Optical properties of oakwood in the near-infrared range of semi-transparency

LEONID DOMBROVSKY,1,2 JEAN-FRANÇOIS HENRY,3 CLARISSE LORREYTE,3 HERVÉ PRON,3 AND JAONA RANDRIANALISOA3,*

1Joint Institute for High Temperatures, 17A Krasnokazarnennaya St., Moscow 11116, Russia
2Tyumen State University, 6 Vолодарский St., Tyumen 625003, Russia
3Groupe de Recherche en Sciences Pour l’Ingénieur - GRESPI (EA 4694), University of Reims Champagne–Ardenne, Reims 51687, Cedex 2, France
*Corresponding author: jaona.randrianalisoa@univ-reims.fr

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The spectral absorption and scattering properties of oakwood are retrieved from the measurements of both the normal-hemispherical reflectance and transmittance in the visible and near-infrared ranges of semi-transparency. We employ two alternative methods for the radiative transfer in wood samples: the modified two-flux approximation and the high-order discrete ordinate method. The modifications of both methods take into account the effect of total internal reflection at both surfaces of the wood samples. The analytical approximate solution of the first method gives very accurate results for the absorption coefficient, but the transport scattering coefficient of wood appeared to be systematically underestimated. Fortunately, this error is between 7% and 12%, and that is acceptable for the estimates. The oakwood samples of four different thicknesses were used in the experiments. The effect of the wood cell orientation appears to be insignificant and can be observed in the reflectance from optically thin samples only. There is a considerable decrease in the transport scattering coefficient of oakwood with the wavelength. This effect is explained by a predominant contribution of micron-sized longitudinal pores in oakwood. The latter is used to develop an approximate theoretical model of scattering based on the rigorous solution for arbitrary-oriented cylindrical pores. The model suggested is in good agreement with the experimental data.

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1. INTRODUCTION

The highly porous layer of oakwood chips has been used as a relatively pure reference material in the Groupe de Recherche en Sciences Pour l’Ingénieur (GRESPI) laboratory studies of heating, drying, and pyrolysis of organic waste. The computational model for the complex multi-stage thermo-chemical process is considered to be a promising procedure for industrial production of a synthesis gas and bio-diesel fuels [1]. Thermal radiation is one of the important heat transfer modes in this technology, and the spectral optical properties of wood are necessary for the complete computational modeling. This was a primary motivation of the present study.

The optical properties of both softwood and hardwood in the visible and near-infrared spectral ranges have attracted permanent attention of researchers during many years because of quite different applications (from the architecture to the bio-fuel industry). Some interesting experimental data can be found in recent publications [2–6]. However, to the best of our knowledge, the broad spectral properties of dried oakwood have not been reported in the literature.

The objective of this particular study is twofold: (1) to retrieve the main absorption and scattering properties of oakwood on the basis of spectral measurements of normal-hemispherical reflectance and transmittance for thin samples in the near-infrared range of semi-transparency, and (2) to develop a simple but sufficiently accurate physical model for the spectral behavior of scattering by wood samples.

The study of the wood optical properties is not a simple task because wood is a complex and variable material with different types of cells aligned mainly along the vertical axis of the tree. It is known that wood is highly organized anisotropic material, containing regular structures of tracheid vessels (composed of lignin and cellulose) that serve the transport of fluids in trees. Numerous high-resolution images of typical wood morphology can be found in well-illustrated work [7].

It is worth noting that softwood exhibits a relatively simple cellular structure, and it would be easier to suggest a theoretical model for a softwood. However, oakwood is of main interest in this study because it is an abundant forest waste in Champagne–Ardenne region, and the authors are studying...
pyrolysis and gasification of this waste. Thermal radiation plays an important role in these thermochemical processes, and optical properties of wood are considered as input values in the computational model.

2. EXPERIMENTAL PROCEDURE
The experimental data for spectral hemispherical transmittance and reflectance were obtained in the wavelength range from 0.4 to 2.5 \( \mu \text{m} \). A setup based on the double-monochromator spectrometer Lambda 9 (Perking Elmer Corp., USA) was combined with a barium sulfate-coated integrating sphere (inner diameter of 60 mm) equipped with a PbS cell detector. The photograph of the laboratory setup is shown in Fig. 1. The normally incident beam of rectangular cross-section (10 mm \( \times \) 4 mm) was used in experiments for all the wood samples.

The overall experimental uncertainty of about 2% comes mainly from the reflectance and transmittance measurements for different irradiated areas due to the sample inhomogeneity.

3. EXPERIMENTAL DATA FOR OAKWOOD
Four samples of thin oakwood slabs cut from the same block of hardwood have been selected for the measurements. These samples with different thicknesses (see Table 1) were characterized by the same measured density equal to 735 kg/m\(^3\).

Some typical fragments of scanning electron microscopy (SEM) images of these wood samples are shown in Fig. 2. The image presented in Fig. 2(a) is the most impressive, and it is similar to those given in paper [7]. One can see the channel-like pores of pentagonal cross section of approximately 10 \( \mu \text{m} \) in size and a wall thickness of approximately 3 \( \mu \text{m} \). This complex structure of the hardwood is formed during the growth of the tree. The SEM image in Fig. 2(b) illustrates that the front (illuminated) side of the sample has a rough surface with some open-pore channels, which are clearly noticeable on this image due to the sample preparation.

The results of spectral measurements of normal-hemispherical reflectance, \( R_{n-h} \), and transmittance, \( T_{n-h} \), in the wavelength range of \( 0.5 \leq \lambda \leq 2.5 \) \( \mu \text{m} \) at the same orientation of anisotropic wood samples with numbers from 1 to 4 (see Table 1) are presented in Fig. 3. The data for shorter wavelengths are not shown in this figure because of very large absorption, which is not interesting for the present study.

For a preliminary analysis of experimental data, it is convenient to introduce the value of normal absorbance:

\[
A_n = 1 - R_{n-h} - T_{n-h}.
\] (1)

The calculated spectral dependences of \( A_n \) (Fig. 4) are similar to each other for all the samples. This can be treated as a confirmation of approximately the same chemical composition of the oakwood samples. The details of chemical composition of wood can be found in the monograph [8].
Note that the case of $\mu_c = 0$ (at $n = 1$) corresponds to the ordinary two-flux model. The intermediate angle interval $-\mu_c < \mu < \mu_c$ gives no contribution to the radiative flux. Therefore, the term "two-flux" is applicable to the modified approximation too.

For convenience of a reader, the analytical solution for both the normal-hemispherical reflectance and transmittance obtained with the use of the modified two-flux approximation is reproduced below:

$$
R_{n-h} = R_{0_{n-h}} + D(1 + B/\zeta + C),
$$  \hfill (3a)

$$
T_{n-h} = T_{0_{n-h}} + D[A/\zeta + (1 + \rho_n)E],
$$  \hfill (3b)

$$
R_{0_{n-h}} = \rho_n + \frac{(1 - \rho_n)^2 C}{1 - \rho_n C},
$$  \hfill (3c)

$$
T_{0_{n-h}} = \frac{(1 - \rho_n)^2 E}{1 - \rho_n C},
$$  \hfill (3d)

$$
A = \frac{k_1 (\varphi s + c) E + k_2}{(1 + \varphi s) s + 2qc}, \quad B = \frac{k_1 E + k_2 (\varphi s + c)}{(1 + \varphi s) s + 2qc},
$$  \hfill (3e)

$$
D = \frac{2 \omega_{\alpha \beta} 1 - \rho_n}{\zeta^2 - 1 - \rho_n C},
$$  \hfill (3f)

$$
\left( \begin{array}{c} k_1 \\ k_2 \end{array} \right) = \left( \begin{array}{c} 1 \\ -1 \end{array} \right) \frac{1}{C} - (1 + 2 \varphi) \left( \begin{array}{c} \rho_n \\ 1 \end{array} \right),
$$  \hfill (3g)

$$
\varphi = \frac{2 \sqrt{1 - \omega_{\alpha \beta}}}{\zeta}, \quad \gamma = \frac{1 - \rho_n}{1 + \rho_n}, \quad \zeta = 2 \sqrt{1 - \omega_{\alpha \beta}}, \quad \rho_n = \left( \frac{n-1}{n+1} \right)^2,
$$  \hfill (3h)

where the values of $R_{0_{n-h}}$ and $T_{0_{n-h}}$ correspond to a contribution of the collimated radiation. $\rho_n$ is the normal reflectivity of the interfaces, $t_{\alpha \beta}^n = \beta_n d$ is the transport optical thickness of the sample with thickness $d$, $\beta_n = \alpha + \sigma_n$ is the transport extinction coefficient, $\alpha$ and $\sigma_n$ are the absorption coefficient and transport scattering coefficient, respectively, and $\omega_{\alpha \beta} = \sigma_n/\beta_n$ is the transport albedo of the medium. The coefficients of Eqs. (3a)–(3h) are spectrally dependent. However, the subscript $\lambda$ is omitted in the designations for brevity. It should be noted that the singularity of the analytical solution at $\zeta = 1$ does not lead to additional difficulties because it is sufficient to use the solution for $\zeta \neq 1$ in practical calculations.

Note that the same expression for $\rho_n$ is used in the above analytical solution, not only for the collimated radiation, but also for the diffuse radiation component, as was done in Ref. [10]. It is not quite correct, and more accurate solution should be based on averaging of the reflectance recommended in Ref. [14] (as in Refs. [15,16]). It was shown in Ref. [17] by comparison with the exact numerical solutions that this correction improves the results in the case of a strongly refracting medium. However, there is no need in this correction for the problem under consideration because both the front (illuminated) and back (shadow) surfaces of wood samples are not smooth [Fig. 2(b)]. The inverse problem solution with the use of the above-presented analytical expressions is not mathematically difficult, and there is no need to discuss the corresponding mathematical procedure [11].

The data for the normal-hemispherical reflectance and transmittance are insufficient to retrieve the scattering phase function. Fortunately, the error of the transport approximation is usually insignificant. However, this error may be comparable with that of the modified two-flux approximation [10].
The transport approximation was also used in the reference solution, based on the high-order discrete ordinate method (DOM) [18–20]. The total internal reflection at the interfaces is taken into account in the DOM numerical solution according to Eq. (2). Separate sets of the Gaussian quadrature points were used in each angular interval. The details of the computational procedure can be found in monograph [11].

5. SPECTRAL PROPERTIES OF WOOD

The resulting spectral dependences of both the absorption coefficient and transport scattering coefficient are presented in Fig. 5. To the best of our knowledge, there are no data in the literature for the index of refraction of oakwood in the spectral range under consideration. Fortunately, the wood is weakly absorbing in a significant part of the spectral range under consideration. Therefore, the index of refraction is almost constant in this range. To estimate the effect of uncertainty in the index of refraction, the calculations were conducted at two constant values of an average refractive index. One can see in Fig. 5 that both \( \alpha \) and \( \sigma_{tr} \) are weakly sensitive to the variation of \( n \).

The DOM calculations confirm good accuracy of the absorption coefficient retrieved using the modified two-flux approximation [see Fig. 5(a)]. However, the error of the approximate analytical method appeared to be considerable (from 7% to 12%) for the transport scattering coefficient [Fig. 5(b)]. Fortunately, this is a systematic deviation over the spectrum, whereas the character of spectral dependences \( \sigma_{tr}(\lambda) \) obtained using two different methods is the same. It should be also recalled that there is a small error of all the calculations due to the transport approximation of the scattering phase function [10].

The peaks of scattering at \( \lambda = 0.85 \, \mu m \) and \( 1.9 \, \mu m \) are not explained by errors of the retrieval procedure. The microscopic heterogeneities in the wood structure are responsible for these peaks.

The transport scattering coefficient varies from one sample of wood to another. It is clear from the comparison of an average difference in \( \sigma_{tr} \) between the samples (approximately 20%) and the above estimate of a relatively small retrieval error. This result seems to be physically correct because of a natural variation of the wood morphology from one sample to another.

Relatively strong scattering obtained for the first sample [Fig. 5(b)] can be partially explained by the effect of anisotropy of oakwood. This effect is more pronounced for thin samples, whereas the anisotropy can be ignored for thick samples because the multiple scattering of light leads to a smooth angular dependence of the light intensity even at small distances from the illuminated side of the sample. In addition, a contribution of rough surfaces shown in Fig. 2(b) (this roughness is not taken into account in the computational models) to the sample scattering is more significant for thin samples. In further analysis, we consider the scattering data for samples 2–4 to minimize the specific effects typical for the thinnest sample.

It is important that the absorption coefficient is practically the same over the spectrum for all the samples. The only difference is in a lower value of \( \alpha \) for sample 4 in the narrow wavelength range from 0.9 to 1.3 \( \mu m \) in the transparency window. This result confirms a close chemical composition of the wood samples. In the case of homogeneous materials, it is convenient to use the dimensionless index of absorption, which is one of the spectral optical constants:

\[
\kappa = \alpha \lambda / (4\pi).
\]
Of course, wood samples are not homogeneous. However, this approach and Eq. (4) enable us to estimate a conventional average value of $\kappa$. The spectral curves $\kappa(\lambda)$ calculated for sample 2 are shown in Fig. 6. Note that even the minimum values of $\kappa$ are not small as compared with the absorption index of pure water that increases from about $10^{-7}$ to $10^{-3}$ in the wavelength range from 0.8 to 2 $\mu$m [21]. It means that the presence of a small amount of water in the wood sample cannot affect the optical properties of oakwood in the spectral range under consideration. Obviously, the effect of water on light scattering is significant if water fills the pores of wood, but it is not the case.

It is interesting that the spectral position of the transparency range obtained for oakwood is approximately the same as that for some other biological tissues. In particular, one can recall the so-called therapeutic window for superficial human tissues in the wavelength range from 0.6 to 1.4 $\mu$m [22–25].

It was shown in papers [26,27] that a simple physical approach is applicable to many semi-transparent materials of different nature. This approach is based on the following major principles: (1) the absorption coefficient is independent of the material morphology and directly proportional to the volume fraction of the absorbing substance, and (2) the scattering is insensitive to a weak absorption and can be predicted by an analysis of the material morphology. In doing so, it is especially important to take into account a contribution of particles and pores of size comparable with the radiation wavelength because of the so-called Mie scattering [11].

A relative mutual independence of absorption and scattering is not a specific property of semi-transparent porous materials. This is a general behavior of disperse systems containing the so-called optically soft particles satisfying the conditions of the Rayleigh–Gans theory [11,28,29].

It was demonstrated in papers [30,31] that there are some cases when scattering really depends on absorption. Of course, the spatially uniform absorption cannot lead to the radiation scattering. On the contrary, the local regions with higher or lower absorption lead to an additional scattering. This effect takes place when the size of these local regions is comparable with the wavelength. Obviously, this effect is not observed for the wood samples. In other words, the transport scattering coefficient does not depend on absorption.

There is a general feature of retrieved spectral dependences of transport scattering coefficient: the value of $\sigma_t$ decreases considerably with the wavelength. It means that very thin longitudinal particles or pores in the wood are mainly responsible for the scattering [11]. This spectral behavior of scattering is also similar to that for other biological tissues, including the human tissues. In the latter case, the micron-sized organelles in the biological cells and thin membranes of the cells determine the spectral behavior of scattering.

Another interesting effect is related to the orientation of wood cells. Of course, the wood sample orientation does not practically affect the normal-hemispherical transmittance because of multiple scattering of light in the sample. As one can expect, the normal-hemispherical reflectance of a thin sample is the most sensitive value with respect to the orientation of wood fibers. This is illustrated in Fig. 7, where the measurements for sample 2 are presented. The effect of different orientation of the wood sample on the values of $R_{n-h}$ indicates the optical anisotropy of wood in the spectral range of semi-transparency.

Strictly speaking, the analytical method employed for the radiative transfer calculations can be generalized for the case of anisotropic materials. However, there is no need in more accurate solution containing additional parameters to be retrieved because of insignificant difference between the values of $R_{n-h}$ at different orientations of wood cells.

6. THEORETICAL MODEL OF SCATTERING

The above analysis showed that there are some longitudinal micron-sized particles or pores in the oakwood responsible for the observed decrease in transport scattering coefficient with the wavelength. However, it is difficult to estimate the volume fraction of these particles. This uncertainty makes reasonable the hypothesis of independent scattering [32–36] to develop a simple theoretical model of the wood optical properties.

The hypothesis of independent scattering means that each particle is assumed to absorb and scatter the radiation in exactly the same manner as if other particles did not exist. In addition, there is no systematic phase relation between partial waves scattered by individual particles during the observation time interval, so that the intensities of the partial waves can be added...
without regard to phase. In other words, each particle is in the far-field zones of all other particles and scattering by individual particles is incoherent.

The analytical solution to the scattering problem for the infinite homogeneous cylinder, illuminated along the normal to the axis, was derived for the first time by Rayleigh in 1881 [37], that is, earlier than the Mie solution for spheres appeared [38,39]. The solution to the scattering problem for cylinders at oblique illumination was derived by Wait in 1955 [40]. One often comes across disperse systems of cylindrical particles randomly oriented in space (isotropic system) or in parallel planes (transversely isotropic system). In these cases, it is convenient to introduce the characteristics averaged over orientations. For randomly polarized radiation, this averaging has been developed by Lee [41,42]. Following the early paper by the first author [43], the relations obtained by Lee are used in modeling of scattering by wood samples. According to the above-reported experimental results, it is sufficient to consider isotropic media with totally random orientation of longitudinal/cylindrical particles.

In the case of some longitudinal particles or pores in a wood, the relative index of refraction (with respect to that of the ambient/host medium), \( n_r = n/\overline{n}_0 \), may be close to 1. Such particles are called “the optically soft,” and the corresponding approximation could be named that of optically soft particles. The condition \( |n_r - 1| \ll 1 \) is insufficient even for weakly absorbing media for radical simplification of the problem to the case of the so-called Rayleigh–Gans scattering because the following two conditions should be satisfied [28]:

\[
|\overline{n} - 1| \ll 1, \quad 2\pi|x|/\lambda \ll 1, \quad \text{(5)}
\]

where \( x = 2\pi a/\lambda \) is the diffraction (size) parameter for the particle of radius \( a \). It is not obvious that the Rayleigh–Gans theory is applicable to the problem under consideration. Therefore, the exact solution to the scattering problem is used in the paper.

The classical solution for the scattering of light by a particle relates to the case when the particle is surrounded by vacuum. It was proved in Refs. [44,45] that the same formulas are also valid for particles immersed in a refracting and absorbing medium with an insignificant modifications of input data and coefficients. In the simplest case of weakly absorbing media, it is sufficient to replace the values of \( n \) and \( x \) by the values of \( \overline{n} \) and \( \overline{x} = \overline{n} \overline{x} \) in the calculations.

The porosity of wood is high even for the oakwood, and three kinds of pores are usually distinguished: the macropores, mesopores, and micropores [46,47]. The calculations based on the rigorous scattering theory showed that contribution of small macropores with a radius of approximately 0.5 \( \mu m \) to the scattering observed in the wavelength range under consideration is the most important. Therefore, one can consider the simplest monodisperse model of thin pores to describe the spectral dependencies presented in Fig. 5(b). The DOM calculations showed the decrease in \( \sigma_n \) with the wavelength from about 27 mm\(^{-1} \) at \( \lambda = 0.5 \mu m \) to 19 mm\(^{-1} \) at \( \lambda = 2.5 \mu m \) for wood samples 2–4. The following simple relation is used for the theoretical prediction of this dependence [11]:

\[
\sigma_n = \frac{2pQ^m}{\pi a}, \quad \text{(6)}
\]

where \( p \) is the volume fraction of pores with radius \( a \), and \( Q^m \) is the transport efficiency factor of scattering. For definiteness, the index of refraction of a surrounding medium is assumed to be equal to the realistic value of \( n_0 = 1.4 \). The results of calculations for randomly oriented pores are presented in Fig. 8. One can see that a 10% or 10.5% volume fraction of cylindrical pores with a radius approximately between 0.55 and 0.6 \( \mu m \) give the predicted spectral behavior of \( \sigma_n \), which is very close to that retrieved from the measurements for oakwood samples.

7. CONCLUSIONS

The spectral optical properties of oakwood were retrieved in the visible and near-infrared range of semi-transparency. It was shown that the error of modified two-flux approximation is insignificant, and this approach can be employed for good estimates. One can also ignore the anisotropy of wood in the case of optically thick samples.

The obtained decrease in scattering with the wavelength is explained by a predominant effect of thin longitudinal pores of the wood. This is confirmed by good theoretical predictions based on the suggested theoretical model. The results obtained are expected to be useful in solving some engineering problems, including complex thermo-chemical processes of bio-fuel production.

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