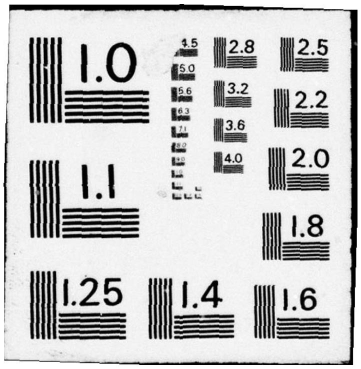
AD-A034 261 DAVID W TAYLOR NAVAL SHIP RESEARCH AND DEVELOPMENT CE--ETC F/G 7/4
DISCRIMINATION OF PARTICULATES FROM OIL IN CONTAMINATED WATER U--ETC(U)
DEC 76 B FRIEDMAN
MAT-76-23
NL

UNCLASSIFIED

1 OF 1
AD-A
034 261

END
DATE
FILMED
2-25-77
NTIS


1.0
4.5
5.0
5.6
6.3
7.1
8.0
9.0
2.8
3.2
3.6
4.0
2.5
2.2
2.0
1.1
1.8
1.25
1.4
1.6

U.S. DEPARTMENT OF COMMERCE
National Technical Information Service

AD-A034 261

DISCRIMINATION OF PARTICULATES FROM OIL IN CONTAMINATED WATER UTILIZING THE ANGULAR INTENSITY DISTRIBUTION OF SCATTERED LIGHT

David W. Taylor Naval Ship Research and
Development Center, Annapolis, Maryland

December 1976

Discrimination of Particulates from Oil in Contaminated Water
Utilizing the Angular Intensity Distribution of Scattered Light

Report MAT-76-23

DAVID W. TAYLOR NAVAL SHIP RESEARCH AND DEVELOPMENT CENTER

Bethesda, Md. 20084

DISCRIMINATION OF PARTICULATES FROM OIL IN CONTAMINATED WATER UTILIZING THE ANGULAR INTENSITY DISTRIBUTION OF SCATTERED LIGHT

by
Bruce Friedman

Approved for public release; distribution unlimited.

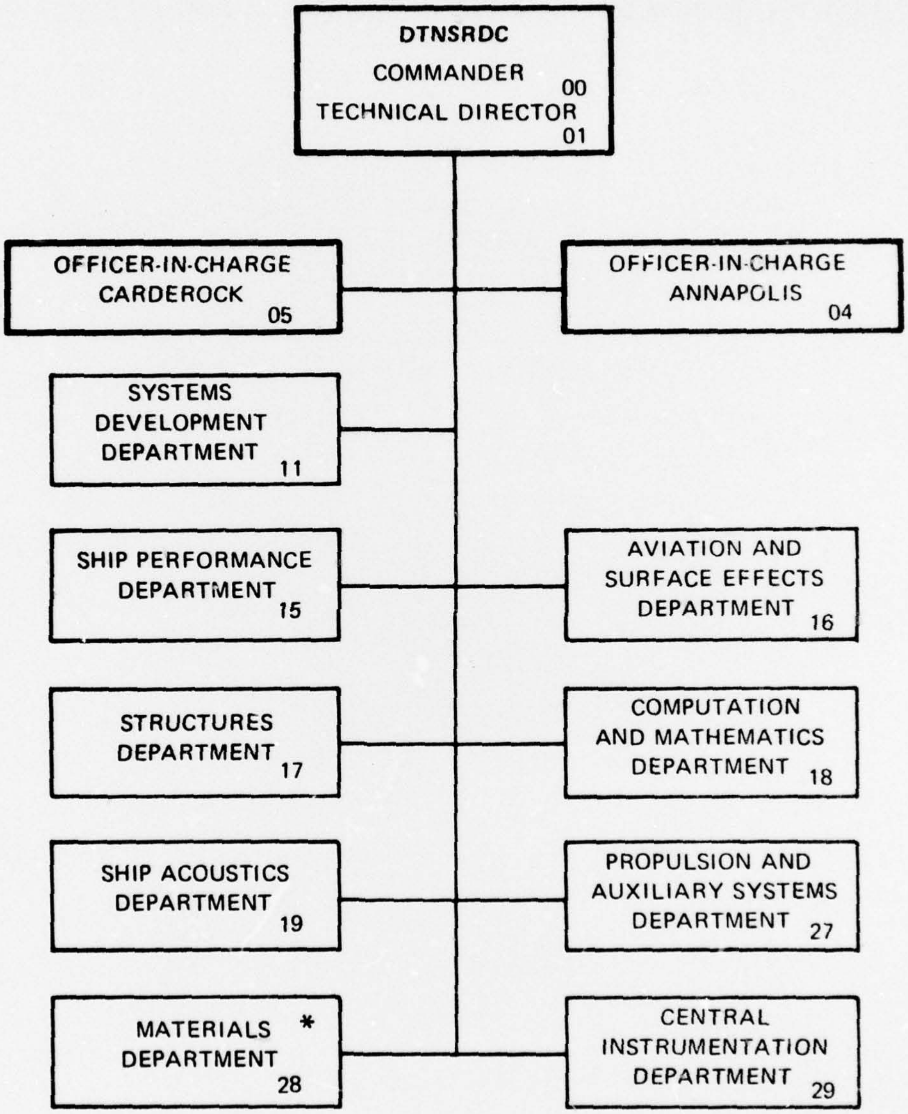
MATERIALS DEPARTMENT
Annapolis

RESEARCH AND DEVELOPMENT REPORT

December 1976

Report MAT-76-23

MAJOR DTNSRDC ORGANIZATIONAL COMPONENTS



UNCLASSIFIED

SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE

READ INSTRUCTIONS BEFORE COMPLETING FORM

1. REPORT NUMBER
MAT-76-23

2. GOVT ACCESSION NO.

3. RECIPIENT'S CATALOG NUMBER

4. TITLE (and Subtitle)
DISCRIMINATION OF PARTICULATES FROM OIL IN CONTAMINATED WATER UTILIZING THE ANGULAR INTENSITY DISTRIBUTION OF SCATTERED LIGHT

5. TYPE OF REPORT & PERIOD COVERED
Research & Development

6. PERFORMING ORG. REPORT NUMBER

7. AUTHOR(s)
Bruce Friedman

8. CONTRACT OR GRANT NUMBER(s)

9. PERFORMING ORGANIZATION NAME AND ADDRESS
David W. Taylor Naval Ship R&D Center
Annapolis, Maryland 21402

10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
Task Area SSL 59001
Task 17509
Work Unit 2862-164

11. CONTROLLING OFFICE NAME AND ADDRESS
David W. Taylor Naval Ship R&D Center
Bethesda, Maryland 20084

12. REPORT DATE
December 1976

13. NUMBER OF PAGES
16

14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)

15. SECURITY CLASS. (of this report)
Unclassified

15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release; distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)
Scattering techniques
Particulate matter
Oily wastes
Angular-intensity distribution

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Light-scattering techniques are used in several oil-in-water monitors, proposed or in existence. Particulate matter which may cause interferences in these monitors is frequenctly found in oily wastes. An analysis is made of the potential of using measurements of the angular-intensity distribution of scattered light for discriminating between oil and particulates. For the purpose of

(over)

cont'd

20. **ABSTRACT** (Cont)

this discussion, the particulate/oil combination in the monitor is approximated by two collections of spheres. It is found that to discriminate between oil and particulates by using only measurements of the angular-intensity distribution of scattered light in a real-life situation would be difficult.

(Author)

ADMINISTRATIVE INFORMATION

This report reflects work carried out under Task Area SSL59001, Task 17509, "Shipboard Oil-in-Water Content Monitor System." The work was performed under Work Unit 2862-164 at this laboratory.

ACKNOWLEDGMENTS

This work was conducted under the sponsorship of NAVSEA; The Program Manager is Dr. F. Ventriglio, NAVSEA (SEA 0331F). Technical management was provided by NAVSEC; Mr. W. Auer, NAVSEC (SEC 6159D), is the Technical Agent.

TABLE OF CONTENTS

ABSTRACT

Light-scattering techniques are used in several oil-in-water monitors, proposed or in existence. Particulate matter which may cause interferences in these monitors is frequently found in oily wastes. An analysis is made of the potential of using measurements of the angular-intensity distribution of scattered light for discriminating between oil and particulates. For the purposes of this discussion, the particulate/oil combination in the monitor is approximated by two collections of spheres.

It is found that to discriminate between oil and particulates by using only measurements of the angular-intensity distribution of scattered light in a real-life situation would be difficult.

INTRODUCTION

A major difficulty in the utilization of light-scattering techniques for the accurate determination of the concentration of oil in water is interference due to the presence of particulate matter. The question which this report addresses is whether or not measurements of the angular-intensity distribution of scattered light can be used to discriminate between particulates and oil in water and thus eliminate such interference.

SCATTERING BY ASSEMBLIES OF SPHERES AND IRREGULAR PARTICLES

For the purposes of this discussion, the particulate matter present is taken to consist primarily of irregularly shaped, opaque particles. The oil present is taken to consist predominantly of free-floating transparent spheres. It is assumed that the particles and spheres are illuminated by a beam composed of parallel rays of light. It is also assumed that the particles and spheres are larger than a few wavelengths of the monochromatic light used. When these size conditions hold for a system of particles having a range of sizes, the combined light scattering can be treated, approximately, as a combination of classical diffraction and gometrical transmission and reflection. This allows a considerable computational simplification as compared to the use of the rigorous electromagnetic treatment of the Mie theory.[1,2]

First, consider scattering by an assembly of irregular, transparent particles. The three contributions to the scattering,

---

[1] Superscripts refer to similarly numbered entries in the Technical References at the end of the test.

diffraction, reflection, and refraction, are treated separately. The discussion of the three contributions follows essentially the treatment by Hodkinson[3].

DIFFRACTION

Diffraction occurs when a system of waves, electromagnetic in this case, travels past the edge of an obstacle. Diffraction is primarily related to the bending of the waves around the edge of the obstacle into the region of shadow. In the region beyond the obstacle, this gives rise to a system of maxima and minima of illumination produced by interference among the waves and known as diffraction fringes.[4]

In the case of a spherical obstacle, the symmetry of the object leads to the main diffraction maximum appearing about a line through the center of the sphere in the direction of the incident wave propagation. If the three-dimensional light distribution is examined, the main diffraction maximum is seen to have the form of a lobe with its narrowest width in the vicinity of the spherical surface. Thus, for a sphere, the forward diffraction lobe refers to the main diffraction maximum of light intensity which apppears in the forward direction.

Consider now, the case of a collection of various-sized particles or spheres. If there is a sufficient range of particle size, 2:1 or more, only the main diffraction maximum is seen; the higher order maxima, which would appear at angles to the forward direction, being smoothed out by the superposition of the patterns of particles or spheres of different sizes.

The diffraction patterns of individual, irregular objects vary considerably with the shape of the objects. However, the resultant forward diffraction lobe for an assembly of irregular particles in random orientation is not expected to differ much from that for an assembly of spheres equal in projected area to the irregular particles.[3]

REFLECTION

Both the surface of a sphere and a random assembly of irregular particle surfaces offer equal probabilities of reflection for all angles of incidence. Thus, the angular distribution of the light scattered by external reflection is the same for an assembly of irregular particles as for spheres.[3]

REFRACTION

The phenomenon of refraction refers to the passage of a portion of the wave incident on the boundary between two media from the first medium into the second medium, in general with a change in direction of propagation of the wave in the second medium.[4]

For the same reason as presented in the section entitled "Reflection," the average angular distribution of refracted light after the first refraction, as it enters the irregular particles, will be the same as for spheres. However, in contrast to the situation for a sphere, wherein a ray of light leaving a sphere at its second refraction deviated further away from its original path by the same amount and in the same direction as at the first refraction, a light ray upon leaving an irregular particle will not necessarily be deviated by the same amount and/or in the same direction as at the first refraction. Consequently, the more irregular the particle, the more the refraction scattering is concentrated forward. For an assembly of transparent, irregular particles, then, the refraction scattering is concentrated more forward than for an assembly of transparent spheres.[4]

Consider now, the case of assemblies of opaque, irregular particles. Here, the scattering differs even less from that of assemblies of opaque spheres, because there is scattering by diffraction and external reflection only.

As a result of the above analysis, the particulate matter is seen as now further approximated to consist of a size distribution of opaque spheres, so that the aggregate of oil, particulates, and water is now considered to be composed of two assemblies of spheres, one transparent and the other opaque, in water. Let the further hypothesis be made that both assemblies of spheres are nonabsorbers of light. Also, let it be taken that the two size distributions for the transparent and opaque spheres are identical.

This discussion leads us to conclude that the contributions to the scattering angular-intensity pattern occurring by difration from the two assemblies of spheres are the same, since the diffraction is independent of the nature of the particle. At this point, the diffraction contribution to the scattering angular-intensity distribution is ignored, because it cannot be used to discriminate between the particulates and oil.

RELATIONSHIP BETWEEN GEOMETRICAL AND DIFFRACTION SCATTERING[1]

As the ratio between the diameter of a sphere and the wavelength of the light incident upon it increases, it becomes increasingly valid to consider scattering to be composed of two components, geometrical (reflection and refraction) and diffraction scattering. However, there is no distinct size at which this division into two types of scattering becomes exactly possible. In principle, the division is always vague. The Mie theory yields the unified pattern of both effects.

A necessary condition for the separation into geometrical and diffraction scattering, besides the ratio of the sphere circumference to the wavelength being much greater than one ($x \equiv 2\pi a/\lambda >> 1$, where a is the sphere radius and λ is the wavelength of the light), is that the phase shift $\phi = (m_{part} - m_{water})\, x >> 1$, where m_{part} is the index of refraction of the sphere and m_{water} is the index of refraction of the medium, i.e., water, in which it is immersed.

Only half of the total scattering from the sphere is due to reflection and refraction. The other half originates from the diffraction around the sphere and composes the Fraunhofer diffraction pattern.

The geometrical optics pattern which arises from the reflection and refraction of the rays that impinge upon the sphere is wide and of moderate intensity. The diffraction pattern which arises from the incompleteness of the wave front passing the sphere is narrow, very intense, and concentrated near the forward direction.

SCATTERING BY OIL AND PARTICULATES

For the hydrocarbons encountered in oil, the ratio of the indices of refraction of the hydrocarbons to that of water, i.e., the relative indices of refraction, lies in the range of about 1.00 to 1.15.[5] This means that almost all of the light incident on a typical oil sphere which is scattered by reflection or refraction is scattered by being twice refracted through the sphere. At least 88% of the light incident on the sphere which is scattered by reflection or refraction would be scattered by being twice refracted.[1]

Expressed in mathematical terms,

$$I(\Theta) = I_o(\Theta) + I_1(\Theta) + I_2(\Theta) \tag{1}$$

where $I(\theta)$ is the total scattering intensity at angle Θ, I_o is the diffraction component, I_1 is the reflection component, and I_2 is the refraction component.[2] Also,

$$I_o = x^2 \frac{J_1^2 (x \sin \Theta)}{\sin^2 \Theta} \tag{2}$$

$$I_1 = I_{1,1} + I_{1,2} \ , \tag{3}$$

$$I_{1,1} = \frac{x}{8}^2 \left\{\frac{\sin\left(\frac{\Theta}{2}\right) - \left[m^2 - 1 + \sin^2\left(\frac{\Theta}{2}\right)\right]^{1/2}}{\sin\left(\frac{\Theta}{2}\right) + \left[m^2 - 1 + \sin^2\left(\frac{\Theta}{2}\right)\right]^{1/2}}\right\}^2 , \tag{4}$$

$$I_{1,2} = \frac{x}{8}^2 \left\{\frac{m^2\sin\left(\frac{\Theta}{2}\right) - \left[m^2 - 1 + \sin^2\left(\frac{\Theta}{2}\right)\right]^{1/2}}{m^2\sin\left(\frac{\Theta}{2}\right) + \left[m^2 - 1 + \sin^2\left(\frac{\Theta}{2}\right)\right]^{1/2}}\right\}^2 , \tag{5}$$

$$I_2 = 2x^2 \left(\frac{m}{m^2 - 1}\right)^4 \frac{\left[m\cos\left(\frac{\Theta}{2}\right) - 1\right]^3 \left[m - \cos\left(\frac{\Theta}{2}\right)\right]^3}{\cos\left(\frac{\Theta}{2}\right)\left[m^2 + 1 - 2m\cos\left(\frac{\Theta}{2}\right)\right]^2}$$

$$\cdot \left[1 + \sec^4\left(\frac{\Theta}{2}\right)\right] , \tag{6}$$

where Θ is the scattering angle from the direction of propagation of the incident wave, m is the relative index of refraction, J_1 is the first order Bessel function, $I_{1,1}$ is the component of the reflected wave polarized in the plane containing the incident and scattered rays, and $I_{1,2}$ is the component of the reflected wave polarized normal to the plane containing the incident and scattered rays. The factor $x = 2\Pi a/\lambda$, where a is the sphere radius and λ is the wavelength of the light.

For the case of oil hydrocarbons, the index of refraction, m, is written as $1 + \Delta$, where Δ is very small compared to one. This gives the approximations

$$I_{1,1} = I_{1,2} = \frac{x}{8}^2 \left\{\frac{\sin\left(\frac{\Theta}{2}\right) - \left[2\Delta + \sin^2\left(\frac{\Theta}{2}\right)\right]^{1/2}}{\sin\left(\frac{\Theta}{2}\right) + \left[2\Delta + \sin^2\left(\frac{\Theta}{2}\right)\right]^{1/2}}\right\}^2 , \tag{7}$$

$$I_2 = 2x^2 \left(\frac{1+\Delta}{16\Delta^4(1+2\Delta)}\right)\left\{\frac{\left(-1 - \cos^2\left(\frac{\Theta}{2}\right) + 2\cos\left(\frac{\Theta}{2}\right)\right]^3 + 6\left[-1 - \cos^2\left(\frac{\Theta}{2}\right) + 2\cos\left(\frac{\Theta}{2}\right)\right]^2\left[\cos\left(\frac{\Theta}{2}\right) - \cos^2\left(\frac{\Theta}{2}\right) - 1\right]\Delta}{4\cos\left(\frac{\Theta}{2}\right)\left(\left[1 - \cos\left(\frac{\Theta}{2}\right)\right]^2 + 2\left[1 - \cos\left(\frac{\Theta}{2}\right)\right]\left[1 - 2\cos\left(\frac{\Theta}{2}\right)\right]\Delta\right)}\right\}$$

$$\left(1 + \sec^4\left(\frac{\Theta}{2}\right)\right) . \tag{8}$$

For small angle scattering in the forward direction, i.e., in the vicinity of 0° the following approximations are obtained

$$I_o^{0°} = x^2 \frac{J_1^2(x\Theta)}{\Theta^2}, \tag{9}$$

$$I^{0°}_{1,1} = I^{0°}_{1,2} = \frac{x^2}{8}\left\{\frac{\frac{\Theta}{2} - \left(2\Delta + \frac{\Theta^2}{4}\right)^{1/2}}{\frac{\Theta}{2} + \left(2\Delta + \frac{\Theta^2}{4}\right)^{1/2}}\right\}^2 \approx \frac{x^2}{8}\left[\frac{\frac{\Theta}{2} - \sqrt{2\Delta}}{\frac{\Theta}{2} + \sqrt{2\Delta}}\right]^2, \tag{10}$$

$$I_2^{0°} = \frac{16\Delta^2 x^2}{\left(8\Delta^2 + \Theta^2\right)^2}. \tag{11}$$

Equation (11) differs from the result of Mie theory analysis, $I_2^{0°} = (4\Delta^2 x^2)/(4\Delta^2 + \Theta^2)^2$ because equation (11) is obtained as an approximation to the geometrical optics theory.

It is interesting to note that as the scattering angle approaches zero, $I_2^{0°}/I_1^{0°}$ approaches $1/\Delta^2$. Furthermore, $I^{0°}_{1,1}/I_o^{0°}$ approaches $(1/2)(x^{-2})$. For the oil hydrocarbons, Δ^{-2} is of the order of 10^2. Also, again as the scattering angle approaches zero, $I_2^{0°}/I_o^{0°}$ approaches $(\Delta x)^{-2}$, so that if x is large enough the diffraction component of the scattered light in the forward direction can become greater than the refraction contribution.

Consider now the scattering as the angle Θ approaches 90°. There is no contribution to the refraction component of the scattering because of the $[m \cos(\Theta/2) - 1]^3$ term which becomes negative when $\cos(\Theta/2) < 1/m$. Hence, for the case of the oil hydrocarbons, there is no refractive scattering for angles greater than about 35° from the forward direction.

For scattering angles in the vicinity of 90°,

$$I_o^{90°} = x^2 J_1^2(x), \tag{12}$$

$$I^{90^\circ}_{1,1} = I^{90^\circ}_{1,2} = \frac{\Delta^2 x^2}{32 \sin^2 \left(\frac{\Theta}{2}\right)} . \qquad (13)$$

In the vicinity of 90°, $I^{90^\circ}_{1,1}/I^{90^\circ}_{o}$ approaches $\Delta^2/(32\ J_1^2\ (x))$. For x considerably larger than one, $J_1(x)$ can be approximated by its asymptotic expansion.[6] In this event, for the purposes of this study, $I^{90^\circ}_{1,1}/I^{90^\circ}_{o}$ is very grossly approximated by $(\Pi\Delta^2 x^3)/(36\ \sin^2(x-3\Pi/4))$. It appears that in the case of right-angle scattering, the reflection component can become much greater than the diffraction component, depending on the size of the spheres.

Now, compare $I^{0^\circ}_{o}$ to $I^{90^\circ}_{o}$. The ratio $I^{0^\circ}_{o}/I^{90^\circ}_{o}$ is given by $x^2/(288\ \sin^2\ (x-3\Pi/4))$. For oil droplets of the size under consideration wherein x is considerably larger than 1.0, the diffraction component of scattering in the forward direction is much greater than the diffraction component of scattering at right angles to the forward direction.

Again, compare $I^{0^\circ}_{1,1}$ to $I^{90^\circ}_{1,1}$. The ratio $I^{0^\circ}_{1,1}/I^{90^\circ}_{1,1}$ is given by $2/\Delta^2$. It is seen that the reflection component of the scattering in the forward direction is much greater (about 10^2) than the reflection component of the scattering at right angles.

One conclusion to be drawn from the discussion to this point is that a light-scattering oil-in-water monitor based on right-angle scattering must be more sensitive than one based on forward scattering. This conclusion arises from the finding that if the scattering pattern of oil in water in the absence of particulates is examined, most of the incident light is found to be scattered in the forward direction with very little at right angles to the incident light.

Consider now the scattering pattern of opaque particulates spheres. Assume that, as is possible in reality, the index of refraction is much larger than 1.0. Then, as is seen from equations (4) and (5), the intensity of the light reflected is essentially independent of direction, i.e., a smooth, totally reflecting sphere with radius large compared to the wavelength scatters light by reflection isotropically. Consequently, the scattering is less forward-directed than in the case of an oil sphere.

Suppose now, than an opaque particulate sphere is composed of metal; that is, it has a complex index of refraction

$m = m_o\ (1+jk)$, where m_o and k are real, $j = \sqrt{-1}$, and k is called the attenuation or extinction coefficient. The absorption represented by k is sufficiently large to absorb any refracted ray completely. In this event, the intensity of the reflection component of scattering again becomes a function of the scattering angle Θ. The strength of the dependence on Θ is a function of the magnitudes of m_o and k to each other. As compared to the isotropic situation for the opaque nometallic sphere, the pattern of reflected light is somewhat stronger in the forward direction. However, the forward directionality is not as pronounced as in the case of the transparent oil sphere.

The intensity of light scattering in a particular direction, $I(\Theta)$, from an oil and particulate mixture in water is summed up by the following equation:

$$I(\Theta) = \sum_i N^i_{oil} \left({}^iI^{oil}_{refl} + {}^iI^{oil}_{refr} \right) + \sum_i N^i_{part}\ {}^iI^{part}_{refl} + \sum_i \left(N^i_{oil} + N^i_{part} \right) {}^iI_{diff} \ , \tag{14}$$

where N^i_{oil} and N^i_{part} are the number of oil droplets and the number of particulate particles of a particular size i, respectively. Also, ${}^iI^{oil}_{refl}$ and ${}^iI^{oil}_{refr}$ are the reflection and refraction components of the scattered light intensity due to an oil droplet of a given size i; ${}^iI^{part}_{refl}$ is the average reflection component of the scattered light intensity due to a typical particulate particle of a size i; and ${}^iI_{diff}$ is the average diffraction component of the scattered light intensity due to an oil droplet or typical particulate particle of size i.

Consider what happens when the particulate concentration increases while the oil concentration remains constant as opposed to the case when the oil concentration increases while the particulate concentration remains constant. It should be possible to discriminate between oil and particulates in both cases, because the scattering for the oil is more forward-directed than for the particles. In the first case, the right-angle scattering should increase more rapidly than the forward scattering while in the second case the opposite is true.

In a real-life situation, the concentrations of oil and particulate can and do vary independently and randomly. This situation renders interpretation of data difficult.

CONCLUSION

As a result of the considerations presented herein, it is reasonable to conclude that there is only a small chance of developing an oil-in-water monitor, based on light-scattering angular-intensity distribution measurements, alone, that would discriminate between oil and particulates.

TECHNICAL REFERENCES

1 - Van De Hulst, H. C. Light Scattering by Small Particles, New York, John Wiley and Sons (1957)

2 - Hodkinson, J. R., and I. Greenleaves, "Computation of Light Scattering and Extinction by Spheres According to Diffraction and Geometrical Optics, and Some Comparisons with the Mie Theory," Jour. Opt. Soc. Amer., Vol. 53, No. 5, p. 577 (May 1963)

3 - Hodkinson, J. R. Light Scattering and Extinction by Irregular Particles Larger Than the Wavelength. In: M. Kerker (ed.), ICES Electromagnetic Scattering, London, Pergamon Press, Vol. 5, pp. 87-100 (1963)

4 - Wood, Robert W., Physical Optics, 3rd Ed., New York, Macmillan Company (1934)

5 - Rossini, Frederick D., et al. Hydrocarbons from Petroleum. New York, Book Division, Reinhold Publishing Corp. (1953)

6 - Watson, G. W., A Treatise on the Theory of Bessel Functions, 2nd Ed., Cambridge, University Press (1966)