Crossflow Electrofiltration of Petroleum Oils

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Abstract

Fine particle removal from nonpolar liquids is increasingly important in industry. Electrical separation methods have shown promise; however, relatively few studies have been published on the subject. This paper discusses the performance characteristics of one such process, crossflow electrofiltration, and the environmental factors which govern it. Experiments were conducted using model systems and oil samples from different types of service. Contaminant removal and pressure drop were monitored while the electric field strength and flow conditions were varied. Removal and service life were greatly enhanced by the application of an electric field for model systems; however, less improvement was observed for field samples. This difference was attributed to viscosity and zeta potential effects. Typically, field samples had moderately high viscosities and low zeta potentials; hence, contaminant electrophoretic mobility was lower than that required for effective separation. The magnitude of the zeta potential appeared to be influenced by a sample's emulsified water concentration, additive package, and contaminant nature. The mechanisms underlying these effects and their implication for electrofilter performance are discussed. Though this study focused on crossflow electrofiltration, many of the findings apply to other nonaqueous electrical separation processes as well.

INTRODUCTION

The removal of fine particles (<10 μm) from hydrocarbon liquids represents a major technological problem of increasing importance to industry. Coal-derived liquids may contain up to 10% (by weight) particulate matter (1). The removal of catalyst fines and other solids from residual fuel oil is of increasing concern to petroleum refiners and engine operators. Hydraulic oil filtration to a nominal 10 μm particle size is no

longer adequate as clearances between moving parts become smaller in high-pressure systems. Similar separation requirements are being faced in certain food and chemical processing applications as well. Cross-flow electrofiltration is one method of achieving fine particle removal which has shown considerable promise in laboratory evaluations (2-5, 16). This paper discusses the use of this process in petroleum oils. However, it should be noted many of the conclusions are applicable to other electrophoretic separation technologies as well.

Crossflow electrofiltration is a hybrid separation process which combines the features of crossflow filtration and electrophoretic separation devices. The crossflow electrofiltration concept is schematically represented in Fig. 1 for positively charged contaminant particles. Like a conventional crossflow filter, the influent flow of contaminated fluid is directed parallel to the filter media surface, in this case a porous metal electrode. In crossflow electrofiltration a dc electrical field is applied normal to this surface. If the field is of sufficient strength and proper polarity, charged contaminant particles will migrate away from the media surface by electrophoresis, giving rise to a clear boundary layer. Particle-free fluid can then be withdrawn through the media.

In theory, the behavior of a crossflow electrofilter toward charged contaminants should approximate that of the mythical ideal filter. Nearly complete separation should be obtained for particles of all sizes without a corresponding increase in pressure drop. From the standpoint of electrophoretic separation, it is immaterial whether the contaminant is an ion or a grain of sand, as long as it has sufficient charge. Since the contaminant does not come in direct contact with the media, no increase in pressure drop should be observed; hence, crossflow electrofilters should be characterized by extremely long service intervals. For reasons discussed in this paper, the crossflow electrofilter's true potential is rarely realized in practice.

A number of investigators have attempted to model the crossflow electrofiltration process. Henry et al. (1) described the filtrate flux, J, in terms of a series of resistances due to the media, contaminant cake, and liquid film. This equation takes the form

$$J = \Delta P(R_f \Delta P + R_m + R_c)^{-1} \qquad (1)$$

where ΔP is the pressure drop, R_f is the liquid film resistance, R_m is the media resistance, and R_c is the cake resistance. At sufficiently high electric field strengths a clear boundary layer should form, preventing cake formation, and R_c can then be ignored.

Lee et al. (3) and Liu et al. (4) have used the clear boundary layer

Filtrate

Porous electrode

Clear boundary layer

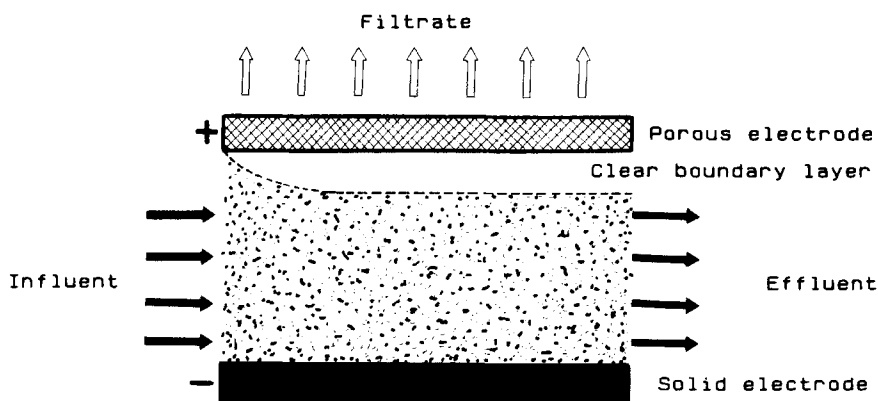Influent

Effluent

Solid electrode

FIG. 1. Crossflow electrofiltration concept.

concept to model various crossflow electrofilter configurations. Under optimum conditions the tendency of the fluid to transport particulates through the porous electrode is opposed by electrophoretic forces. The term "critical field strength" is defined as the field strength such that the electrophoretic and hydrodynamic forces are equal in magnitude and opposite in sign. At higher field strengths a clear, particle-free boundary layer will exist adjacent to the porous electrode. At this critical field strength (E_{cr}), filtrate flux is given by

$$J = ME_{cr} \tag{2}$$

where M is the electrophoretic mobility of the particle. The thickness of the clear boundary layer is a function of field strength and particle zeta potential as well as the fluid velocity components tangential to the porous electrode surface. In petroleum oils, contaminant zeta potentials frequently cover a wide distribution. As a result of this broad distribution and the effects of diffusion, a completely particle-free boundary layer rarely develops in practice and cake formation occurs.

Only a few crossflow electrofiltration studies have been conducted in nonaqueous systems (3–5). As a result of differences between water and nonpolar liquids, there is reason to expect the two types of systems to respond very differently to crossflow electrofiltration. While aqueous systems typically have electrical conductivities in excess of 10^{-4} $\Omega^{-1}m^{-1}$, the conductivity of nonpolar fluids ranges from 10^{-15} to 10^{-7} $\Omega^{-1}m^{-1}$. This difference is reflected in the correspondingly high dielectric strength for

insulating oils (10,000–16,000 kV/m) compared to water, which acts as a conductor. For this reason, electrical fields approximately 2000 times higher may be used in oil than in water, yet the power usage may actually be much less.

Though higher fields can be used in nonpolar liquids, particle velocities are not correspondingly higher. The reason for this is apparent from the basic electrophoresis equation:

$$V = (2\zeta E\varepsilon\varepsilon_0)/(f\eta) \tag{3}$$

where V is electrophoretic velocity (m/s), ζ is the particle's zeta potential (V), E is applied field strength (V/m), ε is the fluid dielectric constant, ε_0 is the permittivity of free space $(C^2 \cdot Nt^{-1} \cdot m^{-2})$, η is the fluid viscosity $(kg \cdot m^{-1} \cdot s^{-1})$, and f is a constant whose value is a function of the electrical double-layer thickness and particle size. For aqueous systems f is usually 2, while for nonpolar liquids it approaches a value of 3. The dielectric constants for the nonpolar liquids examined in this study are ~40 times lower than that of water. Viscosities for petroleum products typically range from 2 to 1000 times that of water. For these reasons electrophoretic velocities in nonpolar liquids are often lower than in corresponding aqueous systems despite the application of higher field strengths.

In contrast to aqueous systems, electrophoretic velocity in nonpolar liquids is not necessarily directly proportional to field strength. At high fields, relaxation effects and field charging may produce velocities in excess of predictions based on Eq. (3). Stotz (7) attributed the dependence of electrophoretic mobility on field strength to the detachment of the diffuse double layer from a particle as its velocity increases. Vincett (8) and Lockhart and Snaith (9) observed temporary increases in mobility and attributed it to the injection of charge into the system by the electrodes. The polarization of particles in an electric field gradient and the resultant dielectrophoretic motion can also be significant (10). These phenomena and their significance in the crossflow electrofiltration of nonpolar liquids have been studied by Verdegan et al. (5). In that study, field-depedent phenomena, particularly relaxation effects, were found to be important for liquids with conductivities less than ~5 × 10^{-8} $\Omega^{-1}m^{-1}$. Many commercially important hydrocarbons have conductivities significantly below this level. Since field-dependent phenomena can increase mobility by a factor of 4 or more, they have potentially great practical utility.

The present study is concerned with the use of crossflow electrofiltration for nonpolar liquids. Past work was often conducted using simplified

model systems such as α-alumina and tetralin (*3, 4*). Though useful for studying the process itself, work conducted under more realistic conditions is required to answer a number of fundamental application questions. The present study addresses a number of these questions. Emphasis was placed on potential problems associated with the selection of electrode polarity and the presence of high solids or emulsified water conditions. An indication of field performance and the factors which affect removal was obtained from tests conducted with samples of various petroleum oils.

EXPERIMENTAL METHODS AND MATERIALS

A radially configured crossflow electrofilter with an inner porous electrode was used in this study. This configuration establishes the maximum field strength adjacent to the porous electrode. As a result, lower applied voltages can be used to create and maintain a clear boundary layer. The test electrofilter is schematically illustrated in Fig. 2(a). In this configuration the influent flow is directed axially through an

(a)   (b)
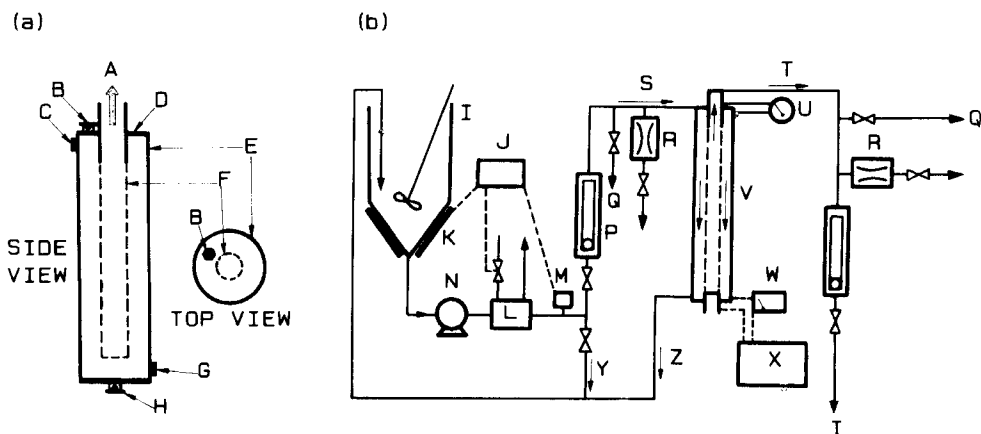
FIG. 2. Crossflow electrofilter test apparatus. (a) Crossflow electrofilter configuration. (b) Schematic of test stand. Key: A, filtrate; B, air bleed; C, influent port; D, insulating end cap; E, outer solid electrode; F, inner porous electrode; G, effluent port; H, drain; I, sump with mixer; J, temperature controller; K, heater; L, heat exchanger; M, thermocouple; N, gear pump; P, flowmeter; Q, sample port; R, particle sensor; S, influent; T, filtrate; U, differential pressure gauge; V, crossflow electrofilter; W, ammeter; X, power supply; Y, bypass; Z, effluent.

annulus formed by the porous and solid electrodes. Filtrate is withdrawn through the porous electrode while a constant voltage (direct current) is applied.

Tests were conducted using a variety of nonpolar liquids. Mil-H-5606 hydraulic oil containing 100 ppm of Shell ASA-3 antistatic additive was used as a model fluid to determine the performance characteristics of the crossflow electrofilter. Depending on the purpose of the test, AC fine test dust (ACFTD) and/or emulsified distilled water were used as contaminant in these tests. The model additive, fluid, and solid contaminant are the same ones used in the multipass method for evaluating hydraulic oil filters (*11*). All other tests were conducted using unmodified samples obtained from the field. The properties of these fluids are summarized in Table 1. Viscosity at the test temperature was determined using a Brookfield viscometer. Water concentration was determined by the Karl Fisher method and contaminant zeta potentials by microelectrophoresis. Electrical conductivity was measured according to ASTM D3114.72 (*12*). Pentane- and toluene-insoluble solids were measured using an adaptation of the gravimetric method described in ASTM F313.70 (*13*).

Experiments were conducted using the test stand illustrated in Fig. 2(b). Depending on its nature, tests were conducted in either a multipass or single pass mode. Multipass tests were used to study the performance characteristics of model systems. In these tests the effluent stream was returned to the sump while the fitlrate stream was collected in a separate receiving vessel. During long-term multipass tests, make-up slurry was continually injected into the influent sump to replace contaminant removed by the crossflow electrofilter. The single pass mode was used to study field samples. In these tests, both effluent and filtrate were collected in separate receiving vessels.

Throughout the testing, applied field strength and polarity were varied. In this paper the polarity of the applied field will be defined with respect to the porous electrode. Thus, a field strength of -1700 kV/m refers to a negatively charged porous electrode with a field strength of 1700 kV/m adjacent to its outer wall. Where feasible, crossflow electrofilter removal was determined by means of on-line particle counting. A Hiac model 320 particle counter with CMH-90 sensors was used. The counter was calibrated using spherical glass beads. When the particle concentration exceeded the saturation limits of the sensors, bottle samples were collected and diluted for later analysis. Throughout this discussion, removal will be expressed in terms of filtration ratios (β_2). For the purposes of this paper, β_2 is defined as the ratio of the number of influent to filtrate particles greater than 2 μm in diameter.

TABLE 1
Properties of Test Fluids

Fluid and description	η^a	Conductivity $(\Omega^{-1}m^{-1})$	Contaminants (mg/L)		
			Water	Pentane insoluble	Toluene insoluble
Hydraulic Oils					
A Mil-5606 (model fluid)	12.5	5.7×10^{-9}	110	—	—
B Industrial metal shears	29.7	2.3×10^{-10}	399	9.4	7.6
C Farm tractor transdraulic oil	44.0	4.45×10^{-8}	1,110	88.3	71.3
D New industrial hydraulic oil	32.8	7.2×10^{-10}	268	35.3	15.2
E New industrial hydraulic oil	13.6^b	6.5×10^{-10}	94	26.5	9.6
Lubricating Oils					
F New diesel engine oil	27.4^b	1.54×10^{-8}	94	556	518
G Compressor oil	36.1	1.50×10^{-8}	715	103.4	86.7
H Stream turbine oil	30.6	7.9×10^{-10}	30,000	9.6	5.1
Fuel Oil					
J #2 Fuel oil	2.8	2.4×10^{-12}	108	1.6	1.53

[a]All viscosities are reported in units of 10^{-3} kg $\cdot$ m$^{-1} \cdot$ s^{-1} (cP) at the test temperature (38°C unless otherwise noted).
[b]A temperature of 66°C was used for these fluids.

PERFORMANCE CHARACTERISTICS

Since it is unlikely that conditions will always be optimal for crossflow electrofiltration, tests using model systems were conducted to determine the performance characteristics under a variety of conditions. In particular, the effects of field strength and polarity, solids concentration, and emulsified water were examined.

In practice, the contaminant zeta potential is not always known and it may even change over time. Therefore, it is important to determine how removal varies as a function of field strength and polarity. Figure 3 shows results obtained for the ACFTD/Mil-5606 model system. Typical plots of β_2 versus field strength were observed to be U-shaped with a minimum at 0 kV/m applied field. The curves are asymmetrical with one polarity demonstrating greater removal than the other. The asymmetry suggests that different removal mechanisms are operating for the two arms of the curve. Microelectrophoresis measurements revealed that ACFTD has a zeta potential of +96 mV under the test conditions. Thus, the contaminant and porous electrode are like-charged for the higher performance arm of the curve. Under these conditions, electrophoretic separation is occurring as was previously discussed and particles are repelled by the porous electrode, resulting in particle-free filtrate. As the field strength increases, more particles attain electrophoretic velocities exceeding the
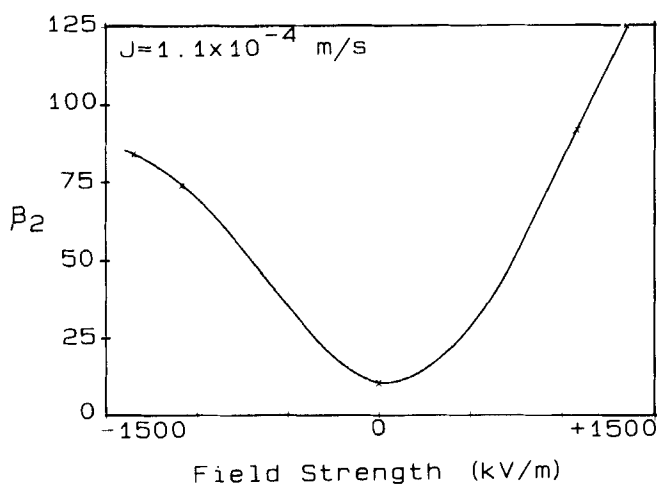
FIG. 3. Effect of electric field strength on separation for ACFTD/Mil-5606 system containing 100 ppm ASA-3.

filtrate flux, and separation improves. In contrast, charged particles are attracted toward the porous electrode for the lower performance arm of the curve. This results in increased capture and higher removal on the outer surface of the electrode than would otherwise be observed. Similar phenomenon have been proposed by Jaisinghani and Verdegan (14) in passive hydraulic oil filtration when the media and contaminant are oppositely charged.

Electrofilter service intervals and dust capacity are also affected by the polarity of the applied field. The service interval is the length of time the unit can operate before the electrode must be cleaned. Dust capacity, as the term is used in this paper, refers to the total mass of ACFTD (per unit area of electrode surface) the electrofilter must be exposed to in order to achieve a terminal pressure drop of 207 kPa (30 psi). Longer service intervals and higher capacity are observed when the porous electrode and contaminant are like-charged than when they are oppositely-charged. For the same fluid and flow conditions, the apparent capacity exceeded 69.2 g/m^2 when the contaminant was repelled by the porous electrode and was only 40.5 g/m^2 when it was attracted toward it. Since cake formation is encouraged when the electrode and particles are oppositely-charged but hindered when they are like-charged, this result is not surprising.

The effect of an applied electric field is particularly evident in the results shown in Figs. 4(a) and 4(b). The results shown were obtained for a filtrate flux of 2×10^{-4} m/s and ACFTD concentration of ~10 mg/L. In the absence of an electric field, β_2s of about 5 were observed and the pressure drop across the porous tube rose from 18 to 207 kPa (2.6 to 30 psi) within the space of 43 min. This corresponds to a dust capacity of 5.61 g/m^2. When a field of +17 kV/m was applied (contaminant and porous electrode same polarity), β_2 increased to ~28. No significant pressure drop increase was observed, even after exposure to twice the amount of ACFTD used in the 0 kV/m test.

In some applications the crossflow electrofilter may be exposed to high contaminant concentrations; therefore, it is useful to know how it will respond to these conditions. A long-term test at a concentration of 1500 mg ACFTD/L was conducted to answer this question and to obtain an indication of the relative service life of the crossflow electrofilter. In general, both β_2 and pressure drop were observed to rise with time, indicating that a contaminant cake was accumulating on the porous electrode; however, the rate of pressure drop increase was still much lower than when no field was applied. Even after 332 min the terminal pressure drop of 207 kPa was not achieved. This indicates that the apparent "capacity" of the crossflow electrofilter is in excess of 4500 g/m^2. Because it is regenerable and most of the contaminant builds up on the
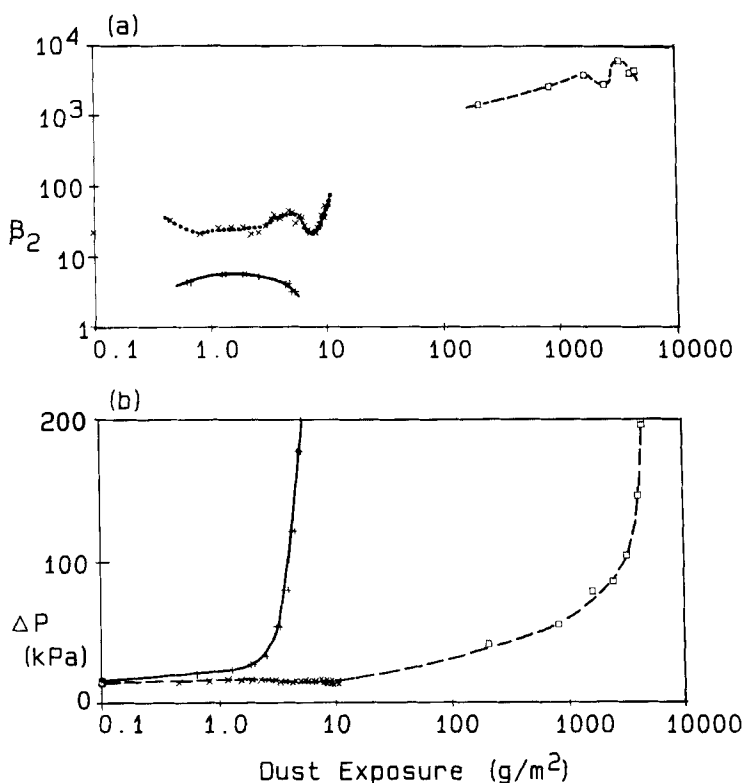
FIG. 4. Performance curves with and without applied electric field. (a) Filtration ratio vs dust exposure, $J = 2 \times 10^{-4}$ m/s. (+) $E = 0$ kV/m, 10 mg ACFTD/L. (×) $E = 17$ kV/m, 7 mg ACFTD/L. (□) $E = 17$ kV/m, 1500 mg ACFTD/L. (b) Pressure drop vs dust exposure.

solid electrode (not the porous electrode), the term "capacity" can be confusing when applied to crossflow electrofiltration. For these reasons, values for capacity should be regarded as indicative of the service interval, not the amount of contaminant held by the media. A comparison of capacities with and without applied field suggests that the service interval can be increased by a factor of 800 or more by proper application of an electric field.

In contrast to other tests, a thick (5×10^{-3} m) cake of ACFTD formed on the negative outer (nonporous) electrode during the high-concentration test. It was found that both electrodes could be readily cleaned by reversing the field and flow. Cleaning would be expected to be much more difficult if soft contaminants such as asphaltenes had been used.

Emulsified water is found at varying concentrations in many petro-
leum oils. To study its effect, tests with the model system containing
emulsified water were conducted. In tests containing ~2000 mg H_2O/L
(instead of ACFTD), 74% of the water was removed when the porous
electrode was negatively charged while no significant removal was
observed in the absence of an applied field. Unlike ACFTD, the water
droplets appeared to be negatively charged. The implications of this
finding are discussed later. During the emulsified water test, the pressure
drop increased very slowly (0.03 kPa/min) when the field was applied, but
the electrical current was 1 mA, three times greater than was observed in
the absence of emulsified water.

In practice, emulsified water is rarely the only contaminant present.
When ACFTD and emulsified water were both present in the model
fluid, crossflow electrofilter performance decreased dramatically. Fre-
quent shorting out of the power supply was observed due to the
accumulation of water. This suggests that a more powerful high voltage
source will be required and higher energy costs will be associated with
crossflow electrofilter applications requiring the removal of emulsified
water. In addition, a synergistic effect on the rate of pressure drop
increase was observed. This effect is shown in Fig. 5. At an ACFTD
concentration of 57 mg/L and water concentration of ~2000 mg/L, the
pressure drop rose at a rate of ~4 kPa/min (filtrate flux of 1.5×10^{-4} m/s)
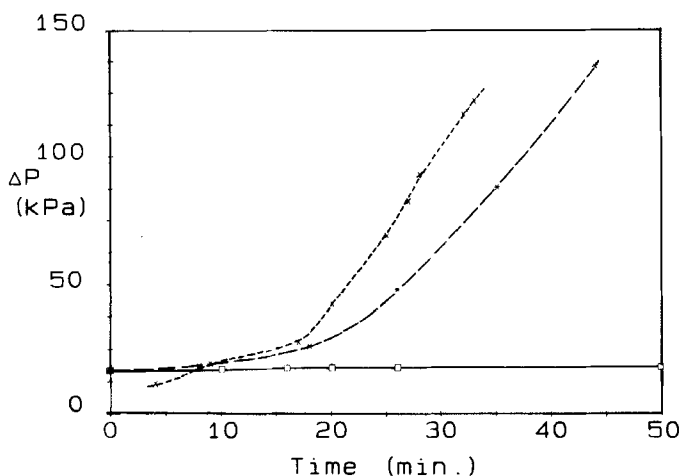

FIG. 5. Effect of emulsified water. ($\times$) $J = 1.5 \times 10^{-4}$ m/s, 57 mg ACFTD/L + 2000 mg H_2O/
L. ($*$) $J = 2.2 \times 10^{-4}$ m/s, 57 mg ACFTD/L. ($\square$) $J = 2.2 \times 10^{-4}$ m/s, 2000 mg H_2O/L.

when a field with positive polarity was applied. This rate is 12 times faster than was observed in the high-concentration ACFTD tests and 150 times faster than was observed in ACFTD-free emulsified water tests.

There are several possible explanations for the synergistic effect of ACFTD and emulsified water. It may be the result of the opposite charges found on the two types of contaminants. Under the test conditions, water drops should be attracted toward the porous electrode while ACFTD should be repelled by it. The rapid pressure drop increase may be the result of the capture of water drops and low-mobility ACFTD particles by the porous electrode and the resultant rapid increase in restriction. The magnitude of the effect is such that this is probably not the only mechanism involved. Positively-charged ACFTD may also form a coating on the negatively-charged water drops. This reduces the effective electrophoretic velocity of both particles, resulting in ACFTD being carried along with the drops toward the porous electrode. Emulsified water may also affect the adsorption of surfactants onto the particles. McGown et al. (15) observed that particle zeta potentials in xylene decrease as water concentration increases beyond its solubility in the bulk phase. This was attributed to competitive adsorption between the two types of contaminants and the corresponding reduction in surface charge and mobility. At present there are insufficient data to determine the relative importance of these mechanisms, though it is expected that all three contribute to the synergistic effect.

The model system results provide an indication of the crossflow electrofilter's performance characteristics. Under ideal conditions (such as the model systems without emulsified water), high removal efficiencies with relatively low pressure drop and power consumption are possible. High capacities and long service intervals were also observed. The apparent capacity of the electrofilter was more than an order of magnitude greater than is observed in most cartridge filters. Upon plugging, regeneration of the electrodes could be readily accomplished by temporarily reversing the polarity of the field and the direction of flow. It is doubtful that this regeneration method would be as effective if soft contaminants such as asphaltenes were involved. Relatively low filtrate fluxes ($\sim 2 \times 10^{-4}$ m/s) were used in these experiments. The flux may be increased to levels approaching that of cartridge filters by increasing the field strength. However, field strengths in excess of $\sim 10,000$ kV/m exceed the dielectric strength of the fluid and may result in arcing.

Several potential problems associated with the presence of emulsified water were observed. When both ACFTD and emulsified water were present, the pressure drop rose rapidly. Increased power consumption and shorting out of the power supply were also observed. Thus, larger

power supplies would be required in applications where both solids and emulsified water are present than would otherwise be the case. In the following section, these and other application problems associated with the electrofiltration of petroleum oils are discussed.

FIELD PERFORMANCE

Petroleum oils represent an extremely diverse collection of fluids and properties. As such, field performance cannot be predicted from the model system alone. This section will discuss crossflow electrofilter performance with different petroleum oils. The results discussed in this section are for unmodified field samples which were tested as received. Care was taken to avoid contamination or alteration of the samples in any way. Because they are so diverse, no effort was made to test the samples at identical flow and voltage conditions. The conditions used are noted at the appropriate locations in the text. Since sample size did not always permit entire β_2 vs field strength curves to be generated, tests were conducted at field strengths of 0 and 1700 kV/m with the porous electrode polarity the same as the contaminant. Other conditions were tested when permitted by the size of the sample.

As indicated in Table 1, the field samples could be divided into three categories: fuel oil, hydraulic oil, and lube oil. The samples differed in terms of their viscosity, contaminants, water content, and fluid chemistry. Like the model systems, the field samples typically demonstrated U-shaped β_2 vs field strength curves. As in the model fluid, two different removal mechanisms were involved; however, in some cases the presence of both positively and negatively charged particles may also have been a contributing factor. Figure 6 summarizes the removal results obtained for the various field samples by application.

The most distinctive U-shaped curve was observed for #2 fuel oil (shown in Fig. 6a). This oil is a relatively additive-free, low viscosity fluid containing primarily toluene-insoluble (i.e., inorganic) solids. When a positive field was applied, the β_2 for #2 fuel oil increased from 15.0 to 38.2 for a filtrate flux of 1×10^{-4} m/s. These β_2s are comparable to the model system even though the contaminant zeta potential is only half as high. A zeta potential of +39.2 mV was observed in #2 fuel oil while a value of +96.2 mV was obtained for the model fluid containing ACFTD. Though one would ordinarily expect the lower mobility of #2 fuel oil contaminants to reduce separation, the low viscosity of this oil compared to the model fluid more than offsets this effect.

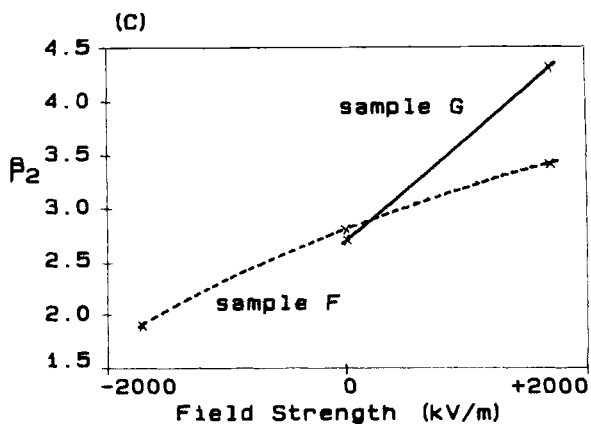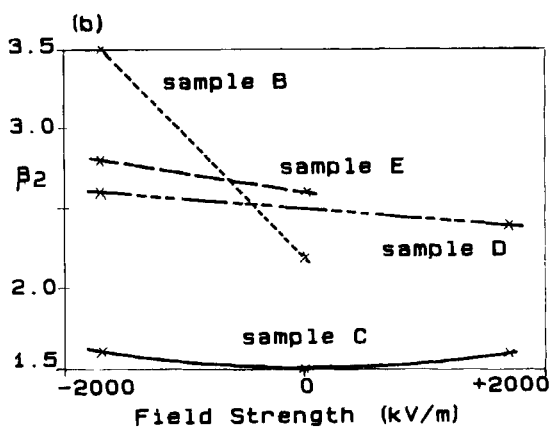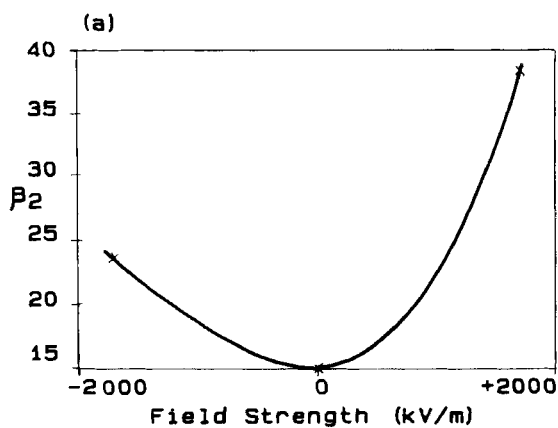The hydraulic oils tested fall into three general categories: new

FIG. 6. Effect of applied electric field on separation for different types of petroleum oils. (a) #2 fuel oil: $J = 1 \times 10^{-4}$ m/s. (b) Hydraulic oils: Sample B, $J = 1.6 \times 10^{-4}$ m/s; Samples C, D, and E, $J = 7 \times 10^{-5}$ m/s. (c) Lubricating oils: $J = 6 \times 10^{-4}$ m/s.

industrial oils, used industrial oil, and used transdraulic oil (i.e., an oil intended to simultaneously serve as transmission fluid and hydraulic oil). Approximately 60% of the solids (weight basis) in the new hydraulic oils (Samples D and E) were toluene-soluble. Since the toluene-soluble (TS) solids typically have specific gravities less than half that of the toluene-insolubles (TI), this suggests that most of the solids were organic in nature. At a filtrate flux of 7×10^{-5} m/s and field strength of 1700 kV/m, no significant removal was observed for these samples. Filtration ratios of ~2.6 were observed regardless of the applied field strength or polarity. Similarly, removal for Sample C (used transdraulic oil) did not improve significantly when a field was applied. A β_2 of ~1.6 was observed. This sample contained a high concentration of emulsified water and the solids were largely TI (inorganic). This system behaved very similarly to the model system containing ACFTD and emulsified water, suggesting that poor removal was due to the emulsified water effect previously discussed. In contrast, the β_2s increased from 2.2 to 3.5 for Sample B (used industrial hydraulic oil) when a field strength of -1700 kV/m and flux of 1.6×10^{-4} m/s were used. This suggests that the particles in this sample were negatively charged. An average zeta potential of -21.6 mV was measured by microelectrophoresis. Primarily TI solids were found in this sample.

Three lubricating oils were also tested: a new oil, a used compressor oil, and a steam turbine oil. All three contained predominantly TI solids, though emulsified water was the major contaminant in the steam turbine oil. Removal from both the new (Sample F) and used compressor oils (Sample G) improved when a field of $+1700$ kV/m and filtrate flux of 6×10^{-4} m/s were used. The β_2 for Sample F rose from 2.7 to 4.3 while it increased from 1.9 to 3.4 for Sample G. In both samples the contaminants appeared to be positively charged. The behavior of the steam turbine oil sample (Sample H) was similar to that of other samples containing emulsified water, i.e., frequent shorting out of the power supply, increased current draw, and poor removal were observed.

Some indication of the factors affecting crossflow electrofilter performance can be obtained from an examination of the field sample results. Perhaps the most obvious factor affecting crossflow electrofilter performance is emulsified water. Poor separation was observed for Samples C and H, both of which contained in excess of 1000 mg H_2O/L. Increased current draw and frequent shorting out of the power supply were also observed. These are the same symptoms which were observed in the corresponding model system tests. Possible explanations for this behavior were discussed in the previous section.

From these results it is apparent that crossflow electrofilter separation in the field is generally much lower than was observed for the model system. The factors contributing to this performance reduction are

apparent from inspection of Eq. (3). Viscosity, dielectric constant and zeta potential are explicit terms in that equation. Superficially, viscosity appears to be the major effect. With the exception of #2 fuel oil and Sample E, the field samples were 2 to 4 times more viscous than the model system and βs were correspondingly 6 to 19 times lower. If viscosity were the only factor involved, the reduced filtrate fluxes for the field samples should have offset this effect. Thus, other factors must also be involved. The dielectric constant was not a significant factor since it was essentially constant for all samples. This suggests that contaminant zeta potentials for the field samples were significantly lower than for the model systems. Microelectrophoresis results indicate that most field samples had zeta potentials less than half that observed for the model system. This effect, coupled with the effect of viscosity, resulted in much lower removal for the field samples.

SYSTEM CHEMISTRY AND PERFORMANCE

Though zeta potential has a major influence on crossflow electrofiltration, the underlying factors controlling its magnitude are not obvious. Previous work in nonaqueous fluids has shown it to be governed by the nature of the contaminant and fluid (*15-17*) as well as the type and concentration of adsorbable species (*5, 15, 18, 19*). In nonaqueous liquids the adsorbable species are usually surfactants, though water also exerts a substantial influence. Ionic surfactants have been shown to exert a particularly large influence on zeta potential. In this section, evidence of the effects of system chemistry on crossflow electrofilter performance are discussed. Though the current study deals with crossflow electrofiltration, it should be noted that these conclusions are not limited to this process. The same effects should exist for other electrokinetic separation processes used in nonaqueous liquids as well.

One of the most important factors affecting contaminant zeta potential is the emulsified water concentration. As previously discussed, three distinct mechanisms may contribute to this effect. The opposite charges observed for water drops and ACFTD in the model system are evidence of the effect of contaminant nature. A less direct effect which results in net zeta potential reduction is the formation of aggregates of water drops and solids. Adsorption competition between the solids and water drops for surfactant may also occur. McGown et al. (*15*) observed that the zeta potential of rutile in xylene containing Aerosol OT decreased as the water concentration increased beyond its solubility. At extremely high emulsified water concentrations, such as was observed in Samples C and H,

surfactants may actually accumulate in the water drops. The net effect is a reduction in the amount of surfactant available for adsorption and a corresponding reduction in zeta potential. The poor separation and service intervals observed for all types of samples containing high emulsified water concentrations demonstrates the importance of this factor.

The properties of the fluid (especially the nature and concentration of the surfactants) may be as important as the emulsified water concentration. Todd and Wild (*16*) observed that a particle may be positive, negative, or uncharged depending on the fluid it is in. Van der Minne and Hermanie (*18*) and others (*5, 15, 19*) have observed that the sign and magnitude of a particle's charge is sensitive to small changes in the amount and type of surfactant. Evidence of the importance of the fluid properties was also found in this study. Results obtained for Samples B, F, and G are shown in Fig. 7. These samples had similar dielectric constants and viscosities. In all three the contaminants were primarily TI solids, and little water was present. Despite these similarities, extremely different β_2 vs field strength curves were observed, suggesting that a difference in contaminant zeta potential exists. For example, the contaminants in Samples F and G (as well as the model system) were positively charged while contaminants in Sample B were negatively charged. In part, this may be due to differences in contaminant nature
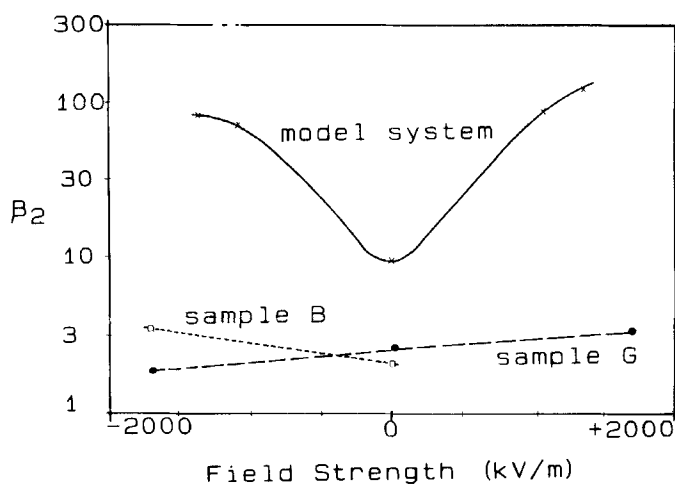

FIG. 7. Effect of fluid composition. Model system: $J = 1.1 \times 10^{-4}$ m/s. Sample B: $J = 1.6 \times 10^{-4}$ m/s. Sample G: $J = 6 \times 10^{-4}$ m/s.

since both wear metal and metal oxide particles are represented in these samples. However, the contaminants in Samples B and G were predominantly wear metals, yet they are charged oppositely. This is a strong indication of the influence of the additive package and other fluid properties on contaminant zeta potential and crossflow electrofilter performance.

Contaminant nature is also an important factor affecting the magnitude of the particle zeta potential and crossflow electrofiltration. In general, separation for TI samples improved upon application of an electric field while TS samples were unaffected. As shown in Fig. 8, removal increased for TI Samples B, F, and G when a field of proper polarity was applied, while TS Samples D, E, and K were relatively unaffected. Removal also improved for the model system and Sample J which contain primarily TI contaminants. In microelectrophoresis experiments conducted using the model fluid, it was found that TI kaolin had a much greater zeta potential than TS asphaltenes obtained from #6 fuel oil. Kaolin had a zeta potential of +74 mV while asphaltenic particles had a zeta potential of only +46 mV. The magnitude of the zeta potential for TI particles is apparently greater than for TS particles.

The differences between the two types of contaminants can be attributed to differences in their surface chemistry and adsorption properties. TS contaminants would be expected to have fewer polar
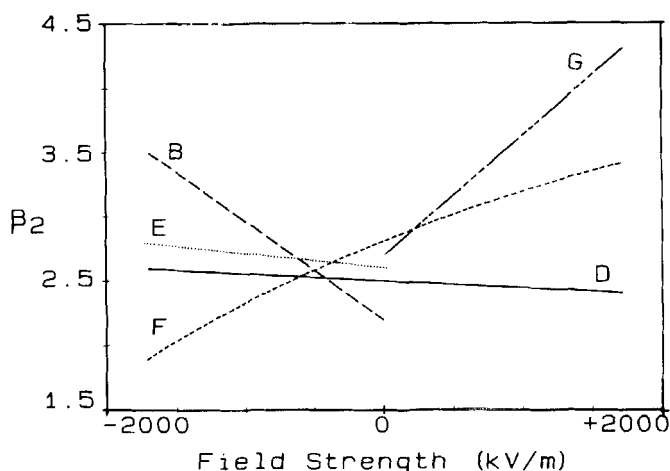

FIG. 8. Effect of contaminant nature. Sample B: $J = 1.6 \times 10^{-4}$ m/s. Samples D and E: $J = 7 \times 10^{-5}$ m/s. Samples F and G: $J = 6 \times 10^{-4}$ m/s.

adsorption sites and be more hydrophobic than their TI counterparts. The relatively low number of adsorption sites for TS contaminants reduces their ability to acquire additional surface charge by surfactant adsorption. Separation for systems with high concentrations of such slightly charged particles would show relatively little dependence on electrical field strength. This explanation is supported by previous work conducted which compared electrophoretic mobilities for a variety of organic and inorganic particles. It has been observed that many types of organic particles are negatively charged in aromatic solvents while metal oxides are positively charged (15, 17). It has also been observed that both types of particles are positively charged in saturated aliphatic solvents. Since petroleum products are typically mixtures of aromatic and aliphatic compounds, it is not surprising that the organic TS contaminants exhibit low electrophoretic mobilities.

Toluene-soluble species, particularly asphaltenes, may also reduce separation by adsorbing onto TI particles. This TS coating may hinder surfactant adsorption by increasing the hydrophobicity of the particles and by rendering the particle surface more acidic. Asphaltene adsorption onto sodium montmorillonite from a model oil has been observed to render it hydrophobic (20). Montmorillonite and the metal oxides found in the field samples of this study should exhibit similar behavior in this regard. McGown et al. (15) suggested that ionic surfactant adsorption is related to a particle's ability to adsorb water. The formation of a hydrophobic TS coating on the particles involved in this study would be expected to reduce adsorption, resulting in the apparently low observed zeta potentials for TS samples. Adsorption has also been related to the acid–base properties of the surface and solvent (17, 19). Most metal oxides would be expected to be more basic than asphaltenes or similar TS material and, therefore, capable of greater adsorption and zeta potential. Adsorption of TS material would hinder adsorption and reduce the zeta potential. Regardless of the mechanism, high concentrations of TS contaminant tended to decrease separation and the magnitude of the contaminant zeta potential.

CONCLUSION

Crossflow electrofiltration has a number of distinct advantages over conventional separation processes; however, the applications where these advantages can be realized are limited. The ideal system for crossflow electrofiltration would have low electrical conductivity ($<10^{-8}$ $\Omega^{-1}m^{-1}$), low viscosity, low emulsified water concentration (<500 mg/L), and

highly charged contaminants (absolute values of zeta potentials > 50 mV). For such fluids, β_2s in excess of 30 could be obtained with extremely long service intervals. Filtrate fluxes comparable to those used for conventional cartridge filters could be obtained and only about 10 W/(L/ min) of flow would be required to achieve separation. For these ideal fluids, crossflow electrofilter behavior approximates that of the consumate filter—high removal for all particle sizes, long life, and minimal power requirements. For ideal systems perhaps the biggest drawback of the crossflow electrofilter is its size. Even without a power supply, the laboratory version used in this study was about 10 times larger than a corresponding cartridge filter. Furthermore, in practice many hydrocarbon systems are not ideal.

Nonpolar liquids containing large amounts of emulsified water present special problems for crossflow electrofilters. Increased power requirements and frequent shorting out of the power supply were observed. When both solids and water drops are present, both removal and service interval are decreased. Thus, performance is reduced and maintenance costs increase when emulsified water is present. To varying degrees, other electrical separation processes used in nonaqueous fluids would face related problems.

The low electrophoretic mobilities observed in the field represent a more serious limitation of the crossflow electrofilter's utility. Contaminants in the petroleum oils examined typically had absolute values of zeta potential less than 60 mV, and the distribution of zeta potentials for individual samples was broad. Both positively and negatively charged particles were observed in some cases. Since removal and service interval are ultimately governed by the concentration of the least mobile particles, high field strengths and/or low filtrate fluxes would be required to obtain effective separation for many applications. This limitation is not restricted to crossflow electrofiltration. Any electrophoretic separation device should be similarly hampered by low contaminant mobility.

Several factors appear to contribute to the apparently low electrophoretic mobilities among the field samples. High concentrations of emulsified water tended to reduce mobility by competing with solid contaminants for surfactant adsorption and by forming aggregates with the solids. The nature of the fluid and surfactants influences mobility. Contaminant nature is also important. Toluene-soluble contaminants tend to reduce separation. This is probably an indication of a relatively low tendency to adsorb ionic surfactants. Further work on the mechanisms by which particles acquire surface charge in nonpolar liquids is needed to confirm the findings of this study and to suggest methods of increasing the inherent charge on contaminants in the field. The need

still exists for practical methods of achieving fine particle and water drop separation from nonaqueous liquids.

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