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Integrated Process for the Removal of Emulsified Oils from Effluents in the Steel Industry

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ABSTRACT

Emulsified oils contained in aqueous effluents from cold-rolling mills of the steel industry can be effectively removed via an integrated process consisting of a coagulation/flocculation stage followed by ultrafiltration of the resulting aqueous phase. The effects of $CaCl_2$, NaOH, and lime on the stability of different industrial effluents were studied in the coagulation experiments. The flocculants tested were inorganic prehydrolyzed aluminum salts and quaternary polyamines. Ultrafiltration of the aqueous phase from the coagulation/flocculation stage was carried out in a stirred cell using Amicon PM30 and XM300 organic membranes. Permeate fluxes were measured for industrial effluents to which the indicated coagulants and flocculants had been added. Oil concentrations in the permeate were 75% lower than the limits established by all European Union countries. Complete regeneration of the membrane was accomplished with an aqueous solution of a commercial detergent.

Key Words. Coagulation; Emulsified oil; Flocculation; Rolling mill; Steel industry; Ultrafiltration.

INTRODUCTION

Petrochemical and metallurgical plants (e.g., rolling mills and mechanical fabrication facilities) generate wastewaters containing excessive levels of oil

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and grease that must be treated before disposal. Emulsified oils are used for two main purposes in metal processing operations: as lubricants to reduce friction between the metal and mechanical equipment, and as coolants to remove friction-generated heat which may be sufficient to cause contact-welding of metal parts.

Over time these emulsions become less effective because of thermal degradation of the oils and accumulation of suspended solids (1). Hence the spent emulsions need to be replaced periodically, and the resulting effluents must be treated before disposal.

In the European Union, regulatory restrictions for the discharge of wastewaters have become more stringent in recent years. At present, treatment of these oily effluents consists mainly of removing the oily phase by physical treatment or by flocculation. However, such processes are no longer able to meet statutory requirements, and further treatment is necessary.

The treatment of wastewaters containing emulsified oils can be accomplished by a two-stage process. The first stage, i.e., the pretreatment stage, consists of breaking up the emulsion, followed by gravity settling and mechanical separation of the oil. The oily emulsion can be destabilized by *coagulation*, adding electrolytes which lower the zeta potential of suspended materials (particles or droplets), or by *flocculation*, which promotes aggregation of small oil droplets into larger drops via addition of polymers of high molecular weight (2). The second stage consists of ultrafiltration (UF) of the aqueous phase resulting from the pretreatment sequence of operations.

Several publications based on laboratory or pilot-scale data describe treatment of oily wastewaters by coagulation (3) and flocculation, either with cationic polymers (4), anionic polymers (5), or dual polymers (6). Ultrafiltration has also been used to treat wastewaters containing oil and grease (7–10). The reduction in volume resulting from ultrafiltration of a typical emulsion of waste-cutting oils in water is of the order of 94–99% (7), while the associated reduction in COD is ca. 90–98% (8). Typical oil rejections are 97–98% (9, 10). However, in spite of the high separation efficiency of UF membranes for the treatment of oily emulsions, their use in industrial applications is limited by the rapid fouling observed for most polymeric and ceramic membranes.

The present investigation is focused on a proposed two-stage process for treatment of emulsified oils. The process consists of either a coagulation or flocculation stage, with mechanical removal of the oil layer, followed by treatment of the resulting water phase by ultrafiltration. Data are reported below for application of this sequence in removal of emulsified oils from wastewaters produced by cold rolling mills in processing steel.

CHARACTERISTICS OF OILY WASTES

The oily waste streams of interest are effluents produced at the cold-rolling mills of the Aceralia plant (Avilés, Asturias), the largest iron and steel company in Spain. This factory has two cold-rolling mills, named Tandem I and Tandem II. Each of these mills utilizes the indicated product as a lubricant and coolant:

- Tandem I: *Quakerol*, a commercial water miscible oil. As employed by industry, this product is emulsified in demineralized water (7–10 vol% Quakerol in water). When this emulsion loses its effectiveness, it must be replaced. Consequently, one obtains an emulsified oily waste which must be properly treated before it can be discharged to the environment.
- Tandem II: *Tinol*, an animal fat which is solid at temperatures below 30°C. In industrial use this material is mixed with demineralized water (6–12 vol% Tinol in water). The resulting suspension serves as a lubricant and coolant for the rolling mills. Tandem II continuously generates an effluent containing Tinol and other free oils.

Associated with the Tandem II rolling mill are three tanks, but only one is in operation at any time. The water in the tank contains Tinol which is used in a closed circuit mode. A supply of 1000 m^3/day of water flows to the system. Each tank contains a skimmer to remove fats and foams. A constant overflow stream, containing suspended particles of Tinol, is mixed with the products removed by the skimmers, and collected in an external tank. Part of the Tinol in the external tank is removed by flotation, but most of it is disposed as a stream which is referred to as *regular effluent*. Tandem II is shut down for maintenance every 15 days. The waste resulting from the cleaning and maintenance operations is referred to as *washing effluent*. The temperatures of both the regular and washing effluents are approximately 60°C.

The Quakerol emulsion employed in Tandem I is replaced from time to time. This waste stream is pumped to the outer tank where it is mixed with the *regular effluent*.

A flow diagram of this process is shown in Fig. 1. Data relating to the quality of the effluents are shown in Table 1.

EXPERIMENTAL

Equipment, Materials, and Methods

Flocculation and coagulation tests were conducted with a jar-test apparatus equipped with four 800 cm^3 beakers. The mixing procedure for the flocculation experiments consisted of 3 minutes of stirring at 160 rpm to bring about the initial dispersion, followed by 30 minutes of mild stirring at 16 rpm. The dispersion was then allowed to settle for 60 minutes. The temperature of the solutions was 60°C for all tests.

The flocculants/demulsifiers used in the jar tests were:

- Inorganic flocculants (*Acideka*): DK-1014 and DK-1018. Both flocculants are prehydrolyzed aluminum salts (PACl).

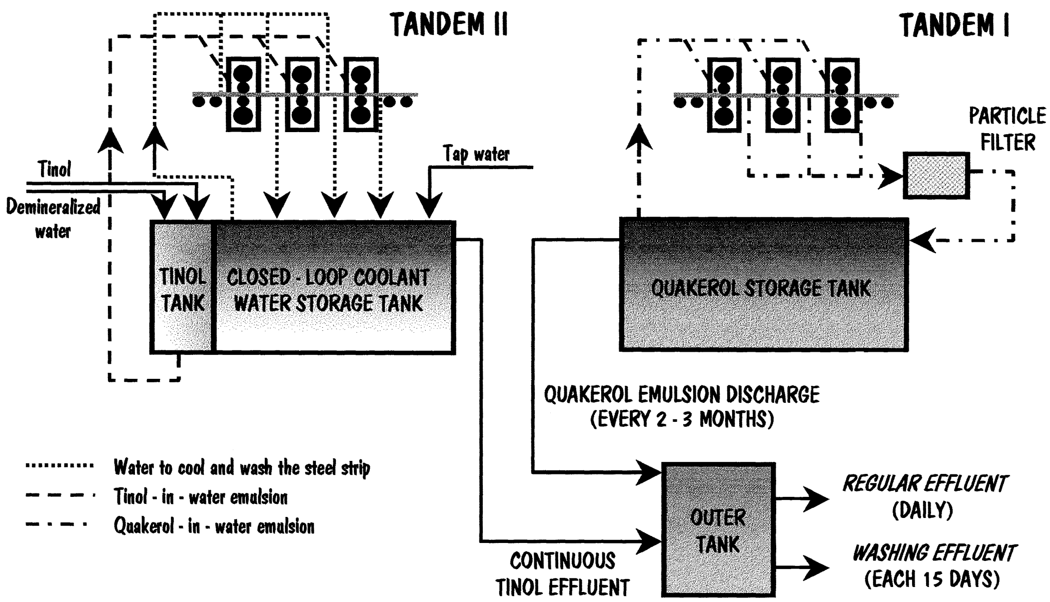

FIG. 1 Industrial cold rolling mill process and oily effluents generated.

- Organic flocculants (*Nalco*): N-7723. This flocculant is a quaternary polyamine and contains $ZnCl_2$ in its formulation.

The inorganic compounds used as coagulants were industrial lime provided by Aceralia, and $CaCl_2$ and NaOH, supplied by Panreac. All these compounds were used without additional purification. The mixing procedures have been described previously (11).

UF experiments were conducted in an Amicon stirred ultrafiltration cell (model 8200) with a volume of 200 cm^3 and an effective membrane area of

TABLE 1
Characteristics of Different Effluents from the Cold Rolling Mills of Aceralia

	Effluents from Tandem II		Effluent from Tandem I
	Regular effluent	Washing effluent	
Flow rate (m^3/h)	30–113	90	Unknown
Oil (mg/L)	200–300	300–400	10,000–11,000
COD (mg/L)	500–600	2800–3200	>20,000
pH	6.5–7.5	11–12	5–6
Suspended solids (mg/L)	0.5	11	—
Fe (mg/L)	0.5–5	9–10	—
Al (mg/L)	—	0.5	—

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28.7 cm^2. Two commercial Amicon ultrafiltration membranes were employed: PM30, made of polysulfone, and XM300, which is a copolymer of polyacrylonitrile and polyvinyl chloride. Both membranes are characterized by *molecular weight cutoff* values of 30,000 and 300,000 Daltons, respectively. Permeate fluxes were measured gravimetrically by an electronic balance (AND Instruments, model FX2000).

All the UF experiments were carried out at the optimum operating transmembrane pressure of 0.1 MPa. Membrane fouling becomes important at higher operating pressures, and an increase in the transmembrane pressure only increases the thickness of the gel layer rather than the transmembrane flux (12).

The membrane was rinsed with distilled water for 30 minutes after every UF test. It was then washed for 15 minutes with a solution of a commercial detergent (2 vol% Derquim+, Panreac) in distilled water and rinsed again for 10 minutes with distilled water. After the membrane had been cleaned, the initial water flux was checked before starting the subsequent experiment (13). In all cases the membrane was completely regenerated in the sense that it was always possible to restore the initial flux of distilled water.

Analytical Methods

Metal ion concentrations (Fe, Zn, and Al) in the effluents were measured by atomic absorption spectroscopy (Perkin-Elmer 2100 spectrophotometer). Turbidity and pH were determined using a Hach Ratio/XR turbidimeter and an Orion-2000 pH-meter, respectively. Chloride concentrations were determined following Volhard's method (14).

The oil content of the wastewater was determined by extraction of the oil with carbon tetrachloride and analysis of the solution by absorbance measurements at 300 nm for Quakerol and at 315 nm for Tinol using an UV/V spectrophotometer (Philips, PU 8720). COD was measured using the method of Crespi and Huertas (15).

RESULTS AND DISCUSSION

Effluents from Tandem II

Regular Effluent

The pH was adjusted to 10 in the flocculation experiments (11). This pH adjustment led to destabilization of the suspended particles of Tinol. Use of NaOH and lime as coagulants produced lower COD and oil concentrations in the effluent than those obtained with flocculants. This destabilization is mainly a result of saponification of Tinol that leads to precipitation of the fatty acids. The addition of $CaCl_2$ did not affect the stability of the suspension.

Results of the treatment of this effluent with lime (0.25 g/L) were compared to those obtained with N-7723 because this flocculant gave better oil removal than the inorganic compounds. This result may be attributed to the presence of $ZnCl_2$ in the N-7723 flocculant. The $ZnCl_2$ destabilizes the colloidal particles so that the cationic polyelectrolyte can act as an aid to coagulation by promoting the formation of bridges between the particles (16).

The clarified water phase obtained in these tests was then ultrafiltered with the PM30 membrane. Figure 2 shows the permeate flux, J, as a function of time for the aqueous phase obtained via different treatments of the *regular effluent*.

When the process fluid was ultrafiltered immediately after pH adjustment with lime (without allowing any time for settling) higher permeate fluxes were obtained than when the flocs that appeared when lime was added were allowed to settle. This result is a consequence of floc deposition on the membrane surface that hinders pore blocking by oil drops, making the membrane more hydrophilic. Therefore the flux decline is smaller (17, 18).

In both cases the permeate fluxes were 4–10 times higher than when no additives were present. In spite of the higher initial fluxes, the addition of the flocculant (N-7723) caused a rapid decline in flux, and also led to the presence of zinc in the effluent. Hence, adjustment of the pH of the emulsion with lime is the recommended treatment.

The experimental results are summarized in Tables 2 and 3.

Washing Effluent

Addition of $CaCl_2$ gave good results with this effluent, but the addition of flocculants gave even better results because the flocs formed in the settler have

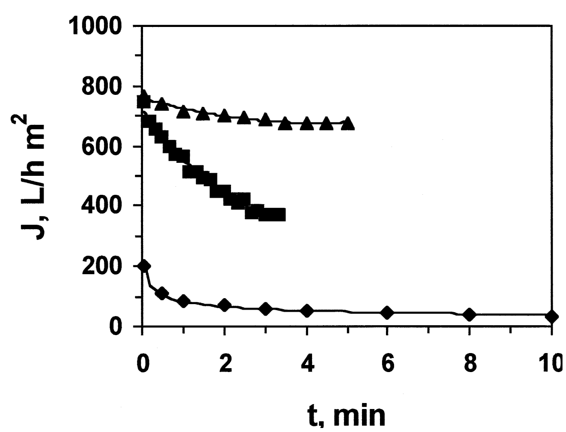

FIG. 2 Comparison of permeate flux for different treatments of the *regular effluent* ($\Delta P = 0.1$ MPa, PM30 membrane): (◆) without additives; (■) N-7723, 2.5 mL/L; (▲) lime, 0.25 g/L.

TABLE 2
Treatment of Regular Effluent with N-7723 Flocculant (2.5 mL/L) and UF of the Resulting Aqueous Phase with PM30 Membrane

	Effluent	Aqueous phase	Permeate	Sludges
pH	7.15	6.78	6.55	—
Oil (mg/L)	236	11	5	580[a]
COD (mg/L)	551	505	303	—
Turbidity (NTU)	—	11	0.15	—
Suspended solids (mg/L)	0.5	—	—	—
Fe (mg/L)	2	0.04	0.04	9.6[a]
Zn (mg/L)	—	298	298	410[a]
Water (%)	—	—	—	94
Permeate flux J (L/h·m^2)	—	—	188	—

[a] Kilograms in 1000 kg of dehydrated sludge.

better settling characteristics (11). Thus, flocculation of these effluents is preferred to coagulation as the pretreatment step prior to ultrafiltration.

In this case a complete study was carried out with each flocculant. In each case the optimum dose of flocculant was close to 1.25 mL/L. Doses below 1 mL/L are insufficient, and doses higher than 1.5 mL/L result in an excess of flocculant being adsorbed on the surface of fat particles. This situation may lead either to a change in the surface charge of the fat particles that again stabilizes the suspension, or to steric stabilization effects.

In the case of inorganic flocculants (DK-1014 and DK-1018), a charge neutralization mechanism presumably takes place (19), although an entrainment mechanism could also be responsible for flocculation (4). When the N-7723

TABLE 3
Treatment of Regular Effluent with Lime (0.25 g/L) and UF of the Resulting Aqueous Phase with PM30 Membrane

	Effluent	Aqueous phase	Permeate
pH	7.15	11.5	11.5
Oil (mg/L)	236	17	13
COD (mg/L)	551	139.5	118
Turbidity (NTU)	—	8	0.4
Suspended solids (mg/L)	0.5	—	—
Fe (mg/L)	2	0.03	—
Permeate flux J (L/h·m^2)	—	—	686

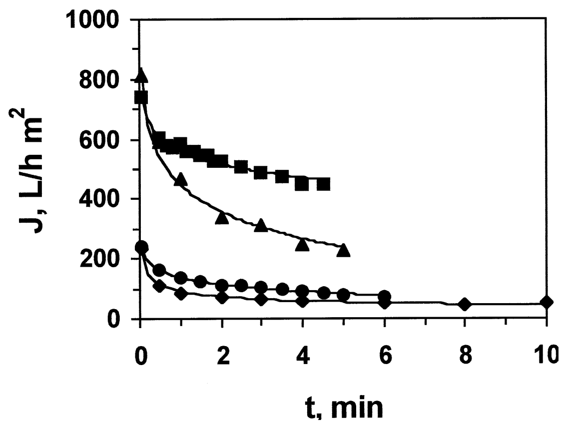

FIG. 3 Comparison of permeate flux for different treatments of the *washing effluent* ($\Delta P = 0.1$ MPa, PM30 membrane): (◆) without additives; (●) N-7723, 1.5 mL/L; (▲) DK-1014, 1.25 mL/L; (■) DK-1018, 1 mL/L.

flocculant is employed, a bridging mechanism is probably responsible for the formation of flocs.

Plots of the permeate flux as a function of time are presented in Fig. 3 for *washing effluents* subjected to different pretreatments. Best results were obtained using the inorganic flocculants (DK-1014 and DK-1018). Membrane fouling was more extensive with the organic flocculant, a result which can be attributed to adsorption of the polymer on the membrane.

The experimental results of the proposed treatments are summarized in Tables 4–6.

TABLE 4
Treatment of Washing Effluent with DK-1018 Flocculant (1 mL/L) and UF of the Resulting Aqueous Phase with PM30 Membrane

	Effluent	Aqueous phase	Permeate	Sludges
pH	11.81	7.07	7.33	—
Oil (mg/L)	380	16	6	887[a]
COD (mg/L)	2925	262	257	—
Turbidity (NTU)	—	1.5	0.11	—
Suspended solids (mg/L)	11	—	—	—
Fe (mg/L)	9.3	0.03	—	21[a]
Al (mg/L)	0.5	0.1	0.1	91[a]
Water (%)	—	—	—	92
Permeate flux J (L/h·m^2)	—	—	534	—

[a] Kilograms in 1000 kg of dehydrated sludge.

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TABLE 5
Treatment of Washing Effluent with N-7723 Flocculant (1.5 mL/L) and UF of the Resulting Aqueous Phase with PM30 Membrane

	Effluent	Aqueous phase	Permeate	Sludges
pH	11.81	7.48	7.7	—
Oil (mg/L)	380	29	12	755[a]
COD (mg/L)	2925	592	395	—
Turbidity (NTU)	—	11.4	0.25	—
Suspended solids (mg/L)	11	—	—	—
Fe (mg/L)	9.3	0.08	0.08	30[a]
Zn (mg/L)	—	298	298	84[a]
Al (mg/L)	0.5	0.1	0.1	83[a]
Water (%)	—	—	—	85
Permeate flux J (L/h·m^2)	—	—	120	—

[a] Kilograms in 1000 kg of dehydrated sludge.

Effluent from Tandem I (Quakerol)

Unlike the effluents from Tandem II, which are suspensions of fat particles in water, the effluent from Tandem I is an oil-in-water emulsion, which presents a milky appearance. This type of emulsion can normally be broken with cationic polyelectrolytes. However, poor flocculation results were obtained when this effluent was treated with DK-1014 or DK-1018.

The addition of $CaCl_2$ and an increase of pH did not affect the stability of the emulsion. Large amounts of lime were used in an effort to promote a *sweep coagulation*. This approach leads to formation of a voluminous precipitate of calcium hydroxide. In this case, clarified water was obtained, but the process

TABLE 6
Treatment of Washing Effluent with DK-1014 Flocculant (1.25 mL/L) and UF of the Resulting Aqueous Phase with PM30 Membrane

	Effluent	Aqueous phase	Permeate	Sludges
pH	11.81	6.81	7.19	—
Oil (mg/L)	380	14	8	810[a]
COD (mg/L)	2925	320	313	—[a]
Turbidity (NTU)	—	5	0.22	—
Suspended solids (mg/L)	11	—	—	—
Fe (mg/L)	9.3	0.03	—	25[a]
Al (mg/L)	0.5	0.1	0.1	83[a]
Water (%)	—	—	—	91
Permeate flux J (L/h·m^2)	—	—	360	—

[a] Kilograms in 1000 kg of dehydrated sludge.

TABLE 7
Treatment of Tandem I Effluent with Lime (20 g/L) and UF of the Resulting Aqueous Phase with PM30 Membrane

	Effluent	Aqueous phase	Permeate	Sludges
pH	5.87	12.8	12.8	—
Oil (mg/L)	10,700	25	17	299[a]
COD (mg/L)	>20,000	476	320	—
Turbidity (NTU)	—	13.3	2.6	—
Water (%)	—	—	—	50.38
Permeate flux J ($L/h \cdot m^2$)	—	—	195	—

[a] Kilograms of oil in 1000 kg of dehydrated sludge.

was more likely the result of adsorption of the oil droplets onto the lime particles.

The experimental procedure consisted of addition of lime, rapid stirring (160 rpm), and settling for 1 hour. The effects of the amount of lime added (which depends on the oil content) and the time during which rapid stirring is employed have been reported previously (11). Good results were obtained with lime concentrations of 10 g/L and mixing times of 15 minutes. These conditions lead to removal of 99% of the oil.

The high concentration of oil in the effluent from Tandem I causes complete blocking of the pores of the PM30 membrane within a few seconds if one attempts to ultrafilter the effluent without pretreatment. Experimental data for the results of treatment of this effluent with lime and subsequent ultrafiltration with a PM30 membrane are presented in Table 7. The main disadvantages of

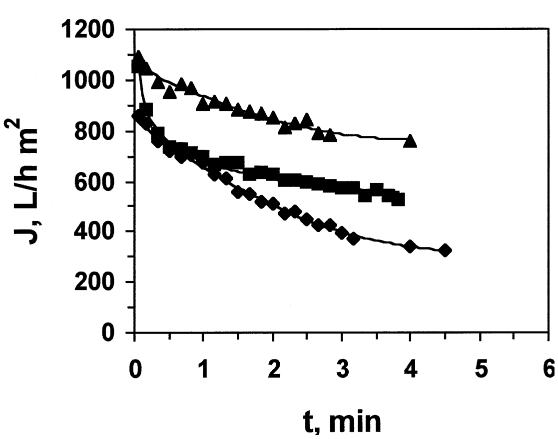

FIG. 4 Permeate fluxes obtained for the treatment of mixtures of *Tandem I* and *regular effluents* with lime (10 g/L) and ultrafiltration with XM300 membrane (ΔP = 0.1 MPa): (◆) 75 vol% Tandem I effluent–25 vol% regular effluent; (■) 50 vol% Tandem I effluent–50 vol% regular effluent; (▲) 25 vol% Tandem I effluent–75 vol% regular effluent.

TABLE 8
Treatment of Tandem I and Regular Effluent Mixtures with Lime (10 g/L) and Ultrafiltration of the Resulting Aqueous Phase with XM300 Membrane

	25 vol% Tandem I effluent + 75 vol% regular effluent				50 vol% Tandem I effluent + 50 vol% regular effluent				75 vol% Tandem I effluent + 25 vol% regular effluent			
	Effluent	Aqueous phase	Permeate	Sludges	Effluent	Aqueous phase	Permeate	Sludges	Effluent	Aqueous phase	Permeate	Sludges
pH	—	12.8	12.8	—	—	12.8	12.8	—	—	12.8	12.8	—
Oil (mg/L)	2,852	58	30	117[a]	5,470	60	40	133[a]	8,084	94	32	224[a]
COD (mg/L)	>20,000	219	245	—	>20,000	234	234	—	>20,000	357	265	—
Turbidity (NTU)	—	18.5	3	—	—	9	4	—	—	50.3	5.3	—
Water (%)	—	—	—	46	—	—	—	46	—	—	—	42.6
Permeate flux J (L/h·m^2)	—	—	785	—	—	—	580	—	—	—	427	—

[a] Kilograms of oil in 1000 kg of dehydrated sludge.

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the treatment with lime are that it leads to high pH of the treated water and the formation of large amounts of oily sludges, both of which are problematic from an environmental point of view.

Because the treatment of this effluent involves mixing with the *regular effluent*, three different mixtures of these effluents were studied. The treatment to which these mixtures were subjected was the same as that previously described for the Tandem I effluent alone, but in the case of the mixtures ultrafiltration was carried out with a XM300 membrane (Fig. 4) because of the ease with which the pores in the PM30 membrane were blocked. The experimental results are shown in Table 8. Inspection of the tabular entries reveals that permeate flux decreases as the concentration of the Tandem I effluent increases.

CONCLUSIONS

- The recommended integrated treatment for the *regular effluent* involves addition of NaOH or lime: 90% of the oils and 75% of the COD are removed. For treatment of the *washing effluent*, use of the DK-1014 flocculant and subsequent UF is recommended. This treatment again removes 90% of the oil, but the resulting COD values exceed regulatory standards. Similar results were obtained for treatment of the *Tandem I effluent* with lime: a significant decrease in the oil concentration is achieved, but the oil level remains above the statutory limits. Moreover, this approach generates large amounts of oily sludges.
- The high COD values obtained when the effluents were treated with lime at 60°C might be a consequence of the presence of organic compounds generated via partial hydrolysis of fats. By contrast, the COD values obtained when N-7723 is used are probably a result of the presence of an excess of the flocculant dissolved in water.
- The oil contents of the permeate from the ultrafiltration stage are 75% lower than the regulatory limits established by all EU countries. Furthermore, all suspended solids were removed during this UF stage.
- The operating costs of the ultrafiltration stage are very low because of the low operating pressure (0.1–0.15 MPa). Membrane cleaning can be easily carried out using a commercial detergent or by backpulsing.

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