



Effect of fibrous filter properties on the oil-in-water-emulsion separation and filtration performance

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ABSTRACT

Separation of secondary emulsions of dispersed droplet size less than 10 μm , by means of fibrous medium is a very complex but important process. The study investigates the influence of thin fibrous filter properties, i.e. surface energy, pore size and porosity on the separation performance of an isooctane in water emulsion (0.2%, mean drop size 2 μm). Experiments were carried out on five different filter media with a wide variation in their pore size (2–51 μm), surface energy (14–46 mN/m) and porosity (0.46–0.87) at similar process conditions. Filter media with different wettability are obtained by applying various hydrophobic and hydrophilic coatings. All the used coatings contain nanoparticles (25 nm) to impart nanoscale surface roughness at the single fiber surface. Besides emulsion properties and operating conditions, the phase separation mechanism and performance highly depends on pore size, surface energy and porosity of the filter media. More complete coalescence takes place at reduced pore size and at a surface preferentially wetted by the dispersed phase. Whereas when the pore size equals to the influent droplet size, then the surface wettability of filter is less effective and the separation mechanism is governed by inflow velocity. The emulsion inflow velocity and pressure drop are significantly affected by the filter media air permeability but do not depend on filter surface energy.

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1. Introduction

Finely dispersed liquid–liquid emulsions occur in various engineering applications and chemical industries. Their separation is very important. Secondary emulsions of droplet size less than 100 μm cannot be separated efficiently just by gravitational or buoyancy forces due to low settling velocity. There are two very important and common mechanisms to separate these emulsions by means of porous fibrous media: (a) coalescence by means of depth filter and (b) separation by means of surface filter [1–9]. The main steps in the coalescence filtration process are (1) transport of small droplets from an immiscible fluid to the filter, (2) attachment of small droplets to the fibers surface, (3) increase in drops size on the fiber (coalescence), (4) merging and transport of drops on the fiber and (5) removal of the drops from the filter [6–13]. While in the case of surface filtration the smaller pore size and the surface geometry of the filter do not let the dispersed droplets to pass through the filter and retain them at filter surface.

A large number of parameters are very crucial and determinative for liquid–liquid separation either by one of these two mechanisms: dispersed and continuous phase properties (droplet size, difference in the density of dispersed and continuous phase, interfacial tension, and presence of surfactants), operating conditions (inflow velocity, pressure drop, and emulsion concentration), the choice of filter properties (pore size, fiber fineness, permeability, thickness, etc.) [4–11]. However, the opinions differ on the effect of filter media surface wettability on emulsion separation. For the emulsion separation by means of coalescence, some authors concluded that the dispersed phase should readily wet the fiber surface [6–8,14], while others concluded that it should not [4,15], yet some have resolved that wetting behavior of the dispersed phase is unimportant and/or intermediate wetting conditions are required [13,15–18].

Besides this divergence about the consequence of filter surface energy, no systematic and comprehensive information is available about the effect of various filter properties on the emulsion separation performance. Due to a wide variation in the decisive operating conditions as well as filter properties, it is very difficult to identify the general dependence of various factors on one another. For example, in most of the done studies very thick coalescer media (>0.5 cm) were used with a mean influent dispersed droplet diameter greater than 10 μm [1–4,8–10,13,17]. Such droplets are bigger than the filter pore size, so are being captured by them easily. These droplets are easily deformable under

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Table 1
Isooctane characteristic properties.

Mean molar mass	114.23 g/mol
Density at 20 °C	692 kg/m ³
Viscosity at 20 °C	0.277 mPa s
Surface tension at 20 °C	18.66 mN/m
Interfacial tension against 100% deionised water at 20 °C	54.7 ± 3.9 mN/m
Interfacial tension against deionised water with 0.00005% sodium dodecyl sulfate surfactant at 20 °C	46.2 ± 2.3 mN/m

sufficient hydrodynamic forces and also exhibit high contact area with the fibers. While in a secondary emulsion of very small drop size (<10 μm), the droplets are nearly spherical and tend to have only point contact with a single fiber instead of contact with several fibers [3,6,19]. The phase separation mechanisms strongly depend on the inflow velocity and pressure drop across the filter. Thus, at such small dispersed droplet size diffusion, sedimentation and London forces predominate at low velocities and interception is most important at high velocity [6]. Furthermore for such smaller droplets, as per Wenzel theory, the surface roughness of the filter media, wettable with the dispersed phase, does not contribute to the drop coalescence by reducing their contact angle ($\cos \theta r = r \cos \theta$), here θ and θr are the intrinsic contact angle on a smooth surface and that on a rough surface made of the same material, respectively, r is the roughness factor [11,13,19], but promotes the drop capture.

Therefore, in the study we have systematically analysed the influences of pore size, filter surface energy, porosity and air permeability on the various phase separation performance parameters of secondary oil in water emulsion, i.e. size of the uncaptured dispersed droplets in the effluent, separation efficiency and mechanism, inflow velocity and pressure drop for very thin nano-coated filter media (<0.2 cm) and very small dispersed oil droplets (<6 μm). At the same time various hydrophobic, oleophobic and hydrophilic nanocoatings were used to alter the fiber wettability of the filter media. All these coatings contain nanoparticles to impart nanoscale surface roughness at the single fiber surface in order to promote the capture and coalescence of dispersed oil droplets by reducing their contact angle.

2. Materials and methods

2.1. Equipment and operating procedure

Isooctane was used as the dispersed phase. The main characteristic properties of the oil are given in Table 1. Its surface tension against air and deionised water was measured according to Wilhelmy plate method (DIN 53914) on Tensiometer instrument, model DCAT-11.

The experiments were performed on a laboratory scale filtration setup with vertical downward fluid flow orientation (Fig. 1). Isooctane in deionised water emulsion (0.2% by weight) was prepared by high speed mechanical agitating (3) followed by ultrasonic treatment. The emulsions were stabilized by adding 0.00005% sodium dodecyl sulfate surfactant. For all the filter media the emulsion was continuously pumped through a peristaltic pump (4) at its constant rpm. The samples for the permeate concentration were collected after reaching steady state condition, where the steady state confirmation was done with the stabilisation of the pressure drop across the filter (Fig. 1) (7).

The dispersed oil droplet size distribution in the influent and effluent stream was determined by the particle counter instrument, model LA-300 based on light scattering principle. The biggest dispersed oil droplet in the influent was always less than 6 μm where

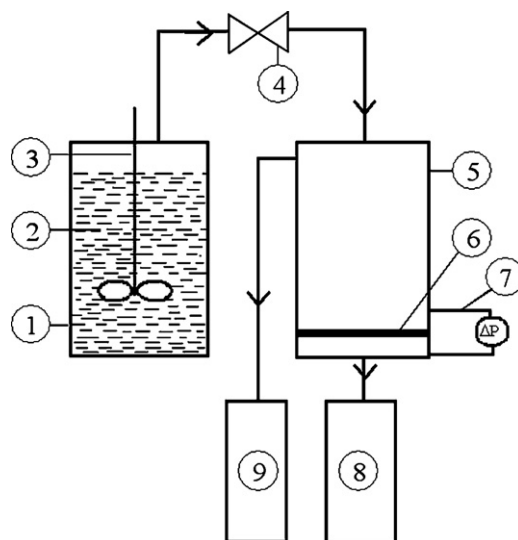


Fig. 1. Experimental installation, 1 – emulsion tank, 2 – oil in water emulsion, 3 – mechanical stirrer, 4 – peristaltic pump, 5 – filtration cartridge, 6 – filter medium, 7 – manometer, 8 – filtrate, and 9 – recovered oil.

50% droplets were less than 2 μm (Fig. 2). The dispersed isooctane concentration was determined by IR spectrometry from a carbon tetrachloride extract, adjusted to pH 1 with HCl to stabilize the oily water samples. Thus, the emulsion separation efficiency (η) was given in Eq. (1), where C_f and C_p represent the organic concentration of the influent emulsion and the effluent, respectively.

$$\eta = \frac{(C_f - C_p) \times 100}{C_f} \quad (1)$$

2.2. Properties of the fibrous filter media

Five different kind of filter media with wide variation in their properties, i.e. pore size, surface wettability, porosity, and air permeability were analysed (Table 2). To obtain filter media with a wide range of surface energy values (15–46 mN/m), they were treated with various hydrophobic, oleophobic or oleophilic coatings on a laboratory scale padding machine (Model-2 Roller Foulard Type HVF) under similar operating conditions (temperature, velocity and pressure). The details of the used coatings are given in Table 3. All the used coatings contain nanoparticles either in situ or in dispersion (25 nm), to impart the nanoscale surface roughness at the single fiber surface in order to promote the isooctane droplets capture and coalescence (Wenzel theory). The surface energy values of various porous filter media were

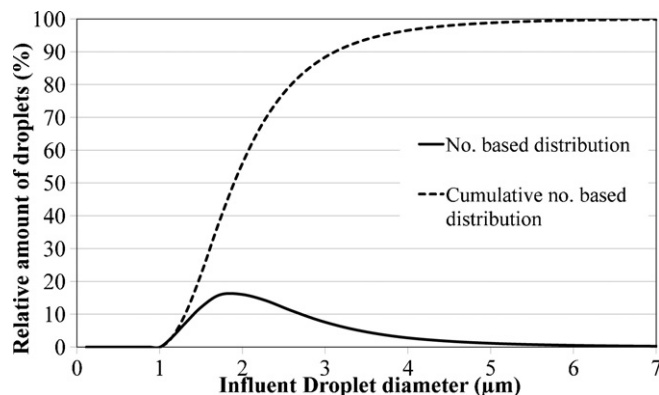
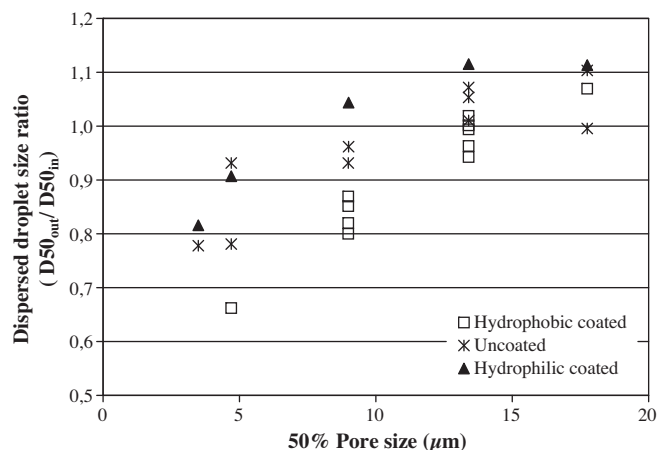


Fig. 2. Dispersed droplet size distribution in the influent ($D_{50,in}$).

Table 2
Physical properties of investigated filter media.

Sample code	Material and polymer type	Pore size (μm) (DIN 58355-2)		Air permeability ($\text{l/m}^2/\text{s}$) (DIN EN ISO 9237)	Thickness (mm) (DIN EN ISO 9073-2)	Porosity
		Range	50% Pore (P50)			
F1	PET-woven	2–19	3.5 ± 0.2	14.5 ± 2	0.86 ± 0.04	0.46
F2	Cellulose-nonwoven	3–11	4.7 ± 0.2	17.8 ± 4	0.60 ± 0.06	0.73
F3	PBT meltblown + PET spunbond	6–19	9.0 ± 0.7	135.0 ± 14	1.64 ± 0.06	0.87
F4	PET-needled nonwoven	9–51	13.4 ± 0.7	152.2 ± 8	1.95 ± 0.05	0.78
F5	PET-spunbond	11–43	17.7 ± 1.1	170.9 ± 19	0.59 ± 0.02	0.65

**Fig. 3.** Effect of pore size (50%) on mean dispersed droplet size in the effluent.

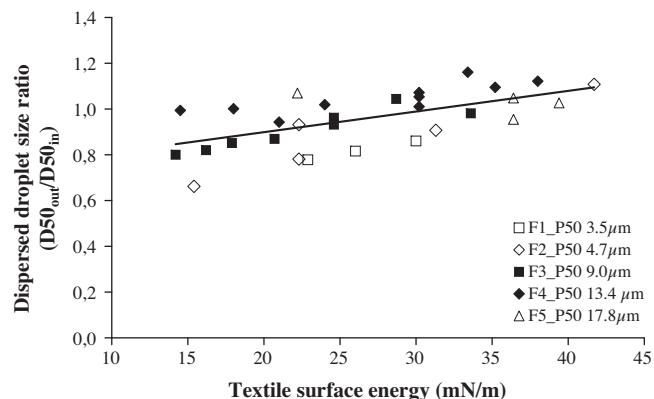
measured according to OWRK-method by powder contact angle measurement (Washburn method) on the DCAT-11 Tensiometer instrument. Various other properties of the filter media were tested according to DIN standards. The filter media air permeability was also determined because other filtration performance parameters, i.e. emulsion flow velocity and pressure drop depend on it. It was found that the air permeability and the pore size did not change after nanoparticles coating treatment. The microstructure and surface morphology of the coated and uncoated fibers and filters were characterized by scanning electron microscopy (SEM). For all the experiments the working temperature was constant, 293 K.

Depending on type of filter medium used and at constant pump rpm the emulsion flow velocity varied from 0.60 to 0.85 m/min with a pressure drop 0.8–2.0 bar.

3. Results and discussions

3.1. Effect of pore size and filter surface energy on the mean dispersed droplet size ($D50_{out}$) in the effluent

The mean dispersed oil droplet size in the effluent is being reduced with decreasing filter pore size for both, the low and high surface energy filters (Fig. 3 and Table 2). It is due to enhanced dispersed oil drops capture by the filter with small pore size. Therefore, the uncaptured dispersed droplets in the effluent are an

**Fig. 4.** Effect of filter surface energy on mean dispersed droplet size in the effluent.

indirect measurement of the amount of coalescence and separation efficiency.

At constant pore size the mean dispersed oil droplet size in the effluent is being increased with increased fiber surface energy (Fig. 4). Depending on liquid surface tension (isooctane 18.7 mN/m, water 72.3 mN/m), the filter with low surface energy (<30 mN/m) is easily wettable with oil but not with water (Fig. 5A-a). Thus, at a low surface energy material the adhesive forces between the oil molecules (isooctane) and the filter surface are much higher than between the water molecules and filter surface, although water is highly polar in nature. Therefore, the dispersed oil droplets displace water easily at the low surface energy filter media and spread out (Fig. 5A-a). While due to high adhesive forces between the polar water molecules and polar surfaces the oil droplets cannot displace water on high surface energy filter media. Oil droplets on hydrophilic surfaces in water exhibit high contact angles being substantially spherical in shape (Fig. 5B-a). These spherical oil droplets pass easily through the high surface energy filter due to little adhesion forces or due to droplet deformation by high hydrodynamic forces (Fig. 5B-b). Thus, for filters with same pore size, bigger dispersed oil droplets from an oil in water emulsion pass through the high surface energy material than through the low surface energy material (Figs. 3–5). Hence the filter wettability (surface energy) and pore size affects the size of the uncaptured dispersed droplets in the effluent significantly.

The flow velocity directly affects the amount of shear forces on the dispersed droplets (therefore droplet deformation) and pressure drop at the filter. The ratio of dispersed droplet size in the

Table 3
Details of nanoparticle coatings used to vary the surface energy of filter media.

Coating name	Functionality	Composition	Particle size (nm)	Concentration applied (%)
x-clean® PK3033 P	Hydrophilic	SiO_2 -nanocomposite in water	25	2.5, 5.0
x-tex® EC 5008	Hydrophobic and oleophobic	Organic fluorine based resin with curing catalyst	25	2.5, 5.0
x-tec® EC 2115	Hydrophobic and oleophilic	Alcohol-based silicone and fluorine free siloxane	In situ	2.5, 5.0

Coatings producer and supplier: Nano-X GmbH, Germany.

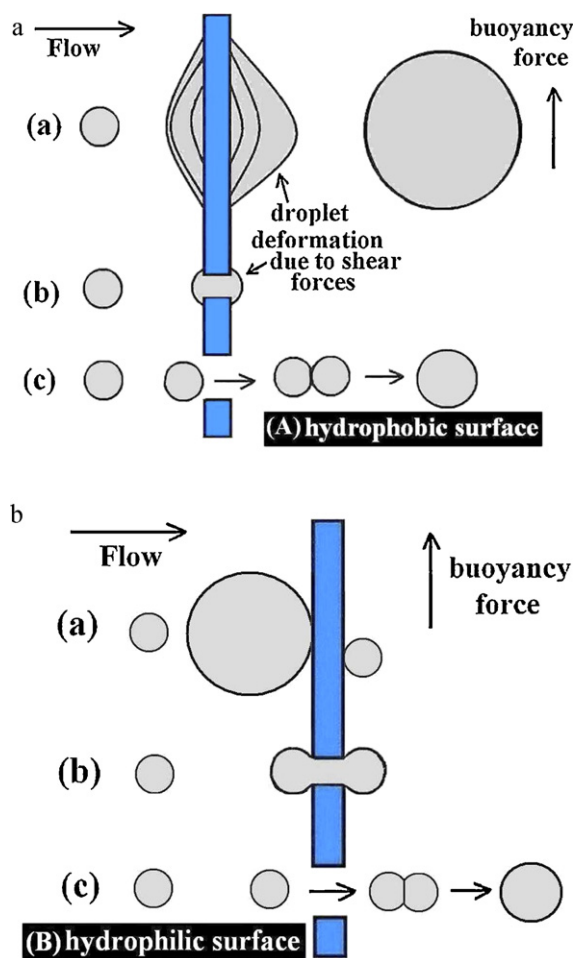


Fig. 5. Oil in water emulsion separation mechanism at a hydrophobic or hydrophilic surface (a) droplet adhesion and coalescence at filter (b) droplet deformation at the pore due to shear forces and (c) droplet escape from the pore and coalescence in the flow stream.

effluent to influent is slightly larger than one for the high surface energy filter (Fig. 4). This slight increase in droplet size is due to superficial coalescence among the droplets in the flow stream (Fig. 5A-c and B-c). At about similar flow velocity, i.e. 0.65–0.8 m/min, the amount of shear forces acting on the dispersed droplets are same at the low and high surface energy filters. But at this similar magnitude of shear forces, the adhesion forces of the oil molecules with the high energy surface (hydrophilic) are much smaller than with the low surface energy material (Fig. 5A-a and B-a). Therefore, easy detachment of the oil droplets takes place from the high surface energy filter, which hinders coalescence of the droplets within the filter. Therefore, depending on pore size to influent drop size ratio, a high number of droplets escape through a high surface energy filter with easy deformation of the droplets, which leads higher possibility of the droplets to coalescence in the flow stream. But these droplets are still small enough to separate themselves from the outflow stream and remain dispersed (Fig. 5A-c and B-b/c).

3.2. Effect of pore size and filter surface energy on the separation efficiency

Since the dispersed droplets capture diminishes with the increasing pore size; the separation efficiency reduces linearly (Fig. 6). Whereas for the similar pore size range (2–51 μm), the rate of fall in separation efficiency is significantly lower for the low

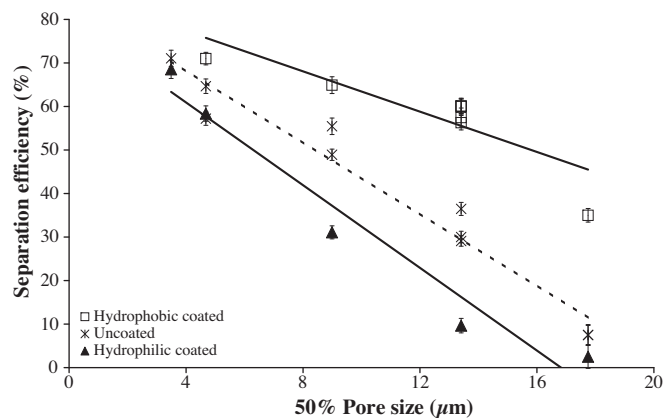


Fig. 6. Effect of pore size (50%) on separation efficiency.

surface energy filter media (hydrophobic coated) than the higher surface energy filter media (uncoated and hydrophilic coated). It is due to spreading and further coalescence of the dispersed oil droplets at the low surface energy materials from an isooctane in water emulsion.

Filter media pore size and surface energy significantly affect the dispersed oil droplet capture and wetting behavior, respectively (Figs. 3–5). Therefore, these parameters affect the size of uncaptured dispersed droplets in the effluent and separation efficiency of the oil in water emulsion (Fig. 7). At the similar pore size the spreading of the dispersed oil droplets on the low surface energy materials is encouraged by the nanoscale surface roughness at the single fiber surface provided by the nanocoatings. Thus low surface energy, surface roughness and small pore size synergistically stimulate the oil droplets capture and coalescence at the filter surface and within the filter. Depending on flow velocity and filter porosity, these coalesced oil droplets leave the low surface energy filter after attaining a critical size and join the outflow stream. These coalesced oil droplets are then big enough to float at the filtrate surface and to be separated easily. Thus similar to the findings of other authors [6–8,14], for the coalescence filtration, the coalescer surface should also be preferably wettable by the dispersed phase but not by the continuous phase.

On a comparison among various investigated filter media, a transition from coalescence filtration to surface filtration or vice versa takes place and depends on the ratio of pore size to the influent droplet size (Fig. 7). Like, when the ratio of pore size to influent drop size (6 μm) is smaller or equal to one (F1 and F2), then the

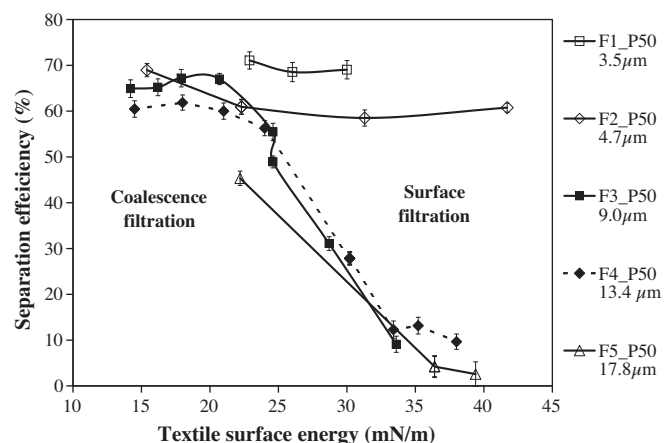


Fig. 7. Effect of filter surface energy and pore size on separation efficiency.

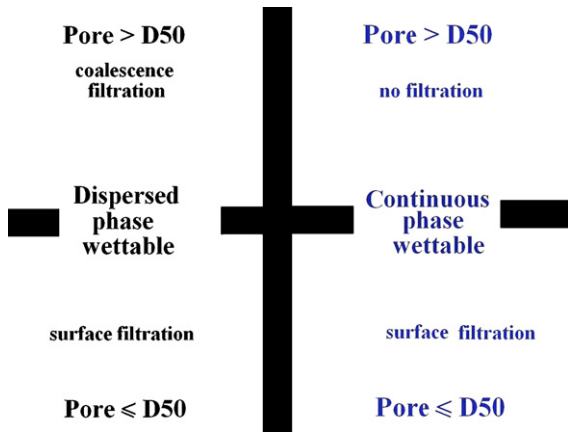


Fig. 8. Separation mechanisms (surface and coalescence) of oil in water emulsion.

surface energy of filter media is less effective (Fig. 7). In the case at low flow velocity the dispersed oil droplets are being retained at the thin filter surface (both higher and low surface energy) and further coalesce there. Thus, at this stage phase separation takes place by surface filtration mechanism and is governed by inflow velocity. Because at high flow velocity, due to high hydrodynamic forces these coalesced oil droplets at the filter surface are either deformed or redispersed into smaller droplets passed through the pores of thin filter (Fig. 5A-b and B-b). Whereas, if this ratio is larger than one, then by means of a surface preferably wetted by the dispersed phase (F3 and F4), high emulsion separation efficiency can be achieved by means of coalescence mechanism at *all the flow velocities* and at *lower pressure drop* (Figs. 6 and 7). Hence, the phase separation strongly depends on the ratio of pore size to influent drop size and filter surface energy (Fig. 8).

Thus, in addition to Davies and Jeffreys findings that for very small droplet size the pore size is much more important than surface wettability of a filter bed [17,19], when the ratio of pore size to influent drop size approaches near one, then the surface wettability of filter is less effective and the separation mechanism is governed by the inflow velocity. Therefore, for the separation of water in oil emulsion at low flow velocity, which is a real application condition of fuel filtration in automobiles by surface filtration, a dense (to retain the dispersed droplets) and rough surface (promotes retained droplets coalescence and easy separation at filter surface, surface roughness contribution to wetting theory) with smaller pore size to influent drop size ratio (≤ 1) and preferably wetted by continuous phase (fuel oil, so that the dispersed water droplets possess large contact angle and can detach easily from the filter surface) is suitable. Whereas a highly porous rough surface preferably wetted by dispersed phase with smaller pore size but bigger than the dispersed droplet size, promotes coalescence and is suitable for deep filtration at all inflow velocities.

3.3. Effect of filtration parameters on the phase separation performance

As mentioned earlier that the phase separation efficiency depends on a large number of variables. At about similar flow velocity (0.65–0.8 m/min) and influent droplet size, the uncaptured dispersed droplet size in the effluent decreases linearly and the phase separation efficiency increases asymptotically with increasing ratio of filter porosity multiplied with influent droplet diameter to the 50% pore size (P50), filter surface energy and inflow velocity (Figs. 9 and 10). Thus, for the investigated filter properties

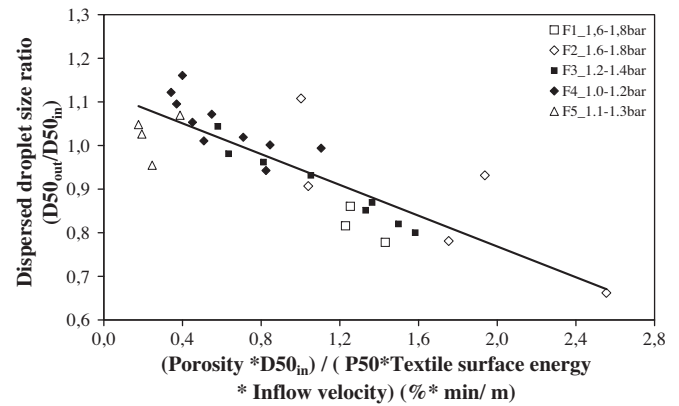


Fig. 9. Effect of filtration parameters on effluent mean dispersed droplet size ($D50_{out}$).

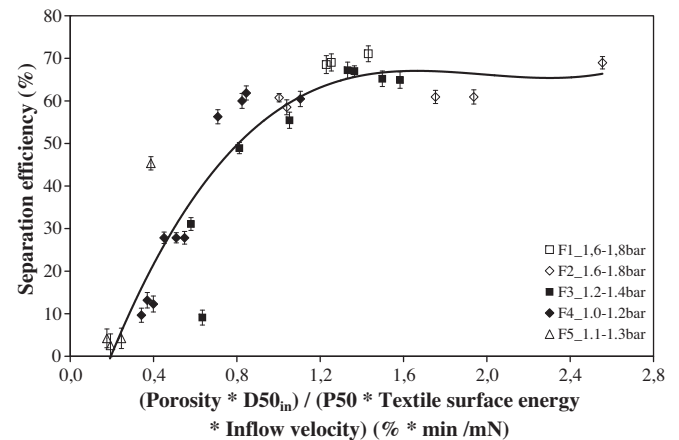


Fig. 10. Effect of filtration parameters on the separation efficiency.

and operating conditions, the optimum value of the investigated parameters function is 1.5 where the highest phase separation efficiency had been achieved (73%) (Fig. 10). Similar to the findings of other researchers [1–9,13–19] for effective phase separation, a coalescer medium (thick/thin) should be preferably wetted by the dispersed phase and possess high porosity and small pore size.

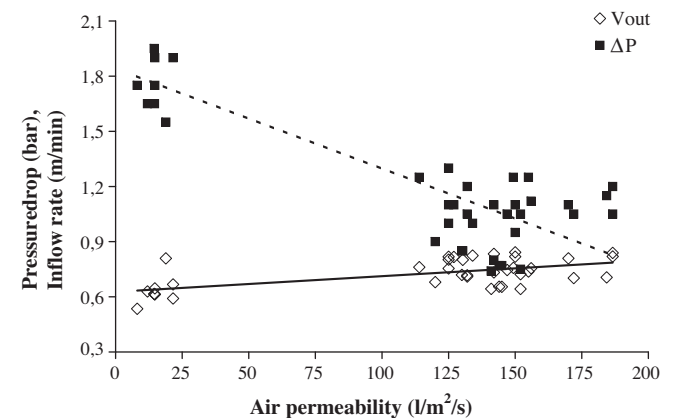


Fig. 11. Effect of filter air permeability on pressure drop and emulsion inflow velocity.

3.4. Effect of filter media air permeability on pressure drop and emulsion inflow velocity

The pressure drop decreases rapidly and linearly while the emulsion flow velocity increases at much slower rate with increasing air permeability of the filter media (Fig. 11). Hence, the air permeability of the filter controls the pressure drop mainly, while the emulsion flow rate mainly depends on the dosing capacity of the pump.

4. Conclusions

The phase separation performance of the various nano-coated porous fibrous media has been analysed and highly depends on the pore size, surface wettability, porosity and air permeability of the fiber filters, besides emulsion properties and operating conditions.

1. At similar surface energy, the droplet capture efficiency diminishes with increasing pore size therefore the separation efficiency decreases. Thus, more complete droplets capture takes place with reduced pore size.
2. The coalescer should be preferably wetted by the dispersed phase, in order to stimulate the droplet capture and coalescence. Whereas when the ratio of pore size to influent drop size approaches near one, then the surface wettability of filter is less effective and the phase separation mechanism depends on the inflow velocity.
3. The coalescence efficiency depends on the influent droplet size, drop residing time in the filter media (which is a function of bed thickness, inflow velocity and drop wettability at the filter media), surface wettability, pore size, fiber fineness, porosity of filter medium.

The presented results give rise to some new issues in directing further studies in this domain. The questions of whether a change of the nature of the oil phase, oil concentration, inflow velocity, and addition of nanofibers of different wettability on different filter media of different surface energy would essentially influence the relationship between the filtration performance for the same and different filter materials, remain to be answered.

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