Electrorheological behaviour at low applied electric fields of microcrystalline cellulose in BP oils

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Electrorheological fluids have been prepared at low applied electric fields from BP oils containing microcrystalline cellulose (MCC); even at a low applied electric field of 500 V mm^{-1}, 10% MCC in oils rich in linoleic or oleic acids behave as electrorheological fluids with the latter displaying significantly higher yield stresses.

Electrorheology describes the rapid and reversible change in viscosity exhibited by certain suspensions of solid particles in electrically non-conducting liquids upon application of an electric field. Electrorheological (ER) fluids can run freely like water, ooze like honey or solidify like gelatine, depending upon the applied electric field. Winslow pioneered the use of ER fluids in the 1940s, with mechanical engineering applications ranging from a simple hydraulic valve[1] to a complex tracking device for copying machines.[2] The appeal of ER fluids is the rapid response, usually on the millisecond time scale, upon application of an electric field,[3–5] and this response is completely reversible upon removal of the electric field.[5,6] Although typically the electric fields necessary to provide an ER effect are in the region of 1–5 kV mm^{-1}, the power consumption is small (mW).[7]

Traditionally, ER fluids are composed of a dispersed particulate phase,[7,8] in the size range 0.5 to 100 μm, in an insulating base fluid.[9–11] In the absence of an electric field, most ER fluids behave, to a first approximation, as Newtonian fluids. When a continuous DC electric field E is applied to an ER fluid and the fluid is sheared in a direction perpendicular to the field, the relationship between the stress τ and the shear rate γ can be described by the Bingham model. According to this equation, flow only occurs once the applied stress exceeds the static yield stress $\tau_\gamma(E)$. The flow equation is given in eqn. (1), where η_B is

$$\tau(E) = \tau_\gamma(E) + \eta_B\gamma \quad (1)$$

termed the Bingham viscosity. We are investigating the feasibility of using ER fluids as controllable drug delivery systems. In the absence of an electric field, a basal level of drug release will occur by diffusion across a mesh electrode. It is envisaged that upon application of an electric field, drug release will be controlled (hindered or halted).

We initially selected the pharmaceutically acceptable tablet excipient microcrystalline cellulose (MCC) in silicone oil as our ER fluid. This ER fluid has been recently reported, but only under high electric fields (in the range 1–3 kV mm^{-1}).[12,13] This project encompasses the search for pharmaceutically acceptable alternatives to traditional (engineering based) ER base fluid components. Thus, silicone oil (100 cS) or an alternative oil was used as the base fluid, together with sieved MCC (size fraction below 45 μm).‡ We have investigated the use of super refined BP oils as substitutes for silicone oil.‡ Oleic acid [(*Z*)-octadec-9-enoic acid] is the major constituent of apricot kernel (68%),[14] safflower (63%),‡ peanut (56%),[15] and sesame seed oils (45%).[15] Linoleic acid [(*Z*,*Z*)-octadeca-9,12-dienoic acid) is the primary constituent of sweet almond oil (75%)[16] and soyabean oil (50%).[15] In pharmaceutics, peanut and sesame seed oils find their uses as vehicles for sustained-release intramuscular injections.[16] Almond oil is also used as a vehicle for injections[17] and soyabean oil has replaced peanut oil in total parenteral nutrition regimens.[15]

A CSL^2 rheometer (TA Instruments, Leatherhead, UK) has been specially modified to allow the application of an electric field across the test fluid. A small electrolyte reservoir (approximately 0.5 ml) containing a 0.1% w/v solution of aq. KCl was used to form the electrical connection between the power supply (Model PS350 High Voltage Power Supply, Stanford Research Systems, Sunnyvale, CA, USA) and the rheometer geometry. The draw rod is insulated except for the threaded portion at the tip which makes contact with the geometry. In our experiments, we used a small volume (*ca*. 3.5 ml) concentric cylinder where the diameters of the cup and bob were 9.33 and 8.60 mm, respectively, resulting in a gap of 730 μm. All ER fluids were prepared using 10% w/w MCC (sieve fraction below 45 μm) in the appropriate oil, then sonicated (Decon FS300b) for two periods of 15 min prior to analysis. Temperature equilibration (37 ± 0.1 °C) of the sample was carried out for 15 min under the influence of an applied electric field (500 V mm^{-1}) prior to measurement. A continuous ramp of shear stresses from 0 to 50 Pa at a rate of 0.1 Pa s^{-1} was applied to each ER fluid with the resultant shear rate measured. A flow curve was plotted (Fig. 1) and the yield stress was determined by extrapolation of the experimental shear stress–shear rate data to zero shear rate using the Bingham model. Five measurements from each sample were taken and the associated mean and standard deviation were calculated (see Table 1).

In these studies, we have shown that, at a low applied electric field of 500 V mm^{-1}, suspensions of 10% w/w MCC in a range of BP oils behave as ER fluids. Furthermore, in general, BP oils afforded a higher Bingham yield stress than silicone oil (see Table 1), typically 12 Pa compared to 9 Pa. Using the ANOVA

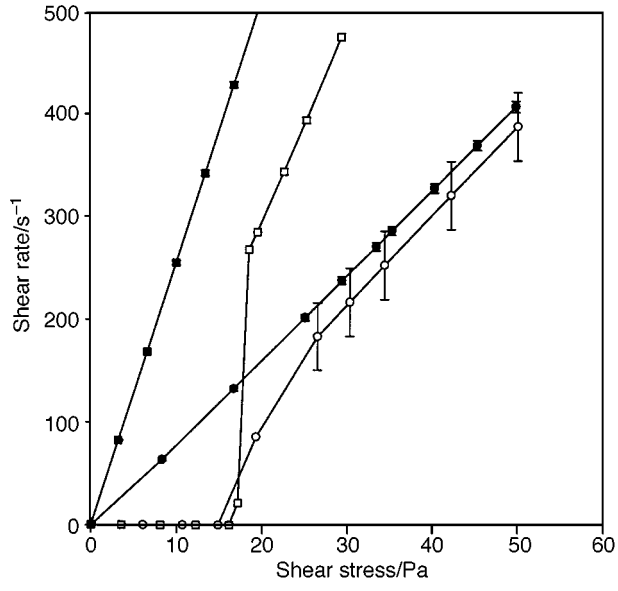

Fig. 1 Comparison of the flow behaviour at 0 V mm^{-1} [(●) silicone oil; (■) sesame seed oil] and 500 V mm^{-1} [(○) silicone oil; (□) sesame seed oil]

Table 1 The Bingham yield stress values for 10% w/w MCC in various oils at 500 V mm^{-1} (n = 5)

Oil	Bingham yield stress/Pa	Standard deviation
Almond	8.79	0.29
Apricot	12.54	0.39
Peanut	12.78	0.33
Safflower	12.17	0.47
Sesame seed	12.27	0.12
Silicone	8.94	0.26
Soyabean	10.53	0.37

test (one way), it was found that there were statistical differences ($p < 0.05$) between the oils. A Fisher analysis was carried out to highlight where differences were present. No significant difference ($p < 0.05$) was found between almond oil and silicone oil. Apricot oil, peanut oil, safflower oil, and sesame seed oils were also found not to be significantly different. In the light of these findings, we conclude that the BP oils which have oleic acid as the major constituent have significantly higher yield stress values compared with the BP oils having linoleic acid as their major constituent. Preliminary studies, using a parallel plate geometry, with 10% MCC in oleic and linoleic acids (*ca.* 95%)‡ also exhibited an ER response at 250 V mm^{-1}. This demonstration of ER responses below 1 kV mm^{-1}, using pharmaceutically acceptable oils, should find ready applications.[18–22]

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Notes and References

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‡ *Reagents*: Microcrystalline cellulose (MCC) (Lot No: E5D7C21, Emcocel 50M, Mendell, Patterson, NY, USA); 100 cS silicone oil, oleic and linoleic acids (Aldrich, Gillingham, UK); super refined BP oils (Croda Oleochemicals, Goole, UK).

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