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Effect of Particle Optical Properties on Size Distribution of Soils Obtained by Laser Diffraction

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Key Terms: ***Absorption Coefficient, Clay-Size Particles, Laser Diffraction Method, Particle Size Distribution, Refractive Index***

ABSTRACT

Particle size distribution (PSD) is essential information for those in the fields of engineering and environmental geosciences, sedimentology, and pedology. The PSD can be determined by conventional sieve, hydrometer, and pipette methods. However, since the 1980s, the PSD of soils has also been determined via the laser diffraction method, which is more reliable and faster and which can be used repeatedly. With this method there are two optical theories commonly used for the determination of PSD: the Mie and the Fraunhofer theories. The main difference between these theories is that the Mie theory requires the refractive index (RI) and absorption coefficient (AC) values of the particles and the RI value of the suspending medium, whereas the Fraunhofer theory does not. Although the Mie theory is more appropriate for soil samples, especially for the determination of clay-size fraction, there are not yet agreed-upon values of RI and AC for soils samples that can be used by researchers and practitioners. In this study, acceptable values for RI and AC were investigated extensively by analyzing 35 natural soil samples that were mainly of sedimentary origin. Based on the results obtained, the values of 1.55 and 0.1 for RI and AC, respectively, are suggested for laser diffraction analysis of naturally occurring sedimentary soil samples with respect to geotechnical practices. Furthermore, in order to demonstrate the pitfalls of using the Fraunhofer theory for the analysis of soil samples, especially for clay-size particles, the PSDs and clay-size fractions obtained from the Mie and Fraunhofer theories were also compared by setting different RI and AC values for the Mie theory.

INTRODUCTION

Particle size distribution (PSD) and clay-size fraction of soils are parameters that control the porosity, permeability, consolidation, shear strength, and volume change behavior of soils with respect to geotechnical practices. PSD also reflects the depositional history of transported soils and evolution of residual soils (Wen et al., 2002). Because size affects the entrainment, transport, and deposition of particles, it therefore provides important clues to provenance, transport history, and depositional conditions (Pye and Blott, 2004). Therefore, PSD is an important parameter for engineering and environmental geosciences, sedimentology, pedology, etc. (Wen et al., 2002). In geotechnical practices, PSD and clay-size fraction of soils can be determined with the conventional hydrometer or pipette techniques that are dependent on Stoke's law. Drawbacks of the techniques associated with this method are that they are time consuming (one measurement takes 24 hours), they require a relatively large mass of sample (at least 20 g), and they lead to significantly variable results depending on laboratory technique, equipment, and operator (McCave and Syvitski, 1991; Percival and Lindsay, 1996). For this reason, new methods, such as laser diffraction, electrical sensing, X-ray absorption (sedimentation), and image analysis (Singer et al., 1988; McCave and Syvitski, 1991; Percival and Lindsay, 1996; and Vitton and Sadler, 1997) have been introduced in recent decades. Of these methods, laser diffraction is the most popular and widely used because it has many advantages when compared to other classical methods, such as short time of analysis (5–10 minutes per sample), small sample size (typically less than 1 g), high repeatability, good reliability, a wide range of measurement limit (typically between 2,000 and 0.04 μm), and the possibility of storing data in a digital format.

The development of the laser diffraction method for particle sizing began during the 1970s, and at

present this method is applied to many products, such as abrasives, ceramics, cement, sand, clay, foods, pharmaceuticals, cosmetics, emulsions, and sprays, and to many processes, including grinding, comminution, classification, crystallization, filtration, and polymerization (Ma et al., 2000).

The laser diffraction method has also been used in geotechnical engineering, environmental geosciences, sedimentology, and pedology since the 1980s. However, no specific standard for the determination of PSD of soils has been set. Although an international standard, namely ISO 13320, was published by the International Organization for Standardization (ISO) at the end of 1999 to set out particle size measurement by laser diffraction methods, only the general principles, validation of inversion procedures, and terminology related to laser diffraction systems were covered by this standard. This standard also did not address the explicit procedures required for particle size measurement of specific materials, such as soils. For this reason, each discipline that is applying the laser diffraction method must determine the analysis principles specifically required for measurement of its materials. Consequently, many investigations have been carried out for the determination of the PSD of soils using the laser diffraction method (e.g., McCave et al., 1986; Loizeau et al., 1994; Buurman et al., 1997; Konert and Vandenberghe, 1997; Muggler et al., 1997; Vitton and Sadler, 1997; Beuselinck et al., 1998; Chappell, 1998; Pabst et al., 2000; Murray, 2002; Wen et al., 2002; Eshel et al., 2004; Sperazza et al., 2004; Arriaga et al., 2006; Özer, 2006; and Özer and Orhan, 2007). Most of these studies compared the results of the laser diffraction method with those of sieve-pipette or hydrometer methods. However, there are many factors that control the PSD and clay-size fraction that are determined with the laser diffraction method, such as sampling, pre-treatment, dispersion, duration of ultrasonication time, optical theory, and optical parameters (i.e., refractive index and absorption coefficient). Of the studies mentioned above, only a few investigated the factors controlling the output of the laser diffraction analysis. For example, Muggler et al. (1997) and Buurman et al. (1997) investigated the sampling, flocculation, pre-treatment, and dispersion method as well as the optical model and refractive index for the analysis of Brazilian oxisols and sand samples, which are commercially available for buildings; Chappell (1998) investigated the optimum dispersion and ultrasonication time for sandy soil samples; Sperazza et al. (2004) introduced a standardized sample preparation procedure for naturally occurring sediments and also investigated the effects of variations in refractive index (RI) and absorption coefficient (AC) settings on the outcome of the laser diffraction analyses; Eshel et al. (2004) investigated the effect of RI of the particles on PSD of soil samples; and Özer and Orhan (2007) suggested a general sub-sampling method for clayey and sandy soil samples in a dry state. However, the studies dealing with the optical theory and optical parameters (RI and AC) did not offer an agreed-upon optical model and optical parameters for soil samples that can be used in practice. This study aims to find possible acceptable values for RI and AC that can be used in geotechnical practice. Within this context, the effects of RI and AC on PSD and clay-size fractions of soils were widely investigated on 35 natural soil samples taken from various locations in Turkey. The origins of these samples are mainly sedimentary, and they were obtained from borehole or trial pits. Detailed information about the samples used in this study and the suggested procedure are given in following sections of this paper.

THEORETICAL BACKGROUND AND PREVIOUS STUDIES

In the laser diffraction method, the sample to be analyzed is dispersed in a bath, where it is stirred and ultrasonically agitated while being circulated through a glass measurement cell. The cell is illuminated with laser beams and forward-scattered light is focused on the multi-element detector with a Fourier transform lens. The signal, which is derived from each detector element according to the intensity of the light falling on it, is collected and digitized for high-speed computer processing to obtain PSD (Allen, 1997). The general principle of the laser diffraction method is that particles of a given size diffract light through a given angle that increases with decreasing particle size (Wen et al., 2002).

To calculate the PSD from light intensity reaching the array of detectors, an optical theory must be applied. The Fraunhofer and Mie theories are the two most commonly used theories. Historically, the Fraunhofer theory is the basis for the first optical model employed for particle size measurement (ISO 13320-1, 1999) and is limited to particles that are opaque or large compared to the wavelength of light (Allen, 1997). In this theory, it is assumed that all particles are much larger (about 40 times) than the wavelength of light, and only scattering at the contour of the particles is considered, which also means that the same scattering pattern is obtained for thin two-dimensional circular disks as well (ISO 13320-1, 1999). With the Fraunhofer theory, only diffraction is considered; hence, no knowledge of the particle refractive index is needed (Allen, 1997).

As for the Mie theory, it is a solution for Maxwell's electromagnetic field equations (i.e., a set of four fundamental equations governing the behavior of electric and magnetic fields) describing propagation of the electromagnetic wave of light in space (Eshel et al., 2004). The theory provides a solution for the case of a plane wave (i.e., the wavefronts of which are planes) on a homogeneous sphere of any size (Jonasz, 1991). In this theory, it is assumed that all particles are spheres and transparent (ISO 13320-1, 1999). While the Fraunhofer theory does not make use of any knowledge of the optical properties of the material, the Mie theory requires knowledge of the RI and AC values of the particles and of the RI of the suspending medium (in this case water). It should be noted that a volume-based (rather than mass-based) PSD can also be obtained using both the Fraunhofer and Mie theories.

Soil samples are generally composed of a mix of minerals (i.e., polymineralic), and many of their refractive indices fall within a range (Table 1). Because the RI values of soil samples are mainly affected by the particle mineralogy and because RI values of the minerals vary, it is difficult to select a representative value of RI for polymineralic soil samples. In order to overcome this drawback, some approximations have been made by several investigators. For example, Buurman et al. (1997) stated that soil samples are generally composed of clay minerals and quartz, and because the RI values of these minerals are within a narrow range (Table 1), they claimed that the effect of variation in RI on soil PSD within this range could be ignored. Jillavenkatesa et al. (2001) argued that both RI and AC values of the materials have a significant effect on PSD, but they did not suggest any value for RI and AC that can be used in practice for the analysis of soil samples. Eshel et al. (2004) showed that the clay-size fraction decreased with increasing RI and that the clay-size fraction is very sensitive to RI for RI values lower than 1.65. Additionally, Eshel et al. (2004) stated that the RI values of the minerals commonly found in soil fall within the range of 1.48 to 1.71, and they claimed that for most minerals an RI value of approximately 1.53 is suitable. However, they also argued that the soil PSD is less sensitive to the AC value, and they chose 0.2 for an AC value for their optical model calculations without performing any analysis to determine the optimum value for their samples. Sperazza et al. (2004) revealed that both RI and AC values can significantly alter the estimated soil PSD, and they performed quantitative X-ray diffraction (XRD) analyses to estimate the most representative RI value of their sediment samples using the volume abundance of each mineral. However, they did not suggest an acceptable value for RI if the XRD analysis was not feasible to perform. In addition, Sperazza et al. (2004) tested the wide range of AC values from 0 to 1, and they found that as the AC value increases, the PSD becomes more unimodal and stable. However, they did not propose an AC value that can be used in practice for the analysis of soil PSD. Arriaga et al. (2006) aimed to find the best optimal value for the RI and AC to match PSD obtained from laser diffraction as close as possible to the sieve-pipette method, and the values they found were 1.42 and 0.001 for RI and AC, respectively.

Table 1. *Refractive indices of some soil-forming minerals (Malvern, 1993; ISO 13320-1, 1999; and CRC Press, 2008).*

Minerals	Refractive Indices (RIs)		
	Malvern (1993)	ISO 13320-1 (1999)	CRC Press (2008)
Albit (feldspar)	1.527–1.538	—	1.527–1.538
Andezin (feldspar)	1.544–1.563	—	1.550–1.557
Anortite (feldspar)	1.577–1.590	—	1.577–1.590
Apatite	—	1.63–1.67	1.645–1.648
Biotite (mica)	1.565–1.696	—	1.595–1.651
Brucite	—	1.56–1.60	1.575–1.590
Calcite	1.486–1.740	1.49–1.74	1.486–1.658
Chlorite	—	—	1.61–1.62
Dolomite	—	1.50–1.68	1.500–1.679
Fluorite	1.433–1.435	1.43–1.44	1.434
Gibbsite	—	1.56–1.60	1.57–1.59
Gypsum	1.519–1.531	1.52–1.53	1.52–1.53
Hematite	2.94–3.22	2.90–3.20	2.91–3.19
Illite	1.54–1.61	1.54–1.61	1.56–1.59
Kaolinite	1.533–1.570	1.53–1.57	1.549–1.565
Magnesite	1.563–1.700	1.51–1.78	1.536–1.741
Mica	1.53–1.70	1.53–1.70	—
Microcline (K feldspar)	1.514–1.539	—	1.522–1.530
Montmorillonite	1.48–1.64	—	1.55–1.57
Muscovite (mica)	1.552–1.616	—	1.563–1.602
Olivine	—	—	1.73–1.78
Opal	—	1.41–1.46	1.44
Orthoclase	1.518–1.539	1.52–1.54	1.523–1.531
Quartz	1.544–1.553	1.54–1.55	1.544–1.553
Serpantine	—	1.53–1.57	1.55–1.56
Talc	1.589	1.54–1.60	1.545–1.595
Topaz	—	1.61–1.64	1.618–1.627
Vermiculite	—	—	1.542–1.556

The main objective of this study was to find suitable RI and AC values that could be used in geotechnical practice for the analysis of naturally occurring soil samples. With this purpose in mind, first the effect of RI and AC values on soil PSD and clay-size fraction was investigated extensively using 35 natural soil samples taken from various locations in Turkey. The RI values of most soil-forming minerals (especially those of sedimentary origin) vary between 1.40 and 2.00 (Table 1), and the AC values of materials range from 0 for perfectly transparent to 1 for perfectly opaque materials (Sperazza et al., 2004).

While the RI values of most soil-forming minerals can be found in the literature, their AC values are not available (Buurman et al., 1997; Sperazza et al., 2004). However, ISO 13320-1 suggests a value of AC ranging between 0.01 and 0.1 for silicon oxides (quartz and its varieties), which are the most abundant sedimentary soil-forming minerals, depending on surface roughness. Based on this statement, it can be argued that the AC values of most soil-forming minerals probably vary between 0.01 and 0.2, considering the fact that they are not completely transparent or opaque. Therefore, the ranges (1.40 to 2.00 for RI and 0.01 to 0.2 for AC) were tested to determine the effect of RI and AC values on PSD and clay-size fraction of soils. Next, the mineralogical compositions of eight samples (out of the 35) were determined by XRD analyses to assign an acceptable RI to the naturally occurring sedimentary soil samples. In addition, clay-size fractions and PSDs obtained with the Fraunhofer and Mie theories were compared to demonstrate how much error will occur if the Fraunhofer theory is used in the analyses of soil samples, especially for clay-size particles.

MATERIALS AND METHODS

A Malvern Master Sizer X (long bed) laser diffraction instrument with a wavelength of 633 nm (He-Ne laser beam source) was used for the analyses. This instrument contains a 31-element solid-state detector array. It measures particle size over the range of 0.1 to 2,000 μm with four focal length lenses that have different measurement limits. A reverse Fourier lens that has a measurement limit ranging from 0.1 to 80 μm and a focal length of 45 mm was used in the study. The software employed was Malvern version 1.2. In order to prepare the samples for laser diffraction analyses, an MSX 17 sample preparation unit was used. The unit has an approximately 1-liter sample bath and comprises a variable-power ultrasonic system with a nominal frequency of 40 kHz. The unit also has a variable speed pump and a stirrer. Tap water was used as a suspending medium in the analyses. For dispersion of the soil samples, Calgon (sodium hexametaphosphate) solution prepared according to ASTM D 422 (1998) was used.

Soil samples were dried in a 110 ± 5°C heated oven and crushed using a plastic hammer to avoid particle breaking and were then sieved through a No. 200 (0.075-mm) sieve, as described in ASTM D 422 (1998). Particles passing from the sieve were collected in a cylindrical container and mixed thoroughly with a small spatula to obtain a homogeneous mixture, as suggested by Özer and Orhan (2007). In order to disperse the soil samples, both mechanical action and chemical agents were used. For mechanical action, a continuously stirring mixer that maintains homogeneous suspension during the analysis and sonication (to facilitate dispersion of the soil sample by breaking the inter-particle forces) was employed.

The sample bath of the sample preparation unit was filled with tap water; 100 mL of Calgon solution was introduced into the sample bath; and then a background measurement was taken. This measurement was used to subtract the non–sample scattering sources, such as daylight, flare off of optical surfaces, etc., from the total scattering received from the sample (Malvern, 1993). Following the background measurement, as suggested by Özer and Orhan (2007), a sub-sample was taken from the cylindrical container with the edge of the spatula and introduced into the sample bath in small increments until the obscuration value fell within the range of 15 percent to 30 percent. In ISO 13320-1 (1999) the following criterion was given for repeatability: for measurements made on at least five different sub-samples from the same sample batch, the coefficient of variation should be smaller than 3 percent for the central value of the distribution (e.g., d_{50}) and 5 percent for the sides of the distribution (e.g., d_{10} and d_{90}). Özer and Orhan (2007) tested the repeatability of the suggested sub-sampling method on two different samples by taking 10 different sub-samples from each sample batch, and values of 2.9, 5.6, and 5.4 from sample 1 and values of 1.5, 3.1, and 4.9 from sample 2 were obtained for d_{50}, d_{10}, and d_{90}, respectively. According to these findings, the sub-sampling method suggested by Özer and Orhan (2007) produces quite repeatable results and provides the criterion suggested by ISO 13320-1 (1999).

The time needed for proper dispersion of the samples under the effects of both mechanical action and chemical agents depends mainly on clay content and mineralogy, and it varies from sample to sample. In this study, the time needed for proper dispersion of the samples was determined using the "inspection" window of Malvern software (version 1.2) that displays the histogram of the PSD on-line during the analyses. Particle breaking and/or flocculation can be noticed simultaneously from this window. In general, the proper dispersion of the samples without particle breakage was achieved at about 6 minutes, which is in agreement with the time (8 minutes) applied by Arriaga et al. (2006).

RESULTS AND DISCUSSION

Effects of Variations in RI on PSD and Clay-Size Fraction of Soils

In order to examine the effects of variations in RI on PSD and clay-size fraction of soils, the absorption

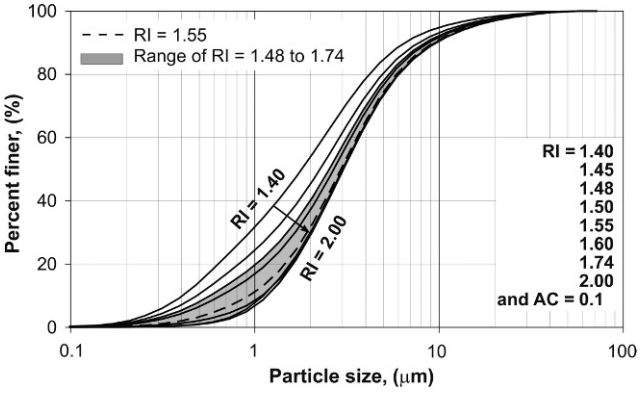

Figure 1. The effects of variations in refractive index on particle size distribution.

coefficient was first held constant at 0.1 while RI values were varied between 1.40, 1.45, 1.48, 1.50, 1.55, 1.60, 1.74, and 2.00, respectively, in the analyses. According to the results obtained from all of the 35 samples, it was observed that the effects of the variation in RI on PSDs were of nearly the same magnitude in all samples that were tested. For this reason, as an example, only the results obtained from sample 1 have been presented in Figure 1.

As can be seen in Figure 1, the RI significantly affected the PSD, particularly for an RI value of less than 1.55, whereas the effect of RI on the PSD was less pronounced for RI values that were greater than 1.55. In addition, Figure 1 shows that as RI increases, the apparent PSD shifts downward (toward larger particles), and clay-size fractions decrease. These results are in agreement with the findings of Eshel et al. (2004), in whose study a Coulter LS-230 instrument with laser beams of wavelength 750 nm was used.

The effects of variations in RI on the clay-size fractions are depicted in Figure 2. For clarity, the results of only four samples have been presented in Figure 2. The results obtained from other samples are similar to those presented in Figure 2. As can be seen

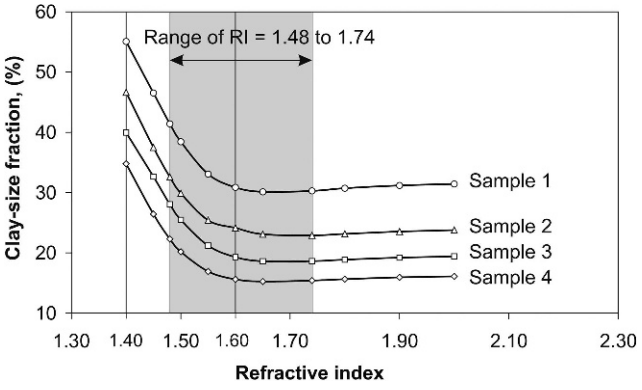

Figure 2. The effects of variations in refractive index on clay-size fractions.

Table 2. *Clay-size fractions calculated by setting the refractive index (RI) at 1.48 and 1.74 while absorption coefficient (AC) was held constant at 0.1.*

	Clay-Size Fractions (percent)		
Sample No.	RI = 1.48	RI = 1.74	Difference (percent)
1	41.4	30.3	11.1
2	32.6	22.9	9.7
3	28.0	18.6	9.4
4	22.3	15.4	6.9
5	40.7	29.0	11.7
6	44.7	27.6	17.1
7	22.9	14.6	8.3
8	29.5	14.6	14.9
9	17.8	11.3	6.5
10	23.4	15.9	7.5
11	28.4	19.7	8.7
12	18.3	8.9	9.4
13	23.5	14.1	9.4
14	30.8	21.4	9.4
15	28.7	18.3	10.4
16	33.5	22.8	10.7
17	36.2	25.0	11.2
18	34.3	22.6	11.7
19	38.0	25.8	12.2
20	33.7	21.4	12.3
21	36.3	23.9	12.4
22	36.1	23.1	13.0
23	37.2	24.0	13.2
24	39.2	25.7	13.5
25	37.3	23.1	14.2
26	41.3	26.7	14.6
27	40.0	25.1	14.9
28	42.5	27.6	14.9
29	45.9	30.9	15.0
30	49.1	34.1	15.0
31	47.3	32.2	15.1
32	45.6	29.9	15.7
33	48.4	32.5	15.9
34	50.2	32.1	18.1
35	57.3	37.2	20.1

from Figure 2, variations in RI between 1.40 and 1.60 significantly affected the clay-size fractions; however, for values of RI that were greater than 1.60, the clay-size fractions were less affected by variations in RI.

According to Table 1, the lower and upper limits of RI values of most soil-forming minerals (except for hematite, which has an extreme value) are 1.48 and 1.74, respectively. In order to exhibit the effect of RI settings on the clay-size fractions, the clay-size fractions obtained by setting the RI at 1.48 and 1.74 and keeping the AC value constant at 0.1 are listed in Table 2 for all of the 35 samples. As can be seen from Table 2, an increase of RI from 1.48 to 1.74 resulted in a decrease in the apparent clay-size fractions, on the average of 12.4 percent. As a consequence, the results depicted in Figures 1 and 2 and Table 2 reveal that the RI value is quite an effective parameter with regard to PSD and the clay-size fractions of soils. In

Table 3. *Index properties, mineralogical compositions, and refractive index (RI) values of the selected samples.*

Sample No.	w_L	w_P	I_P	Percent Passing No. 200 sieve	USCS Group	Mineralogical Composition and RI*
1	89.1	38.3	50.8	81.7	CH	Quartz (1.54–1.55) Calcite (1.49–1.74) Feldspar (≈1.53–1.59) Montmorillonite (1.48–1.64)
2	34.6	20.8	13.8	60	CL	Quartz (1.54–1.55) Calcite (1.49–1.74) Montmorillonite (1.48–1.64) Feldspar (≈1.53–1.59) Illite (1.54–1.61)
3	57.5	31.2	26.3	76.6	MH	Montmorillonite (1.48–1.64) Quartz (1.54–1.55) Feldspar (≈1.53–1.59) Calcite (1.49–1.74) Illite (1.54–1.61)
4	41.5	24.9	16.6	62.8	CL	Montmorillonite (1.48–1.64) Quartz (1.54–1.55) Calcite (1.49–1.74) Feldspar (≈1.53–1.59) Illite (1.54–1.61)
5	88.8	33.6	55.2	99.2	CH	Montmorillonite (1.48–1.64) Calcite (1.49–1.74) Quartz (1.54–1.55) Feldspar (≈1.53–1.59) Kaolinite (1.53–1.58) Illite (1.54–1.61)
6	87.9	30.2	57.7	95.6	CH	Montmorillonite (1.48–1.64) Quartz (1.54–1.55) Calcite (1.49–1.74) Feldspar (≈1.53–1.59) Mica (1.53–1.70) Illite (1.54–1.61) Kaolinite (1.53–1.58)
7	33.5	18.3	15.2	24.4	SC	Quartz (1.54–1.55) Feldspar (≈1.53–1.59) Montmorillonite (1.48–1.64) Illite (1.54–1.61)
8	29	15	14	63.9	CL	Calcite (1.49–1.74) Quartz (1.54–1.55)

*Minerals are listed according to relative abundance, and the values in the parentheses represent the RI ranges of the minerals, which are derived from Table 1.

w_L = liquid limit (percent); w_P = plastic limit (percent); I_P = plasticity index (percent); USCS = Unified Soil Classification System; CL, CH, MH, and SC are the soil groups according to USCS and they represent low plasticity clay, high plasticity clay, high plasticity silt, and clayey sand, respectively.

that case, the following question arises: How should a representative value of RI for a soil sample be chosen? Since most soil samples are polymineralic in nature, unfortunately there is not a clear-cut answer for this question. Nevertheless, the following procedure may be indicated in order to overcome this drawback: first, an XRD analysis should be performed to determine the mineralogical composition of the sample, and then a representative RI value can be selected by considering the RI value of the most abundant minerals in the soil sample. In order to test the applicability of this procedure, XRD analyses were performed on eight selected samples to determine their mineralogical compositions. Index properties, mineralogical compositions, and refractive indices of these samples have been given in Table 3. According to the procedure presented above, the most appropriate RI value representing the samples given in Table 3 seems to be 1.55. However, it should be recognized that in most cases this procedure is not feasible or practical, since it requires the determination of mineralogical composition of the samples with XRD analysis prior to laser diffraction analysis. In these cases, the value of 1.55 for RI can be assigned to most soil samples. This value is quite logical for a number of reasons: (i) it falls within the range of 1.48 and 1.74, which is the range of most commonly found in soil-forming minerals; (ii) the variations in PSD

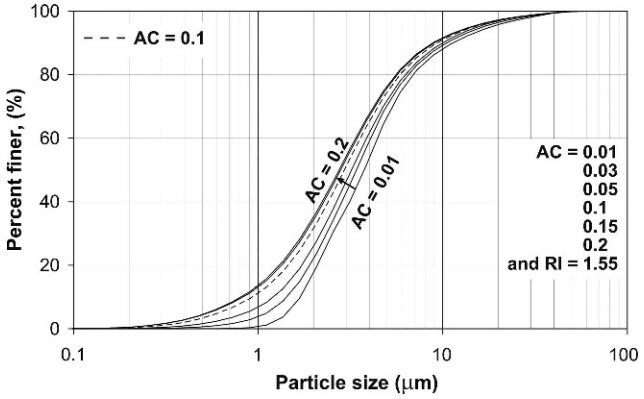

Figure 3. The effects of variations in absorption coefficient on particle size distribution.

and clay-size fraction of soils within the range of between 1.48 and 1.74 can be assessed as quite small for many practical purposes (Figure 1 and Table 2); and (iii) it corresponds to the RI value of quartz, which is a very common and abundant mineral present in most natural soils. In addition, the suggested value of 1.55 for RI is in agreement with the value that was used by other investigators (e.g., Loizeau et al. [1994]; and Buurman et al. [1997]). Besides, Muggler et al. (1997) used the value of 1.56 for RI, and Eshel et al. (2004) concluded that a RI value equal to 1.53 is suitable for most soil samples because the laser diffraction method is likely to overestimate the size of non-spherical particles (Jonasz, 1991). For this reason, they claimed that one should use a somewhat lower RI value to counteract this effect. As a consequence, for many practical purposes and most geotechnical tasks apart from scientific research, the value of 1.55 for RI may be suggested to practitioners in situations in which XRD analysis is absent and/or not feasible to perform.

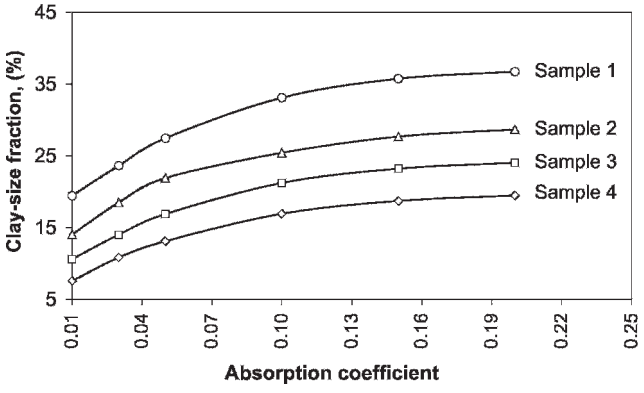

Figure 4. The effects of variations in absorption coefficient on clay-size fractions.

Table 4. *Clay-size fractions calculated by setting the absorption coefficient (AC) value at 0.01, 0.1, and 0.2, respectively, while refractive index (RI) was held constant at 1.55.*

	Clay-Size Fractions (percent) with Absorption Coefficient Shown Below			Differences	
Sample No.	(a) AC = 0.2	(b) AC = 0.1	(c) AC = 0.01	(a) − (b)	(b) − (c)
1	36.7	33.1	19.4	3.6	13.7
2	28.7	25.4	14.0	3.3	11.4
3	24.0	21.2	10.6	2.8	10.6
4	19.5	16.9	8.5	2.6	8.4
5	37.0	33.8	18.7	3.2	15.1
6	37.1	34.4	19.6	2.7	14.8
7	19.3	16.9	8.0	2.4	8.9
8	22.4	20.7	8.6	1.7	12.1
9	14.9	13.0	6.1	1.9	6.9
10	20.3	17.7	8.8	2.6	8.9
11	24.8	21.8	11.6	3.0	10.2
12	13.8	12.4	4.7	1.4	7.7
13	19.2	17.0	8.6	2.2	8.4
14	27.2	24.6	13.6	2.6	11.0
15	24.1	21.4	10.4	2.7	11.0
16	30.0	26.9	14.6	3.1	12.3
17	32.5	29.4	16.0	3.1	13.4
18	29.3	26.2	14.3	3.1	11.9
19	34.1	31.2	16.6	2.9	14.6
20	28.5	25.5	14.2	3.0	11.3
21	30.9	27.8	14.8	3.1	13.0
22	30.6	27.5	15.7	3.1	11.8
23	31.6	28.4	16.3	3.2	12.1
24	33.4	30.3	17.4	3.1	12.9
25	31.1	28.2	15.0	2.9	13.2
26	35.1	32.0	19.0	3.1	13.0
27	33.5	30.6	16.9	2.9	13.7
28	36.1	33.0	19.7	3.1	13.3
29	39.7	36.4	22.7	3.3	13.7
30	43.0	39.6	24.8	3.4	14.8
31	41.0	37.8	22.9	3.2	14.9
32	38.8	35.7	20.9	3.1	14.8
33	41.6	38.5	22.6	3.1	15.9
34	42.3	39.6	23.3	2.7	16.3
35	48.7	46.3	30.7	2.4	15.6

Effects of Variations in AC on PSD and Clay-Size Fraction of Soils

In order to examine the effects of variations in AC on apparent PSD and clay-size fraction of soils, samples 1 through 4 were analyzed by setting the AC at 0.01, 0.03, 0.05, 0.1, 0.15, and 0.20, respectively. In these analyses, the RI value was held constant at 1.55, as suggested previously. Since the results obtained from these analyses were almost the same for the four samples, only the results obtained from sample 1 are depicted in Figure 3. In addition, the sensitivity of the clay-size fraction to the AC is presented in Figure 4 using the results of four samples.

As can be seen from Figures 3 and 4, when AC increases, apparent PSD values shift toward fine grains, and clay-size fractions also increase. In order to

determine the effects of AC on the clay-size fraction for all 35 samples, the RI value was held constant at 1.55 while the AC values were varied between 0.01, 0.1, and 0.2, respectively. The results obtained from these measurements are presented in Table 4.

According to the results in Table 4, an increase in AC from 0.1 to 0.2 did not significantly alter the clay-size fraction; on the average, an increase of 2.8 percent was seen. However, an increase in AC from 0.01 to 0.1 resulted in an increase in the clay contents, on the average of 12.3 percent. As a result, it was demonstrated that the AC value also affected the PSD and clay-size fraction of soils significantly. However, because the AC value of soil-forming minerals changes with size, shape, and chemical alteration of particles; the presence of particle coatings; and the extent of particle surface abrasion (Sperazza et al., 2004), determination of this parameter is a very difficult task. For these reasons, the AC values of soil-forming minerals are not found in the literature (Buurman et al., 1997; Sperazza et al., 2004). Although PSD and clay-size fraction of soils are significantly affected by AC values, unfortunately, there is no key reference as to what the AC values of soil-forming minerals are. The only knowledge that we have about AC has to do with its limits; 0 for optically transparent materials and 1 for perfectly opaque materials, and we know that most soil-forming minerals are not completely transparent or opaque. Altogether, we must use a single value of AC in the analyses, and for this reason we should choose an acceptable value for AC that can be used for the analysis of soil samples. In this sense, one should take into account the suggestions in ISO 13320-1 (1999) and the findings of the present and previous studies dealing with the effects of AC on PSD and clay-size fraction of soils. In ISO 13320-1 (1999), a value of AC of between 0.01 and 0.1 was suggested for cases in which the AC value of the material is unknown. Accordingly, some investigators used a value of AC of near 0.1 for soil samples. For example, Buurman et al. (1997) and Muggler et al. (1997) used a value of 0.15, while Eshel et al. (2004) used 0.20 for the AC value. Sperazza et al. (2004) showed that as AC values are increased from 0 to 1, the PSD of soils becomes more unimodal and stable; however, they did not suggest any AC value to be used in the analysis of soil samples. Arriaga et al. (2006) found that a more appropriate value for AC is 0.001, together with 1.42 for RI, in order to match clay-size fractions obtained by laser diffraction as closely as possible with those obtained using the sieve-pipette method. However, as is known from previous studies (e.g., McCave et al., 1986; Loizeau et al., 1994; Buurman et al., 1997; Konert and Vandenberghe, 1997; Vitton and Sadler, 1997; Beuselinck et al., 1998; Eshel et al., 2004; Arriaga et al., 2006; and Özer, 2006), the laser diffraction method returns a lower clay-size fraction when compared to the pipette or hydrometer methods. In other words, the pipette or hydrometer methods overestimate the clay-size fraction as a result of Stoke's law. For this reason, the AC and RI values that match the results obtained from laser diffraction to the pipette or hydrometer method results should not be considered nominal values. In the present study it was shown that the variations in AC values greater than 0.1 did not significantly alter the PSD and clay-size fraction of soils. However, varying AC values between 0.1 and 0.01 shifted the clay-size fractions on the order of 12 percent. Consequently, according to the results and assessments obtained from the present and previous studies, an AC value of 0.1 seems to be a logical value that can be assigned to soil samples other than the mainly opaque (e.g., graphite, magnetite, and pyrite) or optically transparent (e.g., some quartz) ones.

The Mie Theory Versus the Fraunhofer Theory

Although several theories on light scattering exist, the most comprehensive and rigorous theory is that of Mie, which is based on Maxwell's electromagnetic field equations. However, when laser diffraction instruments were introduced in the late 1970s, the processing power of computers was limited, and since the use of the Mie theory demanded a considerable amount of computing power, manufacturers used the Fraunhofer approximation. Now, the computing power provided by modern PCs ensures that there is no longer a valid reason to continue its use (Jones, 2003). However, ISO 13320-1 (1999) states that although the Fraunhofer approximation can still be used for the measurement of particles that are greater than 50 μm, the Mie theory is recommended for those that measure less than 50 μm. In practice, some investigators (e.g., Loizeau et al., 1994; Konert and Vandenberghe, 1997; Beuselinck et al., 1998; Pabst et al., 2000; Murray, 2002; and Wen et al., 2002) used the Fraunhofer theory in their analyses, whereas others (e.g., Buurman et al., 1997; Muggler et al., 1997; Eshel et al., 2004; Sperazza et al., 2004; Arriaga et al., 2006; and Özer, 2006) preferred the Mie theory for the analysis of soil samples.

The main difference between the Mie and the Fraunhofer theories is that the Mie theory requires the RI and AC values of the samples and the RI value of the suspending medium, while the Fraunhofer theory does not. Because Fraunhofer theory does not make use of any knowledge of the optical properties of the materials, it is recommended by some

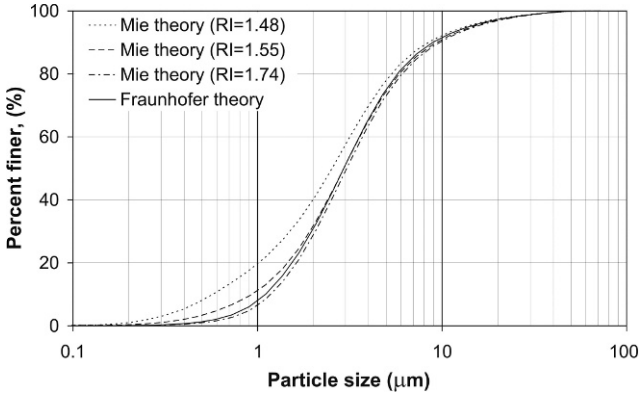

Figure 5. Comparison of PSDs calculated in accordance with the Mie and Fraunhofer theories (AC value was held constant at 0.1 in the Mie theory).

researchers (e.g., Loizeau et al., 1994; Konert and Vandenberghe, 1997; Beuselinck et al., 1998; and Wen et al., 2002) for the analysis of soil samples that are composed of a mixture of different minerals. However, some researchers (e.g., Buurman et al., 1997; Muggler et al., 1997; Eshel et al., 2004; Sperazza et al., 2004; and Arriaga et al., 2006) state that the Mie theory is more appropriate for the analyses of soil samples, especially for the determination of the clay-size fraction. In addition, some investigators (e.g., Bayvel and Jones, 1981; De Boer et al., 1987) indicated that the Fraunhofer theory is inaccurate for particles smaller than 10 times the wavelength.

In order to clarify these obscurities and to show the impact of the choice of optical theory on the output, it was necessary to compare the PSDs and clay-size fractions of soils obtained from the Mie and Fraunhofer theories. For this purpose, sample 1 was analyzed using both the Fraunhofer and Mie theories, and the results obtained are presented in Figure 5. In these analyses, RI values were set at 1.48, 1.55, and 1.74, respectively, while the AC value was held constant at 0.1 for the Mie theory.

According to the results shown in Figure 5, for all RI settings both theories yielded similar results for particle sizes greater than about 10 μm. However, for particle sizes smaller than approximately 10 μm, different particle sizes were obtained. The Fraunhofer theory gave larger particle sizes than the Mie theory when the RI value was set between 1.48 and approximately 1.70. Figure 5 also shows that when the RI value was set at 1.70 with an AC value of 0.1, both theories yielded similar results for all particle sizes.

It should be noted that in the analyses shown in Figure 5, the AC value was held constant at 0.1. However, it is clear from previous findings that if

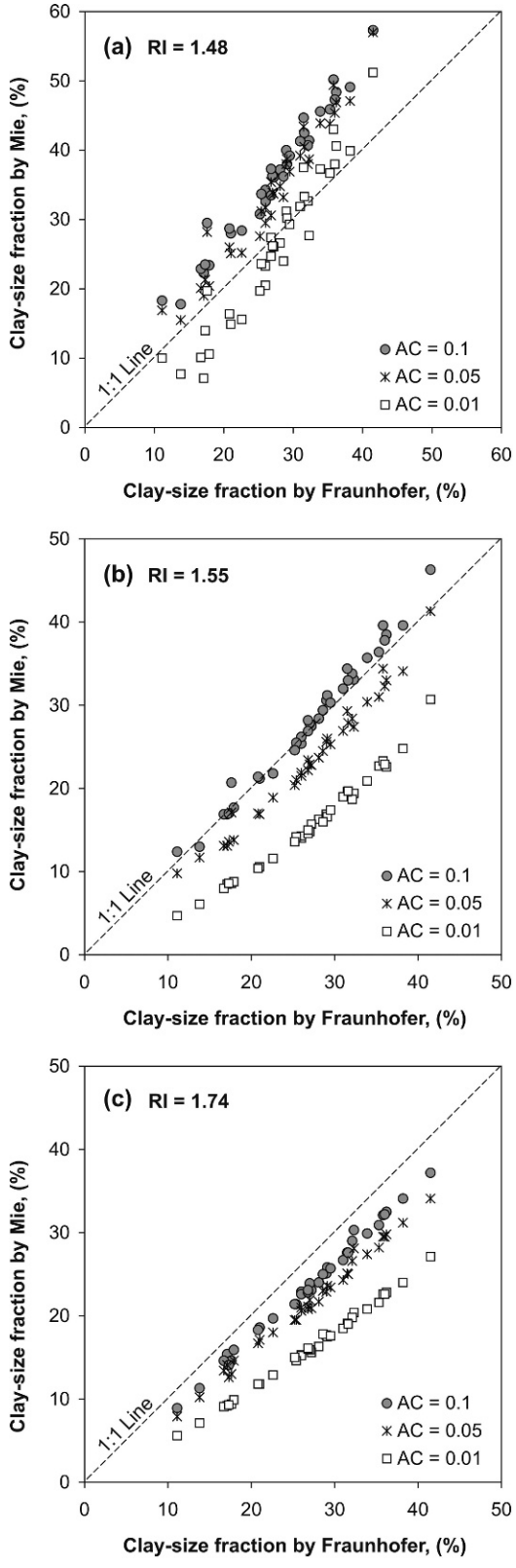

Figure 6. Comparisons of clay-size fractions calculated using the Fraunhofer and Mie theories by setting different AC and RI values. AC values were set at 0.1, 0.05, and 0.01, respectively, while RI values varied from (a) 1.48 to (b) 1.55 to (c) 1.74.

different AC values were used in the analyses, results different from those of Figure 5 might have been obtained. In addition, different results may be obtained from sample to sample. In other words, the effect of optical theory on the calculated clay-size fractions may vary depending on sample and optical parameters. For these reasons, extra analyses were performed on all of the 35 samples. In these analyses, the RI values were taken as 1.48, 1.55, and 1.74, respectively, while AC values were set at 0.1, 0.05, and 0.01, respectively. The results obtained from these analyses are given in Figure 6.

The results presented in Figure 6a indicate that the clay-size fractions obtained from the Mie theory calculations by setting the AC values at 0.1 and 0.05 and the RI value at 1.48 are always greater than those calculated with the Fraunhofer theory. When the AC value was set at 0.01, the Fraunhofer theory gave greater clay-size fraction than the Mie theory for soil samples having clay content lower than about 30 percent. As shown in Figure 6b, the clay-size fractions calculated with the Mie theory by setting the RI at 1.55 and the AC at 0.1 were very comparable to those calculated with the Fraunhofer theory. The clay-size fractions obtained with the Mie theory calculations with AC of 0.05 and 0.01 and RI of 1.55 were always lower than those obtained by the Fraunhofer theory. Figure 6c shows that the clay-size fractions calculated with the Mie theory using an RI of 1.74 and AC values of 0.1, 0.05, and 0.01 were always lower than those calculated with the Fraunhofer theory.

CONCLUSIONS

Based on the results obtained from 35 natural soil samples, it was determined that the variations in RI significantly affected the determined PSD and clay-size fraction of soils; when RI increased, PSD shifted toward larger grains and clay-size fractions decreased. Variations in RI between 1.40 and 1.60 significantly affected the clay-size fractions, whereas for an RI greater than 1.60 the effects were insignificant. The increase of RI from 1.48 to 1.74 resulted in decreased clay-size fraction, on the average of 12.4 percent (up to about 20 percent in some samples). The main difficulty encountered in laser diffraction analysis of soil samples was assigning a representative RI to the soil sample because of the polymineralic nature of soil. In this study, a procedure was suggested to overcome this difficulty. However, according to this procedure the mineralogical composition of the soil sample should be determined by XRD analysis prior to laser diffraction analysis. Unfortunately, for many practical purposes it is not possible or economical to perform an XRD analysis. In situations where XRD analysis was not available or feasible to perform, a value of 1.55 for RI is suggested as an appropriate value for the analysis of naturally occurring sedimentary soil samples.

It was also determined that AC value significantly affected the determined PSD and clay-size fraction of soils; when AC increased, PSD shifted toward the finer grains and, thus, clay-size fraction increased. The clay-size fraction increased on the average of 12.3 percent with an increase of AC value from 0.01 to 0.1. Because AC values of soil-forming minerals are absent in the literature and because there are not any authoritative references that include AC values of soil-forming minerals, it is difficult to assign a representative value to the soil samples. However, because an AC value needs to be assigned to the sample in order for it to be analyzed, this difficulty has to be dealt with. Quartz has a high resistance to physical and chemical weathering and is consequently the most abundant mineral in sedimentary soils. ISO 13320-1 suggests a value of AC ranging between 0.01 and 0.1 for silicon oxides (quartz and its varieties), depending on surface roughness. Therefore, an AC value of 0.1, beyond which no significant change in clay-size fractions takes place, may be suggested as an acceptable value.

In addition, the PSDs and clay-size fractions obtained from the Mie and Fraunhofer theories were compared by setting different RI and AC values for the Mie theory, and it was observed that these theories yielded comparable clay-size fractions only when we set the values of 1.55 and 0.1 for RI and AC, respectively, for the Mie theory.

ACKNOWLEDGMENTS

This work was supported by Gazi University Scientific Research Projects Directorate with Project No. 07/2004-22. The authors would like to thank the staff of this department. In addition, the authors would like to thank Prof. Dr. Reşat Ulusay (Hacettepe University, Geological Engineering Department) and Prof. Dr. Asuman G. Türkmenoğlu (Middle East Technical University, Geological Engineering Department) for their invaluable comments on an earlier draft of the paper.

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