependence on pH was observed for the filtration of a china clay slurry in an acoustic field, however, in this case the use of ultrasound had a predominantly detrimental effect on permeate flux (Figure 8). The sound waves caused membrane fouling to occur at a faster rate and Table 1 shows that the flux reduction was often accompanied by a lowering of the filter cake porosity. The result suggests that the influence of ultrasound in filtration may be dependent not only on the surface properties of the particulates in suspension but also particle shape and orientation.

The typical effects of using an electric field to assist filtration are shown in Figure 9. When no electric field was applied and the filtration initiated, the permute flux was found to fall sharply with rapid fouling of the membrane occurring. As the filtration proceeded, the flux level declined progressively. When the electric field was applied the rate at which the membrane fouled was reduced. Moreover, if the electric field strength was of sufficient magnitude a constant or even

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increasing flux could be achieved after an initial fouling period. The rising flux was presumably due to the action of the potential gradient in removing particulates from the surface of the cake which formed during the first stages of filtration[12,14,18]. The flux enhancement with the electric field was often accompanied by an increase in filter cake porosity (Table 2). Similar results were obtained for the electrofiltration of a china clay slurry although the effects of the electric field were reduced due to the lower zeta potential and larger particle size.

Some experiments were also performed in determine the effects of applying electric and ultrasound fields simultaneously during filtration. Figure 10 and Table 3 show that for an anatase suspension the use of combined fields produced an additional increase in cumulative filtrate volume over that attained with an electric field only. Furthermore, a synergistic effect was noticed as the electric field strength was increased. Whilst the process has not yet been fully investigated it seems that the two fields couple together to change the physical characteristics of the suspension being filtered. These changes may be favourable, as with anatase, but Figure 11 illustrates how ultrasonics can have an adverse effect when used in conjunction with an electric field. Flux levels here were below those achieved with only the electric field and the flux differential was more accentuated at higher potential gradients.

DISCUSSION

The experimental data presented in this paper clearly demonstrate how both electric and acoustic fields can modify the rate at which membranes foul during filtration. However, the analysis of the results is made more difficult by the general lack of understanding of the interaction between solid-liquid mixtures and ultrasound.

The transmission of a two dimensional ultrasonic wave through a non-dissipative, inviscid liquid is described by the equation:

$$\frac{\partial^2 \varepsilon}{\partial t^2} = \left(\frac{K}{\rho}\right)\frac{\partial^2 \varepsilon}{\partial r^2} \qquad (2)$$

Under these conditions, changes in pressure and density take place reversibly and the sound wave propagates through the fluid with undiminished intensity[26,27]. When viscosity is present, the sound wave is attenuated as it passes through the liquid: the depth of penetration given by:

$$U_x = U_0 \exp(-\alpha x) \qquad (3)$$

where α = ultrasonic intensity attenuation coefficient. Should there also be particulate matter present in the fluid the sound wave is likely to be further attenuated to an extent dependent on the acoustic properties of the suspended solid. The complexity of evaluating (or even estimating) the terms ρ, K and α for suspensions means that only a qualitative analysis of the effects of ultrasound can realistically be made.

The passage of sound waves through a suspension is known to generate high inertial forces at the solid liquid interfaces[19]. If these are of sufficient magnitude then particle motion relative to the fluid may result. It has seen postulated that during acoustic separation such inertial forces reduce the effective viscosity of the liquid phase in a suspension to allow for improved dewatering[9]. Whilst this mechanism may partially explain the influence of ultrasound in filtration, the results obtained from the filter tests indicate that several other factors contribute to the process. For the slurries tested, parameters such as particle shape, pH and sound frequency were found to affect filtration performance. By changing the surface electrical properties of the dispersed phase using small amounts of acid or alkali, ultrasound would either enhance, reduce or have no visible effect on permeate flux. The acoustics appeared to have a minimum effect when pH levels corresponded to the points of zero and maximum/high zeta potential. The explanation of these phenomena is difficult. Certainly ultrasound can cause particulate dispersion[28] and agglomeration[29] and one could reasonably expect either to occur close to the iso-electric point (zero surface charge). Equally, at high zeta potential particles are likely to be well dispersed and the influence of further dispersion/agglomeration by ultrasonic agitation seems improbable.

While the effectiveness of ultrasound in the filtration of colloids is closely linked with the surface chemistry of the process stream, the overall picture is somewhat complicated by an apparent dependence on particle shape. Of the two suspensions tested to date only anatase (ellipsoidal particle shape) proved to respond favourably to ultrasonic enhancement. With china clay (plate-like particle shape) any potential improvement by acoustics appeared to be offset by reductions in filter cake porosity. The extent of this behaviour was dependent on pH and sound frequency and may be a consequence of particle orientation (Figure 12). When china clay is filtered normally with no ultrasound an open cake of relatively high porosity results. The unequal surface charging exhibited by the edges and faces of the plate-like particles[30] helps the formation of a network of particulates analogous in structure to a 'house of cards'. If ultrasound is applied it seems that the combination of the sound waves and fluid flow through the cake cause sufficient disturbance for the cake structure to partly collapse and reduce porosity. Should the sound intensity be above the threshold required to initiate cavitation such a mechanism may be aided by the rapid formation and destruction of gaseous bubbles.

Clearly, the effects of combining ultrasound and filtration are numerous and inherently difficult to quantify. It appears that further investigation is required before any firm conclusions can be drawn regarding the mechanisms of interaction. When a better knowledge is gained it should be possible to determine how ultrasonics will affect a given suspension by simply evaluating parameters such as particle shape, particle size and zeta potential.

The use of electric fields in filtration has for several years now been recognised as a method for reducing membrane fouling. As the mechanisms of operation are relatively well understood they are not discussed in great detail here[11,15,18,31]. However, a few aspects of electrofiltration are thought pertinent and worthy of mention. Possibly of most relevant interest is how electric fields affect filter cake formation and prevent flux decline. A potential gradient can cause a filter cake of reduced mass and higher porosity to form: this allowing an increased permeation rate. Moreover, the data presented in Figure 9 suggests that an electric field can not only help prevent particulate deposition during filtration but also assist in the removal of previous deposits from a filter surface. The latter process may prove useful for cleaning 'dirty' membranes where there is a need to remove deposits without applying high backflush pressures or damaging chemicals. The results also implied, and a previous study supports the theory[18], that with a sufficiently high electric field strength little or no membrane fouling by particulate matter occurs and separation proceeds at close to a maximum rate. While this situation may not always be achieved in practice, filtration at high flux levels has been demonstrated.

When an electric field was applied simultaneously with ultrasound a synergistic effect was recorded. Although there is uncertainty about the exact coupling mechanisms, several hypothesis have been proposed. One theory supposes that the passage of the sound waves through a suspension provides for better electrical continuity to induce improved electroosmotic flow during filtration. Another postulates that the ultrasound initiates changes in the suspension properties at the microscopic level to promote more favourable conditions for electrofiltration. Such ideas at this stage seem largely conjectural. Furthermore, the experimental data presented shows that combined electric and ultrasound fields do not always improve filtration performance over that attained with only an electric field. The already complex analysis is required with ultrasonics and filtration is compounded by the addition of the electric field and little benefit would be gained by ‘guessing' the mechanisms of interaction. Whether the synergistic effect can be wholly attributed to the influence of one of the above mechanisms is debatable. It seems more likely that several individual factors contribute to the overall effects observed.

CONCLUSIONS

The fouling of membranes in filtration has been a recursive theme for many years now. Whilst technologists strive to improve the situation with the introduction of new membrane materials it seems likely that the problem will remain for many years to come. The techniques described in this paper may offer an alternative way to alleviate fouling. Both electric and ultrasound fields can reduce membrane fouling caused by the deposition of particulate colloidal material. Increased filter cake porosities and permeation fluxes can readily be achieved when electric fields alone are used to assist filtration, however, the use of ultrasound requires more care. Unless factors such as suspension pH and particle shape are favourable, the application of acoustics can have a detrimental effect on separation performance. Moreover, the synergistic effects observed at combined field conditions illustrate how ultrasonics may either raise or lower the level of fouling in electrofiltration. These results suggest that the use of ultrasound may need to be evaluated on a case-by-case basis. The complexity of the interactions between ultrasound and suspensions almost precludes any quantitative analysis of both acoustic and electroacoustic filtration to the extent that few mathematical theories or models appear to exist. It seems that until a better theoretical knowledge is attained, only extensive experimental investigation can provide an insight into the combined effects of ultrasound, electric fields and filtration.

NOMENCLATURE

K	Elastic modulus (kg m^{-1} s^{-2})
t	Time (s)
x	Distance (m)
U_0	Ultrasonic source intensity (W m^{-2})
U_x	Ultrasonic intensity at a distance x from source (W m^{-2})
V_0	Cumulative permeate volume with no ultrasound (m^3)
V_u	Cumulative permeate volume with ultrasound (m^3)

Greek symbols

α	Ultrasonic intensity attenuation coefficient (m^{-1})
ε	Wave displacement (m)
ρ	Fluid density (kg m^{-3})

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FIGURES AND TABLES

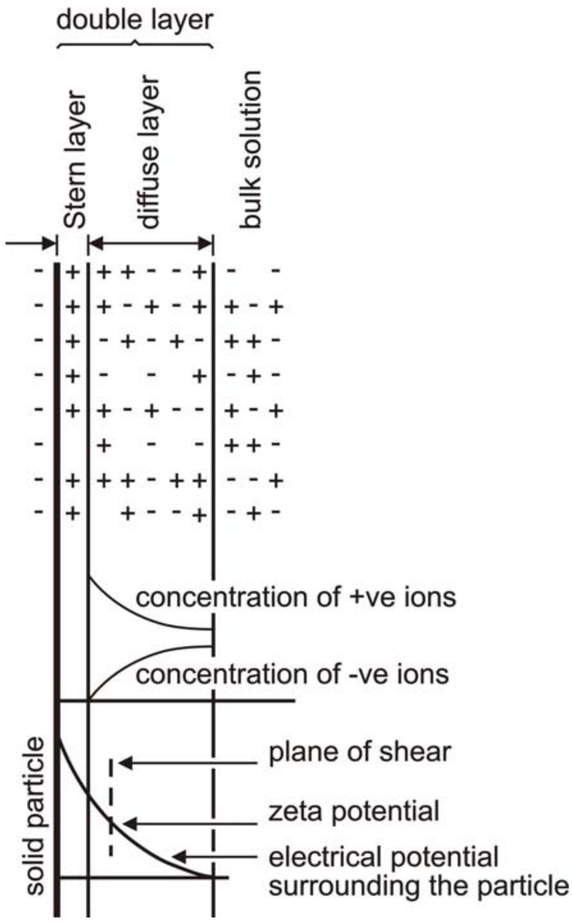

Figure 1: Schematic diagram of the electric double layer.

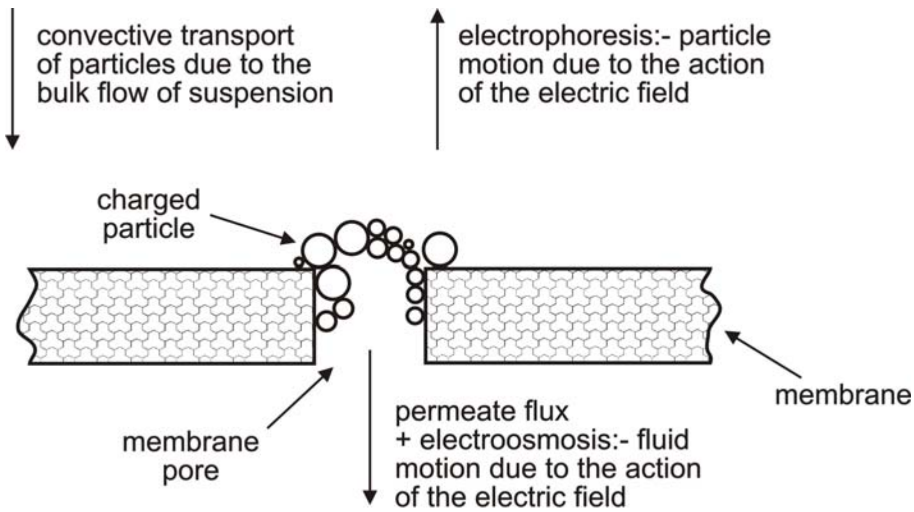

Figure 2: Schematic diagram showing some of the mechanisms involved in electrofiltration.

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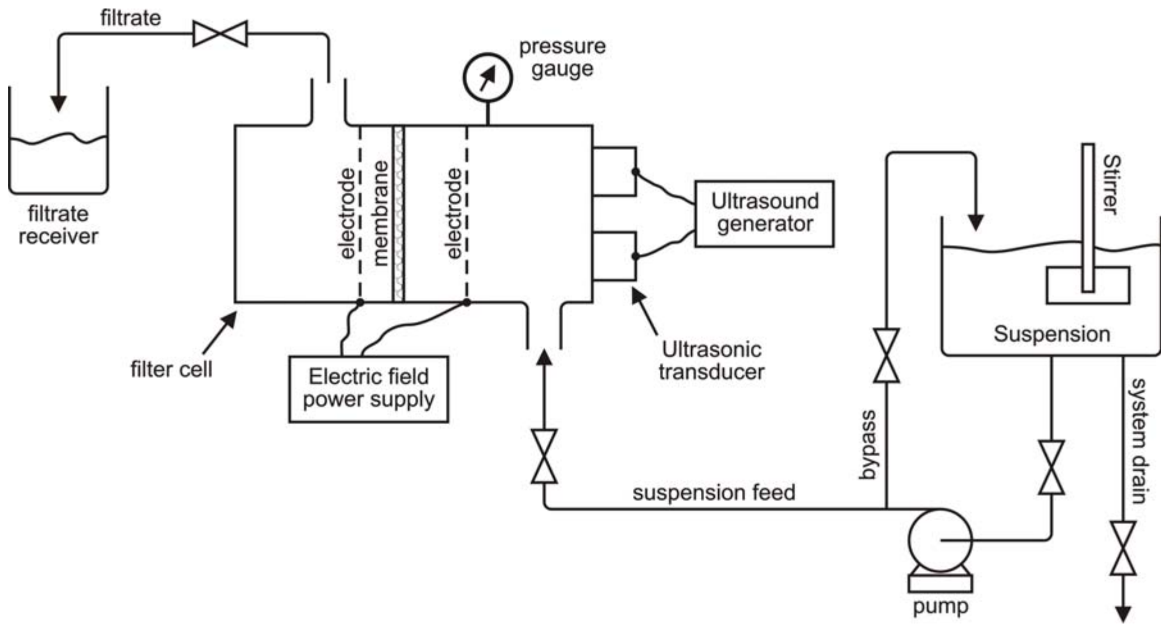

Figure 3: Schematic diagram of the electro-acoustic filter and flow circuit.

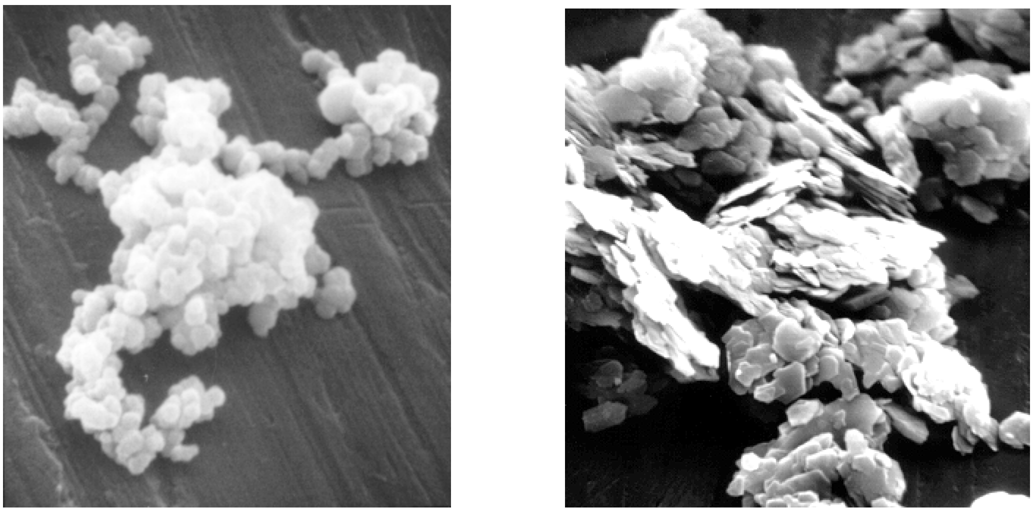

Figure 4: Scanning electron micrographs of, left, anatase (TiO_2) and, right, china clay.

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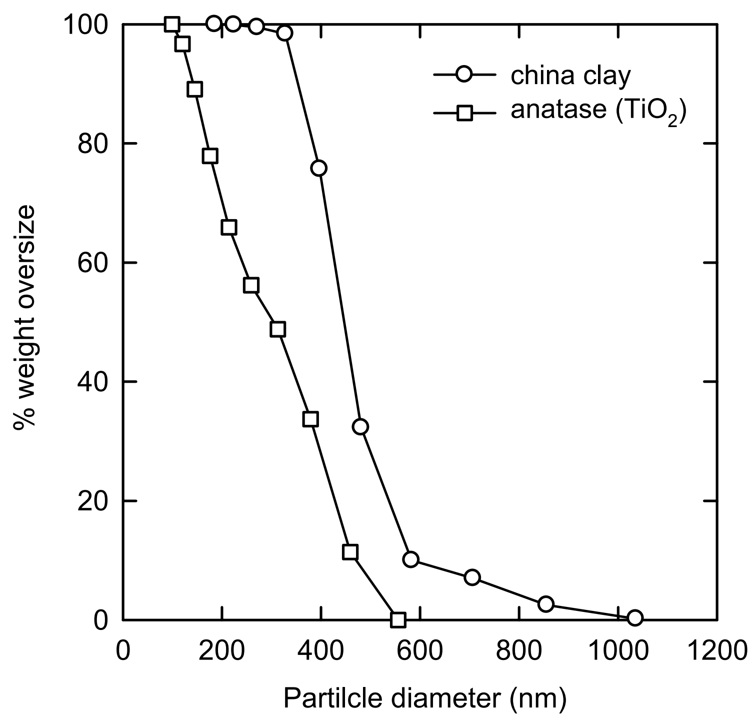

Figure 5: Particle size distributions for anatase and china clay.

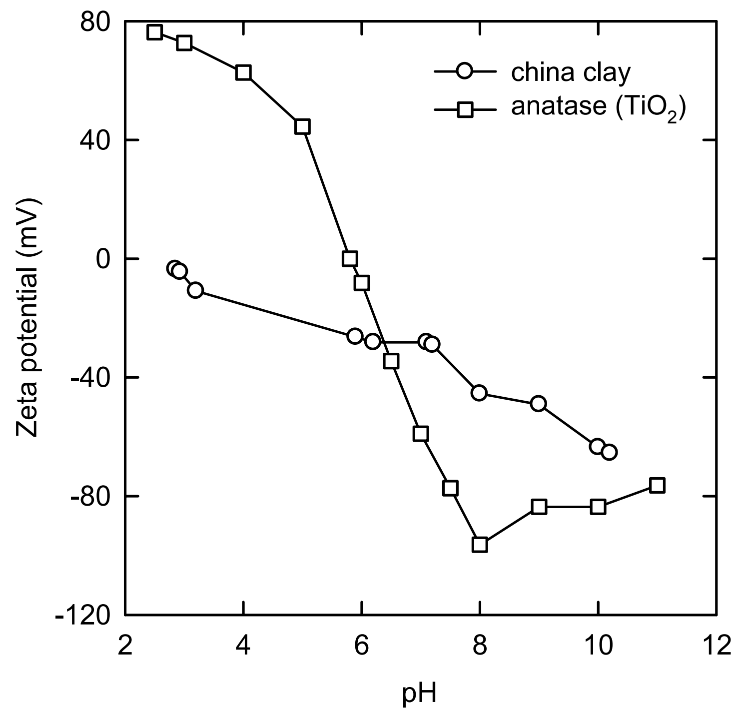

Figure 6: pH – zeta potential curves for anatase and china clay.

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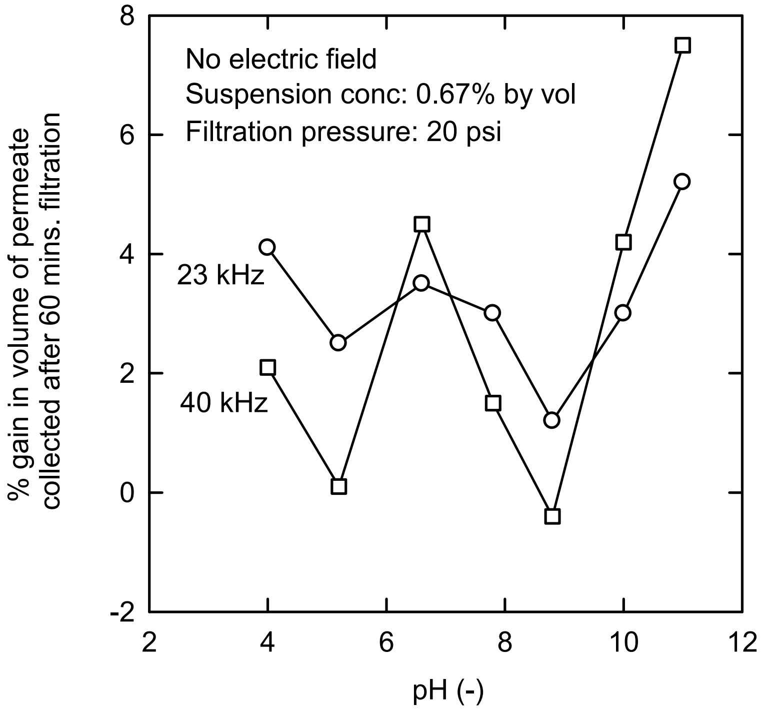

Figure 7: Effect of ultrasound and pH on the filtration of an anatase suspension.

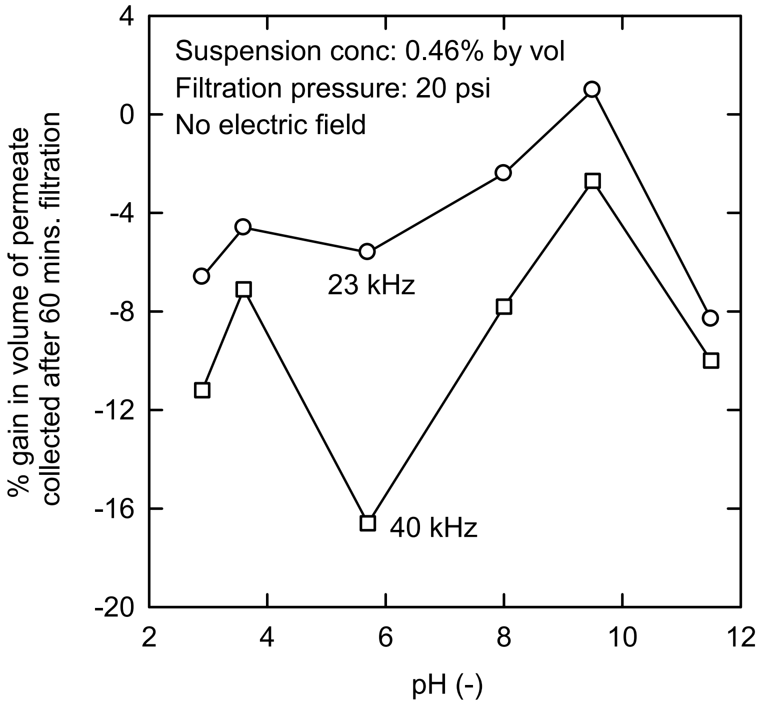

Figure 8: Effect of ultrasound and pH on the filtration of a china clay suspension.

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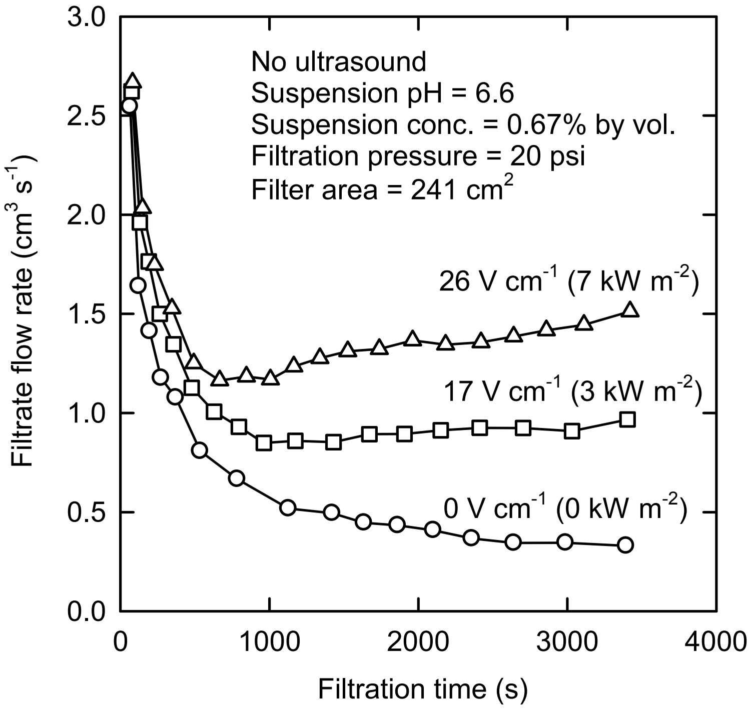

Figure 9: Effect of an electric field on the filtration of an anatase suspension.

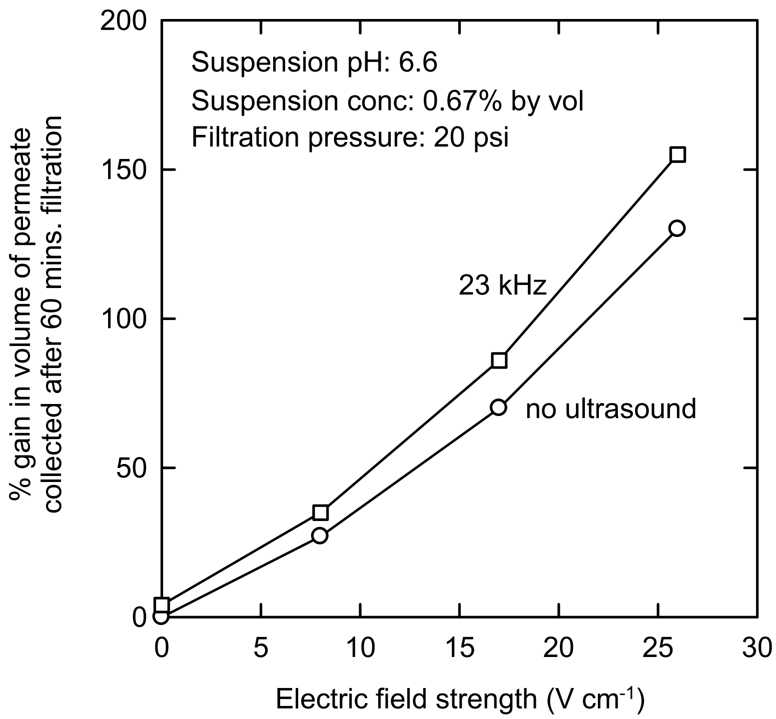

Figure 10: Effect of combined electric and ultrasound fields on the filtration of an anatase suspension.

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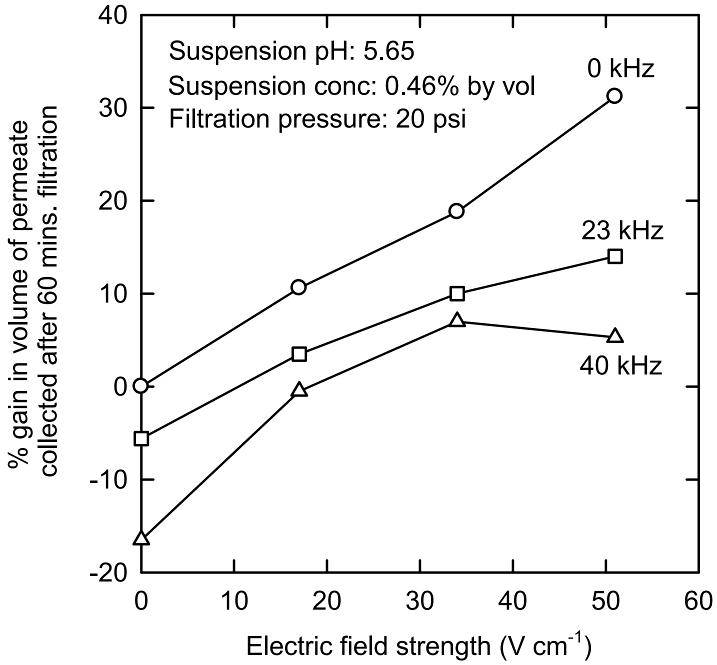

Figure 11: Effect of combined electric and ultrasound fields on the filtration of a china clay suspension.

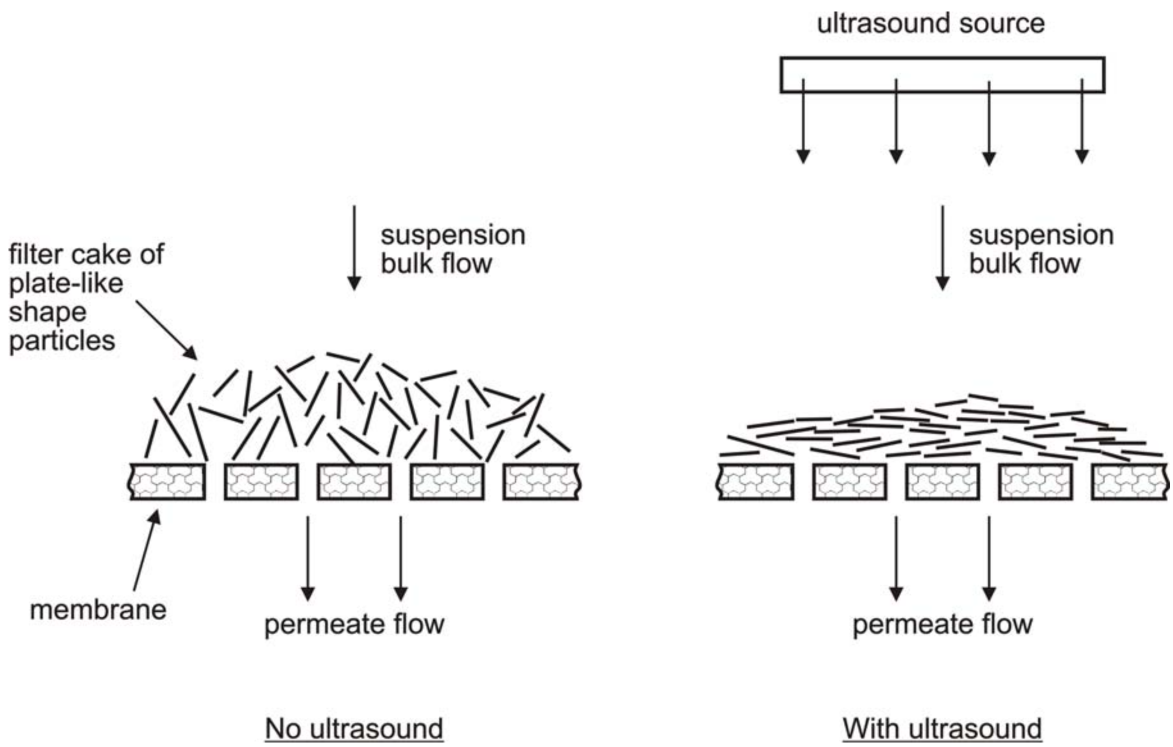

Figure 12: Schematic diagram of the possible effect of ultrasonics on filter cake structure.

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Suspension pH	No ultrasound applied	Ultrasound freq. = 23 kHz	Ultrasound freq. = 40 kHz
2.90	0.79	0.79	0.78
3.60	0.79	0.79	0.79
5.65	0.80	0.78	0.78
8.0	0.79	0.78	0.77
9.45	0.76	0.75	0.72
11.6	0.73	0.72	0.73

Table 1: Some filter cake porosities for china clay.

Electric field strength (V cm^{-1})	Filter cake porosity
0	0.68
8.5	0.69
17	0.71
26	0.69

Table 2: Some filter cake porosities for anatase.

Electric field strength (V cm^{-1})	% gain volume permeate		
	Electric field only	Electric field + 23 kHz acoustics	Electric field + 40 kHz acoustics
0	0	3.74	4.62
8.5	26.8	34.5	38.9
17	70.5	85.1	85.5
26	131.2	155.5	145.3

Table 3: Some data showing the effect of combined electric and acoustic fields for anatase.

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