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DETECTION OF WAVY SEA SURFACE OIL-DERIVATIVE CONTAMINATION WITH FORWARD SPECULAR HIGH-FREQUENCY SCATTERING

Stanisław Pogorzelski
Paweł Rochowski
Maciej Grzegorczyk
Bogumił B. J. Linde
Institute of Experimental Physics, University of Gdańsk, Poland
Ewa Skrodzka
Faculty of Physics, Institute of Acoustics, A. Mickiewicz University, Poland
Łukasz Bielasiewicz
INFO-EKO Design-Consulting Office, Poland

ABSTRACT

A spectrum of low-frequency (20–30 Hz) amplitude fluctuations of the ultrasonic (10 MHz) signal specularly scattered from water surfaces covered with monomolecular and thicker crude oil origin films of well-defined, oceanographically relevant viscoelastic properties was examined in laboratory and at-sea conditions. The relationship between the surface water wave (30 Hz) damping coefficient and the oil layer thickness was established, and compared to the one predicted by the classical Stokes theory. The depression of the spectral energy density of wind-driven waves by surface films was inferred from the ratio of acoustic signal fluctuations spectra with/without films, and compared to that resulting from the Marangoni damping theory applicable to monolayers of particular surface viscoelasticity. The agreement between the theory and experimental data was satisfactory. As shown in at-sea experiments performed with a free-floating, buoy-like acoustic system, and an artificial oil slick spread over the Baltic Sea surface, the film's rheological surface properties can be recovered from acoustic surface probing, as well as oil spill edge detection. Simultaneous statistical analyses of the scattered signal amplitude distribution parameters turned out to be unequivocally related to the oil substance fraction weight, oil layer thickness, and the form of oil contamination.

Keywords: crude oil contamination,capillary wave damping,acoustic surface scattering,statistical-frequency analyses,at-sea pollution detection

INTRODUCTION

A crude oil derivative introduced on a water surface may form a number of structural arrangements [4]. One of them can be a monomolecular layer with very diverse viscoelastic properties [10]. If the amount of petroleum derivatives is quite large, then their excess will form a lens-shaped layer in equilibrium, as shown in Fig. 1. The spreading coefficient is defined as $S_{0/W} = \gamma_W - \gamma_0 - \gamma_{W/O.}$ Oil spreads on the water surface (complete wetting), for $S_{0/W} > 1$ [11], where: γ – surface tension of water (w), surface tension of oil substance (o), and interfacial tension of water–oil (w/o). For $S_{0/W} < 1$, a lens-shaped oil layer of particular thickness d_∞ (a few millimetres [4]) is formed. The equilibrium thickness of the lens is strictly determined by the value of surface and interfacial tensions and the densities of both liquids [1]. The crude oil spreading phenomenon existing in the sea is much more complex to evaluate quantitatively. In particular, the steady-state signatures of oil contamination are affected by the presence of the sea surface's natural surfactant films having 2D thermo-elastic properties [2, 29], and the spreading coefficient is a quantity that is dependent on surfactant concentrations, surface activity and temperature. Moreover, the oil spreading kinetics is mediated by the viscoelasticity modules of all the interfaces (O/W/A) [3].

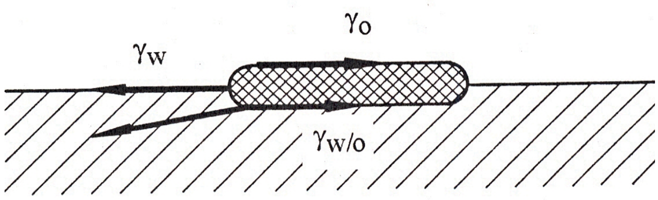

Fig. 1. Lens of oil on water surface

An intense undulation process will result in the formation of several other structural arrangements, in particular an emulsion, in which the fraction of the dispersed phase of the organic compound varies in time and space.

The attenuation coefficient of the sine surface capillary (frequency > 13 Hz) wave of low amplitude, propagated along such an oil-coated surface, cannot be described by the Stokes equation (1),

$$\alpha_m = \frac{4k^3\eta}{3\rho\omega} \qquad (1)$$

where:
$k = 2\pi/\lambda$, λ – wavelength, η – liquid viscosity, ρ – density, $\omega = 2\pi f$, and f is the frequency.

Such a relation refers to the waves running on the surface of an ideal liquid [21]. The proximity of both interfacial surfaces creates different conditions of the liquid flow, and facilitates the exchange of energy between adjacent areas. The dependence of the surface wave attenuation coefficient as a function of the thickness of the layer consisting of various petroleum compounds was obtained by the ultrasonic pulse reflection method [23, 24]. The determined structural physico-chemical properties of the investigated liquids, obtained in an accompanying study, supplied interesting information on the damping process of water surface wave motion in a two-layer type system. In further studies, high-frequency scattering signal amplitude characteristics (spectral and statistical) were evaluated for the wind-driven surface of a clean (pollution-free) and affected by crude oil layer of water [20, 27, 28].

DAMPING RATIO MEASUREMENTS IN NATURAL CONDITIONS

The spectral analysis of acoustic signal amplitudes scattered specularly from a clean sea surface and one covered with artificial oil films was performed for Gasoline 94, Gasoline 86 and Selectol plus engine oil. The effects of the damping by an artificial monolayer sea slick in the frequency range of 2–20 Hz on short gravity and capillary waves were demonstrated by a number of authors [12, 13, 22].

The relative spectrum $D_r(f)$ expressed by the spectral density was obtained for the vegetable oil slick and slick-free areas, respectively, the frequency range (2–20 Hz) clearly corresponding to the wave damping by a film in the presence of light breeze (1.0–1.5 m/s) [7–9]. The spectral ratio of wind-generated waves of a clean sea surface and a polluted surface can be presented by the low-frequency-range damping ratio of the amplitude of high-frequency scattered signals. The analysis of the signals scattered from open-sea formed slicks with oceanographically relevant elastic properties was discussed in terms of the $D_r(f)$ pattern damping ratio with the derived static and dynamic surface film properties [5, 6].

EXPERIMENTAL CONDITIONS

The influence of oil layers of different properties on the amplitude fluctuation of scattered ultrasonic signals from a wind-driven water surface was examined under natural conditions in Gdynia's Naval Port [19]. The air and the sea water temperatures during the measurements were 285 and 287 K respectively. A high-frequency (10 MHz), two-transducer acoustic system based on a forward specular scattering geometry as a free-floating buoy-like arrangement was used in these investigations [18, 23, 25, 28] (Fig. 2).

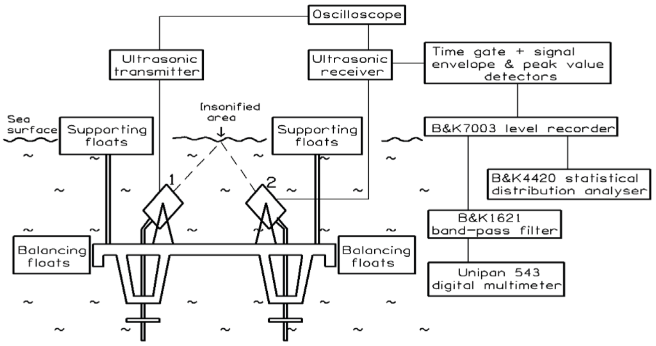

Fig. 2. Block diagram of the experimental setup. Numbers "1" and "2" denote two ultrasonic transducers

The registered acoustic signal amplitude fluctuations were played back by a level tape recorder and analysed using a Brüel & Kjaer 1621 analogue tuneable band pass filter with a width of 1/3 octave (23%) and a V-543 digital multimeter over a frequency range of 1–50 Hz. The crude oil substances were used as an oil-film-slicks-forming mono-layer on the sea surface. The oils applied in these investigations were dissolved in hexane to make a volatile solution, and spread carefully on the sea surface. It should be noted that the intensity of high-frequency scattered signals Is is inversely proportional to the square of the slope of the surface, $I_S \sim 1/(h/\lambda)^2$, and linearly related to the scattered signal amplitudes $(I_S \sim A^2)$. The damping ratio $D_r(f)$ of the oil films spread on the open sea surface can be measured on the basis of the following theoretical expression, for the spectral density proportional to the surface wave height $(S(\omega) \sim h^2)$, as described by Phillips [18, 22].

$$D_r(f) = \left[\frac{S_0(\omega)}{S_o(\omega)}\right]^2 = \frac{h_0(\omega)}{h_o(\omega)} = \frac{I_o(\omega)}{I_0(\omega)} = \left(\frac{A_o}{A_0}\right)^2 \qquad (2)$$

The physical and visco-elastic properties of an oil-covered sea surface were obtained for the selected oil substances from additional measurements performed using the Langmuir trough system [17].

The light oils were treated as monolayers exhibiting positive spreading coefficients. The Selectol plus oil had a negative spreading coefficient, and appeared on the sea water surface as floating lenses or spots with an equilibrium thickness of 0.39 cm.

The static and dynamic oil film 2D properties characterised by the elasticity modulus E_0, the film surface pressure Π, and the relaxation time τ, were introduced in Eq. (3) over the frequency range of 1–35 Hz, for two oil substances (Gasoline 94, Gasoline 86) for a better comparison.

RESULTS AND DISCUSSION

The damping ratio results were obtained in the presence of oil substances under natural conditions at a constant wind velocity above the sea surface. The damping ratios within a frequency range of 1–35 Hz were analysed. The frequency relationships of the damping ratio k (~ $D_r(f)$) are illustrated in Fig. 3, where the theory predicted relation together with the scattered signals ratio experimentally determined are presented.

It is clear from the plots that the wave damping ratios appear to have low values (0.5–1.5) in the frequency region of $f >> 20$ Hz. The theoretical damping ratio (contrast) values of $D_r(f)$ were computed for Gasoline 94 and Gasoline 86, by means of Eq. (3).

The damping ratio of short surface waves in the presence of insoluble oil films [5, 6, 14–16] is given by:

$$D_r(f) = \alpha_{o} / \alpha_0 = \frac{1 - 2\tau - 2\tau^2 - X_0 - Y_0(X_0 + \tau)}{1 - 2\tau + 2\tau^2 - 2X_0 + 2X_0^2} \tag{3}$$

where:

α_∞ and α_0 are the respective damping coefficients, for a film-covered and film-free surface,

$$X_0 = \frac{E_0 K^2}{\rho(2\eta\omega^3)^{1\backslash 2}}, \qquad Y_0 = \frac{E_0 K}{4\eta\omega\rho}, \qquad \tau = \left(\frac{2t_r}{\omega}\right)^{1/2}$$

where:

$E_0 = -A\frac{d\pi}{dA}$ – the dilational elasticity modulus accounting for film-forming material adsorption or desorption variations during a compression–dilation film area cycle, A – the film-covered area, η – the viscosity of water, ρ – density of water, K = 2π/λ – wave number of surface water wave, ω = 2pf – angular frequency, t_r – relaxation time.

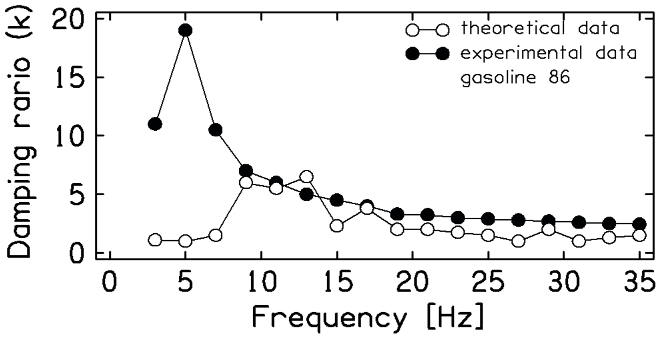

Fig. 3. The damping ratios k (contrast) of sea surface relationships versus frequency in the presence of Gasoline 86 film

We believe that the not close agreement between the theoretical and experimental curves could result from other environmental factors (temperature, currents, non-uniform slick deposition, etc.). This disagreement may have arisen from the fact that the static and dynamic oil film parameters, introduced in the theoretical relationship in Eq. (3), were taken from laboratory measurements made under different conditions. In the open sea investigation, the monolayer films were not completely and uniformly distributed over the sea surface. In addition, the elastic films spread on the sea surface with different physical properties may break any holes that occur, and the surface oil films will move slowly as a result of Stock's wave drift and currents. The other responsible factor is the roughness of the sea water surface modified by an oil film at lower wind velocities. From the damping ratios

$$D_r(f) = \frac{S_{clean}}{S_{dirty}} \tag{4}$$

obtained in the presence of the insoluble oil substances *in situ*, which are depicted in Fig. 4, it can be seen that a peak in the frequency region of 5–10 Hz is observed in all cases.

The damping intensity (peak height) and peak frequency may be introduced, as the maximum of the damping ratio $D_{r,max}(f)$ and the resonance frequency, f_m (in the so-called Marangoni damping phenomenon). From the theoretical relationship $D_r(f)$, one can see that the respective maximum damping ratio values for Gasoline 94 and Gasoline 86 are 20 and 13, occurring at a maximum frequency of 5 Hz.

The similar peaks of the amplitude fluctuation spectra of scattered acoustic waves on capillary waves are easier to notice for both clean and polluted water and for different wind speeds. This process, some kind of relaxation, can be explained as a physico-chemical nature of the sea surface films, but to clarify the process it is necessary to have more experimental data from *in situ* measurements.

The amplitudes of the acoustic signal as a function of the surface capillary wave's frequency are shown in Fig. 5 in the frequency range 1–50 Hz. The measurements were performed for a clean surface and a surface covered with the oils. All the frequency dependencies of the amplitudes are similar in shape. They have a maximum at relatively low frequencies, between 5-10 Hz, whereas the amplitude above these frequencies is rather low and constant. However, the measurements of the statistical parameters of the scattered signals are performed under the same surface conditions, and they are presented in the following chapter.

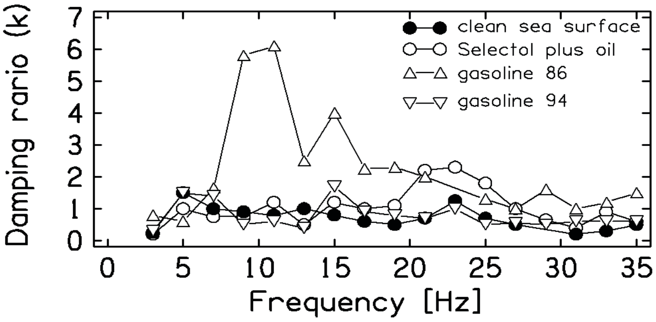

Fig. 4. Acoustically obtained damping ratios, for the oil films studied versus frequency in natural condition and at low wind speeds of 2-2.3 m/s

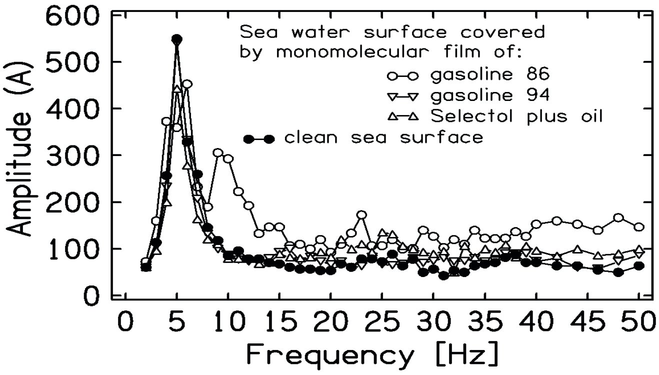

Fig. 5. Spectra of amplitude fluctuations of the signal scattered from water surface coated with oil films at low-wind conditions

To sum up, it can be stated that the measurements of the $D_r(f)$ or the amplitude fluctuation spectra pattern may enable the nature of the surface film to be characterised. It is of course too early to deduce the physical and chemical nature of the sea surface films from the intensity and frequency of this characteristic peak, but it seems feasible, in principle, to characterise surface films by this method.

The mechanism of the water wave damping effect by surface films is not fully understood. The direct influence of the surface film should be considered in terms of the chemical structure of the film's hydrophobic-hydrophilic balance of film-forming compounds.

EFFECT OF OIL LAYER THICKNESS ON THE ATTENUATION OF THE SURFACE WAVES ON WATER

The model material used consisted of five crude oil derivatives; ethyl gasoline 78, lamp oil, Diesel oil, Extra 15 engine oil and Marinoll 111 gear oil. In order to characterise 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 collected in Table 1. The two last columns in the table contain the wettability coefficient $S_{o/w}$, and equilibrium thickness d_∞. The values of $S_{o/w}$ for the first three liquids are positive, which suggests the ability to form on a water surface a layer with a thickness ranging from monomolecular to one resulting from the amount of the liquid and the surface area available.

Extra 15 and Marinoll 111 oils have negative values of $S_{o/w}$; 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 an oil layer thickness, before spreading an oil film, a surface-active agent (Fotonal) was dissolved in water phase. Undergoing absorption, this substance reduced the interfacial water-oil tension, changing the value of wettability coefficient $S_{o/w}$ into a positive one. Thus, it was possible to form a permanent film of both oils with thicknesses ranging from 0.00001 m to the equilibrium thickness d_∞.

Tab. 1. Physical properties of crude oil derivatives and an oil-water system at T= 293 K and pressure of 101.325 Pa

Petroleum derivative	$\rho_b \cdot 10^3$ [$kg \cdot m^{-3}$]	$\eta_b \cdot 10^{-3}$ [$Pa \cdot s$]	$\gamma_b \cdot 10^{-3}$ [$N \cdot m^{-1}$]	$\gamma_{b/a} \cdot 10^{-3}$ [$N \cdot m^{-1}$]	$S_{b/a} \cdot 10^{-3}$ [$N \cdot m^{-1}$]	$d_\infty \cdot 10^{-2}$ [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

RESULTS

The attenuation coefficient of a surface wave with a frequency f = 30 Hz propagating on a water surface covered with a film of crude oil derivative, as a function of its thickness d, is presented in Fig. 6. The value of the coefficient α was determined by means of an instrument described in [23, 24]. Table 2 presents the theoretical values of α_m (1) and the condition of applicability (4) of this expression, for p_d – the penetration depth of the eddy component [25] based on the measurement results from Table 1.

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

when the following condition

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

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

DISCUSSION

The nature of function α(d) presented in Fig. 6, although complicated, has some common features for ethyl gasoline 78, lamp oil, Diesel oil and Extra 15 oil.

The values of α for the thinnest film (0.00001 m) are greater than those predicted by the theory, being equal to $2.44\alpha_m$ for ethyl gasoline, $1.573\alpha_m$ for lamp oil, $1.378\alpha_m$ for Diesel oil and $16.36\alpha_m$ for Extra 15 oil.

For larger thicknesses, the value of α decreases steadily and approaches the values predicted by theory, although they remain higher by several per cent: $1.04\alpha_m$ for ethyl gasoline, $1.015\alpha_m$ for lamp oil, $1.0004\alpha_m$ for Diesel oil and $1.028\alpha_m$ for Extra 15 oil.

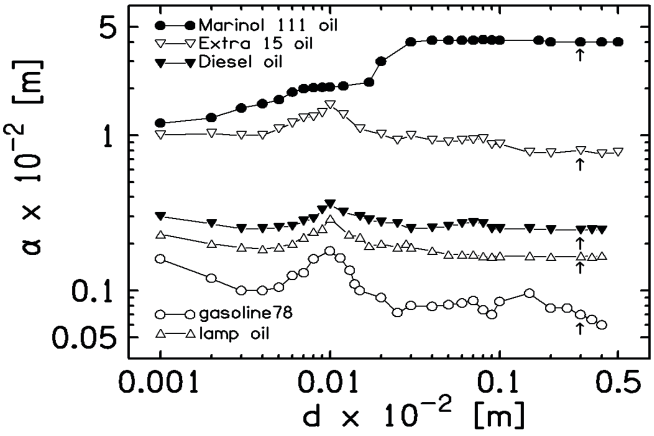

Fig. 6. Attenuation coefficient of surface capillary waves on water covered with a film of crude oil derivatives versus layer thickness. The arrows indicate the thickness of oil layers equal to $l_b/2$

The nature of the relationship α(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). These are collected in Table 2 and determined on the basis of Kelvin's dispersion equation [26]. Their closeness facilitates the process 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 $\lambda_b/2$, i.e. corresponding to the conventional depth of undulatory processes of penetration beneath the surface, the attenuation only slightly (by several per cent) exceeded the theoretical values. The dependence α(d) for Marinoll 111 oil had a completely different character. For an oil film of thickness d = 0.001 m, the value of α = 0.286am is relatively low. For d = 0.0003 m, α reaches the value of 0.989am, and continues to increase until it reaches $1.0013\alpha_m$ for the equilibrium thickness d_∞ (Table 1).

Tab. 2. Applicability condition of Stokes approximation (5)

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

Higher than theoretical values of α most likely result from the partially eddy-like, duct-similar flow of liquid in the vicinity of the undulating surface, as was suggested by the applicability condition of the Stokes expression (4) found in Table 2, for all the investigated liquids. It is fulfilled in the case of the first three liquids of 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 ≈ 0.0001 m.

Recently, several other methods have been used to determine the spatial, seasonal and temporal variability of the elastic and spreading characteristics of oil layers in contact with seawater [29, 30].

CONCLUSIONS

The attenuation of a surface wave propagated on the surface of water covered with an oil film attains higher values than those assumed by the classical hydrodynamic theory. When the thickness of the film approaches or exceeds the generally assumed value which determines the depth of penetration of the undulatory motion $d \geq l_b/2$, the values of the coefficient tend to approach the Stokes expression αm, exceeding it only by several per cent.

The observed instances of the 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 lead to violation of the applicability condition of the Stokes approximation for an ideal liquid.

The depression of the spectral energy density of wind-driven waves by surface films (the relative spectra, so-called contrast K) was inferred from the ratio of acoustic signal fluctuations spectra with/without films, and compared to that resulting from the Marangoni damping theory applicable to monolayers of particular surface viscoelasticity (determined in a separate measurement) and a moderate agreement was found.

The high-frequency two-transducer buoy-like free-drifting scattering system based on a specular forward scattering geometry was successfully applied in at-sea experiments to recover the viscoelastic oil film properties and turned out to be an effective oil pollution detection system.

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CONTACT WITH THE AUTHORS

Stanisław Pogorzelski
e-mail: fizsp@ug.edu.pl
Institute of Experimental Physics, University of Gdańsk ,
Wita Stwosza 57, 80-308 Gdańsk,
Poland

Paweł Rochowski
e-mail: p.rochowski@ug.edu.pl
Institute of Experimental Physics, University of Gdańsk ,
Wita Stwosza 57, 80-308 Gdańsk,
Poland

Maciej Grzegorczyk
e-mail: maciej.grzegorczyk@ug.edu.pl
Institute of Experimental Physics, University of Gdańsk ,
Wita Stwosza 57, 80-308 Gdańsk,
Poland

Ewa Skrodzka
e-mail: afa@amu.edu.pl
Faculty of Physics, Institute of Acoustics,
A. Mickiewicz University,
Umultowska 85, 61-612 Poznań,
Poland

Łukasz Bielasiewicz
e-mail: lbielasiewicz@infoeko.com.pl
INFO-EKO Design-Consulting Office,
Łódzka 50A/3, 80-180 Gdańsk,
Poland

Bogumił B. J. Linde
Adres e-mail: fizbl@ug.edu.pl
Institute of Experimental Physics, University of Gdańsk,
Wita Stwosza 57, 80-308 Gdańsk,
Poland