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# 1 Dry powders reflectance model based on 2 enhanced back-scattering: case of hematite 3 $\alpha - \text{Fe}_2\text{O}_3$

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9 **Abstract:** By performing Bidirectional Reflectance Distribution Function (BRDF) measure-  
10 ments, we have identified back-scattering as the main phenomena involved in the appearance of dry  
11 nano-crystallized powders. We have developed an analytical and physically based BRDF model  
12 which relies on the enhanced back-scattering theory to accurately reproduce the measurements  
13 performed on optically thick layer of dry powders with various grains' morphology. Our results  
14 are significantly better than the ones obtained with previously existing models. The model has  
15 been validated against the BRDF measurements of multiple synthesized nano-crystallized and  
16 mono-disperse  $\alpha - \text{Fe}_2\text{O}_3$  hematite powders. We also discuss the ability of our model to be  
17 extended to other material or more complex powder morphology.

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## 19 1. Introduction

20 Accurate reproduction of powdered material appearance is an important issue in many domains  
21 such as environmental sciences, teledetection, digital prototyping, entertainment and art preserva-  
22 tion. Powder materials are ubiquitous in real life, either used with a binder like pigments in most  
23 painting techniques and in some cosmetics such as lipstick, or dry such as spices, dry soils, dust,  
24 and pigments. The latter are particularly important as they are involved in parietal paintings from  
25 paleolithic to neolithic era. Some of them have been subject to many degradations processes to  
26 the point of becoming almost illegible. The digital reproduction of such cultural heritage artifacts  
27 through photo-realistic images synthesis will be of great interest for their preservation and will  
28 promote their broadcasting to a large audience.

29 Their appearance and more specifically the color of a powder material depends on both its  
30 chemical composition and on its morphometry, which refers to the size and the shape of its  
31 grains. This hypothesis is well known and particularly well justified in the case of pure  $\alpha - \text{Fe}_2\text{O}_3$   
32 hematite powders that can show various shades going from light red to purple [1,2]. The chemical  
33 composition of the material is involved in the absorption phenomena, while the morphology of  
34 the grains influences the way light is scattered by the material. Such light/matter interactions at  
35 the nanometric scale result in the macroscopic material appearance an observer perceive.

36 The light scattering by a material is numerically described using a *Bidirectional Scattering*  
37 *Distribution Function* (BSDF) model. Most of them assume that the light is mainly scattered at  
38 the surface of an object, around the specular direction that is to say in the direction opposite to  
39 the incident direction. Depending on the roughness of the surface, a more or less diffuse look  
40 is obtained. These models are not suitable in the case of dry powders for two reasons. First,  
41 powders are a medium composed of a collection of randomly oriented scatterers, so light is  
42 scattered multiple times in the volume of the medium, which is not taken into account by the  
43 surface models. To overcome this issue, one may use the Mie scattering theory [3]. This is  
44 relevant for powders diluted into a binder, and takes into account volume scattering depending on

45 the size and shape of the grains. But in the absence of a binder the grains composing the powder  
46 are too close to each other to actually be modeled using such model [2]. The second issue is that  
47 surface scattering models do not account for any scattering in the backward direction.  
48

49 In this paper, we show that contrary to the prediction of most BSDF models the major  
50 contribution to a dry powder BSDF is not the specular peak but the back-scattering peak. We  
51 propose an analytical and physically-based BSDF model involving six parameters able to simulate  
52 the appearance of optically thick layer of dried pigments. We separate the model into two  
53 independent contributions: volume scattering and surface scattering. The former decomposes  
54 into single scattering and multiple scattering responsible for the sharp back-scattering lobe while  
55 the latter accounts for the specular scattering. This model has been developed and experimentally  
56 validated for seven hematite  $\alpha - \text{Fe}_2\text{O}_3$  powders of different morphology. Our model reproduces  
57 the color variations of the studied samples, and can be applied to other pigments.  
58 In Section 2 we will review previous works on BSDF models suitable for the description of dry  
59 powder materials, and more particularly on models that take into account the back-scattering  
60 contribution. The theoretical background necessary for the modeling of light scattering in a  
61 particle medium is introduced in Section 3. The  $\alpha - \text{Fe}_2\text{O}_3$  samples and their measurements are  
62 described in Section 4. The description of our appearance model is done on Section 5 while in  
63 Section 6 we present its results, showing that it fits well the experimental BRDF of the studied  
64 samples. We conclude and give some future work directions in Section 7.

## 65 2. Previous work

66 Most of the BSDF models that are used in physically based renderers assume that the light is  
67 mainly scattered at the surface of an object, and in the specular direction which is to say the  
68 direction opposite to the incident direction. According to these models, the color of the rendered  
69 materials only depends on their chemical composition through the optical constants. In the case  
70 of participating media, the appearance also depends on physical properties and more particularly  
71 on the morphology of the involved scatterers. For diluted media, this is well described by the  
72 Lorenz-Mie scattering theory [3,4].

73 Dry powders and pigments are even complex materials composed of a quite dense collection of  
74 particles with nanometric to micrometric grains' size. In the absence of a binder, the dry grains  
75 are too close to each other to be modeled using Mie scattering theory [2]. Their appearance is the  
76 result of absorption and scattering phenomena, so its description suggests the full understanding  
77 of the interactions of light with the material at both its surface and in its volume.

78 The incoming light is partially scattered at the surface of the material. This phenomenon can  
79 be empirically modeled using the microfacet model [5] by considering that the surface is made  
80 of small but larger than wavelength specular micro facets with known orientations distribution.  
81 Alternatively, the generalized Harvey-Shack theory [6] allows to predict the reflectance of a  
82 rough surface while taking into account the diffraction effects caused by surface asperities. It  
83 accurately simulates the appearance of opaque and homogeneous materials but only considers  
84 single scattering.

85 As we will show in this paper, in the case of pigments an important amount of the incoming  
86 light is scattered backwards, responsible for a high peak back into the incident direction. This  
87 scattering phenomena, known as *back-scattering*, has already be observed on other dry powders  
88 such as meteorites soils [7], and has already been taken into account in some BRDF models: the  
89 spectralon model, the RTLSR model and the enhanced back-scattering model.

### 90 2.1. Spectralon

91 An empirical one has been developed by [8] to model the BRDF of the Spectralon®, a material  
92 that resembles a powder since it is made out of a collection of compressed polymer particles.

93 This model is assumed to be the summation of four independent scattering phenomena, occurring  
94 at both the surface and in the volume of the material. As it involves a large number of material-  
95 dependent parameters optimized in the specific case of Spectralon, it can be adapted to reproduce  
96 with a high fidelity a large range of materials. Nevertheless, these parameters has no explicit  
97 relationship with the chemical composition nor the physical parameters of the considered material  
98 so they cannot be predicted, but only deduced from measurement fittings.

## 99 2.2. *RTLSR*

100 The RTLSR (Ross-Thick Li-Sparse Reflectance) model [9, 10], is a semi-empirical model used in  
101 teledetection for simulating the BRDF of forest canopies. It decomposes the BRDF into a volume  
102 scattering contribution deduced from an approximation of the radiative transfer theory (RT) [11],  
103 and surface scattering which is inferred according to purely geometrical considerations [12].  
104 An optimization factor allows to control the proportion of each of the contributions. Initially  
105 developed for thick and sparse canopies, extensions of this model have been proposed for thin and  
106 dense media [13]. [14] brought modifications to the model for the study of snow, and multiple  
107 works [12, 15, 16] agree that it can be used for bare soils although they do not explicitly introduce  
108 the changes that should be done in such a case. Because of its unique controllable parameter, this  
109 model is too rigid and remains unable to reproduce fine appearance variations.

## 110 2.3. *Enhanced Back-scattering*

111 A deeper analysis of the back-scattering phenomena (see Section 3.5) occurring in a scattering  
112 medium refers to *Enhanced Back-Scattering (EBS)*, which describes it as the result of constructive  
113 interferences [17], [18]. Contrary to the RT theory, the scattered fields are supposed to be  
114 correlated. As a result, the EBS is the sum of the two terms of RT theory (*single and multiple*  
115 *scattering*) and a third one accounting for the constructive interferences in the backward direction.  
116 As the analytical results are deduced within the diffusion approximation, only isotropic particles  
117 are considered. Nevertheless, [17] extended the model to account for anisotropic scattering in the  
118 case of normal incidence.

## 119 3. Background

120 As a dry powder layer involves a large number of scatterers with relatively high density, it  
121 is complex to describe the scattering of light in such a medium. Most of the time, it can be  
122 done using the radiative transfer theory (RT) [19], [20], which accounts for both single and  
123 multiple scattering inside the medium. As it relies on the assumption that the scattered fields  
124 are uncorrelated, RT is unable to predict the appearance of materials which exhibits a strong  
125 back-scattering contribution. In this section, let's overview the theoretical aspects and hypotheses  
126 useful for an extension of RT known as the enhanced back scattering theory (EBS), that takes  
127 into account correlated scattered fields to describe the back-scattering phenomenon.

### 128 3.1. *Medium description*

129 Let's consider a region 0 free of scatterers with wavenumber  $k_0 = 2\pi/\lambda$ , and a medium 1  
130 characterized by an effective wavenumber  $K = K' + iK''$  containing  $N$  scattering spherical  
131 inclusions per unit volume. All scatterers have the same radius  $r$  and scattering properties (Figure  
132 1).

133 The medium is illuminated in the incoming direction  $\theta_i$  by the light coming from a point source  
134 at the position  $\mathbf{r}_i$  in the region 0. The light scattered by the slab is observed at a point in region 0  
135 in the direction  $(\theta_o, \varphi)$  at a distance  $r_o \gg 1$  from the interface. To ease the description of the  
136 scattering inside the layer, point scatterer approximation is made: the scattering events occur a  
137 the center point of the spherical scatterers.

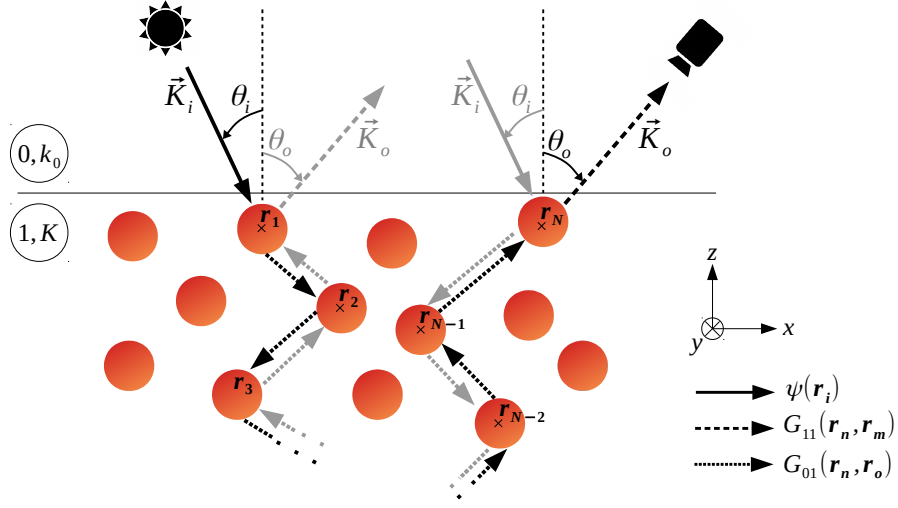


Fig. 1.  $N$ -th order multiple scattering in a medium composed of particles. The incident ray follows a path going from the particle at  $\mathbf{r}_1$  to the one at  $\mathbf{r}_N$ . Interferences occur with the ray from  $\mathbf{r}_N$  to  $\mathbf{r}_1$ . The incoherent part of the resulting field (denoted as  $\mathcal{L}_N$  in Section 5.1) contributes to the ladder term while its coherent part (denoted as  $C_N$  in Section 5.1) accounts for the cyclical term.

### 138 3.2. Foldy's approximation

139 For a sufficiently small density of scatterers, the effective wavenumber  $K$  can be expressed using  
 140 the Foldy's approximation [21]:

$$K \approx k_0 + i \frac{2\pi N}{k_0} F \quad (1)$$

141 where  $F$  is the complex scattering amplitude, describing the scattering properties of the particles.

### 142 3.3. Light propagation

143 The propagation of light inside a medium containing scattering particles is illustrated by the  
 144 Figure 1. The light ray coming from region 0 without being scattered is described by the incident  
 145 field :

$$\psi_m(\mathbf{r}_i) = \exp(i\mathbf{K}_i \cdot \mathbf{r}_i) \quad (2)$$

146 where  $\mathbf{K}_i$  is of magnitude  $K$  given by the Foldy's approximation, and oriented in the incident  
 147 direction.

148 After it has been scattered by a particle, light travels in the region 1 towards another scatterer, as if  
 149 it was in a space free of scatterer. Under this hypothesis, the propagation of light from a position  
 150  $\mathbf{r}'$  towards  $\mathbf{r}$  is given by the Green's function  $G(\mathbf{r}, \mathbf{r}')$ , that satisfies the Helmholtz equation :

$$(\Delta + k^2)G(\mathbf{r}, \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}') \quad (3)$$

151 By averaging over the particles, the mean Green's function referring to the propagation of light  
 152 from an inclusion to another is given as follows:

$$G_{11}(\mathbf{r}_n, \mathbf{r}_m) = \frac{\exp(i\mathbf{K} \cdot |\mathbf{r}_n - \mathbf{r}_m|)}{4\pi|\mathbf{r}_n - \mathbf{r}_m|} \quad (4)$$

153 where  $\mathbf{r}_m$  and  $\mathbf{r}_n$  are the positions of two distinct scatterers in the medium 1. In the same way,

$$G_{01}(\mathbf{r}_n, \mathbf{r}_o) = \frac{\exp(ik_0 r_o)}{4\pi r_o} \exp(-i\mathbf{K}_o \cdot \mathbf{r}_n) \quad (5)$$

154 describes the propagation from a scatterer at position  $\mathbf{r}_n$  in region 1 towards the camera at  $\mathbf{r}_o$  in  
155 region 0. The vector  $\mathbf{K}_o$  is of magnitude  $K$  and oriented towards the camera.

### 156 3.4. Light scattering by one particle

157 The scattering events caused by the encountering of light with the  $n$ -th scatterer is described  
158 using the transition operator  $T_n$  that we introduced as :

$$T_n(\mathbf{r}_n) = 4\pi F(\Theta_n) \delta(r_{in} - r_{on}) = 4\pi F(\Theta_n) \quad (6)$$

159 where  $\Theta_n = \theta_{on} - \theta_{in}$  is the scattering angle between the incident direction on the  $n$ -th scatterer  
160  $\theta_{in}$  and the outgoing direction  $\theta_{on}$ .

161 The complex scattering amplitude  $F(\Theta_n)$  is linked to the scattering phase function using the  
162 relationship:

$$\rho p(\Theta_n) = \frac{4\pi N}{\kappa} |F(\Theta_n)|^2 \quad (7)$$

163 where  $\rho$  is the single scattering albedo and  $\kappa$  the extinction rate.

### 164 3.5. Enhanced back-scattering

165 The EBS theory offers a physical description of the back scattering phenomenon observed  
166 in quite dense media. It is an extension of radiative transfer (RT) theory [19], [20]: the  
167 so called *ladder terms* accounting for both single scattering  $BRDF_{ebs, \mathcal{L}_1}$  and some multiple  
168 scattering  $BRDF_{ebs, \mathcal{L}_{2+}}$  takes into account the contribution of uncorrelated scattered fields  
169 already described by the RT theory. This model also accounts for coherent scattered fields  
170 that induce interference effects as light undergoes multiple scattering events in the medium. It  
171 is described by a *cyclical term*  $BRDF_{ebs, \mathcal{C}}$  and results in a sharp peak in the back-scattering  
172 direction.

173 The BRDF is then composed of three contributions:

$$BRDF_{ebs} = BRDF_{ebs, \mathcal{L}_1} + BRDF_{ebs, \mathcal{L}_{2+}} + BRDF_{ebs, \mathcal{C}} \quad (8)$$

174 This model has been developed for thick layers of a medium composed of isotropic spherical  
175 scatterers of radius  $r$  with a constant phase function  $p(\Theta) = p$  embedded in a host medium  
176 [17, 18, 22, 23].

177 Analytical expressions of the multiple scattering terms are obtained by considering that for  
178 optically thick media with high albedo the resulting overall scattering statistically tends to be  
179 isotropic. This is known as the *diffusion approximation* and leads to results in Eqs. 9.

$$\left\{ \begin{array}{l} BRDF_{ebs, \mathcal{L}_1} = \frac{\rho p(\Theta)}{\mu_o + \mu_i} \left[ 1 - \exp\left(-\kappa d \left(\frac{1}{\mu_o} + \frac{1}{\mu_i}\right)\right) \right] \end{array} \right. \quad (9a)$$

$$\left\{ \begin{array}{l} BRDF_{ebs, \mathcal{L}_{2+}} = \frac{3\rho^2}{1 - \mu_i^2 c^2} \left[ \frac{2Q}{1 - \mu_o c} - \frac{\mu_i^2}{\mu_i + \mu_o} \right] \end{array} \right. \quad (9b)$$

$$\left\{ \begin{array}{l} BRDF_{ebs, \mathcal{C}} = \frac{3\rho^2 [c + v(1 - \exp(-2c\tau_o))]}{2\mu_i \mu_o c v [(c + v)^2 + u^2]} \end{array} \right. \quad (9c)$$

The parameters  $u$ ,  $v$ ,  $c$  and  $Q$  in Eqs. 9 vary with the isotropy of the scatterers and with the geometry under which the medium is illuminated and observed. It has been described by [18] for

isotropic scatterers such as:

$$\begin{cases} u = \frac{k_0}{\kappa}(\mu_i - \mu_o) & (10a) \\ v = \frac{1}{2}\left(\frac{1}{\mu_i} + \frac{1}{\mu_o}\right) & (10b) \\ \alpha = \frac{k_0}{\kappa}f & (10c) \\ \kappa_{tr} = 1 - \rho & (10d) \\ c = \sqrt{3(1 - \rho)\kappa_{tr} + \alpha^2} & (10e) \\ Q = \frac{[1 + \mu_i c - (1 - \mu_i c)\exp(-2c\tau_o)]}{4c} & (10f) \end{cases}$$

180 where  $f$  is the norm of the vector  $\mathbf{f}$  defined as difference between the projections of the normalized  
 181 incoming and outgoing direction vectors (Figure 2) which allows the BRDF modelization in any  
 182 azimuthal plane.

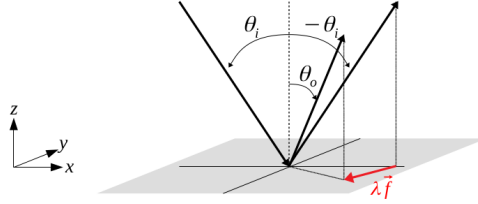


Fig. 2. Definition of the vector  $\mathbf{f}$ .

183 Only the single scattering term involves the phase function in such a case. [17] introduce  
 184 the consideration of anisotropic scatterers but only with normal incidence. We will discuss the  
 185 extension of the model for anisotropic scattering in the Section 5.

187 The last parameter involved in the BRDF expressions (Eqs. 9) is the extinction ratio  $\tau_o$  that is  
 188 defined as intrinsic to the scatterers' material. It accounts for internal reflections that may occur  
 189 between the interface of the medium. A definition is given by [24]:

$$\tau_o = \frac{2}{3} \frac{1 - \bar{R}}{1 + \bar{R}} \quad (11)$$

with  $\bar{R} = \frac{3C_2 + 2C_1}{3C_2 - 2C_1 + 2}$  and  $C_i = \int_0^{\frac{\pi}{2}} R(\theta) \cos^i \theta \sin \theta d\theta$

190 In the previous equations,  $R(\theta)$  is the Fresnel's angular reflection coefficient [25]. The  
 191 parameter  $\tau_o$  thereby depends on the optical constants of the material the particles are made of  
 192 and on the wavelength.

#### 193 4. Sample description and BRDF measurements

194 The physical phenomena at stake in the appearance of a material can be inferred through the  
 195 BRDF measurement.

196 **4.1. Sample description**

197 For the study of dry powder appearance we focused on synthetic nano-crystallized hematite  
198  $\alpha - \text{Fe}_2\text{O}_3$  powders with various colors, that we obtained using different routes allowing to control  
199 the nucleation and growth of  $\alpha - \text{Fe}_2\text{O}_3$  crystallites in solution. These samples are presented on  
200 Figure 3

201 The samples we have denoted from 1 to 5 have been prepared following methods described  
202 by [26]. 1, 2 and 5 have been obtained by forced hydrolysis of  $\text{Fe}^{III}$  salts solutions in acidic  
203 conditions, while 3 and 4 are the results of the transformation of a ferrihydrite precipitate in  
204 aqueous suspension under basic conditions. We generated peanuts-like hematite particles 6  
205 following the routes proposed by [27],  $\text{NaH}_2\text{PO}_4$  salts. Sample 7 is a commercial Iron(III) oxide  
206 powder sample purchased from Puratronic®.

207 The color and the morphometry of each sample are respectively deduced from naked eye  
208 observations completed with optical microscopy using a Nikon Eclipse LV100ND microscope,  
209 and by scanning electron microscopy observations performed with a Zeiss Ultra+ microscope.  
210 Various grains' shape and size ranging from 10 nm to 10  $\mu\text{m}$  are obtained. The small grains are  
211 characterized by a lighter red color (samples 1 to 5) while a darker purple hue is observed for the  
212 pigments with larger grains (samples 6 and 7). We may also note that samples 5, 6 and 7 can be  
213 described by two grain sizes: either two different size populations (sample 5), either large grains  
214 that are agglomerates of smaller ones (samples 6 and 7).

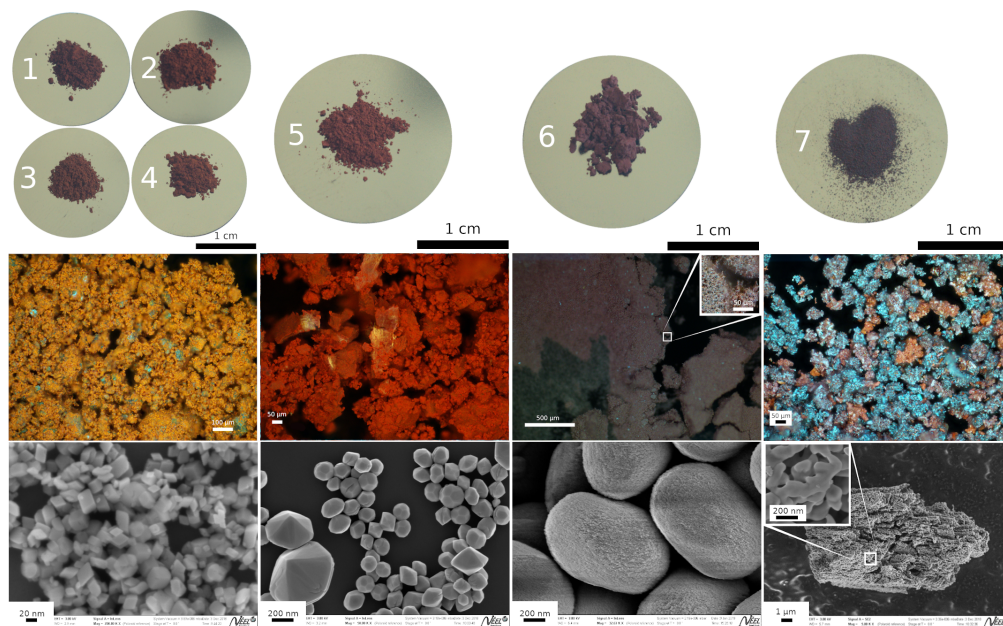


Fig. 3.  $\alpha - \text{Fe}_2\text{O}_3$  hematite samples under study, showing various colors and grains' morphology. Upper row: The photographs are calibrated using an X-rite 24 patches Classic Colorchecker. Middle row: Observations using optical microscopy. Lower row: Samples' morphology observations using Scanning Electron Microscopy. The samples 1 to 4 show similar grain shape, with slightly different sizes from 10 to 50 nm in diameter, and light red hue. Sample 5 presents two size populations and shows a red-pinkish hue. Samples 6 and 7 have a more complex morphology with grains that seem to be agglomerations of smaller ones. The grains are larger and a darker purple hue is observed. According to optical microscopy, 7 is even composed of different types of grains, with different colors.



215 **4.2. BRDF measurements**

216 The whole BRDF of each sample is measured by combining two distinct measurements, depending  
217 on which scattering geometry is at stake. The first one does not account for the back-scattering,  
218 while the second one is specifically performed to account for this contribution.

219 **4.2.1. Out the back-scattering**

220 The main part of the BRDF is measured using the SHADOWS instrument [28], over 180 geometries:  
221 the scattered light is observed for  $\theta_o \in [-70^\circ, 70^\circ]$ , for incidences  $\theta_i \in \llbracket 0^\circ, 20^\circ, 40^\circ, 60^\circ \rrbracket$   
222 and azimuthal angles  $\varphi \in \llbracket 0^\circ, 30^\circ, 60^\circ, 90^\circ \rrbracket$ , and  $135^\circ$ . The spectral range has been limited to  
223 to 360 nm - 860 nm with a step of 20 nm. The reflectance measurements are calibrated with a  
224 Spectralon. Every point is measured with an integration time of 300 ms and averaged over 10  
225 spectra. Such measurement takes about 18 hours.

226 When it comes to a geometry where incident and observation directions are identical, the  
227 instrument's design causes the sensor to be obstructed by the light source. A specific device  
228 needs to be designed to evaluate the amount of light scattered in these directions.

229 **4.2.2. In the back-scattering direction using an in-house-built set-up**

230 Our design for measuring the back-scattering contribution to the BRDF is inspired from the  
231 work of [29] and [8]. As these only deal with vertically held samples, they cannot be used for  
232 studying dry powders. We propose a new design to allow the measurement of any horizontally  
233 held sample.

234 We chose an halogen light source to normally illuminate the sample after being reflected by a  
235 beam splitter at  $45^\circ$ . The size of the illumination spot is minimized by focusing the light with a  
236 50 mm focal lens. An additional diaphragm is added to clean the focused beam. The direction of  
237 the incident light  $\theta_i$  is adjusted by positioning the beam splitter at an angle of  $45^\circ + \theta_i/2$ . As the  
238 light interacts with the powder, it is scattered in every directions and preferably back into the  
239 incident direction. It is then partially transmitted through the beam splitter and collected by a  
240 Thorlabs F950FC-A collimator. The latter is fixed on a vertical rotating plate so that we can scan  
241 different observation directions around the exact back-scattering one in the incident plane. We  
242 use a fiber spectrometer OceanOptics USB650 along with the SpectraSuite software in order to  
243 detect and visualize the measured signal.

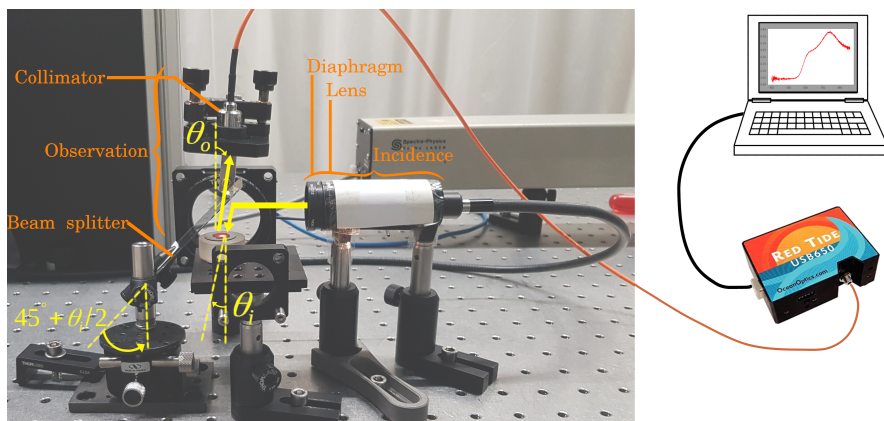


Fig. 4. Back-scattering measurement device. The beam of a white source is focused on the sample's surface at an incidence  $\theta_i$  using a 50 mm focal lens and a beam splitter. The reflected light is collected through a collimator in different observation directions  $\theta_o$ .

244 In the incident plane ( $\varphi = 0^\circ$ ), we scan the observation angles in the range  $[\theta_i - 10^\circ, \theta_i + 10^\circ]$   
245 with a step of  $2^\circ$ , for incidences  $0^\circ, 20^\circ$  and  $40^\circ$ . The reflectance measurements are calibrated with  
246 a Spectralon. The integration time is chosen to maximize the detected signal and an averaging  
247 over 5 spectra is performed to minimize the noise. Less than 20 seconds is necessary to measure  
248 one spectrum.

#### 249 4.2.3. Complete measurement

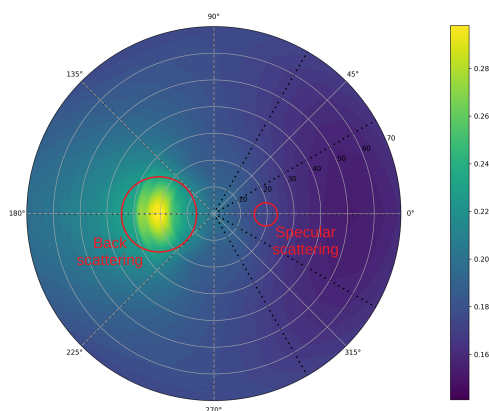


Fig. 5. Complete BRDF of the hematite powder sample 1 at the wavelength 700 nm for an incidence  $\theta_i = 40^\circ$ . The measured geometries are spotted with black dots, and the intermediate values are interpolated. Light is mostly reflected in the backward direction, and a very weak specular scattering peak may be observed as well. The spreading of the back scattering peak with the azimuth angle is due to the colormap interpolation between the measured points.

250 As the two measurements are performed independently, they happen to be shifted in amplitude  
251 from one another. This may be caused by the underestimation of the back scattering of the  
252 spectralon used for photometric calibration. Since the difference is smaller than 5%, a re-scaling  
253 of the back-scattering measurement is done so it matches the goniometer results.

254 A typical experimental BRDF is shown figure 6 after merging the measured data. The  
255 measurements show that the BRDF of dry hematite powders is composed of an overall diffuse  
256 contribution, and two directional scattering phenomena including mainly back-scattering and  
257 some specular scattering (Figure 5). The BRDFs of characterized samples vary in intensity and  
258 width of the specularly and back scattered lobes. We give results for the six other samples under  
259 study in Supplementary Work.

## 260 5. Powder BRDF Model

261 As shown by the measurements on Figure 5, powders' BRDF mainly involves back-scattering  
262 and almost no specular scattering which makes the usual specular BRDF models ineffective to  
263 reproduce their appearance.

264 Although our model is derived from a study on pure nano-crystallized hematite powders, we  
265 chose a physically-based approach so that it can be applied to any other pigment. It involves  
266 both chemical composition and morphological parameters to accurately account for the color  
267 variations observed on such pigments. Moreover it allows to consider either isotropic or  
268 anisotropic scatterers. Inspired by both RTLSR and Spectralon's models, our model is divided

269 into two contributions respectively accounting for volume and surface scattering.

$$BRDF = A_v BRDF_v + A_s BRDF_s \quad (12)$$

270 These two contributions are weighted with the amplitude parameters  $A_v$  and  $A_s$ .  
 271 The main difference with the previously introduced models is that, based on the EBS theory the  
 272 back scattering is considered as part of the volume contribution. The surface contribution is then  
 273 reduced to specular scattering caused by surface roughness.

### 274 5.1. Volume scattering

275 As EBS theory is established within the diffusion approximation, the averaged scattering is  
 276 assumed to be isotropic. This greatly simplifies the multiple scattering contribution by considering  
 277 the scattering phase function as constant over the whole scattering hemisphere. The anisotropic  
 278 scattering of each particle is accounted for by choosing the phase function parameter  $g$  involved  
 279 in the single scattering contribution. The extension of the model for taking into account this  
 280 anisotropy in multiple scattering has only been done for a normal incidence by Tsang et al. [17].

281 In the following we expose the calculations that we made in order to consider anisotropic  
 282 scattering in both single and multiple scattering terms for any  $\theta_i/(\theta_o, \varphi_o)$  geometry. The final  
 283 model only involves two variable parameters ( $g, \kappa$ ) that rely on the physical and chemical  
 284 properties of the material that is under study.

285 **First order ladder term  $\mathcal{L}_1$ :** This contribution accounts for light that is only scattered once  
 286 before being observed and is similar to the one introduced in Section 3.5. The anisotropy is taking  
 287 care of by the scattering phase function involved in the analytical expression of  $BRDF_{obs, \mathcal{L}_1}$   
 288 Eq. 9.

289 **Multiple ladder term  $\mathcal{L}_{2+}$ :** The phase function is involved at every scattering event that  
 290 light encounters as it travels the particulate medium. We denote  $P_i$  and  $P_o$  respectively the first  
 291 and the last particles that are responsible for light scattering along a light path.

292 At the second scattering order, the light is first incoming on  $P_i$ , scattered with an angle  $\theta_i$   
 293 towards the particle  $P_o$ , and finally scattered towards the observer with an angle  $\theta_o$  as illustrated  
 294 by the Figure 6.

295 For the  $n$ -th scattering order, light undergoes  $n$  scattering events. As for the second order, it  
 296 undergoes a first one described by the angle  $\theta_i$  while encountering the particle  $P_i$  and last one  
 297 with the particle  $P_o$  and the angle  $\theta_o$ . But it also interacts with  $n - 2$  other particles in between,  
 298 resulting in  $n - 2$  scattering events respectively described by the scattering angles  $\theta_k = \theta_{ik} - \theta_{ok}$ ,  
 299 difference between the incoming direction  $\theta_{ik}$  and outgoing direction  $\theta_{ok}$  on the  $k$ -th particle,  
 300 with  $k = \llbracket 2 ; \infty \rrbracket$  as it is shown on the Figure 6.

301

By averaging over all the possible paths that light can follow inside the medium while  
 encountering  $n$  scatterers, we can write the  $n$ -th order ladder term as follows:

$$\begin{aligned} \mathcal{L}_n &= \frac{(4\pi)^{n-2} (\rho\kappa)^n}{r^2} \\ &\times \int |p(\theta_n)|^2 |G_{11}(\mathbf{r}_n, \mathbf{r}_{n-1})|^2 \dots |G_{11}(\mathbf{r}_2, \mathbf{r}_1)|^2 |p(\theta_1)|^2 \exp\left(\kappa \left(\frac{z_1}{\mu_o} + \frac{z_n}{\mu_i}\right)\right) d\mathbf{r}_1 \dots d\mathbf{r}_n \end{aligned} \quad (13)$$

302 with  $r$  the distance from the sample to the observation point.

303 The total ladder term accounting for multiple scattering can be written in a recursive way,  
 304 giving:

$$\mathcal{L}_{2+} = \int \int \exp\left(\kappa \left(\frac{z_i}{\mu_o} + \frac{z_o}{\mu_i}\right)\right) A(\mathbf{r}_i, \mathbf{r}_o) d\mathbf{r}_i d\mathbf{r}_o \quad (14)$$

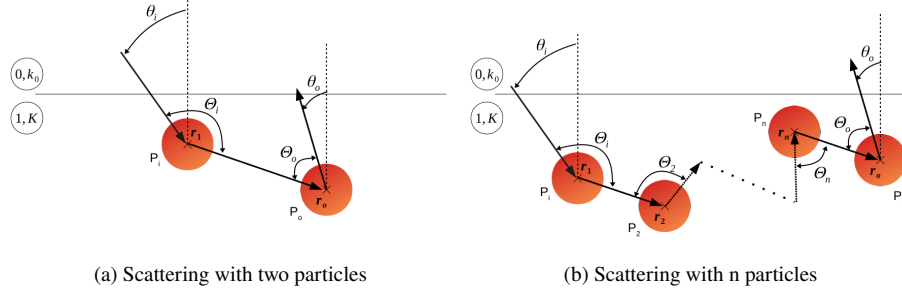


Fig. 6. Multiple scattering with two and  $n$  particles. The first scatterer that light encounters as it travels inside the medium is denoted as  $P_i$  and involves a scattering event described by the angle  $\theta_i$ . The last scattering event before light goes out of the medium is caused by the scatterer  $P_o$  with an angle  $\theta_o$ . In between these two scatterers, light is scattered with an angle  $\theta_k$  at each interaction with a particle  $k$ .

305 with  $A(\mathbf{r}_i, \mathbf{r}_o)$  defined such as:

$$A(\mathbf{r}_i, \mathbf{r}_o) = \left(\frac{\rho\kappa}{4\pi}\right)^2 p(\theta_1)p(\theta_o) \frac{\exp(\kappa|\mathbf{r}_i - \mathbf{r}_o|)}{|\mathbf{r}_i - \mathbf{r}_o|^2} + \int \left(\frac{\rho\kappa}{4\pi}\right) p(\theta_n) \frac{\exp(\kappa|\mathbf{r}_i - \mathbf{r}_n|)}{|\mathbf{r}_i - \mathbf{r}_n|^2} A(\mathbf{r}_n, \mathbf{r}_o) d\mathbf{r}_n \quad (15)$$

306 Note that contrary to the ladder term defined by the Eq. 9, our model involves a varying  
 307 scattering phase function whose expression is chosen to better model the material under study. If  
 308 one wants to describe isotropic scattering a constant phase function such as  $p(\theta) = p$  should be  
 309 used which leads back to the expression originally introduced in Section 3.5.

310

311 The resulting BRDF of the ladder contribution is then given by:

$$BRDF_{\mathcal{L}_{2+}} = \frac{4\pi r^2 \mathcal{L}_{2+}}{a\mu_i\mu_o} \quad (16)$$

312 where  $a$  is the area of the sample under studied.

313 **Multiple cyclical scattering  $C$ :** As for the ladder term, we are aiming to take into account  
 314 the anisotropic scattering of particles in the cyclical term. We will use the same assumptions as  
 315 illustrated by the Figure 6. We consider a ray of light that is scattered by  $n$  particles as it follows  
 316 a path inside the medium. By accounting for the correlation between this path and its reversal  
 317 and averaging over all the possible paths, the  $n$ -th order cyclical term can be written:

$$C_n = \frac{(4\pi)^{n-2} (\rho\kappa)^n}{r^2} \int |p(\theta_n)|^2 |G_{11}(\mathbf{r}_n, \mathbf{r}_{n-1})|^2 \dots |G_{11}(\mathbf{r}_2, \mathbf{r}_1)|^2 |p(\theta_1)|^2 \\ \times \cos\left(k(\mu_i - \mu_o)(z_1 - z_n)\right) \exp\left(\frac{\kappa}{2}\left(\frac{1}{\mu_o} + \frac{1}{\mu_i}\right)(z_1 + z_n)\right) d\mathbf{r}_1 \dots d\mathbf{r}_n \quad (17)$$

318 The complete cyclical contribution is obtained by summing over the  $n$ -th orders and can be  
 319 simplified replacing  $G_{11}$  by its definition (Eqs. 4). By using the new expression of  $A(\mathbf{r}_i, \mathbf{r}_o)$   
 320 given by Eq. 15:

$$C = \int \int \cos(2k(\mu_i - \mu_o)(z_o - z_i)) \exp\left(\frac{\kappa}{2}\left(\frac{z_i}{\mu_o} + \frac{z_o}{\mu_i}\right)\right) A(\mathbf{r}_i, \mathbf{r}_o) d\mathbf{r}_i d\mathbf{r}_o \quad (18)$$

321 Once again, the scattering properties of each particle that plays a role in the  $n$ -th order  
 322 scattering is explicitly involved through the varying phase function. The use of a constant phase  
 323 function  $p(\theta) = p$  brings back to the isotropic case already discussed in Section 3.5.

324  
 325 Finally, the cyclical term contributes to the complete appearance of the dry powder through  
 326 the partial BRDF:

$$BRDF_C = \frac{4\pi r^2 C}{a\mu_i\mu_o} \quad (19)$$

327 Under the *Diffusion approximation*, the multiple scattering terms 16 and 19 are reduced to  
 328 the analytical formulas given by the Eqs. 9. By extending the work from [17] the parameters  
 329 involved in the model are defined such as in Eqs. 10 so that the model is valid for any geometry  
 330 including various incident directions, but using  $\kappa_{tr} = 1 - \rho g$  to also account for the phase function  
 331 anisotropy in the multiple scattering.

### 332 5.2. Surface scattering

333 In our model, surface scattering is reduced to the specular contribution. The light that is incoming  
 334 on a rough surface such as the interface between air and a dry powder is scattered in a lobe  
 335 around the specular direction.

336 Two options have been considered to include such a phenomenon in our model. The former is  
 337 to model the specular peak by a gaussian function. According to the measurements performed  
 338 in Section 4 the specular scattering is very weak so this simple method is reliable enough to  
 339 reproduce the specular contribution of dry powders, although it is empirical. By assuming a  
 340 uniform distribution of the surface normals, it requires only one parameter which is the gaussian  
 341 half width  $\sigma$ . With this modeling the surface scattering BRDF is written as:

$$BRDF_s = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{\theta^2}{2\sigma^2}\right) \quad (20)$$

342 The second option, which is the one we chose to use, relies on the physical approach from  
 343 Harvey-Shack [6] that accounts for diffraction effects caused by the surface asperities. Using  
 344 this theory, the incoming light on a rough surface is then scattered around the specular direction  
 345 according to the following BRDF:

$$BRDF_s = \frac{4\pi^2}{\lambda^4} (\cos\theta_i + \cos\theta_o)^2 PSD(\mathbf{f}) \quad (21)$$

346 where the power spread function ( $PSD$ ) models a rough surface topography. We chose a gaussian  
 347 power spread function, involving a roughness parameter  $\sigma_s$  and a correlation length  $l_c$  that  
 348 quantifies the mean distance for which the surface roughness of two points on the surface are  
 349 completely independent.

$$PSD(\mathbf{f}) = \pi\sigma_s^2 l_c^2 \exp(-\pi^2 l_c^2 (f_x^2 + f_y^2)) \quad (22)$$

### 350 5.3. Complete model

351 Our complete model is the weighted sum of volume and surface contributions according to  
 352 Eq. 12 respectively given by the adapted enhanced back scattering model and a specular model.  
 353 Note that the roughness parameter involved in the surface contribution has been included in the  
 354 amplitude parameter  $A'_s = A_s\sigma_s^2$  in order to reduce the number of optimization parameters.

$$BRDF = A_v [BRDF_{\mathcal{L}_1}(g) + BRDF_{\mathcal{L}_{2^*}}(g) + BRDF_C(g, \kappa)] + A'_s BRDF_s(l_c) \quad (23)$$

355 The model depends on a total of 5 empirical and physical parameters to accurately simulate  
 356 the BRDF of dry powder. As it is shown in the Section 6, our model works even better while  
 357 including another physical parameter to refine which is the *effective radius* of the grains, defined  
 358 as the mean size of the grains actually seen by the incoming light and therefore responsible for  
 359 the scattering. The influence of such parameter is shown on Figure 7. The parameters involved  
 in our model are summarized in Table 1.

Volume scattering	
$A_v$	Intensity of volume scattering
$g$	Scattering anisotropy
$\kappa$	Extinction rate
$r$	effective radius (optional)
Surface scattering	
$A'_s$	Intensity of surface scattering
$l_c$	Correlation length

Table 1. Parameters involved in our BRDF model. The model can be optimized by refining only 5 parameters while the grain radius  $r$  is fixed, or by refining 6 parameters including the same 5 previous ones and an effective radius.

360

## 361 6. Results

362 In order to check on the ability of the model to simulate the pigments' BRDF, we applied it to  
 363 the hematite powders measurements by optimizing the variable parameters of Table 1. We used  
 364 the least squares method to fit the measurement data. A first option is to optimize while using a  
 365 fixed radius. The average dimensions of the grains of a sample is deduced from SEM images and  
 366 reduced to only one dimensional parameter by computing the radius of a sphere that is equivalent  
 367 to the grain. To better account for the shape anisotropy this radius is computed so that the ratio of  
 368 the surface over the volume of the equivalent sphere and of the grain remains constant. The final  
 369 mean radius is obtained by averaging on the radii of multiple grains that are in the field of view  
 370 of a single SEM image. However, in the following we chose to optimize on the grains' effective  
 371 radius as it leads to better results. Indeed it allows a nicer modeling of the back scattering peak  
 372 amplitude, while keeping values pretty close to the radius deduced from microscopy observations  
 373 (see Table 2). The comparison of the two optimizations performed on sample 2 is shown on  
 374 Figure 7. The optimization is performed for all geometries of incidence whose back scattering  
 375 contribution has been measured, which corresponds to incidence smaller than  $60^\circ$ . We only  
 376 account for the measurements at the wavelength 700nm, the spectral dependency of the model  
 377 being discuss later on.

378 For comparison, the same optimization process is performed using both Spectralon and RTLSR  
 379 models adapted to be relevant in the case of dry powders. Such adaptations to the original models  
 380 are detailed in Supplemental work. Results of the optimization process for the sample 1 are given  
 381 on the Figures 8, 9, and 10. The results obtained for other samples are listed in Supplementary  
 382 work.

383

384 Our model is able to reproduce the measured BRDF of hematite powders with an accuracy  
 385 comparable to that of the Spectralon mode ( $< 8\%$  of the reflectance factor), but with a much lower

386 complexity: only 6 parameters related to physical properties of the samples are required against  
 387 12 empirical ones for the Spectralon's model. It is particularly efficient in the case of samples  
 388 whose morphology is easily characterized, that is to say for the samples with well-crystallized  
 389 single-crystals. Both the back-scattering and the specular contributions are detected and modeled,  
 390 even though the latter is sometimes too weak for the modeling to be reliable. Errors are  
 391 mostly spotted around the back scattering peak because of the measurements' poor sampling  
 392 and interpolation at these geometries. Let's note that we have encountered difficulties in the  
 393 modelization of samples 3 and C's BRDFs that we attributed to the more complex morphology  
 394 of these powders. These powders are not monodisperse since they show two size populations  
 395 and agglomerations of grains. An extension of our model is proposed in supplementary work to  
 396 deal with powders presenting two grain size populations.

Sample	Radius SEM (nm)	Effective radius (nm)		
1	13 ± 3.0	17		
2	12 ± 1.6	16		
3	22 ± 3.7	16		
4	25 ± 4.1	15		
Sample	Large radius SEM (nm)	Effective radius (nm)	Small radius SEM (nm)	Effective radius (nm)
5	83 ± 34	83	47 ± 5.3	46
6	687 ± 64.0	687	10 ± 0.4	43
7	2222 ± 1054	2222	49 ± 13	49

Table 2. Comparison of the radius deduced from SEM observations and the effective radius obtained by the optimization of our model. For samples 5 to 7, the model extension for two size populations presented in Supplementary work is used. As it takes two different radii to optimize, two effective radii are obtained.

397 Since the refined parameters permit our model to quite precisely fit with the measurements in  
 398 any geometry, it appears that none of them depends on the incoming, outgoing and azimuthal  
 399 directions. The model is then able to predict the appearance of the studied powder in any other  
 400 geometry that hasn't been considered during its optimization. The results are convenient even for  
 401 more grazing angles such as incidence  $\theta_i = 60^\circ$  as shown on Figure 7.

## 402 7. Discussion & Conclusion

403 From BRDF measurements, we highlighted that in the particular case of dry powders the light  
 404 is mainly back scattered. We proposed a new BRDF model accounting for three different  
 405 scattering interactions: single scattering, incoherent multiple scattering and coherent multiple  
 406 scattering responsible for the sharp peak in the backward direction. It is a semi-empirical and  
 407 analytical model based on the propagation of light inside a granular medium that involves the  
 408 physico-chemical properties of the powders connected only six refinement parameters.

409 The model has been validated against the BRDF measurements of seven  $\alpha - \text{Fe}_2\text{O}_3$  hematite  
 410 powders, and compared to two adapted existing models. Ours is able to reproduce the measured  
 411 BRDF with an accuracy comparable to the one of the Spectralon's model, but with a much  
 412 lower complexity. It can be used in its original form to reproduce the appearance of a mono

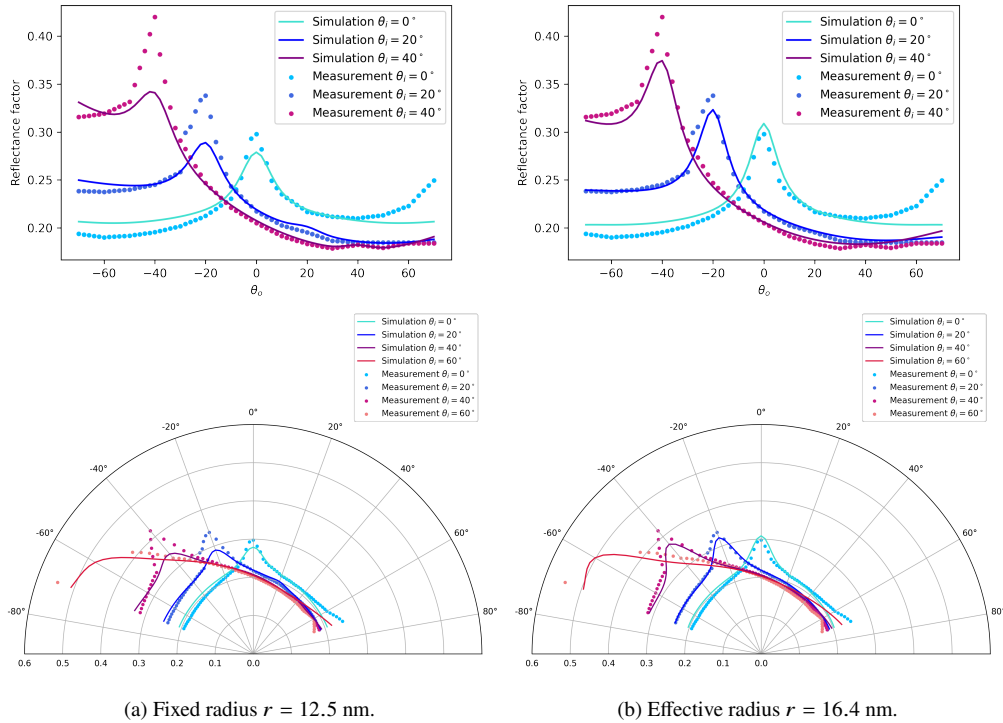


Fig. 7. Influence of the grain radius optimization on the results of sample 2, in the incident plane ( $\varphi = 0^\circ$ ) for incidences  $\theta_i = 0^\circ, 20^\circ$  and  $40^\circ$ . The effective radius found by optimization ( $r = 16.4$  nm) is pretty close to the theoretical result deduced from SEM observations ( $r = 12.5$  nm). On the polar representation, the prediction of our model for incidence  $\theta_i = 60^\circ$  is also given.

413 disperse powder with spherical grains, or in its extended form presented in Supplementary work  
 414 to account for more complex morphology involving multiple grains size populations.

415

416 Our model still has some limitations. The results obtained for specular scattering are quite  
 417 questionable since the specular contribution is very weak and almost unnoticeable on the BRDF  
 418 measurements. A more accurate model should be established by studying powders showing shiny  
 419 and sparkling reflections such as eye shadow.

420

421 More generally, the model is not able to predict the appearance of any powder whose BRDF  
 422 has not been measured beforehand. This would require to establish a relationship between the  
 423 variable parameters of the model and the physico-chemical parameters of the material the powder  
 is composed of, which requires to study much more samples with more variability in sizes.

424

425 The validity of the model's wavelength dependency is also a main issue that couldn't be  
 426 solved throughout this work. At smaller wavelengths, the optimization is not robust enough to  
 427 lead to nice fitting results (see Figure 11). This might be caused by a poor knowledge of the  
 428 optical constants of the studied material which is the only fixed parameter known as spectrally  
 429 varying. Indeed, it is mainly described by the optical constant at stake whose value appears to be  
 430 quite doubtful according to the discrepancies between the results in the literature [30, 31]. Such  
 431 uncertainties are often ignored in both material science and computer graphics whereas it surely  
 affects the results.



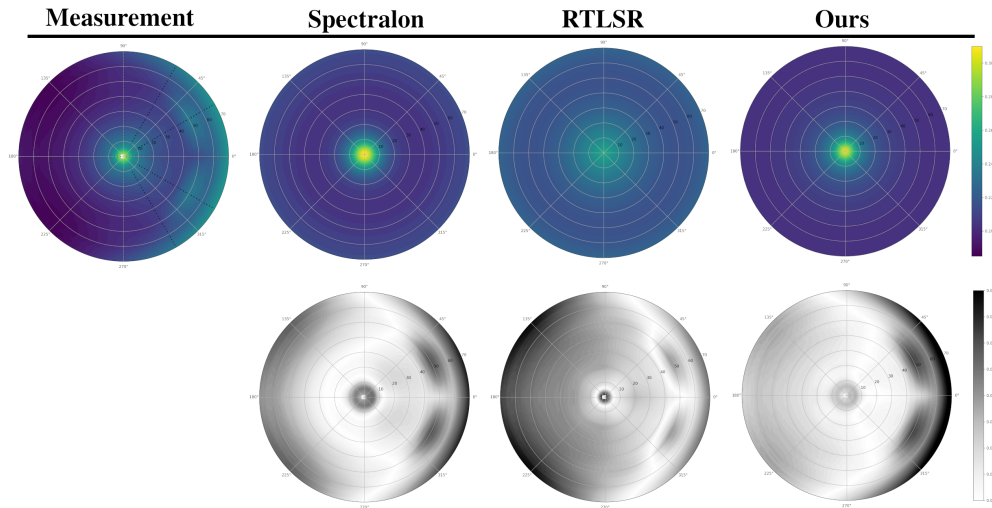


Fig. 8. Upper row: Simulation of sample 1’s BRDF using our model, spectralon and RTLSR models adapted to dry powders, at incidence  $\theta_i = 0^\circ$ . Lower row: difference between the measurement and the simulation.

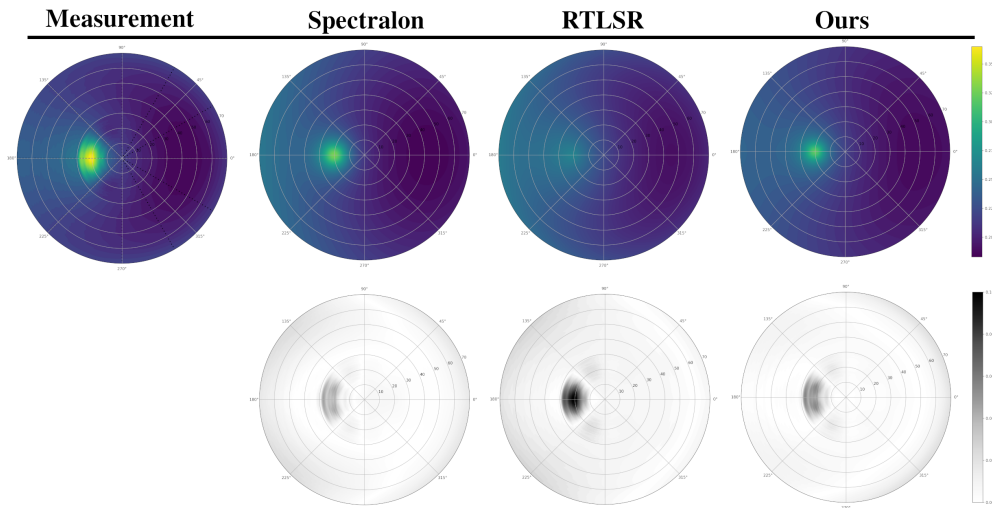


Fig. 9. Upper row: Simulation of sample 1’s BRDF using our model, spectralon and RTLSR models adapted to dry powders, at incidence  $\theta_i = 20^\circ$ . Lower row: difference between the measurement and the simulation.

432

433 As a future work we would like to implement our dry powder BRDF model in a physically  
 434 based renderer so that photo-realistic renderings of the powders can be made. This would first  
 435 allow the validation of the model by comparing the rendered image with photographs of the actual  
 436 samples. It would also permit, by letting the user choose the values of the variable parameters to  
 437 either reproduce the appearance of other powder materials, or to create new ones.

438 Although the model has been tested for only one material, we believe it can be used for any  
 439 other pure pigment. In the case of parietal paintings, ochers (mixture of one iron-based coloring

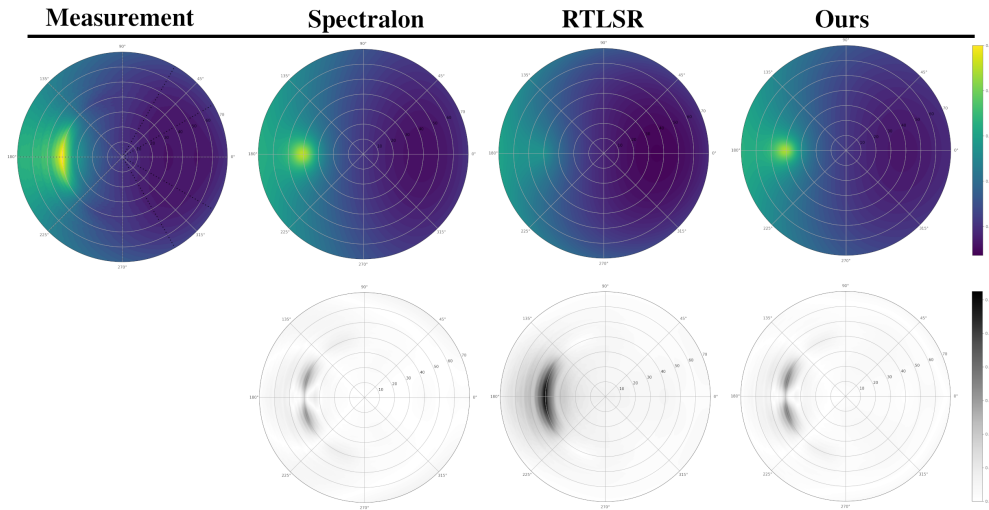


Fig. 10. Upper row: Simulation of sample 1's BRDF using our model, spectralon and RTLSR models adapted to dry powders, at incidence  $\theta_i = 40^\circ$ . Lower row: difference between the measurement and the simulation.

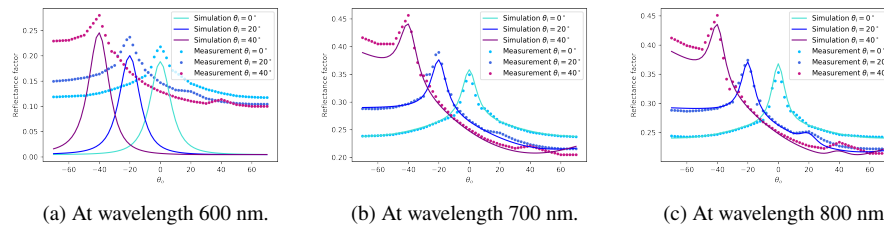


Fig. 11. Optimization of our model at different wavelengths using the optical constants from [30]. For smaller wavelengths, our model is not able to accurately fit the measurements.

440 pigment and colorless powders such as clays) and mixtures of different pigments were more  
 441 commonly used than pure and almost pure iron oxide. In order to reproduce photo-realistic  
 442 appearance of such paintings, we would like to extend our model to more complex powders.

443 The assumption of optically thick layer of powder is also too restrictive. An extension of the  
 444 enhanced back scattering theory for thin films and isotropic scatterers has been proposed by [18]  
 445 and may be a good starting point for our model adaptation, leading to a better reproduction of the  
 446 appearance of painting layers.

447  
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456 **Supplemental document.** See Supplement 1 for supporting content.

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