Liquid–liquid separation using steady-state bed coalescer

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Abstract

This paper presents a literature review on the current understanding of liquid-liquid separation that is immensely widespread in practice, highlighting the steady-state bed coalescer being a good solution in various engineering application. Generally, the fibre bed coalescence has proven to be very effective separation method in the industry. Due to the complexity of bed coalescence phenomenon coalescer design and sizing procedure relies on experimental test. This review provides a research overview of the key phenomena essential for the efficient bed coalescence, such as mechanisms of droplet coalescence and emulsion flow through the fibre bed. In addition to this provides an overview of the current knowledge about coalescer´s design properties and variables such as: fluid velocity, fluid flow orientation/flow mode, fibre bed geometry, and bed length.

Keywords: liquid–liquid separation, droplet coalescence, fibre bed, bed coalescers.

During the crude oil production on oil fields, two types of separation must be carried out. Since the crude oil always contains a certain amount of water, the first step is to remove this water. Two fractions are formed after this separation: crude oil with a residual dispersed water and water with a residual amount of dispersed crude oil (formation water). Further preparation of crude oil for transport consists of additional dehydration, whereas the optimal care of formation water implies its injection into abandoned wells [1–5]. In order to prevent clogging of the pore space, prior to the injection the suspended particles and the remaining content of crude oil must be removed from the water. For all of these separations regarding the liquid––liquid systems, bed coalescers have been commonly used.

Oily water is extremely widespread in the industry. Water is often the main cooling fluid. Maintenance of vehicles and heavy machinery also cause the formation of significant quantities of oily wastewater. Oily wastewater can also be formed in specific operations that use oil or other organic solvents such as quenching, extraction and other similar operations [1–5].

In addition, the application of bed coalescence significantly improves the productivity of the process furnaces during the steam reforming process. In this way, the control and reduction of the emission of waste gases is achieved. In order to eliminate the water droplets from the output material flow of gas oil, modern facilities for hydrodesulphurization of gas oil incur-

porate coalescers. Since the petroleum products are often highly acidic in the oil and gas industry, their neutralization with base solvents is needed. The base treatment is applied in the following processes: during the mercaptans oxidation process, removal of hydrogen sulfide, the production of LPG (liquefied petroleum gas), kerosene, FCC (fluid catalytic cracking) gasoline, olefins, alkylates and etc. The residual base solvent affects the petroleum product turbidity and increases the sodium concentration. The bed coalescers are used to remove the excess base solvent from the petroleum products [3,5].

MECHANISMS OF COALESCENCE

Introduction

Coalescence is defined as droplets drowning either in its formed continuous phase or as merging two droplets into a larger one. Three main types of coalescence can be distinguished: droplet coalescence at the liquid-liquid interface, coalescence of the adjacent droplets and droplet coalescence in the porous bed.

The beginning of the investigation of coalescence phenomenon is based on the analogy with the DLVO theory. In 1941 Derjaguin and Landau presented a theory of stability of colloidal particles as a function of the attractive and repulsive forces [6]. Seven years later, independently of the previous theory, Varwey and Overbeek proposed the same theory of stability [7]. The DLVO theory takes into account only the attractive van der Waals forces and the repulsive force of the electrical double layer. The authors concluded that the colloidal particles stability depends on the total potential energy, which has three components. One component represents the energy contribution of

attractive forces; the second component is the energy contribution of repulsion forces, whereas the third is potential energy of a solvent. The potential energy of the solvent can be neglected due to the low intensity and a short action radius of a few nanometers. The most important attractive forces between particles are the London-van der Waals forces. The energy contribution of the repulsive forces comes from the presence of the electrical double layer. With the increase of ion concentration the thickness of the electrical double layer decreases and thus the system becomes unstable. Another group of repulsive forces that occur between particles are the Borne forces. These forces are the result of interactions between electrons in the electron shells that occur at shorter distances being the main difference from the electrical double layer force [6,7].

Elimelech *et al.* pointed out that all the forces acting between the particles are of electromagnetic nature [8,9]. However, there are also other forces that influence the particles/droplets interactions and are responsible for their capture, coalescence, aggregation, detachment, and redispersion. These forces are called external forces and include: the force caused by the Brownian motion, hydrodynamic forces, friction forces etc. [10–15].

Coalescence at the liquid-liquid interface

The coalescence of droplets at the liquid–liquid interface consists of the following: the droplet transport from bulk liquid to the interface, the continuous phase film formation between the droplets and its established phase, the rupture, film breakage, drainage and coalescence of droplets into the surface of continuous phase. The developed mathematical models for the droplet coalescence at the liquid–liquid interface allow monitoring the kinetics of the film drainage trapped between the droplets and the liquid–liquid interface. The time to reach the film thickness for its rupture is given in some mathematical relations [16–23].

Coalescence at the liquid–liquid interface can be performed with or without deformation [3,5,16–24]. The coalescence without deformation is shown in Figure 1a where a rigid droplet and the rigid interface exist. This form of coalescence is generally efficient, as it provides effective rupture and drainage of the film. Droplet and phase interface interact in only one point at which the film has the minimum thickness. In that way the film breaks up more efficient.

Deformation of the droplet can be single or double. The deformation can appear only on the liquid interface but the droplet remains rigid, Figure 1b, or droplet deformation could occur leaving the liquid-liquid interface unchanged, Figure 1c. Also the droplet may be further deformed due to its own weight, Figure 1d. The double deformation includes both the liquid interface deformation, as well as droplet contact surface deform-

ation. The proper deformation of both surfaces forms a radial shaped film, Figure 1e, or a corrugated surface of droplets, Figure 1f.

The mentioned deformations are present and possible, either in the dispersed oil in water, or dispersed water in oil.
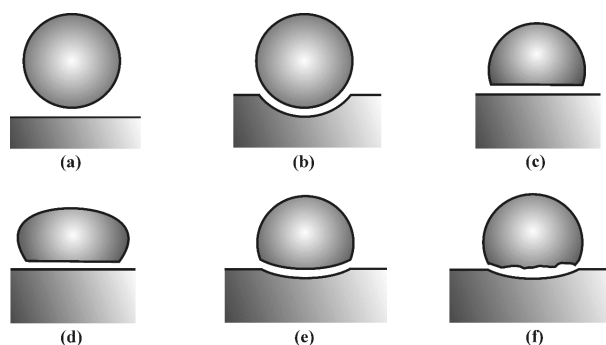

Figure 1. The coalescence models in the phase interface: a) without deformation, b) rigid sphere, c) rigid interface, d) droplet deformation due to own weight, e) proper deformation of both surfaces forms film with shape of bow, f) deformation of corrugated surface of the droplets.

Coalescence of the adjacent droplets

Coalescence of the adjacent droplets comprises of the following steps: mutual approach of droplets, formation of the continuous phase film between droplets, film rupture, drainage of the film, droplet–droplet coalescence and formation of enlarged droplets. Coalescence of the adjacent droplets may be complete or partial. Partial coalescence involves in addition to large droplet formation, the formation of a smaller droplet, secondary droplet, so-called the "daughter" [3,16].

Many authors acknowledge various forms of coalescence of the adjacent droplets and only a few will be specified: coalescence of the same size droplets, coalescence of different size droplets, the coalescence between large or coalescence of small droplets, as well as the coalescence without or with droplets deformation [3,5,16–24].

Ivanov *et al*. [16–24] investigated the interaction between two droplets of radius *a* having a narrow gap (thin film) between them. In this system the hydrodynamic and intermolecular forces are present. The hydrodynamic forces exist due to the viscous friction and they are usually in a rather long range. The interaction between two droplets in the emulsion favours the role of the following intermolecular forces: van der Waals, repulsive electrostatic double-layer forces, steric, etc. The authors claim that depending on the resultant of all the forces when two droplets collide, it may lead to droplet rebound, aggregation and coalescence. Aggregation is a process of the formation of droplet packets, aggregates, of several droplets that do not lose their individuality. If the resultant of all the forces is suffi-

cient to maintain the droplets in a small equilibrium distance, the aggregation will occur. These processes can take place through a number of mechanisms depending on the droplet size and the surfactant properties. The authors also state that the total coalescence time, being the drop lifetime; depend on the time required for the mutual approach of two droplets and rupture of the film.

During the droplet–droplet coalescence, the droplets can retain the spherical shape, initiating the coalescence without deformation having the model of rigid spheres, Figure 2a. This coalescence model can be attributed to droplets with small diameters. In other cases, during the coalescence between two droplets, deformations primarily occur in both droplets forming a film of different shapes: planar film, Figure 2b, and film model of large/small droplets, Figure 2c. Truncated droplets and the planar film can be formed in the zone of contact during the coalescence of large droplets. In the dispersed systems, where the emulsifier is present, a planar thin film is formed between the droplets. The formation of a thin film between the two droplets greatly enhances the role of surface forces and opposes the film drainage [3,16–24]. The emulsifier concentration in the deformed surface is non-uniform, and therefore the interaction forces are not of uniform intensity. The surfactants do not only affect the intermolecular interactions across the film, but, at the same time, favour the formation of the gradient of interfacial tension. Various surfactants amounts bond on the surface of droplets in a monolayer consequently forming the gradient of interfacial tension. This phenomenon occurs due to the surface non-uniformity of the droplet and is called the Marangoni effect. In this case the surfactant presence opposes the film drainage and reduces the velocity rate of the droplet approach.

Coalescence of irregularly shaped or crescent-shaped droplets, Figure 2e, is also considered to be one of the types of coalescence having significantly different interaction between these mentioned drop-

lets. During the deformation process, two droplets can come into contact. In these circumstances, multipoint surfaces of the droplets participate in the interaction. When smaller drops are present, considering that the interaction is at only one point, then the deformation does not occur and therefore the effect of surface non-uniformity is not dominant.

Due to the van der Waals attractive forces, droplet deformation could occur in the zone of their contact. Ivanov et al. [16–24] concluded that if the droplets diameter is larger than 80μm due to the action of the hydrodynamic pressure they twist towards the inside, leading to the formation of a flat film.

When the droplet diameter is less than 80 μm a different type of strain occurs. If the distance between the drops is $h = h_P$ the formation of bulges occurs. The bulge formation mechanism was suggested by Yanitsios and Davis [25] during the investigation of modeling the behavior of pure liquid emulsions without the presence of surfactants. Cristini *et al*. [26] found in their research on systems in the presence of surfactants, that effective and rapid coalescence takes place at a distance of $h < h_P$. The cause of the bulge formation is due to the disjoining pressure that rises faster than the hydrodynamic pressure when the distance reduction, h, occurs. At a distance of $h < h_P$ the bulges spontaneously grow until the drop contact and the coalescence starts. Modern techniques of high-speed video camera recorded this scenario of the bulge formation during coalescence between two droplets [27–30].

Danov *et al*. [16,20,21] investigated coalescence of different size droplets, with radius, R_1 and R_2 ($R_1 < R_2$). The film that was formed in these circumstances, in the zone of contact was curved with uniform thickness. The film length is R and thickness h. The given situation can be analyzed either thermodynamically and/or kinetically. In the thermodynamic approach due to the interaction between the droplets over the film, additional disjoining pressure occurs within the film. The disjoining pressure depends on the thickness of the film. Posi-
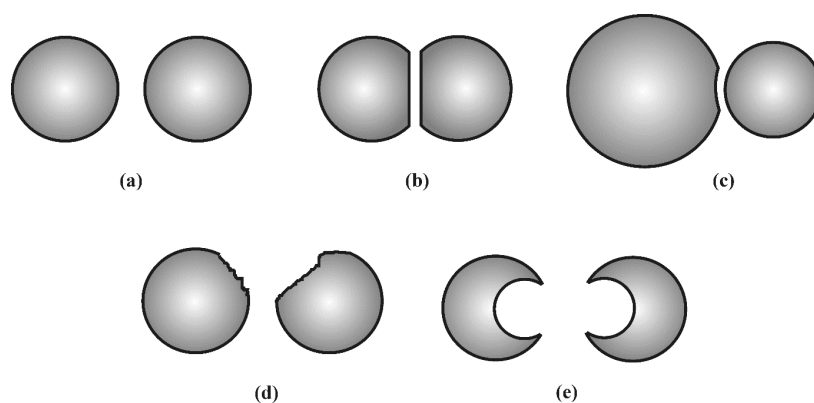

Figure 2. The droplet models when in mutual contact: a) model of rigid spheres, b) model of planar film, c) model of large/small droplets, d) irregular deformation, e) model of crescent-shaped droplets.

tive values of the pressure correspond to the repulsion of two droplets, while the negative values of the pressure indicate that the attractive forces are dominant. As a consequence of the disjoining pressure influence, the difference increases between the surface tension of the continuous phase, and interfacial tension of both liquids.

The film length increases with the increase of the contact angle. When the contact angle is zero, the equilibrium length of the film is also zero, and in such situation there is no possibility of the formation of equilibrium doublets, aggregates or coalescence. The disjoining pressure is the dominant thermodynamic factor that stabilizes the droplets and hinders coalescence. In addition to the mentioned thermodynamic factors, kinetic factors in the system may also occur. In this case, the effect of hydrodynamic forces must be taken into account. If the intensity is high enough, the hydrodynamic forces can cause film rupture before it reaches the thermodynamic equilibrium. During the contact of two droplets, with any disorder, including hydrodynamic, a capillary wave is generated on the surfaces of droplets. The resulting wave is usually formed in the upper and lower part of the film. If the amplitude of one or both waves becomes equal to the film thickness in any region, the film ruptures and the coalescence occurs [16,20,21].

The effect of hydrodynamic force can be reduced by the effect of kinetic stability. The most significant kinetic stabilization factors are: Gibbs surface elasticity, surface viscosity and surfactant adsorption relaxation time [20,21].

Gibbs surface elasticity tends to eliminate the gradient of the surface tension and suppress the fluctuations of the capillary waves. Surfactant and electrolyte concentration influence the mentioned elasticity. With the increase in their concentration the elasticity increases, which obstructs the coalescence. It should be noted that the electrolyte presence, may have a double effect on coalescence. An increase in electrolyte concentration may trigger an increase or decrease in the electrostatic barrier. This barrier increase may be caused by the increase of the film elasticity that hinders the coalescence. However, the barrier reduction may be the consequence of the reduction of electrical double-layer thickness that is more compact at higher concentration of ions, and therefore is less influential, which promotes the coalescence.

The film viscosity is often different from the viscosity of the continuous phase due to the local impacts, such as the capillary phenomenon and the surface roughness of the capillary wall. Therefore, the film viscosity is called the surface viscosity. Due to the dominant influence of the viscosity force in the capillaries, the behaviour of the fluid flow is different than in the

free volume of the liquid. Consequently, an increase in surface viscosity slows the drainage of the film down. If the surfactants are present and if there is film thickness deformation due to delamination, then there is an additional effect on the surface viscosity [20,21].

Mechanisms of film rupture

There are a number of mechanisms and models of film rupture. Most of the models are based on the assumption that the film is planar. Models that simulate the film drainage of the continuous phase starts from two droplets submerged in an incompressible viscous fluid.

Danov *et al.* claim that the film rupture can unfold through four main mechanisms: capillary-wave, film rupture by nucleation of pores, the solute transport across the film and the barrier mechanism [3,24]. Derjaguin published that Vries with his research group first proposed the capillary-wave mechanism [31,32]. This mechanism explains the rupture of the film under the influence of attractive forces. Since the space between the two droplets is narrow, it corresponds to the dimensions of the capillaries. The upper and lower capillary film surfaces oscillate as sinusoidal phase-shift. The capillary waves are generated due to the kinetic energy of the molecules. The mentioned phenomenon accelerates and facilitates the rupture of the film in several ways. The reduction of the film thickness is achieved due to the presence of capillary waves and disjoining pressure. The disjoining pressure enhances the oscillation amplitude. Increasing the oscillating amplitude of the capillary waves can lead to the film rupture when the critical thickness is reached.

The mechanisms of the film rupture by nucleation of pores have been investigated by Derjaguin [31,32]. The pores are created by breakage of the film due to the departure of the ruptured parts of the film. The resulting empty space between the ruptured parts the authors called pores. The liquid film is thereby torn into two parts being the left and right wall of the pores that could be also called the pore edges of the pore periphery. Ivanov is of the opinion that not every rupture will lead to the pore formation [16]. Pore formation is favoured by the decrease in surface energy, but it is opposed by the edge energy obtained during the formation.

Edge energy of the pores can be explained macroscopically and microscopically [16–21]. Macroscopic approach describes the edge energy of the pore as a line tension that has the tendency to reassemble the two parts of the film. The microscopic approach defines the edge energy as an effect of the spontaneous curvature and bending elasticity of the wall that does not have uniform distribution of surfactants that are present in the monolayer. This is defined as the Marangoni effect. For small ruptures or cracks of the film, the edge

energy is predominant, while for the larger ruptures the surface energy is dominant. Therefore, larger pores cause the rupture of the film enabling the spontaneous pore formation, while smaller pores shrink and disappear. Regarding the mechanism of film rupture due to the formation of numerous pores, the film can rupture triggering droplet formation. Due to the development of bulges from the dispersed droplets in the pore space, the newly formed droplets of the continuous phase are trapped and cannot be drained. In this way, the double emulsion can be formed.

Ivanov *et al.* [16–21] have investigated the rupture mechanism due the solute transport across the film. The solute transport across the film amplifies the Marangoni effect, and therefore the chemical entropy increases. Marangoni effect is favoured by simultaneous mass and heat transport.

The mechanism of the barriers results from the explanation of the equilibrium states of film thickness h_1. When the pressure is increased to the value higher than the barriers, then the film rupture can be caused, initiating coalescence or the transition to another stable state with defined film thickness h_2. The increase of the adsorbed surfactant stabilizes the secondary films. During the pressure increase, the accumulated mechanical energy is released overcoming the barrier. The lower the barrier energy is, the less chance of film rupture is present. If the barrier is not too high, the film rupture might not occur. Overcoming the barriers is usually achieved by reducing the film thickness in several places, rather than by a sudden decrease of the whole film thickness [16–21].

Droplet coalescence in the porous bed

As already mentioned, a number of forces coexist in the solid matrix. From the resultant of the dominating forces in the system, the drop can be either captured or detached. The hydrodynamic forces are present due to the fluid flow. Depending on the interstitial velocity, these forces can predominantly be viscous or inertial. If the hydrodynamic forces are low intensity then the retaining, capturing and agglomeration of the droplets in the porous bed primarily occur. On the contrary, when the intensity of hydrodynamic forces is higher, then the dispersed phase leaves the porous bed, and the droplets detachment or even re-dispersion could happen.

Mechanisms of drop capture can be classified into two main groups: transport mechanisms and mechanisms of attraction [33,34]. The transport mechanisms include: screening effect, interception, sedimentation, inertia, diffusion, hydrodynamic forces and motion due to temperature gradients. Mechanisms of attraction forces include electrostatic interactions, van der Waals forces, adhesion and mutual adsorption.

Hubbe, Ryan and Elimelech with coworkers [35–41] investigated the detachment of colloidal particles/ /droplets from solid surfaces influenced by shear stress, Figure 3. It should be emphasized that the particles/droplets are exposed to the adhesive force and the hydrodynamic forces. Hydrodynamic forces consist of two components, horizontal and vertical. The horizontal component is the viscous force acting parallel to the surface where colloidal particles are situated, while the vertical component is the lifting force. Usually the intensity of the horizontal component is greater than of the lifting force. The horizontal component has a significant impact on the moment when colloidal particle/ /droplet will detach from the solid surface. The rotation and rolling of the drops appear due to the influence of the torque force. Hubbe claims that the drop detachment predominantly depends on its size and shear stress.
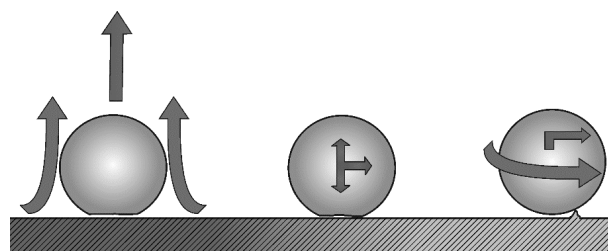

Figure 3. Schematic view of forces that influence the droplet trapped on a solid surface.

According to Spielman, there are three forms of the dispersed phase in the pores of the bed: capillary-conducted phase, globules and small droplets [42–46]. Capillary-conducted phase is formed quickly in the bed. Only this form of the dispersed phase influences the pressure drop during the flow of both phases through the bed. The amount of capillary-conducted phase in the pores of the bed is predominantly determined by its geometry, the volume of interconnected pores and the intensity of hydrodynamic and adhesion forces. Emulsion separation by bed coalescence can occur in two conditions: unsteady-state and steady-state conditions. The flow of relatively unstable emulsions through the bed initiates increasing filling of the pores in the bed by the dispersed liquid phase, resulting in a permanent rise of the pressure drop with time. Due to the accumulation of the dispersed phase in the pores of the bed, there is a cross-section reduction of flow capillaries, causing an actual increase of the fluid velocity and the flow resistance, by which the bed permeability reduction is achieved. In the case of deep bed filtration the amount of capillary-conducted phase changes in the pore volume, increasing quantity of the disperse phase. This regime is unsteady-state due to the change of pressure drop with time [47]. The filter cycle in the case of deep bed filtration ends when reaching the

maximum allowable pressure drop or when penetration of the dispersed phase through the bed is achieved. The filtration ends when the bed cannot accumulate any new amount of the dispersed phase and at this point the washing could begin. During the bed washing additional oily wastewater is formed due to which this regime seem unfavourable for the liquid–liquid separation. A more appealing application of the bed coalescence for the liquid-liquid separation is the steady-state regime [2,3]. Fluid velocity determines the intensity of the hydrodynamic forces, and the quantity of a capillary-conducted phase in the pores of the bed. According to Spielman the maximum amount of this phase is 30% of the pore volume, while the coalescence on its surface already begins at the amount of 10% of the pore volume. Simultaneously, new droplets of the disperse phase enter the bed, while the other droplets//globules leave the porous bed. Depending on the achieved conditions of the resultant forces, larger or smaller droplets than the ones entering will leave the porous bed. If larger droplets leave the bed, then the goal is reached. These droplets will be separated by sedimentation behind the porous bed, in the settling section, Figure 4.
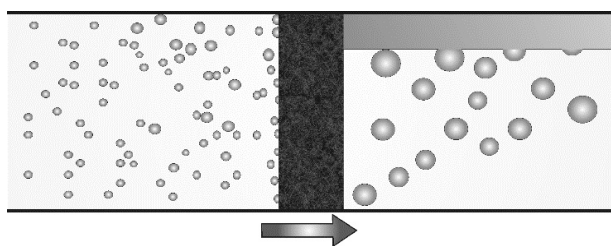

Figure 4. Operating scheme of bed coalescer.

The steady-state regime is achieved when the constant pressure drop is established, meaning that the pressure drop does not change with time anymore. Steady-state regime exists in an indefinite period of time. The steady-state regime eliminates filter cycle and bed washing requirements. Coalescence of droplets that enter into the bed occurs on the surface of the capillary-conducted phase, leading to their detachment and emerging from the bed as enlarged droplets.

Three coalescence mechanisms can occur in the porous bed: coalescence of the droplets on the fibre surface, coalescence of the adjacent droplets in the pore space and coalescence of the droplets on the surface of the capillary-conducted disperse phase, saturated liquid [3,42–46]. With the coalescence of droplets on the surface of the solid material the formation of capillary-conducted phase in pores is obtained. Coalescence in the capillary-conducted phase is similar to coalescence at the liquid–liquid interface. All previous discussion about the droplet–droplet coalescence also applies to their mutual coalescence in the volume of

the pores. Mutual drop coalescence in the pores may be enhanced by the bed properties that influence the distance between drops and hence favouring this type of coalescence [3,5,9,48–54].

DESIGN VARIABLES OF BED COALESCER

The porous bed that exists in the practice may have drastically different properties [3,5,46–49,55]. The two bed properties that mostly differ are: bed porosity and bed length. The bed porosity can be from close to zero to up 98%. Such a wide range of bed porosity inevitably influences different behaviours of disperse and continuous phase flows and droplets coalescence. Granular bed has a porosity interval of up to around 50%, while the fibre beds are able to form a high-porosity bed [47].

The bed length also can be found in two extreme situations, being the finite and infinite bed length. The finite bed length can be defined as expressively low bed length ranging from a few millimetres to a few centimetres, which corresponds to the membrane, while high bed length can range from half meter to several meters. The porous bed with infinite length is the petroleum reservoir. Taking into account that gas, oil and water are exploited from the earth depth, the phenomena for this porous bed are extremely important because it significantly influences the global economy [3].

Bed can be formed from natural or artificial materials, inorganic or organic. When the bed is formed out of polymeric fibres, it has the ability of such a compression that the bed can have completely different properties, although formed out of the same polymer fibres [48,49,55,56].

Selection of the filter media, as well as its packing conditions is essential for design of bed coalescer. When designing the steady-state bed coalescer it is necessary to determine the ratio of droplet size to pore size. If it is possible, the conditions should be chosen in such a way that the pore size is greater than the size of droplets. In most cases, the mentioned relation is usually achieved. In these circumstances the basic preconditions are provided, enabling the formation of the capillary-conducted phase in the pores of the bed where the coalescence of the newly arrived droplets is realized.

Šećerov Sokolović *et al.* published several papers on the subject of the design of steady-state bed coalescer [2,48,56–65]. Common design targets were small unit with low energy spending. For high working velocity, size of coalescer is small, but at the same time consumption of energy increases. Energy consumption and pressure drop can be modified with variations in bed properties. By reducing the size of the coalescer, its weight is also reduced. Therefore, less structural material for the coalescer production, less material for foundation, as well as less space for its accommodation

are needed. These specifics are of particular importance on ships and oil platforms where the carrying capacity and space are drastically limited.

Bed coalescers can be installed in the industry to perform separation of oil from water in two different situations. One is to separate same oil during its lifetime, and the other with the task to separate different oils during time [65]. The same oil is separated by the coalescer in the oil fields and on ships. These coalescers can be designed on the basis of experiments on the model emulsion of this oil and optimal working conditions can be selected. In petroleum refineries and petrochemical industry coalescers, as part of a central facility for wastewater treatment, are able to separate oils of different quality over time. The coalescer design for such circumstances is the most complex.

Many factors affect steady-state bed coalescence and separation efficiency: fluid velocity, bed properties, fibre properties, properties of liquids, surface phenomena, set-up of the unit and interdependences of many variables.

Fluid velocity

Numerous authors have investigated the role and importance of the fluid velocity related to the coalescence and the flow of heterogeneous systems through the porous media [42–46,66–77]. Very early in the investigation the scientists observed that there is a limit value of the fluid velocity at which the behaviour of the system changes. According to available literature, this velocity, was first mentioned by Voyutskii *et al.* and was called the critical velocity [66]. Evidently, the interpretation of the critical velocity depends on whether the researchers focused on unsteady-state or the steady-state regime. In the first one the critical velocity is the velocity below which a complete retention of the dispersed phase in the pores of the bed is achieved.

Sareen *et al.* defined the critical velocity as the velocity at which the re-dispersion of the dispersed phase in the bed occurs. Therefore the droplets leaving the bed have smaller diameters than the droplets that enter the bed and cannot be separated by settling. For fluid velocity below the critical, the complete coalescence is achieved of large output droplets that can be easily separated. It is more than obvious that the coalescence in this case is in the steady-state regime [67].

Spielman studied the effect of the fluid velocity on the flow of two immiscible fluids through the porous bed [42–46]. The investigation of fluid velocity focused on the filtration coefficient, the total pressure drop and the amount of the capillary-conducted phase in the bed. His experiments covered a region of critical velocity, but the author did not mention, nor analyse or define this value.

Rosenfeld gave numerical values of critical velocity for Spellman's experiments [68]. He defined the critical velocity as the velocity at which the filtration coefficient begins to rapidly decrease, Figure 5. According to Rosenfeld, the critical velocity is the velocity at which the hydrodynamic forces become dominant compared to the other forces, leading to the detachment of a part of the dispersed phase. Rosenfeld studied the flow through the interior of reservoirs and the following phenomenon was observed. The flow of the fluid carries the dispersed phase further through the bed causing the clogging of pore channels and the decrease of bed permeability. Additional increase of the velocity leads to the increase of the dispersed phase amount that is detached moving in the direction of the liquid flow. In contrast, if the bed length is low, then the detached dispersed phase emerges from the bed, increasing the bed permeability due to the decrease of the capillary-conducted phase amount in the pores.
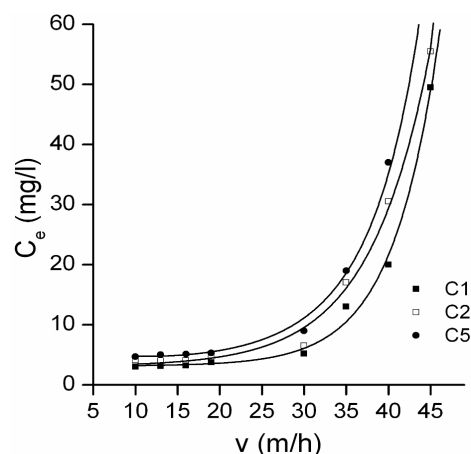

Figure 5. Illustration of the existence of the critical velocity.

One of the key information essential for investigation is whether the fluid velocity below or above the critical value. Considering that this information is not provided in numerous cases, incorrect interpretations could occur by comparing the dependencies that exist above and below the critical velocity. Sometimes, the working conditions that are selected provide an extremely low critical velocity, leading to a misconception that the investigated area cannot be above the critical velocity.

Grilc, Golob and Modić published a series of papers on the effect of fluid velocity on coalescence and investigated the critical velocity as the phenomena [69–72]. These authors studied both water-in-oil and oil-in-water emulsions and the influence of following parameters: bed length, interfacial tension and pH on the critical velocity and modified Spielman adhesive number. Authors modified the Spielman equation for dimensionless filtration coefficient and adhesive num-

ber by introducing the critical velocity. Furthermore, the critical velocity was defined as the velocity at which the effluent concentration of the dispersed phase is no longer constant and begins to rise exponentially.

Soo and Radke studied the influence of the fluid velocity on the flow of the oil-in-water emulsion through bed of sand, simulating the conditions in the underground collectors. Unlike other authors, their research focused on the events that take place in the bed for diluted, stable emulsions with the dominant retention mechanisms, such as filtration and interception [73––76]. During the flow of a stable emulsion through the bed, each single drop is captured on the surface of the filter media, but there is no coalescence between the droplets, or formation of capillary-conducted disperse phase that was reported by Spielman. These authors define the critical velocity as the velocity above which the capturing possibility of the droplets is zero, meaning that there is no capturing of new drops, initiating the detachment of the previously "captured" ones.

Maini *et al.* emphasized the importance of the fluid velocity during the investigation of suspended particles migration in the oil fields [77]. In the case of the suspension flow the term critical velocity also occurs. They noted that the permeability decrease due to the particle migration occurs only when the fluid velocity becomes higher than the critical velocity. Low velocity initiates the flow of a small number of particles, which move towards the collector, one by one. At high fluid velocity a larger number of particles are torn from the surface of the reservoir rock, and collide with each other, interact and migrate in another way.

Ryan and Elimelech studying the transport of colloids in the groundwater also accepted the concept of critical velocity [37]. In theory, the mobilization of colloids by physical disturbance is based on the torque moment, which affects the particles that are stored on a solid surface, and is the result of fluid flow that initiates the particle detachment. The torque moment, which influences the detached particles, includes the adhesive force operating normal to the solid surface, the drag force operating tangential to the surface, and lift force operating normal to the solid surface.

Šećerov Sokolović *et al.* studied the importance of the influent concentration, bed length and fluid velocity on the effluent concentration for the system of oil-in-water emulsion and polyurethane bed [57]. In this paper the importance of the critical velocity as a boundary of behaviour of the system was underlined. The effect of the influent concentration and bed length in the observed range on the effluent concentration is negligible for the fluid velocity below the critical; while for the velocity above the critical this effect is significant. According to these authors, critical velocity is a key variable for designing the coalescers, by which the

capacity and dimensions of the bed coalescer are determined. The fluid velocity need to be high, but below the critical value for the given system. The critical velocity can be determined from the experimental data concerning the dependence of the effluent concentration on the fluid velocity. The authors proposed that the critical fluid velocity should be the velocity at which the effluent oil concentration reaches a value of 15 mg/l, which is often the recommended limit of oil concentration in wastewater, Figure 6.
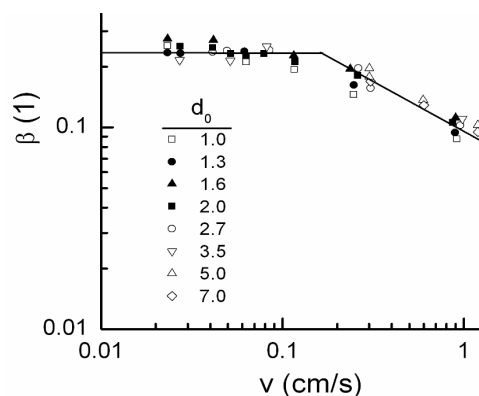

Figure 6. Dependence of effluent concentration on fluid velocity for polyurethane bed length 15 cm over influent oil concentration: C_1 = 500 mg/l, C_2 = 800 mg/l and C_5 = 2000 mg/l.

Šećerov Sokolović *et al.* in all of their later published work applied the critical velocity as a key variable for the investigation of the influence of many factors.

Researchers are still investigating the influence of fluid velocity using other parameters, such as efficiency, quality factors and etc. They also concluded that over lower flow velocity droplet separation performance is better than over higher flow velocity in all investigated circumstances [78,79].

Fluid flow orientation-flow mode

Fluid flow orientation can be horizontal (H), vertical down (VD) and vertical up (VG). Most frequent type of fluid flow mode for liquid-liquid separation is vertical down that has probably been taken from deep bed filtration. Investigation of fibre bed coalescence phenomenon often neglects fluid flow orientation. However, in term of coalescence efficiency the fluid flow mode in relation with the fibre bed coalescence has additional significance [3,59,60,78,79].

Šećerov Sokolović *et al.* in earlier stages of their research spotted the importance of the flow mode on the coalescence efficiency [59,60]. In their comprehensive research, as well as in their equipment development activities, they investigated the possibilities for increasing the coalescence efficiency.

Šećerov Sokolović *et al.* experimentally determined that the separation efficiency of steady-state bed coalescer is highly influenced by the flow mode. The flow of the dispersion in through the filter media and coalescence efficiency are conditioned by the balance of acting forces. This balance is mainly determined by the density of the dispersed phase, and fluid flow mode. The dominant forces that are of influence in the porous bed are: gravity, F_g, hydrodynamic forces, F_h, buoyant force, F_p, and adhesion, F_a. The adhesion is present in the pore space in all directions equally, having a short radius of action, and for this investigation can be ignored when the same filter media is used.

For H fluid flow mode the forces act at two planes, in the direction of *x*- and *z*-axis. Under these circumstances, it is defined as a two-dimensional system. The gravity force, F_g, and the buoyant force, F_p, act opposite to each other in the direction of the *z*-axis. Their resulting force of the higher density dispersed phase than the continuous phase will be directed downward, whereas lower density dispersed phase will be directed upwards. The hydrodynamic force, F_h, is oriented in the direction of the *x*-axis, and is at an angle of 90° when compared to the resultant force of F_p and F_g. The resulting force of these three forces is the diagonal of a parallelogram with the direction determined by the density ratio of existing liquids. Such orientation of the resulting force promotes settling of droplets leaving the bed, which is desirable.

The fluid flow mode VD and VG differ to each other in direction and intensity of the observed resulting force. If the dispersed phase has lower density than the continuous phase in case of VD flow mode, the hydrodynamic and gravity forces are added, and the buoyant force reduces this total. For VG flow mode, the hydrodynamic force is added to the buoyant force and the gravity force reduces the intensity of resulting force.

As previously emphasized, the critical velocity should be regarded as the maximum possible fluid velocity of the device. In the experiments of Šećerov Sokolović *et al.* the highest critical velocity was realized in the H flow mode, for all observed bed lengths and bed permeability. The value of the critical velocity varies up to 100% for different conditions.

Šećerov Sokolović *et al.* analyzed the separation efficiency of two commercial coalescers with different geometry marked as "04" and "H" [60]. Experiments were carried out over a wide range of oil properties and fluid velocities. Operation of coalescer "04" was characterized by an extremely low working velocity. This coalescer involved both vertical flow modes (VG and VD), while coalescer "H" had a horizontal flow mode.

This phenomenon can be explained by the amount of the capillary-conducted phase in the bed for the steady-state operating conditions of the coalescer. For the H flow mode the amount of the capillary-conducted phase was the highest in relation to the other two flow modes. In the H flow mode high amounts of the dispersed phase retained in the bed, since only the hydrodynamic force tends to drain out the part of dispersed phase pushing it outside the bed. The other two forces, being vertically oriented, tend to retain the dispersed phase in the bed.

The most unfavourable situation is when vertical up, VG, flow mode existed. In these circumstances, the hydrodynamic force tends to push the dispersed phase from the bed. When the dispersed oil is less dense than water, quantity of the saturated oil in the pores is low and the contact time is insufficient for the efficient coalescence. Published results confirmed that separation efficiency is the lowest for vertical up flow mode [59].

The study of Vigneaux *et al.* observed the flow of the emulsion through inclined pipes and presented the distribution of the dispersed phase through the cross-section of the pipe, confirming the above explanation, Figure 7 [80]. Based on the mentioned analysis it can also be pointed out that the amount of the capillary-conducted phase in the bed with the H flow mode is not uniform over the cross-section of the pipes. When the dispersed phase is of lower density than the continuous phase, then the upper parts of the bed is enriched by it. On the contrary, when the dispersed phase has higher density than the continuous phase, then higher amounts of the capillary-conducted phase is present in the lower part of the bed. It could be concluded that the amount of dispersed liquid phase, as well as the uniformity of its distribution in the bed volume, is a function of the flow mode.

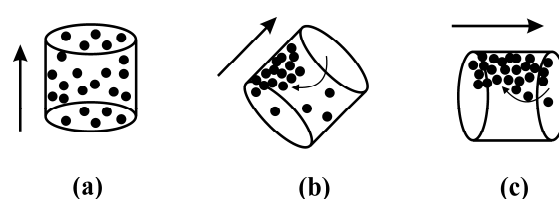**(a)**   **(b)**   **(c)**

Figure 7. Distribution of the dispersed oil in pipes with different orientations: a) vertical pipe, b) slightly tilted pipe and c) considerably tilted pipe.

The literature search shows that only one research group have addressed the problem of fluid flow mode. Burganos *et al.* investigated this problem but in the domain of deep bed filtration for a solid-liquid system. On the basis of their simulation, the importance of the flow mode in this operation was explicitly pointed out [81].

The fibre bed geometry

The fibre bed geometry is defined by two sets of properties: fibre properties and bed properties. The

fibre properties are considered to be: the fibre diameter, fibre length, shape of cross-section, tortuosity, interconnected fibre structure and surface roughness. The bed properties are: porosity, bed permeability, bed length, shape and tortuosity of pores, pore size, the fibre orientation, active surface area and the packing density of fibres.

The fundamental problem for defining the bed geometry is the absence of a representative characteristic that give the overall description. Although this is an important feature, most authors do not give enough data on the bed geometry and thereby the bed properties for the conducted investigation are not precise. The authors usually give the bed length, sometimes the fibre diameter, but very rarely bed permeability and/or bed porosity. The most investigated effects in the literature were the fibre diameter and the bed length. Most authors who have studied the influence of the bed length claim that reducing the fibre diameter increases the separation efficiency [82–85], while Dalquist and Setterwall noted that with the reduction in the fibre diameter the bed length could be reduces [86]. Golob *et al.* pointed out that the required fibre diameter is determined by the droplet size of the dispersed phase and that these two values should be close to each other [72].

Investigation of fibre diameter effect is usually irregular because there is no report whether the changes in fibre diameter in the same time initiated the variation of the solid phase ratio. The bed which Hazlett used in the study consisted of several segments of glass fibres with different diameters [82–84]. The author used glass fibres of smaller diameter (0.75 and 1 μm) and glass fibres with larger diameter (2.8 and 4.6 μm). Fluid flow mode was vertically down, VD. The bed was formed in such a way that on the top were usually segments of thinner fibre, followed by the segments of

thick fibres. The author detected that the stacking order of the segments with different fibre thickness affects the coalescence efficiency. The fibre diameter at the bed exit has a predominant influence on the dimensions of detached droplets. The fibres of smaller diameters at the bed exit may reduce the coalescence efficiency, and therefore thicker fibres are suggested to be on the bed exit. If the fibre diameter of the last segment is not well defined, all the positive effects of the droplet capture and their aggregation can be annulled.

Daiminger *et al.* introduced a microporous membrane of nano fibres with low thickness in front of the polypropylene bed, having vertically upward, VG, fluid flow mode, in order to enable enlargement of the smallest droplets, and thus improve the separation of the dispersed oil and allow higher fluid velocity [87].

Šećerov Sokolović *et al.* [56] reported that the porosity dependence on the bed permeability and fibre diameter completely determined the fibre bed geometry. In addition, the authors indicated that the existence of the critical bed permeability, below which the system becomes unstable. Authors have pointed out that the operation range in terms of the permeability is suggested to be above the critical value, Figure 8. The authors also claimed that it is necessary to maintain the same bed geometry when investigating other effects such as: bed length, the droplet size, the properties of the filter media, the properties of the dispersed phase, and etc. The authors noted that only comparable results are those that have been realized in the same fluid flow mode and the same bed permeability higher than the critical one.

Bed length

Bed length is one of the most important design variables in bed coalescence. Sareen and Hazlett both investigated bed coalescence of water-in-oil emulsion
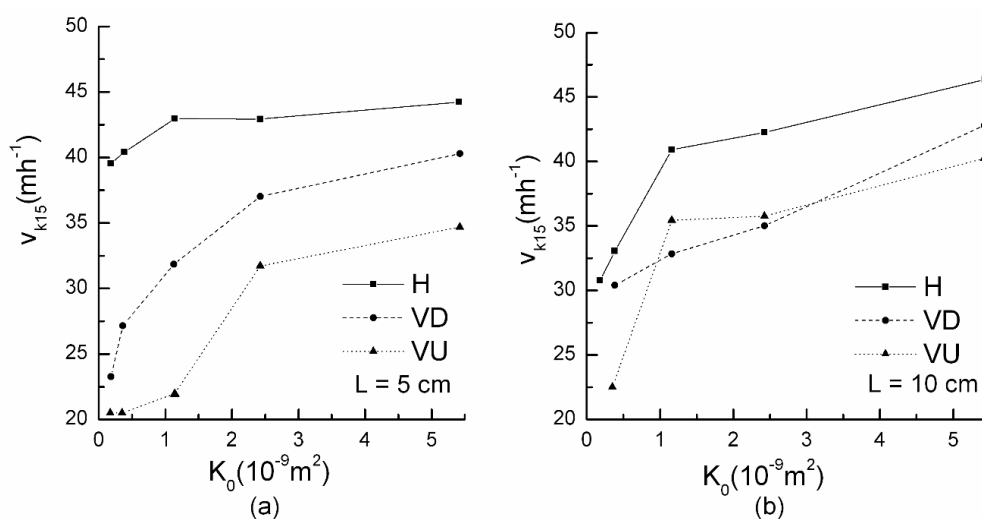

Figure 8. Dependence of critical velocity on bed permeability for all flow modes for the bed length of: a) 5 and b) 10 cm.

[67,82–84]. In connection with bed length Sareen concluded that there is an optimal bed length which enables maximal value of the critical velocity [67]. Hazlett thought that the bed length, above the minimal value, has no influence on coalescence efficiency [82––84]. It is important to point out that investigations of both researchers were conducted in a very narrow range of bed length around its minimal value, between 0.5 and 3.2 cm.

Fahim and Akbar investigated the effect of a wide range of bed lengths, from 10 to 40 cm, on separation efficiency of oil from wastewater, over a wide range of fluid velocity, using high surface energy fibre materials at vertical up flow mode [88]. They concluded that separation efficiency is very high for all bed lengths. The authors observed that drop diameter decreases with increase in fibre bed length. At higher velocity and relatively high bed lengths, the droplets enlarged by coalescence can be re-dispersed while passing through the fibre bed.

Deshamps *et al*. [89] explored the possibility of separation of dispersed vegetable oil from water. The bed was formed out of cotton fibres and the bed length was varied from 9 to 12 cm. The authors claimed that with the increase of the bed length, the coalescence efficiency also increases.

In general, it could be concluded that with the increase of the bed length the coalescence efficiency increases. Magiera and Blass compared experimental data with the simulation data and they also concluded that longer fibre beds have better separation efficiency [85]. High and low surface energy fibre materials and three bed lengths over two fluid velocities were included in this investigation.

Šećerov Sokolović *et al*. established that the effect of bed length on the bed coalescence is dominantly determined by the applied range of fluid velocity over the investigated bed length (3–15 cm). The effect of the bed length on the separation efficiency is negligible for the fluid velocity below critical. When the fluid velocity exceeds the critical value, the bed length exhibits an unexpected effect on effluent oil concentration [61,65].

Shin and Chase, on the basis of two glass micro-nanofibre bed length (3 and 5 mm), over horizontal flow mode concluded that the capture efficiency and quality factor were higher for the 5mm bed, but the overall coalescence efficiency decreased with the increase in the bed length applied [89,90].

Šećerov Sokolović *et al*. explained all aforementioned observations analyzing interdependences between the bed lengths, flow mode and bed permeability [61]. The effect of the bed length on the critical velocity was monitored. Critical velocity increases with the increase in bed length, ranging from 3 to 10 cm, over all three flow modes (H, VD and VG). These results

were obtained at the maximal bed permeability, 5.39×10^{-9} mm^2. This dependence under the applied circumstances is expected, because at high bed permeability and the maximum bed length, highest pore size, pore volume and contact time were obtained. Corresponding to this, the amount of the capillary-conducted phase is maximal, and the droplet coalescence on its surface is optimal. Dependence of the critical velocity on bed length is similar over all three flow modes, but numerical values are very different as a consequence of resulting forces which are acting in the system, Figure 9.
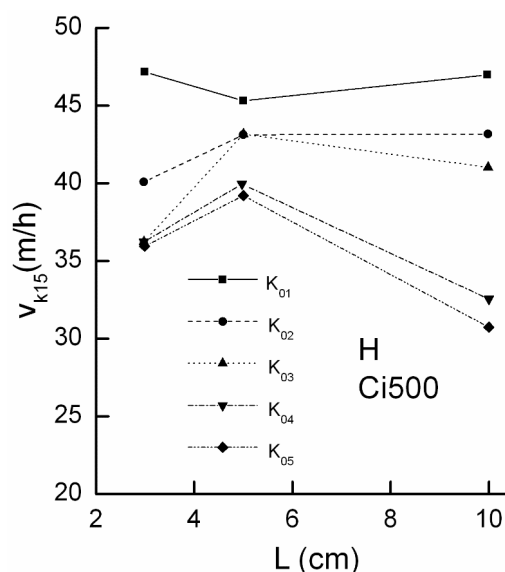
Figure 9. Illustration of existence of the optimal bed length.

Šećerov Sokolović *et al*. also recognized the presence of the minimal bed length in the steady-state bed coalescence [61]. This property could be defined as the critical bed length. The effect of the bed length above the critical value on steady-state bed coalescence is dominantly determined by bed permeability and influent oil concentration, independent on the flow mode. At low bed permeability and with H and VD flow mode, the authors detected the existence of the optimal bed length. Under the applied experimental conditions this optimal value of the bed length was 5 cm for polyurethane fibre bed.

DISCUSSION AND OUTLOOK

Up till now, there has been no systematic approach to the problem of the complex bed coalescence phenomena. Experimental conditions differ from study to study over different bed properties and fluid flow orientations. Very often, the chosen conditions do not allow studying the desired phenomenon because of the simultaneous influence of several variables. In these

circumstances it is difficult to set up any general conclusions. On the other hand, the existing knowledge of coalescence mechanisms, mechanisms of film rupture and flow of two immiscible liquids through porous bed is still not adequately connected to each other. Limitation of current experience about phenomena on the liquid–liquid–solid interface also prevents further progress of fibre bed coalescer design.

Acknowledgment

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IZVOD

Separacija tečno–tečno primenom koalescentnog filtera u stacionarnom stanju

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(Pregledni rad)

Ovaj rad daje literturni pregled postojećeg znanja i razumevanja separacije tečno–tečno, koja je izuzetno rasprostranjena u praksi, ističući da je koalescer koji radi u stacionarnom stanju odlično rešenje za ovakvu separaciju. Koalescentna filtracija je metoda koja se pokazala kao izizetno efikasna separaciona tehnika u industriji. Zauljene vode zastupljene su u čitavoj procesnoj industriji. Voda je često rashladni fluid. U izmenjivačima toplote može da dođe do proboja organske faze koja se hladi i da voda bude kontaminirana. Sve pumpe i kompresori koriste ulja za podmazivanje. Zbog lošek zaptivanja, hlađenjem, pranjem i održavanjem kako te opreme tako i prostora gde je ona smeštena nastaju zauljene vode. Održavanje vozila i teških mašina takođe uzrokuju formiranje značajnih količina zauljenih otpadnih voda. Posebne operacije koje koriste ulja ili druge organske rastvarače kao što su solventna ekstrakcija, kaljenje, i slično izvor su nastajanja zauljenih otpadnih voda. Kod proizvodnje nafte postoji potreba separacije sistema tečno-tečno i to kako vode od nafte tako i nafte od ležišne vode. U procesima naftno-petrohemijske industrije koalescer sa slojem se sve češće primenjuje ne samo za tretpan otpadnih tokova nego i u procesima proizvodnje. Zbog složenosti fenomena koji se odigravaju tokom ove separacije projektovanje koalescera još uvek zahteva eksperimente. Ovaj literaturni pregled oslikava ključne fenomene koji uslovljavaju efikasnu separaciju kapi dispergovane faze kao što su koalescencija na granici faza, koalescencija između kapi, koalescencija u poroznom sloju, kao i proticanje emulzije kroz porozni sloj. Pored toga dat je pregled postojećeg znanja projektovanja koalescera. Opisane su najvažnije projektne veličine: brzina fluida, geometrija sloja, debljina sloja i orijentacija toka fluida kao do sada proučavane veličine. Zbog mogućih međusobnih zavisnosti i interakcja, površinskih pojava koje se događaju na granici faza tečno-tečno-čvrsto i dalje su neophodni eksperimenti za dimenzionisanje uređaja, odabiranje materijala za sloj i usvajanje radnih uslova.