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Investigation into hydro- and oleophilic porous medium saturation with two-phase liquids during the imbibition process

Introduction

The processes of spontaneous imbibition as a physical phenomenon occurs in porous media due to the capillary suction. The appearing capillary force is mainly conditioned by interactions between surface energies of a porous material and a wetting liquid [*Zhmud et al., 2000; Masoodi et al., 2007*]. The imbibition is regarded as relevant to a wide range of processes in the environmental and chemical engineering, medicine, pharmaceutical industry, i.e. oil recovery, application of antimycotic emulsions in concrete structures, hydrological regime in soils, composite materials processing etc. Therefore, it explains the presence of a continuing interest in imbibition and a plurality of efforts to describe this process and to define the main influential factors [*Zhmud et al., 2000; Alava et al., 2004; Masoodi et al., 2007*].

The common feature of these publications is the investigation of single-phase liquid imbibition. Meanwhile the main objective of our research work is to study a process of porous medium imbibition with multiphase liquids, i.e. stabilized emulsions.

The change of medium saturation with an oil-in-water emulsions vs height of their penetration was discussed issue in this publication. Such investigation gives the possibility to estimate the efficiency and peculiarities of the emulsion penetration in hydrophilic-oleophilic porous media compelled by the capillary force.

Experimental

Characteristics of emulsions and porous medium

The refined vegetable oil *Uniwersalny* (*FHPU Marlibo Z.P.Chr*, Poland) represented the basic oily component of the investigated emulsions. The viscosity of this oil was equal to 53.12±0.44 mPa·s, its density was 922.00±0.60 kg/m³, and value of surface tension was 32±1.7 mN/m. Ethoxylated oleic acid (*Rokacet O7*) is a non-ionic surfactant used to stabilize the prepared two-phase liquids. It was obtained from *PCC Exol SA*, Poland. The fraction of the added emulsifier was equal to 2 vol. %.

Three types of oil-in-water emulsions with different dispersed phase concentrations, i.e. 10 vol. %, 30 vol. % and 50 vol. %, were used in the current experiments. They were correspondingly referred as: VP–10, VP–30, and VP–50. The emulsions were prepared by means of stirring during 600 s at maximal rate of intensity, using a homogenizer MSM–67170 (*Bosch*, Germany). The properties of the prepared emulsions are shown in the Tab. 1.

The porous medium was represented by a commercial modified nonwoven polypropylene material, obtained from *Sintac© Polska Sp.z o.o.* It belongs to universal group of sorbents, i.e. measured sorption capacity of vegetable oil was 24 g/g, while for water it equaled 12 g/g. The hydrophilic-oleophilic sorbent was investigated because a great deal of presence porous media reveals an ability to absorb both oil and water. The average calculated porosity was 0.95, while density of the investigated sorbent was equal to 50.23 kg/m³.

Sheets of the polypropylene sorbent were cut into stripes (i.e. size

Tab. 1. Emulsion properties measured at T = 23±1°C

Property	Type of emulsion		
	VP–10	VP–30	VP–50
Density, [kg/m³]	923.1±3.19	958.1±0.68	962.7±0.81
Viscosity, [mPa·s]	1.20±0.22	3.25±0.05	8.82±0.28

of 0.035×0.20 m and mean thickness of 1.5·10⁻³ m), which were used for the purpose of the experiments.

Imbibition and saturation

The process of imbibition was investigated experimentally and its procedure has been described in our previous publication [*Sęk et al., 2015*]. The prepared stripes of polypropylene material were submerged in beakers with prepared emulsions. The depth of immersing was 0.02 m and total contact square equaled $1.4 \cdot 10^{-4}$ m². The experiments were conducted at 23±1°C. The changes of an emulsion mass in a beaker and its height h_{im} were observed and registered during imbibition process. Time, when mentioned parameters became unchangeable was defined as the maximal time of the process t_{max}.

After a stripe was removed from a beaker, the height of an emulsion front was registered and mass of left emulsion was measured using an analytical balance (*Radwag WTB 200*, Poland). The obtained height was defined as the equilibrium height h_{max}, while the maximal imbibed mass (referred as m_{max}) was calculated as the difference of the initial mass and mass of left emulsion in a beaker.

Afterward, the sample of sorbent imbibed with an emulsion was cut into fragments with a height of 0.02 m. Each fragment represented the defined height of the soaked polypropylene stripe, thus: for the first fragment, f_1 it was 0÷0.02 m; for f_2: 0.02÷0.04 m; for f_3: 0.04÷0.06m; and for f_4: 0.06÷0.08 m. After that, a part of the emulsion was mechanically extracted and its volume V_{ex} and mass m_{ex} were determined. The weight of sorbent pieces after extraction, *viz.* m_{wf}, was also measured. Such procedure was necessary to be fulfilled because the properties of an imbibed emulsion can differ from its initial ones due to the changes of the emulsion concentration within the imbibition process.

The saturation, S_f is an important parameter that characterizes a level of free porous volume filling and can be used to predict the sorptive material efficiency. The saturation of polypropylene samples with the emulsions was calculated in accordance with the following expression:

$$S_f = \frac{V_{im}}{V_f \varepsilon}, \tag{1}$$

where V_{im} is the volume of an imbibed emulsion in a fragment of polypropylene sample, [m³]; V_f is the total volume of this fragment [m³]; ε is the porosity of a sorbent sample. The total volume of an imbibed emulsion in each fragment was computed by means of the equation:

$$V_{im} = V_{ex} + \frac{V_{ex}(m_{wf} - m_{f_0})}{m_{ex}}, \tag{2}$$

where: m_{f_0} is the mass of a dry fragment, [kg].

This experiment allowed to define the saturation changes of polypropylene sample vs the height during the imbibition and to evaluate influence of the dispersed phase concentration and process duration.

Results and discussion

The value of the maximal imbibed mass was achieved for a time of 900 s for all investigated emulsions, while for pure liquids such time equaled 660 s. The obtained results are shown in Tab. 2.

The highest value of total imbibed mass was observed in case of pure liquids, i.e. $3.30 \cdot 10^{-3}$ kg for oil. The same tendency was noticed for the mass imbibed by unsubmerged part of a polypropylene. The highest value of total imbibed mass was observed in case of

Tab. 2. Values of the maximal mass of the imbibed liquids

Value of mass ×10³ [kg]	Type of investigated liquid				
	VP–10	VP–30	VP–50	Oil	Water
Total mass	2.28	2.32	2.98	3.30	3.08
Mass at $h_{im} \geq 0.02$ m	1.12	1.24	1.6	2.1	1.39

pure liquids, i.e. $3.30 \cdot 10^{-3}$ kg for oil. The same tendency was noticed for the mass imbibed by unsubmerged part of a polypropylene sample and it equaled $2.1 \cdot 10^{-3}$ kg. For water, values were a few lower, in particular $3.08 \cdot 10^{-3}$ and $1.6 \cdot 10^{-3}$ kg, relatively. Among two-phase liquids, the highest values of imbibed mass were registered in case of 50% emulsion, i.e. 2.98×10^{-3} and $1.6 \cdot 10^{-3}$ kg. Thus, the initial dispersed phase concentration was defined as an influential factor that determined the imbibition process.

Fig. 1 presents the alteration of saturation level with height of imbibed liquid front for the emulsions with different dispersed phase concentration.
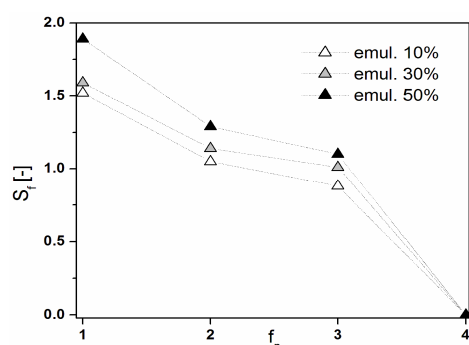
Fig.1. Saturation vs height of an imbibed emulsion front (for t_{max} = 900 s)

The analysis of obtained data showed a significant exceeding of free volume of pores in case of the unsubmerged part of a polypropylene sample (f_1). Thus, the saturation values were 1.89 for 50 % emulsion, 1.59 for 30% and the lowest was 1.52 for 10% emulsion (Fig. 1). It can be explained by an extra sorption of a liquid on the external surface of the samples. In further fragment at $0.02 \text{m} \leq h_{im} \leq 0.04$ m, the sorptive material was oversaturated and S_f level was in a range 1.05÷1.29. The full saturation of f_3 was observed only in case of VP–30 and VP–50, i.e. 1.01 and 1.1, while for VP–10 its level was equal to 0.88 (Fig. 1).

The previously described experimental procedure was repeated for shorter time: 300 s and 600 s. The purpose was to define the changes of the polypropylene samples saturation with the height for different time values. The results obtained for VP–50 are shown in Fig. 2.

Obviously, the saturation levels increasing was observed with growing of imbibition time, but such changes are not large for f_1: for 600 and 900 s, S_f was correspondingly 7% and 17% higher than for t_{im}=300. For the fragment f_2, this difference slightly grew and equaled 10÷17%. In case of f_3, saturation at 900 s was 35% higher
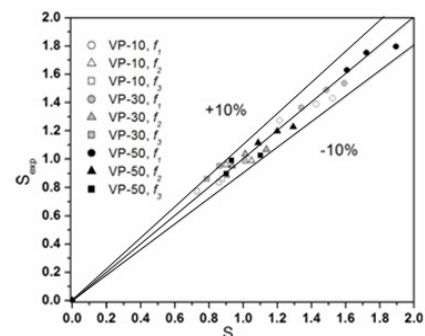
than at 300 s, while its difference between minimal time and 600 s was the smallest, i.e. 3% (Fig. 2). The analysis of the obtained data gave the possibility to conclude that the same tendencies were representative for other emulsions. To sum up, most of the emulsions was imbibed and consequently, depending on fragment up to 65÷85% of registered sorbent saturation was achieved during the first 600 s of the process.

The experimental data concerning saturation changes in each fragment with time were approximated by the following equation:

$$S_f = S_{max} \frac{t_{im}}{k + t_{im}}, \qquad (3)$$

where: S_{max} is the maximal saturation level in each fragment, [-]; t_{im} is the time of imbibition, [s]; k is the coefficient of the given equation.

The results of such approximation for VP–50% (as an example) are plotted on Fig. 3. Correlations between experimental data and those calculated analytically from Eq. 3 for all investigated emulsions are presented in Fig. 4. The straight lines on the plot refer to the boundaries within which solutions were found. Thus, results are in a range of ±10% that proved the appropriation of the proposed equation.

Conclusions

The saturation level of the sorbent changes with the height of the imbibed emulsion front. The phenomenon of sorbent oversaturation was observed mostly at $h_{im} \leq 0.04$ m due to the formation of a layer on the external surface of a sorbent sample.

The saturation level depends on the initial concentration, thus the highest value was observed for 50% emulsion and can be explained by twice higher sorption capacity of the refined oil comparing to water.

The level of S_f also varies with time, but up to 65÷85% of the registered sorbent saturation level was obtained within the first 600 s.

In addition, the proposed equation provides an accurate approximation of the experimental data and a precise description of the process of the saturation changes with time.

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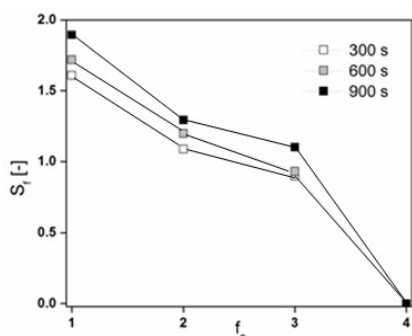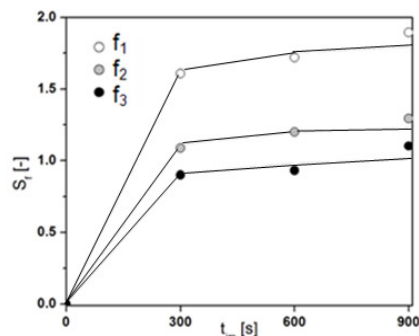
Fig. 2. Saturation vs height for 50% emulsion (t_{im} = 300, 600 and 900 s)

Fig. 3. Approximation of the experimental data obtained for 50% emulsion from Eq. (3)

Fig. 4. Correlation between experimental values (S_{exp}) and calculated ones (S_{cal}) from Eq. (3)