Experimental measurements of light scattering from samples with specified optical properties

Khadija Tahir\textsuperscript{1} and Christopher Dainty\textsuperscript{2}

\textsuperscript{1} Photonics Group, Blackett Laboratory, Imperial College, London SW7 2BW, UK
\textsuperscript{2} Applied Optics Group, Department of Experimental Physics, National University of Ireland, Galway, Republic of Ireland

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Abstract

We have measured the photon intensity transmitted through cylindrical and slab phantoms of different concentration and size scattering particles. Our continuous wave (CW) measurements show quantitatively that the mean cosine of the scattering angle ($g$) must be taken into account unless the phantoms have very high scattering coefficient. We have compared the results for phantoms made of titanium dioxide in resin, which is commonly used to simulate tissue despite its low $g$ value (0.56), with those for silica in resin phantoms of high $g$ value (0.95). The comparison has shown different angular scattered intensity, both in form and magnitude, for phantoms that have the same reduced scattering coefficient but different $g$ value. The results show that, in some cases, using the reduced scattering coefficient and absorption alone for comparing phantoms or for reconstruction using inverse methods based, for example, on diffusion theory would lead to incorrect interpretations.

Keywords: scattering, diffusion theory

1. Introduction

During the process of light–tissue interaction, scattering causes photons to travel a longer distance in tissue than the geometric spacing between source and detector positions. The beam that is transmitted into the tissue undergoes processes of multiple scattering and absorption. Multiple-scattering events occur for virtually all photons propagating through tissue, producing a wide range of paths varying with absorption and scattering. Thus, most photons have been randomly scattered hundreds or thousands of times before emerging to be detected.

The idea of using optical radiation to penetrate highly scattering media, combined with image reconstruction methods for recovering optical parameters inside the media is an old concept and has been used for over a century [1–4]. Early time windows have been used to image phantom objects through chicken breast [5] and fish body [6], and for optical localization of bone in fingers [7]; also certain features of the time-of-flight curve have been collected and used to image whole animals, animal organs and infant brain and fluid [8] and also for real time imaging of the brain in neonatal piglets [9]. During the last decade, with the advances in measurements, and also in theoretical and practical understanding [10, 11] of the image reconstruction problem, tremendous progress has been made in using diffuse optical tomography in medicine, including the detection and classification of tumours from breast tissue [12–14], monitoring of infant brain tissue oxygenation level [15–17] and functional brain activation studies [18–21]. Some of these applications are based on the fact that absorption of near-infrared light in biological tissue depends on the oxygenation level of the tissue [22–24]. Three-dimensional optical tomography of the premature infant brain was published by Hebden et al [25], whilst Chen et al [26] tried to find the correlation between near-infrared spectroscopy and magnetic resonance of rat brain oxygenation modulation, and Dahghani et al [27] have used multi-wavelength three-dimensional tomography of the breast. Gibson et al [28] describe a method for generating patient-specific finite element meshes for head modelling. Siegel et al [29] describe the temporal comparison of
functional brain imaging with diffuse optical tomography and functional magnetic resonance imaging during rat forepaw stimulation. Barnett et al. [30] highlighted the growing interest in diffuse optical tomography (DOT) as a noninvasive tool for neuro-imaging as well as breast tumour detection, tracking muscular oxygenation and arthritic joint imaging. They placed this method somewhere between MEG (magnetoencephalography) and EEG (electroencephalography) as regards both the fast temporal resolution and moderate spatial resolution.

Kolehmainen et al. [31] proposed a new numerical approach to the nonstationary optical (diffusion) tomography problem. Prince et al. [32] studied the time-series estimation of biological factor optical diffusion tomography. Martelli et al. [33] presented a procedure for retrieving the optical properties of a two-layered diffuse medium based on an exact analytical solution of the diffusion equation and on relative multistatence time-resolved reflectance measurements. Zaccanti et al. [34] presented results of measuring optical properties of high density media. Li et al. [35] introduced a modified Tikhonov regularization method to include three-dimensional x-ray mammography as a prior in the diffuse optical tomography reconstruction and have shown through a simulation study that spatial information provided by another imaging modality (such as x-ray or MRI) can be used as a prior in the diffuse optical image reconstruction to improve the image contrast to noise ratio and resolution. The results they have presented in their paper are for reconstruction of absorption images and they claim that the same framework can be used to image the scattering coefficient. Culver et al. [36] presented volumetric diffuse optical tomography of brain activity in a rat. Corlu et al. [37] derived conditions for the unique and simultaneous recovery of chromophore concentration and scattering coefficients in multispectral continuous wave (CW) diffuse optical tomography.

A study of the papers that have been published in optical tomography during the last few years shows that the scattering theory employed is diffusion theory. Diffusion theory has been used for both forward and reconstruction techniques as it is simple and fast, and it is widely believed to be appropriate for highly scattering media such as the human tissue. Although diffusion theory is an important tool, it does have some well known limitations, which have been highlighted by several researchers. Dorn [38] has published the transport–back-transport (TBT) algorithm for calculating the difference between the computed and the physically given measurements for a fixed source. He has shown that the TBT method can be used to reconstruct and to distinguish between scattering and absorbing objects in the case of large mean free path. In his summary and conclusion he stated that reconstruction methods that use the diffusion approximation to model photon propagation in the medium have two main disadvantages:

First, diffusion models generally do not consider the directional behavior of the photon density inside the medium. So, part of the information about the hidden objects is unavailable for the reconstruction process. Second, diffusion models are reliable only some distance away from the boundary. If there are also some regions of large mean free path, diffusion models cannot be used at all to model photon propagation in these regions.

Arridge [39] has concluded that although the diffusion approximation is by far the most prevalent approach, it is not completely accurate and its ability to differentiate between two coefficients appears to be very limited. Firbank et al. [40] have shown that diffusion theory cannot handle a sharp discontinuity of scattering properties in regions that are relatively clear, as in the case of cerebrospinal fluid. Hielscher [41] has shown the influence of boundary conditions on the accuracy of diffusion theory in time-resolved reflectance spectroscopy of biological tissues. Researchers in the field of optical imaging [4, 10, 42, 43] have considered a higher order radiation transport solution but the most practical application has been restricted to relatively low order P3 approximation. Dahgiani et al. [44] have shown that in the presence of a clear region, the diffusion approximation of photon transport is no longer valid and therefore they used the transport code DANTSYS [45].

In optical tomography, near-infrared light is directed through tissue or a tissue-like phantom and measurements of the transmitted light are made on the tissue or phantom boundary. Data acquisition can be achieved with time domain, frequency domain and continuous wave (cw) techniques. The CW approach is the simplest of the three methods. The inverse problem associated with CW diffuse optical tomography does not have unique solutions [10, 11] and multiple sets of optical parameters can yield identical data for CW measurements [37]. In this study we are not dealing with human tissue but with phantoms made with different concentration and different particle size. The CW method is ideal for this study as it is simple and easy to interpret compared with others.

Our measurements reported below demonstrate the extent of the ‘well known’ limitations of the diffusion approximation, which we attribute to the fact that in the diffusion approximation there is insufficient information about the scattering medium. This is shown by comparing two phantoms of different scattering particles but the same reduced scattering coefficient. The other important point we have highlighted in our results is that using low g value scattering particles to represent the human tissue in phantoms is not adequate unless the reduced scattering is higher than 1.0 mm\(^{-1}\). As far as we know, there are no published experimental results to show that using the reduced scattering cross section \(\mu_s'\) as the optical property (used in diffusion theory to replace both the scattering cross section \(\mu_s\) and the mean cosine \(g\) of the scattering angle) can give erroneous results in some cases. Our experimental results reported below strongly suggest that diffusion theory, which uses only the reduced scattering cross section, is inadequate for such cases. It could be argued that human tissue has a large value of the reduced scattering coefficient, so there is no need to worry about the limitations that the diffusion approximation imposes for lower values. However, the literature [46] shows that there are some organs with values of \(\mu_s'\) less than 1.0 mm\(^{-1}\), for example at 633 nm bladder tissue \(\mu_s' = 0.26–0.35\) mm\(^{-1}\), white matter \(\mu_s' = 0.2\) mm\(^{-1}\), heart \(\mu_s' = 0.36\) mm\(^{-1}\) and lung \(\mu_s' = 0.18\) mm\(^{-1}\). Our conclusion is that diffusion theory should not be used blindly for all cases, and also that, in phantoms, the \(g\) value should be considered; for example TiO\(_2\) phantoms (\(g = 0.558\)) could give incorrect results if they are used to represent tissue of less than 1.0 mm\(^{-1}\) reduced scattering coefficient.
2. Experimental work

We have constructed scattering samples of simple geometry and carried out careful measurements of the scattered light. The mean intensity of scattered radiation has been measured over a large dynamic range using photon counting techniques. Our results show that using the reduced scattering coefficient \( \mu'_s \) for comparing phantoms of different scattering particles and concentration is not always correct as in that case we are neglecting their difference in mean cosine \( g \) of the scattering angle. The reduced scattering coefficient \( \mu'_s \), the scattering coefficient \( \mu_s \) and the mean cosine \( g \) of the scattering angle are related by

\[
\mu'_s = \mu_s (1-g).
\]  

Figure 1. Schematic diagram of the experimental set-up.

We have used a simple photon counting system as shown in figure 1. It consists of a helium–neon laser (Edmund Optics Ltd) attenuated by neutral density filters (Optima Research Ltd) for different concentration phantoms. A single-mode fibre coupler (Newport, UK) is used to connect and couple the beam into a single-mode fibre (Elliot Scientific Ltd, UK). The other end of the fibre is fixed at the centre of the slab or at a fixed position, defined as the zero angle, around the cylinder. The cylinder was surrounded by a circular collar divided into 20 positions at 18° intervals for the detection fibre. A plastic multimode fibre (Comar Instruments, UK) collects the signal along the slab or at different angles around the cylinder and this is connected to a cooled avalanche photon counting module (EG&G). A counter/timer module (CT2, Electron Tubes, Ruislip, UK), converts the PC into a high performance pulse counting instrument. The intensity measurements for cylindrical samples and slab samples are relative, but all measurements for the cylindrical samples were made with the same incident intensity (approximately \( \pm 2\% \)) within one series. Several solid phantoms of cylindrical and slab geometry shown in figure 2 were built using a solution of epoxy (MY753) and hardener (XD716) from Robnor Resins Ltd and according to the Mie equation [47–50] \( g = 0.558 \). In the case of silica spheres (Bangs Microspheres, USA), two types were used: the first was a solution with average particle diameter 0.81 \( \mu \)m and refractive index of 1.37, giving \( g = 0.930 \). The second silica sample is a powder with average particle diameter 0.97 \( \mu \)m, standard deviation 0.10 \( \mu \)m and refractive index of 1.37; \( g = 0.945 \). It should be noted that for most tissues [46], \( g > 0.9 \), so the embedded silica samples provide a better approximation to tissue scattering properties. Several concentrations of the \( \text{TiO}_2 \) and silica were used to give phantoms of different reduced scattering, which were moulded into cylinders of diameter 32 \( \pm \) 1 mm or into large (110 by 100 mm) slabs of thickness 15 \( \pm \) 1 mm.

3. Results

The values of the reduced scattering coefficient that we quote below are based on experimentally measured values of the mean particle diameter, which have an error of the order of 2%, the published values of refractive index for the various materials and the weighed values of the particles and embedding medium. The resultant errors in the reduced scattering coefficient are less than 2% in the worst case (for the \( g = 0.95 \) case). It is very unlikely that experimental error accounts for the large observed departures we observe below from the predictions of diffusion theory. Absorption is negligible in our samples.

Figure 2. Cylindrical and slab phantoms.

In figures 3(a) and (b) show the scattered intensity, as measured at 18° around a cylinder which is loaded with different concentrations of \( \text{TiO}_2 \) particles, giving reduced scattering coefficients \( \mu'_s \) in the range 0.25–1.48 \( \text{mm}^{-1} \) and scattering coefficients \( \mu_s \) in the range 0.56–3.36 \( \text{mm}^{-1} \). In figure 3(a), the symmetrized data are plotted and in figure 3(b) the data are normalized to the first value (measured intensity at 18°). Note that, for the most weakly scattering sample, there is a peak intensity in the forward direction (180°), as one would expect intuitively.

In figures 4(a) and (b), we compare scattered intensities in cylindrical samples made with \( \text{TiO}_2 \) and silica particles. Figure 4(a) shows the symmetrized data and in figure 4(b) the scattered intensity is normalized as in figure 3(b). Note...
to match the silica sample, we need to make a TiO$_2$ sample.

The scattered intensities are completely different. In fact, in order to match the silica sample, we need to make a TiO$_2$ sample with TiO$_2$ particles.

When the particle concentrations are adjusted to have the same reduced scattering coefficients of 0.25 mm$^{-1}$, the scattered intensities are completely different. In fact, in order to match the silica sample, we need to make a TiO$_2$ sample with a reduced scattering coefficient of 0.37 mm$^{-1}$ (solid squares in figures 4(a) and 4(b)), approximately 50% higher than that for the silica sample (whose $g$ value has a better match to tissue).

It is clear from this result that any reconstruction method that deduces optical scattering coefficients based on diffusion theory must lead to unreliable results in such a case. Of course the difference will become less pronounced as the absolute value of the reduced scattering coefficient increases, and the values here are fairly low compared to those for many tissues.

Figures 5(a) and (b) show some scattering measurements for the slab geometry. We have chosen the slab geometry as this allows analytical solution for the diffusion approximation (apart from complications at the boundaries), and may therefore be of more value for computer modelling. The scattered relative intensities for different concentrations of TiO$_2$ particles in slabs are shown in figure 5(a): note that all the curves were divided by their value at the centre of the slab for comparison purposes. Figure 5(b) shows the results from TiO$_2$ particles in slabs and silica particles in slabs. As in the case for the cylindrical samples (figure 4(b)), note that the scattered intensities are quite different for slabs of equal reduced scattering coefficient and that the best match between TiO$_2$ and silica particle slabs is when the reduced scattering coefficient for TiO$_2$ is much greater than that for the silica particle slab.

Several phantoms of 32 mm diameter were made with cylindrical holes of 8 mm diameter and the hole in each cylinder was filled with a scattering mixture of different reduced scattering coefficients to the original main cylinder. Intensities were measured in each case for the cylinder alone (without the inhomogeneity) and with the inhomogeneity. Figure 6 shows the cross section of a cylindrical phantom with the four chosen positions for the inhomogeneity. P1 is where the inhomogeneity is close to the source, in P2 it is close to the 180° detector, in P3 it is close to the 90° detector and in P4 it is close to the 270° detector. Three sets of samples were prepared. In the first set, a highly scattering mixture of TiO$_2$ in resin was chosen for the main cylinder and a much lower scattering TiO$_2$ mixture was chosen for the internal cylinder. In the second set, a lower scattering TiO$_2$ mixture than the first case was used for the main cylinder with the same TiO$_2$ scattering internal cylinder as in the first case. In the third case, the main cylinder has silica particles (whose $g$ value is more typical of tissue), while the internal cylinder is TiO$_2$ of the same reduced scattering but different scattering coefficient due to the difference in the mean cosine of scattering $g$.

Figure 7(a) shows the results for the first case when the main cylinder is TiO$_2$ in a resin mixture and has a
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Figure 5. Scattering from slab geometries. (a) Resin slabs with TiO2 particles (g = 0.56). Key to symbols: □ μs = 3.36 mm\(^{-1}\), μs’ = 1.48 mm\(^{-1}\); ○ μs = 1.68 mm\(^{-1}\), μs’ = 0.74 mm\(^{-1}\); ▲ μs = 0.84 mm\(^{-1}\), μs’ = 0.37 mm\(^{-1}\); ▼ μs = 0.64 mm\(^{-1}\), μs’ = 0.28 mm\(^{-1}\); * μs = 0.56 mm\(^{-1}\), μs’ = 0.25 mm\(^{-1}\).

(b) Comparison of the relative intensity of scattered light for resin slabs with TiO2 particles (g = 0.56) and silica particles (g = 0.95). Key to symbols: □ TiO2 μs = 3.36 mm\(^{-1}\), μs’ = 1.48 mm\(^{-1}\); * TiO2 μs = 0.56 mm\(^{-1}\), μs’ = 0.25 mm\(^{-1}\); ○ silica μs = 4.47 mm\(^{-1}\), μs’ = 0.25 mm\(^{-1}\); ▲ silica μs = 1.61 mm\(^{-1}\), μs’ = 0.09 mm\(^{-1}\).

Figure 6. Cross section of a cylindrical phantom with immersed cylinders.

Figure 7. Relative intensity of scattered light for a resin cylinder with TiO2 particles (μs = 1.68 mm\(^{-1}\), μs’ = 0.74 mm\(^{-1}\)) with a lower scattering cylinder also with TiO2 particles (μs = 0.25 mm\(^{-1}\), μs’ = 0.11 mm\(^{-1}\)), as in the geometry of figure 6. (a) Symmetrized data, (b) symmetrized data normalized to the 18° value, (c) plot between scattering angles of 72° and 288°. Key to symbols: □ no internal cylinder; ○ internal cylinder at position 1; ● internal cylinder at position 2; ▲ internal cylinder at position 3; ▼ internal cylinder at position 4.

The scattering coefficient \(\mu_s\) of 1.68 mm\(^{-1}\) and a reduced scattering coefficient \(\mu_s'\) of 0.74 mm\(^{-1}\), while the inner cylinder has a scattering coefficient \(\mu_s\) of 0.25 mm\(^{-1}\) and reduced scattering \(\mu_s'\) of 0.11 mm\(^{-1}\). It should be noted that in this case the reduced optical depth of the inner cylinder is less than one, and therefore diffusion theory would not be valid. The four curves in this figure represent different positions for the inner cylinder during the photon counting measurements. Figure 7(b) shows normalized results after dividing each curve by the outcome at its first detector (18°). Figure 7(c) shows the results between the 72° and 288° detectors, where the effect of the position of the inner cylinder in the outcome is more noticeable. Note that, when the inner cylinder is closest to the point of entry of the light, there is the greatest effect on the measured intensity for all of the detectors. In contrast, when the inner cylinder is furthest from the source, i.e. at position P2, only the intensities close to P2 are altered (small peak at 180°). It is from curves such as these, where the differences are relatively small, that one might attempt to solve the inverse problem in the CW case, and clearly this is a difficult problem. It is improbable that the small differences can be explained by diffusion theory.
The second case, where the main cylinder is TiO$_2$ in a resin mixture and has a scattering coefficient $\mu_s$ of 0.70 mm$^{-1}$ and reduced scattering coefficient $\mu'_s$ of 0.31 mm$^{-1}$ while the inner cylinder is also TiO$_2$ in a resin mixture and has a scattering coefficient $\mu_s$ of 0.25 mm$^{-1}$ and reduced scattering coefficient $\mu'_s$ of 0.11 mm$^{-1}$, is shown in figure 8(a). The three curves in this figure represent the results for the main cylinder alone and with the inner cylinder at two different positions P1 and P2. Figure 8(b), shows normalized results after dividing each curve by the outcome at its first detector (18°). Figure 8(c), shows the results for the detectors between 72° and 288°. As in figure 7, when the inner cylinder is closest to the point of entry of the light (open circles in figures), there is the greatest effect on the measured intensity in all of the detectors, and when the inner cylinder is furthest from the source, i.e. at position P2, only the intensities close to P2 are altered.

Figures 9(a) and (b) show the light scattering results for the third case when the main cylinder is of silica, which has a scattering coefficient of 2.00 mm$^{-1}$ and reduced scattering coefficient of 0.11 mm$^{-1}$. The inner cylinder is still made of TiO$_2$ particles of scattering coefficient 0.25 mm$^{-1}$ and reduced scattering coefficient of 0.11 mm$^{-1}$. This means that the outer and inner cylinders have the same reduced scattering ($\mu'_s$) but different scattering ($\mu_s$) coefficients, as the value of $g$ for silica is 0.95 while $g$...
for TiO$_2$ is 0.56. Figure 9(b), shows the normalized results (obtained by dividing each curve by the first detector’s signal).

The above curves represent the main cylinder alone, then with inner cylinder at four different positions P1, P2, P3 and P4. Figure 9(c), shows the results between the detectors at 72° and 288°. The results in figure 9 differ from those in figures 7 and 8 in that the inhomogeneity has a different concentration and is not comprised of the same particles (different $g$ values) as the original phantom although it has the same reduced scattering coefficient. A calculation using diffusion theory would not detect the presence of the inner cylinder, since the reduced scattering coefficients of the inner and outer cylinders are the same, yet figure 9(c) shows the effect of the inner cylinder in each position very clearly. When the inhomogeneity is in position P1 (near the source) the effect is seen in all the detectors (even the first and last detectors, 18° and 342°). When the inhomogeneity is at P2 (180°) the effect is seen only at and close to the 180° detector. When the inhomogeneity is placed at P3 (near 90°) or P4 (near 270°) the effect on the detector at 180° is very small (in comparison with the results for the uniform phantom), but in the case of the P3 position the effect is mainly in the detectors of the upper part of the axis and vice versa for the P4 case.

All the data presented in figures 3–9 are available in Excel on request from the authors. Modelling our experimental arrangement accurately is not trivial, since various reflections have to be taken into account, but we hope this will be the subject of future work by others.

4. Conclusion

The application of diffusion theory is very widespread in optical tomography, particularly for solving the inverse problem where Monte Carlo techniques are slow and cumbersome and use of full radiation transport theory is difficult. Our experimental results show that, for the range of reduced scattering coefficients studied, the use of diffusion theory is incorrect, both for forward and inverse problems. Different tissues have a wide range of reduced scattering coefficients [46]. The values used in our experiments are undoubtedly on the low side for real tissues but still may be encountered. If such low values of the reduced scattering coefficient are encountered, a theory other than diffusion theory must be used to explain the results, or used to solve the inverse problem. One hopes that a full radiative transfer solution would provide a better description of the forward scattering process, although it may be necessary eventually to use a full electromagnetic approach. A positive aspect of our measurements is that scattering parameters of immersed bodies do produce measurable and distinguishable signals, as for example in figures 7–9, where the different positions of the immersed cylinders give different scattered intensities: in principle, both $\mu'_s$ and $g$ could be separately determined. We believe it is important to carry out further simple and well defined scattering experiments of the kind described in the paper. It is tempting to use “more realistic” shapes of phantoms but the scattering properties of the very simplest geometries (slabs and cylinders) need to be properly understood first.

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