The effect of thickness of crude oil layers on the attenuation of the surface capillary wave*

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Abstract

The paper presents the results of investigations of absorption of a capillary wave with a frequency of 30 Hz propagated on a water surface covered with a layer of crude oil derivatives. The relationship between the value of absorption coefficient and the thickness of the pollutans layer was investigated by the acoustic pulse method. The values of α determined experimentally are greater than those predicted by the Stokes' theory. Applicability of the Stokes' equation was considered on the basis of physical properties characterizing the investigated petroleum derivatives.

1. Introduction

A crude oil derivative introduced onto a water surface may form a number of structural arrangements. One of them can be a monomolecular layer with very diverse viscoelastic properties [2]; if the amount of petroleum derivatives is quite large, then their excess will form a lens-shaped layer after forming a monolayer. The equilibrium thickness of the lens-shaped layer is strictly determined by the value of surface and interfacial tensions and the densities of both liquids [1].

Intense undulation processes will result in the formation of another structural arrangement – namely an emulsion – in which the fraction of the dispersed phase of the organic compound varies.

The attenuation coefficient of the surface capillary wave propagated along such surface cannot be described by the Stokes' equation [3], which refers to the waves running on the surface of an ideal liquid. The proximity of both interfacial surfaces creates different conditions of the liquid flow and facilitates the exchange of energy

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between adjacent areas. The dependence of surface wave attenuation coefficient as a function of thickness of the layer consisting of various petroleum compounds, obtained by the pulse method [4, 5], together with the determination of structural parameters of the investigated liquids may supply interesting information on the process of undulatory motion in systems of a two-layer type.

2. Mutual wettability of liquids

The phenomenon of mutual wettability of liquids can be described by means of one parameter (relation between the wettability coefficient of substance 'a' and substance 'b') $S_{b/a}$, defined by the values of surface and interfacial tensions of these substances [1]:

$$S_{\mathrm{b/a}}=\gamma_{\mathrm{a}}-\gamma_{\mathrm{b}}-\gamma_{\mathrm{ab}}, \tag{1}$$

where:
γ_a – surface tension of the solvent (water),
γ_b – surface tension of the solute ('oil'),
γ_{ab} – interfacial tension (water – 'oil').
If this coefficient assumes a positive value, the expansion of liquid 'b' is accompanied by a decrease in the thermodynamic potential of the system, and this process takes place spontaneously [2]. Its negative value leads to the formation of a system composed of a monomolecular layer at an equilibrium with the excess of liquid in a lense-shaped layer with a strictly defined equilibrium thickness. If the densities of liquids used and the values of surface and interfacial tensions are known, we can determine this thickness from the relationship [1]:

$$d_0^2=-\frac{2S_{\mathrm{b/a}}\rho_{\mathrm{a}}}{g\rho_{\mathrm{b}}\Delta\rho}, \tag{2}$$

where:
g – acceleration due to gravity,
ρ_a – density of solvent (water),
ρ_b – density of solute ('oil').

3. Attenuation of surface waves in liquids

Surface capillary waves propagating on the surface of a clean liquid are attenuated with distance according to the classical hydrodynamic wave theory. The value of the attenuation coefficient was given for the first time by Stokes in the following form [3]:

$$\alpha_m=\frac{4k^3\eta_{\mathrm{b}}}{3\rho_{\mathrm{b}}\,\omega}, \tag{3}$$

where:

k – angular wave number of the surface wave $\left(k=\frac{2\pi}{\lambda_b};\ \lambda_b \text{ – surface wavelength}\right)$,

η_b – liquid viscosity,

ρ_b – liquid density,

ω – angular frequency of surface wave.

The equation obtained is derived from the solution of the dispersion equation for surface capillary waves propagating on the surface of an ideal liquid [6]. We thus assume that the flow of liquid is mostly laminar potential and disregard the influence of the eddy component. The penetration depth of the eddy component may be expressed by a conjugate parameter m [4]:

$$m^2=k^2+\frac{i\omega\rho}{\eta}, \tag{4}$$

when the following condition:

$$k^2 \ll \frac{\rho\omega}{\eta} \tag{5}$$

is met, then the flow is mainly laminar and the attenuation of the wave may well be approximated by the Stokes' equation.

4. Material

The material used consisted of five crude oil derivatives: ethyl gasoline 78, kerosene, Diesel oil, extra 15 engine oil and Marinoll 111 oil. In order to characterize the materials used, the following additional measurements of structural parameters were performed: density, viscosity, surface and interfacial tensions in contact with water. The results are presented in Table 1. The two last columns in Table 1 contain the wettability coefficients $S_{b/a}$ (1) and equilibrium thickness d_0 (2). The values for the first three liquids are positive, which suggests the ability to form on the water surface a layer with a thickness ranging from monomolecular to one resulting from the amount of the liquid and the surface available.

Table 1. Physical properties of crude oil derivatives and a system water – 'oil' at 293 K and pressure of 101, 325 Pa

Petroleum derivative	$\rho_b \cdot 10^3$ [$kg \cdot m^{-3}$]	$\eta_b \cdot 10^{-3}$ [Pas][Pa·s]	$\gamma_b \cdot 10^{-3}$ [$N \cdot m^{-1}$]	$\gamma_{b/a} \cdot 10^{-3}$ [$N \cdot m^{-1}$]	$S_{b/a} \cdot 10^{-3}$ [$N \cdot m^{-1}$]	$d_0 \cdot 10^{-2}$ [m]
Ethyl gasoline 78	0.724	0.676	20.3	50.4	2.04	–
Kerosene	0.761	1.641	21.8	50.8	0.15	–
Diesel oil	0.847	3.540	30.6	29.3	12.94	–
Extra 15 oil	0.854	11.93	35.7	48.36	–11.33	0.4242
Marinoll 111 oil	0.853	82.06	35.85	51.54	–15.64	0.5091

Extra 15 and Marinoll 111 oils have negative values of $S_{b/a}$; introduced onto the water surface; they will form a lens with a thickness of 0.004242 and 0.005091 m, respectively.

In order to obtain intermediate values of oil layer thickness, before distributing an oil film, a surface-active agent (Fotonal) was introduced. Undergoing absorption, this substance reduced the interfacial tension of water–'oil', changing the value of $S_{b/a}$ wettability coefficient (1) into a positive one. Thus, it was possible to form a permanent film of both oils with a thickness ranging from 0.00001 m to the equilibrium thickness d_0.

5. Results

The attenuation coefficient of a surface wave with a frequency $f = 30$ Hz propagating on the water surface covered with a film of crude oil derivative, as a function of its thickness d is presented in Figure 1. The value of the coefficient α was determined

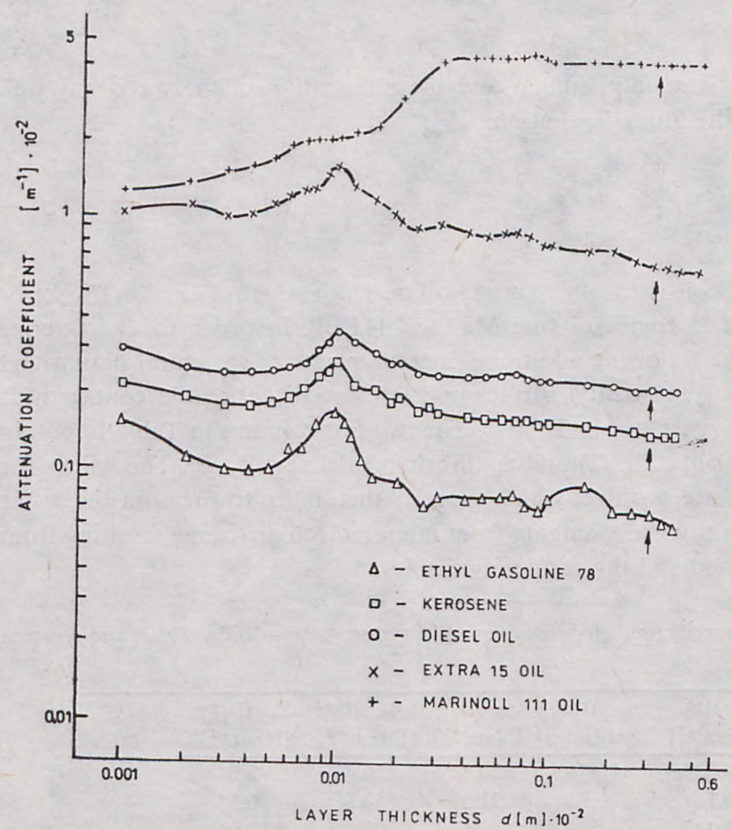

Fig. 1. Attenuation of a surface capillary wave on a water surface covered with a film of crude oil derivative

by means of an apparatus presented in earlier studies [4, 5]. Table 2 presents the theoretical values of α_m (3) and the condition of applicability (4) of this expression, based on the measurement results from Table 1.

6. Discussion

The nature of function $\alpha(d)$ presented in Figure 1, although complicated, has some common features for ethyl gasoline 78, kerosene, Diesel oil, and Extra 15 oil.

The values of α for the thinnest films (0.00001 m) are greater than those predicted by the theory, being equal to 2.44 α_m for ethyl gasoline, 1.573 α_m for kerosene, 1.378 α_m for Diesel oil, and 1.636 α_m for Extra 15 oil.

Table 2. Applicability condition of Stokes' approximation (5)

Petroleum derivative	$\alpha_m \cdot 10^2$ $[m^{-1}]$	$\eta_b \ll \frac{\rho_b \omega}{k_b^2}$	$\lambda_b \cdot 10^{-2}$ [m]	$\lambda_{ab} \cdot 10^{-2}$ [m]
Ethyl gasoline 78	0.0602	$0.00676 \ll 1.461$	0.65	0.85
Kerosene	0.1379	$0.0164 \ll 1.5358$	0.65	0.83
Diesel oil	0.2155	$0.0354 \ll 1.9827$	0.7	0.7
Extra 15 oil	0.6418	$0.1193 \ll 2.144$ (!)	0.725	0.8
Marinoll 111 oil	4.3641	$0.8206 \ll 2.1409$ (!)	0.725	0.84

For larger thicknesses the values of α decrease steadily and approach the values predicted by theory, although they remain higher by several per cent: 1.04 α_m for ethyl gasoline, 1.015 α_m for kerosene, 1.0004 α_m for Diesel oil, 1.028 α_m for Extra 15 oil.

The nature of the relationship $\alpha(d)$, especially for small values of d, is probably a result of the existence of two interfacial surfaces, along which propagate two surface waves with different wavelengths λ_b (oil surface) and λ_{ab} (interfacial surface oil-water) (Table 2), determined on the basis of the Kelvin's dispersion equation [6]. Their closeness facilitates the processes of energy exchange carried by the wave and leads to a change in the character of flow. An increase in the distance between the two surfaces diminishes these effects; thus, when d was approximately equal to $\lambda_b/2$, *ie* corresponding to the conventional depth of penetration of undulatory processes, the attenuation only slightly (by several per cent) exceeded theoretical values. The dependence $\alpha(d)$ for Marinoll 111 oil is of a completely different character. For an oil film of thickness $d=0.001$, the value $\alpha=0.286\,\alpha_m$ is relatively low. For $d=0.0003$ m, α reaches the value of 0.989 α_m and continues to increase until it reaches 1.0013 α_m for the equilibrium thickness d_0 (Table 1).

Higher than theoretical values of α most likely result from the partially eddy-like flow of liquid in the vicinity of the undulating surface, as suggested by the applicability condition of the Stokes' expression (4) found in Table 2 for all investigated liquids. It is fulfilled in the case of the first three liquids with low viscosity, while the values on both sides of inequality (5) for the remaining two liquids differ by not more than one order of magnitude. When the thickness of the film increases, the values of α decrease and pass through a well-pronounced maximum at $d \approx 0.0001$ m.

7. Conclusions

(i) The attenuation of a surface capillary wave propagated on the surface of water covered with an oil film attains higher values than those assumed by the classical hydrodynamic theory.

(ii) When the thickness of the film approaches or exceeds the generally assumed value which determines the depth of penetration of the undulatory motion $d \geqslant \lambda_b/2$, the values of the coefficients tend to approach the Stokes' expression α_m, exceeding it only by several per cent.

(iii) The observed instances of departure of α from the theoretical value are probably caused by the short distance between interfacial surfaces and the excessive viscosity of heavier fractions of crude oil, which leads to the unfulfillment of the applicability condition of the Stokes' approximation for an ideal liquid.

(iv) The acoustic pulse method applied for the determination of attenuation of a surface capillary wave may be useful when investigating the exchange processes of undulatory energy in stratified systems.

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