ADAMA SCIENCE AND TECHNOLOGY UNIVERSITY

SCHOOL OF APPLIED NATURAL SCIENCE,

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LASER SPECTROSCOPY STREAM

LASER LIGHT SCATTERING AS AN ALTERNATIVE METHOD OF INVESTGATING COFFEE QUALITY

BY

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(RTE) radiations transport equation.

(LLS) laser light scattering.

(PM) particulate matter.

d_{50}=smallest sieve opening through which 50% or more of the material passes.

(EMR) Electromagnetic radiation.

(EM) electromagnetic.

(LASER) Light Amplification by Stimulated Emission of Radiation.

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ABSTRACT

The main focus of this work was to investigate particle size dependency of quality of coffee using laser light scattering. In these experiments a continuous wave frequency He-Ne (633nm) laser, lock-in amplifier, chopper were used as an apparatus. Green coffee powder was used as sample; the reason we choose green coffee as a sample is that to minimize the effect of roasting on the quality of coffee. Arranging all the apparatus as needed as the experiment was carried out and necessary data were recorded, and then analyzed to get different optical properties and particle size of coffee sample. After the determination of the optical properties of the diffusing medium (coffee) such as the absorption, scattering, reduced scattering, effective attenuation coefficients and anisotropy factor it was tried to relate with the quality of coffee. In this research also, efficient and objective discrimination of coffee quality determination method is developed by investigating particle size dependency of quality of coffee using Laser light scattering technique. The scattering coefficient has direct relation with the particle size as well as the quality; the larger the scattering coefficient is the finest the particle size and the highest the quality of coffee. Thus, since the developed method have wider economic and scientific applications; light scattering method is recommended as the most preferable for classification of coffee quality standards.

1. INTRODUCTION

Coffee has been for century the most commercialized food product and most widely consumed beverage in the world. Since the opening of the first coffee house in Mecca at the end of the fifteenth century, coffee consumption has greatly increased all around the world. In 2010, coffee production reached 8.1 million tons worldwide [1].

Particle size influences many properties of particulate materials and is a valuable indicator of quality and performance. This is true for powders, suspensions, emulsions, and aerosols. The size and shape of powders influences flow and compaction properties. Larger, more spherical particles will typically flow more easily than smaller or high aspect ratio particles. Smaller particles dissolve more quickly and lead to higher suspension viscosities than larger ones.

For many years measurement techniques based on laser light scattering theory have been applied to obtain information about the size, structure, shape, composition and concentration of substances [1, 2]. In this research, laser light scattering experiment is carried out in the following manner. That is, scattered light intensity (at different scattering angle) and transmitted light (at 0°angle) were measured experimentally and the result was calculated and analyzed by using Beer-Lamberts law and radiation transfer equation (RTE) to determine the optical properties of coffee samples, and then using angular distribution scattering intensity measurements, particle size determination was performed. Therefore, this research work will initiate a model for coffee variety classification which is consistent, efficient, time saving, cost effective and direct by exploring the technology of laser light scattering through the scattering coefficient and particle size parameter analysis.

Natural phenomenon include altitude, duration and severity of rainfall, type of soil, pH, genetic origin, location of producing area, chemicals and pesticides, harvesting methods and timing, packing and so forth [2]. Chemical compositions of coffee like caffeine chlorogenic acids and other aromatic and active components are also an internal factor that contributes to the quality and flavor of coffee beverage.

In different country coffee is graded for export with the objective of producing the best cup quality and there by securing the best price possible. However, there is no universal grading system. Each producing country has their own national standard which fulfills the minimum export quality requirement suggested by the market [3, 4, 5]. Most probably the flavor and quality of coffee depends on the coffee beans selected, the roasting process, the brewing method and the fineness of the ground coffee used. Each brewing method will brew better if the beans ground to the optimal size in the first place. The objective of the grinding is the reduction of the size of the roasted bean to facilitate the extraction of the flavor and of the soluble compounds during the preparation of the beverage. The time of contact between water and coffee determines how much flavoring material of the coffee could be extracted from the ground coffee. To avoid a low development or an excess of extraction of the components of the flavor, one should use the correct size of the particles of the ground coffee. This is why it is necessary to adjust the grinders to guarantee this effect. In commercial grinding, the process is well developed and monitored by the use of laser diffraction particle size analyzers. The speed, consistency, and ease of use have made it the most popular method for process control [6]. Particle size influences many properties of particulate materials and is a valuable indicator of quality and performance. This is true for powders, suspensions, emulsions, and aerosols. The size and shape of powders influences flow and compaction properties. Larger, more spherical particles will typically flow more easily than smaller or high aspect ratio particles. Smaller particles dissolve more quickly and lead to higher suspension viscosities than larger ones.

1.1 Objective

1.1.1General Objective:

- To introduce laser light scattering techniques one method of quality control.

1.1.2 Specific Objectives:

- To introduce a model of direct ease and precise method for coffee quality investigation.
- To introduce a method of measuring quantitative absorbance of a specific green coffee sample solution by collimated transmission intensity measurement;
- To determine the scattering particles size by angular distribution of intensity measurement.

1.2 Organization of the Thesis

Chapter-1 Introduction of the study is presented. In Chapter-2 historical, socio-cultural and economical aspects of coffee are presented, theory of electromagnetic radiation and its interaction with isotropic dielectric material medium is discussed briefly In this chapter Properties of electromagnetic radiation, Lasers and their properties and applications, light propagation theories like transmission, absorption, reflection and scattering of light with their respective quantitative and qualitative definitions as Beer-Lamberts law, Snell's law and Fresnel's laws are presented,. Model of light propagation through isotropic dielectric material medium is presented. Here radiations transport equation (RTE), methods of solving RTE in which analytical method of diffusion approximation equation and its assumption is discussed in detail. Solution of Diffusion approximation equation for infinite medium and fluency rate measurement equation are also presented in this chapter. In Chapter -3 Different methods used in this study are presented In Chapter-4. Result and discussions are presented. In Chapter -5 Conclusions and Recommendation for future works of the research will be presented. Finally, Reference materials used in this research will be reported as bibliography.

2. LITERATURE REVIEW

2.1 Historical, socio-cultural and economical aspects of coffee

Coffee presents one of the world's most favorite beverages. It is greatly appreciated for its delightful smell and flavor as well as for the stimulating effects of caffeine. While the beverage is consumed mainly in Europe, North and Central America, the coffee plant grows at elevated altitudes in tropical and subtropical regions all around the world. More than 5 million tons of green coffee beans are annually produced worldwide. Among all internationally traded food commodities, coffee holds a unique position with the greatest trade volume in financial terms. Some 20 million people earn a living directly from coffee production. Post-harvest processing is accomplished in the producer countries, resulting in green coffee beans ready for shipping. In the consumer countries roasting is the most important unit operation in roast coffee manufacturing.

A good quality cup of coffee is depended on many factors, such as the quality of green beans, the roasting conditions, the time since the beans are roasted, and the type of water used for brewing. More than 800 volatile compounds have been identified in roasted coffee, whereof around 30 compounds are responsible for the main impression of coffee aroma [25]. The overall quality and chemical composition of green coffee beans are affected by many factors, such as the composition of the soil and its fertilization, the altitude and weather of the plantation, the cultivation, and the drying methods used for the beans. Coffee plants are mainly grown in tropical and subtropical regions of central and South America, Africa and South East Asia, in temperate and humid climates at altitudes between 600 and 2500 m [37].

The genus coffee belongs to the botanical family of Rubiaceae and comprises more than 90 different species [30]. However, only C. arabica, C. canephora, and C. liberica are of commercial importance. As a result of modem breeding techniques some hybrids of C. arabica and C. canephora have recently been introduced with success. Usually roasted coffee beans from different origins are blended at specific ratios to provide coffee of unique flavor profiles. Often time, coffee beans are blended for the purpose of cost saving. Coffee cherries are harvested each year when they are bright-red, glossy, and firm. After removing the outer hull, the seeds inside of the cherry are commonly called "green coffee beans". The quality of the green coffee beans is dictated by a number of parameters, including bean size, color, shape, method of drying, crop year, and presence of defects (crack, withered bean, bean in parchment, mouldy bean, etc.).

The unique aroma profiles of coffee are closely related to the time-temperature profile used during roasting. The roasting profiles are chosen to produce high quality coffee which are unique to specific brands and must be strictly controlled to meet consumers' expectations. Coffee producers rely on sensory and physicochemical characteristic evaluations to assure that roasting takes place at the target process parameters. Industrial scale roasting of coffee beans is mainly achieved by conventional drum roasting, in which beans are heated with hot gas in a horizontal drum, or vertical drums equipped with paddles. Roasting time can range from 3 to 12 min, depending on the temperature used, which is typically between23^o C to250^oC. By contrast, fluidized bed roasting is achieved by directing high velocity hot air towards the beans, usually from the bottom of the roaster, to suspend the beans in turbulent air.

The hot air temperature ranges from 23^oC to 360^oC [31]. The roast temperature determines both flavor formation and structural product properties. Different temperature profiles affect dehydration and the chemical reaction conditions in the bean which control gas formation, browning and flavor development. In general, the use of roasting temperature of greater than 200°C is required in order to result in desirable chemical, physical, structural, and sensorial changes in the coffee beans [25, 29, 37, and 38]. Color change and weight loss are frequently used as a measure of the degree of roast, and both are directly related to the final roasting temperature [33]. Other methods, such as the ratios of free amino acids and chlorogenic acids content [33] have also been used.

External and internal factors can contribute to the quality of coffee beverage. Mainly man and nature are external factors. Each brewing method will brew better if the beans are ground to the optimal size in the first place. The objective of the grinding is the reduction of the size of the roasted bean to facilitate the extraction of the flavor and of the soluble compounds during the preparation of the beverage. The time of contact between water and coffee determines how much flavoring material of the coffee could be extracted from the ground coffee. To avoid a low development or an excess of extraction of the components of the flavor, one should use the correct size of the particles of the ground coffee. This is why it is necessary to adjust the grinders to guarantee this effect. In commercial grinding, the process is well developed and monitored by the use of laser diffraction particle size analyzers. The speed, consistency, and ease of use have made it the most popular method for process control [24].

Coffee variety identification is very useful in encouraging good quality coffee production, ensuring dependable and competent exporters as well as creating lasting business relationship with overseas clients. The basis of coffee variety identification is often subjective with attributes color, size, shape and flavor frequently examined by human inspectors [22, 23]. It is also found that human perception could easily be biased. As a result, objective discrimination of coffee varieties and quality determination is necessary which is consistent, non-destructive and cost effective for commercial purposes.

In light of these, it is useful to explore the possibilities of adopting the most economical and faster system which saves time and is more accurate in classification of quality of coffee by reducing observer effects of biases pertaining to the quality standard that enhances the commercial needs. One of such best method is scattering of laser light using roasted, grounded and dissolved coffee solutions. Scattering experiment is an applied spectroscopic technique that used to study the physical system when it interacts with electromagnetic radiation. More over it is a spectroscopic technique that studies about the production, transmission and detection of light with relation to the optical properties of a material medium.

2.2 The green coffee beans

The overall quality and chemical composition of green coffee beans are affected by many factors, such as the composition of the soil and its fertilization, the altitude and weather of the plantation, and the final cultivation and drying methods used. Coffee plants are grown in tropical and subtropical regions of central and South America, Africa, and South East Asia, mainly in regions with temperate and humid climates [38].

Brazil is by far the largest grower and exporter of green coffee beans in the world followed by Vietnam, Colombia, Indonesia, Ethiopia and India – producing nearly 2.5 million tons of green coffee beans per year [32]. The genus coffee belongs to the botanical family of Rubiaceae and comprises more than 90 different species [30]. However, only Coffea Arabica (Arabica), Coffea canephora (Robusta), and Coffea liberica are of commercial importance [38]. Arabica accounts for approximately 64% while Robusta accounts for about 35% of the world's production; other species with not much commercial value like Coffea liberica and Coffea excelsa represent only 1% [35]. Due to its more pronounced and finer flavor qualities, Arabica is considered to be of better quality and accordingly. The genus coffee belongs to the botanical family of Rubiaceae and comprises more than 90 different species [30]. However, only Coffea Arabica (Arabica), Coffea canephora (Robusta), and Coffea liberica are of commercial importance [38]. Arabica accounts for approximately 64% while Robusta accounts for about 35% of the world's production; other species with not much commercial value like Coffea liberica and Coffea excelsa represent only 1% [35]. Due to its more pronounced and finer flavor qualities, Arabica is considered to be of better quality and accordingly command higher prices [39, 33].

Coffee cherries are harvested when they become bright-red, glossy, and firm, either by selective hand-picking or non-selective stripping of whole branches or mechanical harvesting. The hand-picking method is very time-consuming, but results in a superior product quality because only ripe cherries are selected. After harvesting, the coffee fruits are separated from the pulp, which is carried out by dry or wet processing [28, 33]. The dry process is simple and inexpensive. The whole cherries are dried under the sun in open air, followed by the separation of the hull (dried pulp and parchment) mechanically to yield the green beans. On the contrary, the wet process requires greater investment and more care, but results in a superior coffee quality. In the wet process, the pulp of the coffee cherries, which is made up of exocarp and mesocarp, is removed mechanically, but the parchment remains attached to the beans. After drying either under the sun or in a dryer, the parchment is removed to produce the green coffee beans. Bean size, color, shape, processing method, crop year, and presence of defects, are some of the parameters used to evaluate the quality of green coffee beans [26].

Green coffee beans have minimal flavor. However, upon roasting, characteristic coffee aroma is developed due to the complex reactions that take place in the beans. To develop a unique flavor, green coffee beans are roasted according to different time-temperature profiles [27]. The aroma profile of roasted ground coffee is also related to the origin and variety of the beans. In general, blends with greater Arabica content tend to carry more fruity notes due to the aldehydes, acetaldehyde, and propanal, while the pyrazines give the earthy odor. In comparison, Robusta beans carry stronger "roasty" and "sulphury" note due to the presence of greater amount of sulphur-containing compounds [36]. Thus, Arabica is often added for the aroma effect while Robusta is used for enhancing the body, earthy and phenolic notes of the coffee blend [31]. Besides contributing to balanced flavor profiles, Robusta coffee is often blended with Arabica for cost reduction purpose. Robusta beans are lower in cost since the crops are more hardy to grow (more resistant to infestation) and easier to harvest (grown in regions of low elevation) than the Arabica counterpart.

2.3 Size and Properties of Particles

Nearly all, industries and research fields that handles materials needs or demands to analyze Particle Size and/or Shape. The flow characteristics of solid particles in a gas-solid suspension vary significantly with the geometric and material properties of the particle. The geometric properties of particles include their size, size distribution, and shape. Particles in a gas-solid flow of practical interest are usually of non spherical or irregular shapes and poly dispersed sizes. The geometric properties of particles affect the particle flow behavior through an interaction with the gas medium as exhibited by the drag force, the distribution of the boundary layer on the particle surface, and the generation and dissipation of wake vortices. The material properties of particles include such characteristics as physical adsorption, elastic and plastic deformation, ductile and brittle fracturing, solid electrification, magnetization, heat conduction and thermal radiation, and optical transmission. The material properties affect the long- and short-range inter-particle forces, and particle attrition and erosion behavior in gas-solid flows. The geometric and material properties of particles also represent the basic parameters affecting the flow regimes in gas-solid systems such as fluidized beds.

2.3.1 Particle Size and Sizing Methods

The number of techniques employed for particle size determination exemplifies its importance Microscopy Methods, Light Interaction Methods, Electrical Property Methods, Sedimentation Methods, Sorting and Classification Methods and perhaps, indicates the difficulty in acquiring this information [40].

Of the five methods techniques listed above, this research addresses only the light interaction method and only laser light scattering (LLS) technique within that category. To fully describe light scattering phenomenon, familiarity with several supporting theories is required. That background is provided here, but only to the extent necessary to provide continuity in the discussions that follow. Detailed information is available from various sources and the reader will need to research those sources if technical details are desired. This study focuses only on details pertinent to the main subject which is particle size dependency of quality of coffee by deconvolution of the light scattering using Mie theory.

Ambient particulate matter (PM) is a complex mixture of particles suspended in the air that vary in size and composition depending on their sources. Particles emitted from mobile sources tend to fall into a bimodal distribution referred to as nuclei mode and accumulation mode depending on the mechanism of formation. Nuclei mode particles are less than 50 nm in diameter and are generally made of hydrocarbons, sulfur, or metallic ashes. Accumulation mode particles range in size from about 50 nm to 500 nm and may contain elemental and organic carbon, nitrate, sulfate, and various metallic ashes.

Ambient particles are generally categorized into three size groups: ultrafine (< 100 nm), fine (between 100 and 2,500 nm), and coarse (>2,500 nm and up to 10,000 nm). Ultrafine PM includes nuclei mode and a fraction of accumulation mode PM.

Ultrafine particles: Particles operationally defined (mainly within the health-sciences community) as those having diameters less than 0.1μm

Fine particles: Particles operationally defined as those smaller than 2.5μm aerodynamic diameter. Fine particle measurements include the accumulation mode (nominally 0.1 to 2.5μm), where most of the submicron mass is found and, depending on measurement technique, may include ultrafine particles, where most of the particle number concentration is found. Because filter-based PM 2.5 sampling techniques collect all particles smaller than 2.5μm, such “fine particle” samples implicitly include ultrafine particles. However, because the properties and effects of ultrafine particles are different from those of larger particles, it is often useful to separately identify “fine” and “ultrafine” particles as distinct fractions of PM2.5.

Coarse particles: Particles extending through the high end of the aerosol size distribution. This assessment adopts the proposed regulatory definition, which includes those particles between 2.5μm and 10μm aerodynamic diameters (PM10-2.5). Many "coarse-particle" sampling techniques also collect particles in the finer ranges. Thus reported data designated as "coarse-particle" data may or may not include contributions from finer modes, and these contributions can be significant [10,18].

2.3.2 Particle size analysis

The particle size of coffee has traditionally been measured using sieve-based methods. This methods classifies particle as in table 6.4.1.2

Table 2.3.1: d_{50}=smallest sieve opening through which 50% or more of the material passes

Classification of powders	d_{50} -sieve opening(μm)
Very coarse	$>$ 1000
Coarse	355-1000
Moderately fine	180-355
Fine	125-180
Very fine	90-125

Although sieving has provided a means of differentiating between different grinds, it is both laborious and does not provide an accurate assessment of the amount of fine particles produced during milling. This lack of ability to resolve changes in the fine fraction limits the capacity of the technique to help control the quality of the final product.

Given the limitations of sieving, laser scattering/diffraction has become widely accepted for the characterization of ground coffee. The measurements are also rapid, providing full size distributions in a few seconds. The technique is therefore well suited to the requirements of production and quality control [45].

2.4 The Nature of Light

Energy transmitted by waves through a medium is called radiation or radiant energy; it includes electromagnetic, sound, and elastic waves. Electromagnetic energy includes cosmic rays, γ rays, X-rays, light, heat, microwaves, and radio waves. This continuum of energy is called the electromagnetic spectrum. Energy residing in a specific and very narrow band of this spectrum is referred to as ’light’. Wave theory often is used to describe electromagnetic radiation and therefore to describe light. Light waves, sound waves, water waves, etc. share some characteristics and can be studied using the same theories, but only in limited cases. Although similar in some respects, there are considerably more differences than similarities between sound waves and electromagnetic waves. The same holds true when studying details of electromagnetic waves in different bands of the spectrum; theories that apply to one band of wavelengths may not apply well to another band. This document is concerned only with light. Even within this narrow band of electromagnetic radiation, interaction between light and matter may produce very different effects depending on the wavelength of the light or the characteristics of the material; these variables may be referred to as the conditions under which an interaction takes place. Interactions that result in what is called *light scattering* are no exception. So, any description of light scattering must be accompanied by qualifying statements that define the conditions under which the statements (theories, explanations, or conclusions) are believed to be factual. The discussions that follow hold fast to this caveat; terms are defined and statements qualified as they pertain to the theoretical discussion. These definitions and qualifications may be referred to as limitations, conditions, constraints, or boundary conditions. Regardless, they are what ultimately will separate the theoretical ideal from real-world applications of light scattering theory.

A few additional facts about light should be mentioned. The smallest bundle in which electromagnetic energy is packaged is called a *photon*. All photons of light of a specific wavelength have a specific quantity of energy. The intensity of light of a specific wavelength is a function of the total number of photons emitted (or collected per unit area) per unit time. The study of optics also deals with light beams and light waves. A *ray* is the imaginary line that corresponds to the direction in which radiant energy flows. A *light beam* can be thought of as a group of parallel rays and as having finite thickness; a light beam also may be considered a stream of photons all moving in the same direction. To express it another way, a light ray is the limit of narrowness of a light beam. The branch of optics that is concerned with the path of light rays through optical systems is called *geometrical optics*. Light and, in fact, all electromagnetic radiation has a dual personality. Sometimes it behaves like a particle and sometimes it behaves like a wave, the wave most often being depicted as sinusoidal. This is the particle-wave duality characteristic often attributed to photons. In this document, light will be treated either as a continuous wave or as a photon, depending on which view best serves the current technical discussion [47].

2.5 Theory of light propagation in an isotropic dielectric medium

Propagation of light through material medium is the most important and interesting aspect of light. In this section the theories of interactions of light with matter are discussed. Starting from the definition and properties of electromagnetic radiation this chapter presents how and what phenomenon take place when light and matter interacts. More over it deals about mathematical representations that can often be used for qualitative and quantitative expressions of varied optical phenomenon exhibited by medium like reflection, absorption and scattering of light.

2.5.1 Electromagnetic Radiation

Electromagnetic radiation (EMR) is radiant energy that travels outward in all directions from its source through space with or without the need of a material medium. There are different types of EMR, some very familiar, and others rarely mentioned outside scientific discussions. Electromagnetic radiation includes AM/FM radio waves, microwaves, visible light (red to violet), ultraviolet radiation, X-rays, and gamma rays. Light is a form of electromagnetic radiation in the visible range from its broadened spectrum [41].

Electromagnetic radiation types are identified based on their frequency, wavelength, and source. The energy sources that produce EMR vary greatly, from nuclear reactions in the Sun, which generate gamma radiation, to chemical reactions in the human body that generate infrared radiation (heat) [41]. The electromagnetic spectrum is the term applied to all the types of EMR considered together in terms of frequency, wavelength, or energy. All parts of the spectrum are found, with varying intensity, in our natural environment. We are most familiar with the visible spectrum since we sense it directly with our eyes. And it ranges from 390nm to 780nm wavelength.

The infrared spectrum in which it's Wavelengths $\lambda > 780$ nm is sensed as heat and the ultraviolet spectrum ($\lambda < 390$ nm) includes radiation that can damage living cells, often causing a physiological response such as sun burn. Other parts of the spectrum may be present as background radiation. Natural background radiation originates from two primary sources: cosmic radiation and terrestrial sources. Cosmic radiation from deep space interacts with atoms in the atmosphere producing X-rays and unstable isotopes. Terrestrial sources of gamma radiation include radioactive isotopes of uranium, radon, potassium, and carbon [41]. The relative energy of the different types of EMR varies with frequency across the spectrum. Here below figure (2.5.1) shows the distribution of electromagnetic radiation spectrum.

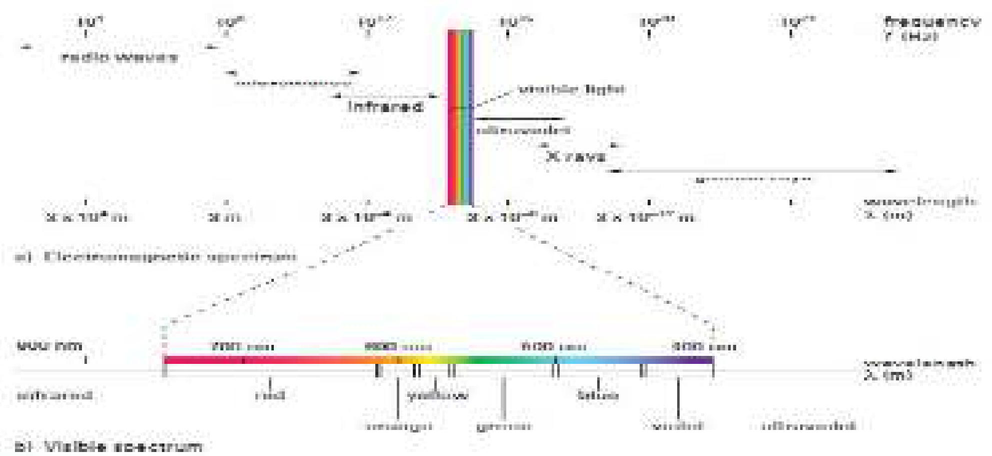

Figure 2.5.1: The electromagnetic spectrum showing the visible range.[61]

The actual light wave may be treated as a collection of plane monochromatic polarized waves of various frequencies. It may be more correct physically to represent an actual light wave as stream of micro particles, called photons [42]. Depending on the former nature of light, propagation of Electromagnetic Radiation or light through the material medium can be described by Helmoltz wave equation derived from four Maxwell's equations. The dual natures of light categorize its properties as wave like properties, (interference, polarization etc) and particles like properties, (photoelectric effect, and Compton Effect). The study of EMR specifically light propagation through material medium needs special properties of light which can be maintained constant or variable depending on the objective and the method of study.

2.5.2 Laser and Laser Applications

Most spectroscopic measurement techniques use a special light which has special properties for special conditions. The most known special light is laser light. The word LASER is an acronym for the most significant feature of laser action: Light Amplification by Stimulated Emission of Radiation. There are many different kinds of lasers, but they all share a crucial element: Each contains material capable of amplifying radiation. Thus Laser is a device that produces a beam of intense, directional (linearly polarized) and monochromatic light in which all the waves are in phase or are coherent. Lasers have three basic components: laser active medium, energy source (or pumping system), and resonant optical cavity with reflecting mirrors [43]. Active medium: contains atoms or molecules whose electrons may be excited to an elevated energy level by an external energy source (pumping source) to form population inversion. The excitation mechanism functions by pumping energy into the active medium by one or more basic methods such as optical, electrical, chemical, etc. Many scientific, military, medical and commercial laser applications have been developed since the invention of laser in 1958. The coherency, high mono-chromaticity, ability to reach extremely high powers with narrow band width, large frequency tuning range over considerable values and all other properties of Laser light allows it to play tremendous specialized applications [44]. The numerous fields of laser applications may be broadly categorized in to two groups.

One group involves applications where laser beams play a role of high power beam. This is exploited to produce a targeted effect on material. These applications include material processing (e.g., welding, heat treatment, cutting, hole burning, isotopes separation etc.) [43].

The other group includes data transmission and processing, measurements and quality control applications [43]. The high purity/mono-chromaticity of laser makes it as a very useful source for spectroscopy. For many years measurement techniques based on laser light scattering theory have been applied to obtain information about the size, structure, shape, composition and concentration of substances. Thus, for this scattering experiment, a linearly polarized continuous He-Ne laser with 35mW power at 632.8 nm wavelength is used to investigate scattering properties and particle size dependency of quality and flavor of grounded coffee.

2.6 Light Matter Interaction

When a beam of light is incident on material medium and propagates through it, various light-mater interaction phenomenons' takes place. These interactions can occur at the surface or inside the medium components and the rest of light may penetrate the medium (transmission). Due to this interaction of light with the medium components there might be reflection on the surface and attenuation due to scattering and absorption inside the medium by internal suspended components [7].

The absorption properties are strongly dependent on the wave length of the incident light while reflection and scattering have negligible effect on the wave length of the incident light. All these processes give concrete information about the size, concentration, motion and shape of the constituents of the medium [40]. The information we will obtain from this experimental work has a paramount importance in calculating the optical properties of the medium (coffee) and its particle size effects on the quality.

The interaction of light with material medium depends on the composition & concentration of the medium and the wavelength & energy of the light. A solution of coffee is turbid, and suspended coffee particle in the solution may scatter most of light energy in all directions. And it may have a little absorption peak on the corresponding wave length. In any conditions of material medium, when light tries to propagate through the given medium the following properties of light can happen.

- Reflection and Refraction
- Absorption
- Scattering

Here in figure (2.7.2) a typical situation is shown, where laser light beam is incident on material medium (specifically coffee solutions for this typical work) and it's interaction inside with suspended particles of coffee.

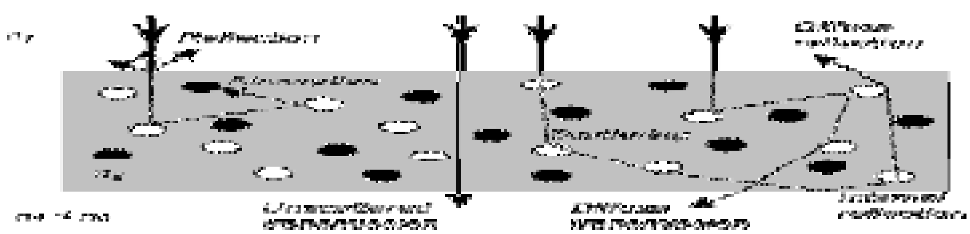

Figure 2.6.2: Geometry of Reflection, Refraction, Absorption and scattering

2.6.1 Transmission of He-Ne laser beam through coffee Solution

When collimated He-Ne laser beam impinges on a sample solution, a fraction of light energy is transmitted through the sample and the rest fraction is lost by three processes. These processes are reflection, absorption and scattering. Figure (2.6.2) above illustrates the interaction between Laser light and sample medium. Assume that the total incident light intensity is,I_O, the reflected light intensity is I_r, absorbed light intensity is I_a, transmitted light intensity is $\mathrm{I_t}$ and the scattered intensity of light is I_S. According to the conservation principle the normalized total incident intensity of light fulfills the condition;

$$I_O = I_r + I_a + I_s + I_t \qquad 2.6.1.1$$

And divide both sides of the above equation by I_O to write as

$$1 = R + A + T, \qquad 2.6.1.2$$

Where R is Reflectance (the ratio of reflected to incident intensities), T is Transmittance (the ratio of transmitted to incident intensities) and A is Absorbance in which the sum total of absorbed and scattered fraction of light intensities.

2.6.2 Reflection and Refraction

Reflection and refraction are the most common phenomenon when light and matter interacts with each other. Light intensity will be reflected or/and refracted because of a refractive index mismatch between the air and the sample boundary. The intensity of both reflected and refracted light doesn't only depend on the difference in refractive index mismatch but also depend on polarization, Angle of incidence, wavelength of light and structure & shape of the surface.

Reflection and refraction are strongly related to each other by Fresnel's law. Reflectance or the fraction of reflected intensity R, for unpolarized light is defined as the ratio of the reflected intensity from the medium to the incident intensity. This can be expressed as Fresnel's law [8],

$$R = \frac{1}{2}\left[\left(\frac{n_1 cos\theta_i - n_2 cos\theta_t}{n_1 cos\theta_i + n_2 cos\theta_t}\right)^2 + \left(\frac{n_2 cos\theta_i - n_1 cos\theta_t}{n_2 cos\theta_i + n_1 cos\theta_t}\right)^2\right] \qquad 2.6.2.3$$

Where n_1 and $n_2$are the refractive indices for the external medium (air) and material respectively, while θ_i and $\theta_t$are the incident and the transmitted angle respectively. Using Snell's law of refraction

$$n_1 \mathrm{Sin}\theta_i = n_2 \sin\theta_t \qquad 2.6.2.4$$

$$\theta_t = sin^{-1}\left(\frac{n_1}{n_2}\sin\theta_i\right) \qquad 2.6.2.5$$

When the light incident in the direction normal to the medium surface, i.e. $\theta_i = \theta_t = 0^o$, and if the external medium is air (n_1 = 1), and n_2 = n, equation (2.6.2.3) becomes.

$$R=\left(\frac{n-1}{n+1}\right)^2 \quad \text{……………………………………………………..2.6.2.6}$$

2.6.3 Absorption

Absorption is due to a partial conversion of light energy into heat, motion or certain vibrations of molecules of the absorbing material. Instead of completely transmitting light, an object or a substance can absorb part or the entire incident light, usually by converting it into heat. A substance is said to show general absorption if it reduces the intensity of all wave lengths of incident light nearly with the same amount. Instead many materials absorb some wavelengths while transmitting others, which are called selective absorption [9]. Laser light is monochromatic (i.e. single wavelength), which is responsible for its selective effects on the given sample medium.

Absorption occurs when the photon frequency incident on the medium matches with the frequency associated with the molecules transition. The ability of a medium to absorb electromagnetic radiation depends on a number of factors, mainly the electronic constitution of its atoms and molecules, the wavelength of incident radiation, the thickness of the absorbing layer, and the internal parameters such as the temperature and/or concentration of absorbing agents. The relationship between the absorption of light in a purely absorbing medium of thickness x, can be found using Beer-Lambert law [9]. For plane waves, the fractional reduction of the beam intensity $\frac{dI}{I}$ is directly proportional to an infinitesimal path length dx in its direction through a homogeneous medium with absorption coefficient μ_a i.e.

$$\frac{dI}{I} = -\mu_a dx \quad \text{……………………………………………………..2.6.3.7}$$

Integrating and evaluating it as definite integral in traversing a finitesimal thickness x we can find

$$\int_{I_O}^{I} \frac{dI}{I} = -\mu_a \int_0^x dx \quad \text{…………………………………………………2.6.3.8}$$

And the reduced intensity (I) can be written as

$$I = I_O \exp(-\mu_a x) \quad \text{………………………………………………... 2.6.3.9}$$

The absorption coefficient μ_a at a particular frequency can be related with the absorption cross-section σ_a and the number density of molecules ρ in the sample as follows

$$\mu_a = \rho\sigma_a \quad \text{.......} \quad 2.6.3.10$$

And it can be related again with the concentration c and absorptive as $\mu_a = c\epsilon_a$ then using the above two relations, the reduced intensity (I) can be rewritten as

$$I = I_O \exp(-\rho\sigma_a x) \quad \text{.......} \quad 2.6.3.11$$

$$I = I_O \exp(-c\epsilon_a x) \quad \text{.......} \quad 2.6.3.12$$

The absorption coefficient μ_a in equations above can thus be interpreted as the probability that the photon will be absorbed by the medium per unit length. The reciprocal $\frac{1}{\mu_a}$ is called the absorption path length (absorption depth) and equal to the mean free path a photon travels between successive absorption events. This is the distance required for the intensity (I) of the beam to fall to $\frac{1}{e}$ of the initial intensity (I_O) value.

2.6.4 Scattering

The scattering of light may be thought of as the redirection of light that takes place when an electromagnetic (EM) wave (i.e. an incident light ray) encounters an obstacle or non-homogeneity, in our case the scattering particle [11]. It is a fundamental factor in our visual perception of this world's phenomenon, such as the blue sky, red sunsets, clouds, rainbows, snow, fog, milk, and white paint are striking examples of the influence of scattering. When a cloud of smoke suspended particles, which has elastically bound charged micro particles, are exposed to electromagnetic waves, the particles are set into motion or vibration by the electric field.

Unlike absorption, scattering of light occurs at a non-resonance frequency, in which it takes place at frequencies not corresponding to those natural frequencies of particles. The oscillation or perturbation of the electron cloud on the particle results in a periodic separation of charge within the molecule, which is called an induced dipole moment.

The oscillating induced dipole moment is manifest as a source of EM radiation, thereby resulting in scattered light. The majority of light scattered by the particle is emitted at the identical frequency (v_o) of the incident light, a process referred to as elastic scattering. Rayleigh and Mie scattering are two types of elastic scattering, while Raman and Brillouin scattering are inelastic scattering in which the frequency of the scattered wave differs from that of incident wave. Rayleigh scattering is applicable to small (smaller as compared to incident wavelength), dielectric (non-absorbing), spherical smoke particles suspended in the solution, where as theory of Mie scattering encompasses the general spherical scattering solution (absorbing or non-absorbing) without a particular bound on particle size, most preferably structures with the same scale to the wavelength of the incident light wave [11]. The criteria for Rayleigh scattering is that x ≫1 and $|m|$ ≫1, where x is the dimensionless size parameter given by the expression

$$X = \frac{2\pi a}{\lambda} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots 2.6.4.13$$

Where a is the spherical particle radius, and λ is the relative scattering wavelength defined as

$$\lambda = \frac{\lambda o}{mo} \ldots\ldots\ldots\ldots\ldots\ldots\ldots 2.6.4.14$$

where λo is the incident wavelength in vacuum, m_o represents the refractive index of the surrounding medium and m is the refractive index of the scattering particle. Assuming the sample is homogeneous so that the scattering particles are uniformly distributed throughout its entire body. The non-scattered components of light I_x through a non-absorbing sample of thickness x=L, can be related with scattering coefficient μ_s as

$$I_x = I_o \exp(-\mu_s x) \ldots\ldots\ldots\ldots\ldots\ldots\ldots 2.6.4.15$$

The scattering coefficient μ_s at a particular frequency can be related with the scattering cross-sections and the number density of molecules in the sample as

$$\mu_s = \delta_s \rho \ldots\ldots\ldots\ldots\ldots\ldots\ldots 2.6.4.16$$

The scattering path length 1/ μ_s is the average distance the photon travels between successive scattering events which is known as mean free path.

When a photon that is incident along a direction described by a unit vector, ŝ experiences a scattering event, the angular probability of its being scattered in to the direction,$\hat{s}'$, with an angle θ is given by the normalize phase function, P (ŝ , $\hat{s}'$). It can be expressed as a function of the cosine of the scattering angle, P (ŝ,$\hat{s}'$) = cosθ. The mean cosine of the scattering angle called anisotropy factor, g, and it is a measure of degree of scattering which can be expressed as follow [12];

$$g\langle cos\theta\rangle = \int_{4\pi} (\text{p}\cos\theta)\, \text{d}\, (\text{p}\cos\theta) \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \ 2.6.4.17$$

If g is positive, the phase function is predominantly forward scattering; if g is negative, backward scattering dominates. A constant phase function results in an isotropic scattering (g = 0). Hence the transport (or reduced scattering coefficient), $\acute{\mu}_s$ is defined as

$$\acute{\mu}_s = \mu_s(1\text{- g}) \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots .\ 2.6.4.18$$

In most turbid medium absorption and scattering may be present simultaneously. Their total attenuation coefficient can be expressed as a combination of scattering and absorption coefficients of the medium by

$$\mu_t= \mu_a+ \mu_s \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 2.6.4.19$$

This tends us to express the total mean free optical path of incident photons in the medium as

$$L_t = \frac{1}{\mu_t} = \frac{1}{\mu_a+ \mu_s} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots .\ 2.6.4.20$$

In some cases, either μ_a or μ_s may be negligible with respect to each other, but it is important to realize the existence of both processes and the fact that usually both are operating. Also, it is very convenient to define an additional parameter, the optical albedo, α

$$\alpha= \frac{\mu_s}{\mu_t} = \frac{\mu_s}{\mu_a+ \mu_s} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 2.6.4.21$$

For α = 0, attenuation is exclusively due to absorption only, whereas in the case of α = 1, only scattering occurs.

For $\alpha = 1/2$, the total attenuation coefficient $\mu_t = 2\mu_a = 2\mu_s$ in equation above or/and which $\mu_a = \mu_s$ means the coefficients of absorption and scattering are of the same magnitude. In general, both effects may take place but they may occur in variable ratios [13]

Intensity Distribution of scattering of light and Airy Disk

In most turbid mediums photons are preferably scattered in the forward direction. The intensity of the scattered light is a function of the wavelength λ, the scattering angle θ, the particle size diameter d, and the relative index of refraction n of the particle and the surrounding medium. Symbolically, then, $I_s = I(\lambda, \theta, d, n)$. During the interaction of laser light with particles of the medium, either the particle has larger or smaller in size as compared to the incident light wavelength, Mie scattering theory describes the phenomenon by considering the optical properties (refraction and absorption) of the particles. The smaller the particles are the higher is the contribution of refraction and absorption to the light scattering pattern. For the measurement of particle size distributions inside the given sample medium, therefore, the scattering intensity distribution pattern should be detected over the entire angle range around the sample, and it will looks like as shown in figure (2.6.4.3).

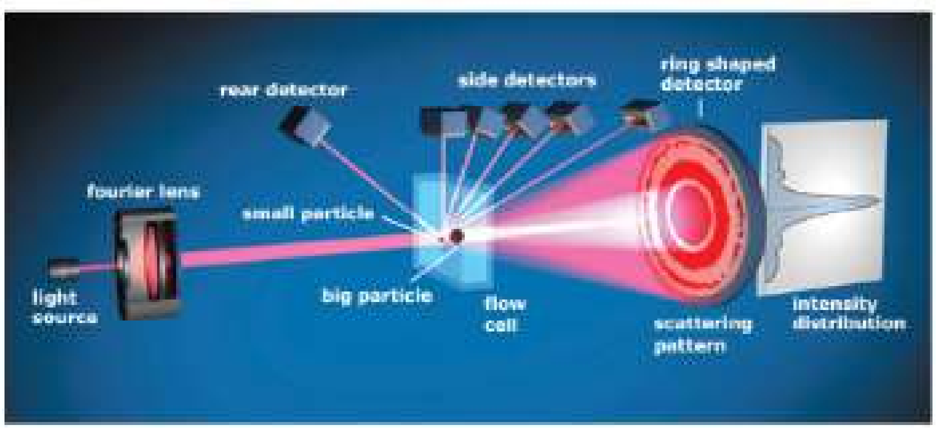

Figure 2.6.4.3: Intensity distribution profile of light propagation through medium (airy disk) [8]

The scattering intensity distribution pattern resulting from a scattering of laser light has a bright region in the center, known as the Airy disk which together with the series of concentric bright rings around is called the Airy pattern. The dark ring pattern between two bright regions shows the detected minimum intensity distribution. The diameter of the airy disk, d, at the center of the distribution corresponds to the particle size of the scatterer in the medium [9], that is helpful for calculating the dimensionless size parameter x, which can be expressed using diffraction equation as follows

$$d = \frac{1.22\lambda}{\theta} \quad \text{2.6.4.22}$$

Where the angle θ is measured from the incident transmission line to the first detected minima. The particle size calculation from the scattering intensity distribution pattern is useful for investigation of the optical properties of the medium especially here coffee medium samples.

Most researches announce that scattering coefficient calculations for coffee rather than Ethiopian coffee is in the order of $\sim 10^{-2}$ [15]. But here in Ethiopia there is no such research which can show the scattering or absorption coefficients of coffee, where as scattering experiment are done effectively and efficiently for different types milk sample mediums to find the particle size as well as concentrations of fats and proteins. Thus it is necessary to investigate a model for particle size and optical properties of coffee.

2.7. Model of light propagation through turbid medium

2.7.1 Introduction

When beam of light is passing through sample medium, its propagation is affected in two important ways:

(1) The intensity will decrease as the light penetrates farther in to the medium, and

(2) The velocity will be less in the medium than in free space. The loss of light intensity is chiefly, due to absorption, although under some circumstances scattering may play an important part [9]. In this chapter we shall discuss the model how beam of light propagate through the sample medium (specifically through medium of coffee sample solution for this typical experimental work). The model we used here is Radiative Transfer Equation (RTE), which can be solved analytically with Diffusion Approximation.

RTE model is used as good by other researchers for scattering experiment to determine fat and protein concentrations of milk and to investigate optical properties and particle size of milk sample solution with Indian ink absorber. But, here for coffee sample solutions, we used variable concentration method which makes the experiment easier and faster. Even though Photon transport in turbid medium can be equivalently modeled numerically with Monte Carlo simulations, analytical solution of RTE with diffusion approximation is more efficient computationally. Thus in this section mathematical model of photon transport through turbid medium and its solution will be presented. More over quantitative and qualitative expressions which can be used for determination of scattering, absorption reduced scattering coefficients and anisotropy factor of the medium will be discussed in detailed.

2.7.2 Radiative Transfer Equation (RTE)

The physical phenomenon of energy transfer (photon transfer) in the form of electromagnetic radiation is called Radiative Transfer. The propagation of radiation through medium is affected by absorption, emission and scattering processes. The equation of radiative transfer describes these interactions mathematically. The fundamental quantity which describes a field of radiation is nowadays called the spectral radiance, traditionally called the specific intensity. The flow of radiation energy through a small area element in the radiation field can be characterized by radiance L ($\vec{r}$, ŝ, t) (W/m^2sr), which is defined as energy flow per unit normal area per unit solid angle per unit time. Here, r denotes position, ŝ denotes unit direction vector and t denotes time [16].

The RTE simply says that as a beam of radiation travels, it loses energy to absorption, gains energy by emission, and redistributes energy by scattering. In RTE, Optical properties such as refractive index n, absorption coefficient, μ_a scattering coefficient, μ_s and scattering anisotropy, g, are taken as time-invariant but may vary spatially, and Scattering is assumed to be elastic. The integro-differential form of the equation for radiative transfer is given as [11]:

$$\partial L\frac{(\vec{r}, \hat{s},\ t)/v}{\partial t} = - \hat{s}.\nabla L(\vec{r}, \hat{s}, t) - \mu_t L(\vec{r}, \hat{s}, t) + \mu_t \int_{4\pi} L(\vec{r}, \hat{s}, t)\ P\ (\hat{s}'.\hat{s}) d\Omega' + S(\vec{r}, \hat{s}, t) \ldots\ldots. 2.7.2.23$$

Equation (2.7.2.23) describes the change of radiance, $L(\vec{r}, \hat{s}, t)$ at position in $\vec{r}$ the direction of the unit vector ŝ where v is the speed of light in material medium as determined by the relative refractive index n =$\frac{v}{c}$, μ_t= μ_a+μ_s is total attenuation coefficient, SL$(\vec{r}, \hat{s}, t)$ is a radiation source and P $(\hat{s}, \hat{s}')$ is scattering phase function representing the probability of light with propagation direction ŝ being scattered into solid angle dΩ around $\hat{s}'$. In most cases, the phase function depends only on the angle between the scattered $\hat{s}'$and incident ŝ directions, i.e.

P $(\hat{s}, \hat{s}')$ = P $(\hat{s}', \hat{s},)$ the scattering anisotropy can be expressed as

$$g= \int_{4\pi} L(\vec{r}, \hat{s}, t)\ P\ (\hat{s}'.\hat{s}) d\Omega' \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 2.7.2.24$$

The radiation transport equation above [equation (2.7.2.23)] can be derived by considering the radiation energy balance in arbitrary elemental volume of the medium. It is a balance equation relating the change of the energy radiation, $L(\vec{r}, \hat{s}, t)$ the left hand side term of equation (2.7.2.23) is the time dependent term referred to as temporal variance of radiance. The first term in the right hand side of the equation is the change of variance in the direction of the unit vector, $\hat{s}$. The expression $\mu_t L(\vec{r}, \hat{s}, t)$ represents the total attenuation of the intensity due to scattering and absorption. The integral term in right hand side of the equation is the gain due to scattering source, and the last term S $(\vec{r}, \hat{s}, t)$ is the gain due to the radiation source. Based on the definition of radiance, there are other important physical quantities like [10];

- Fluency rate or intensity $\Phi(\vec{r}, t) = \int_{4\pi} L(\vec{r}, \hat{s}, t)\ d\Omega W/m^2$
- Fluency $F(\vec{r}) = \int_{-\infty}^{+\infty} \Phi(\vec{r}, t) dt \vec{j}\ /m^2$
- Current density (energy flux) $\vec{j}\ (\vec{r}, t) = \int_{4\pi} \hat{s} L(\vec{r}, \hat{s}, t)\ d\Omega$W/m2

This is the vector counterpart of fluence rate pointing in the prevalent direction of energy flow. There are no general solutions to solve the radiation transport equation due to the inherent in homogeneities and irregularities with the physical shape of the scatterer in the medium. Even if RTE has application in wide variety of subjects including optics, astrophysics, atmospheric science and remote sensing, its Analytic solution exist for simple cases by introducing approximations. A common approximation summarized here is the diffusion approximations. But for more realistic media with complex multiple scattering effects numerical methods are required.

2.7.3 Diffusion Approximations for RTE

Light propagation in a highly turbid medium, can be considered as a process of energy diffusion because the properties of light wave (polarization, interference and diffraction) are lost due to multiple scattering events. Appropriate assumptions about the behavior of photons in a scattering medium per-minutes to reduce the number of independent variables in RTE equation (2.8.2.23). The following assumptions lead us to the diffusion theory (and diffusion equation) for photon transport in the medium. It should be noted that the following assumptions require a high-albedo (predominantly scattering) medium [16].

a) The spectral radiance will become nearly isotropic due to relative occurrence of absorption and scattering events, i.e. relative to scattering there are very few absorption events. Likewise, after numerous scattering events, few absorption events will occur.

b) In primary scattering medium the time variation of the radiant flux density over a length of $\frac{1}{\acute{\mu}_s}$ (which is one transport mean free path) is assumed to be negligible with respect to the flux density itself. Thus, over one transport mean free path, the fractional change in current density is much less than unity.

In diffusion theory, radiance, which can be expanded on the basis set of spherical harmonics $Y_{l,m,n}$, is taken to be largely isotropic, so only the isotropic and first-order anisotropic terms are used [23]:

$$\mathrm{L}(\vec{r}, \hat{\mathrm{s}}, t) = \sum_{n=0}^{1} \sum_{m=-n}^{n} \mathrm{L}_{\mathrm{n,m}}(\vec{r}, \mathrm{t})\mathrm{Y}_{\mathrm{n,m}}(\hat{\mathrm{s}}) \quad \ldots\ldots\ldots 2.7.3.25$$

Using properties of spherical harmonics and the definitions of fluence rate $\Phi\ (\vec{r}, t)$ and current density $\vec{\mathrm{J}}(\vec{r},\ \mathrm{t})$, the isotropic (for n = 0) and anisotropic (for n = 1) terms can respectively be expressed as follows:

$$\mathrm{L}_{0,0}(\vec{r}, \mathrm{t})\mathrm{Y}_{0,0}(\hat{\mathrm{s}}) = \frac{\Phi(\vec{r},\mathrm{t})}{4\pi} \quad \ldots\ldots\ldots 2.7.3.26$$

$$L_{1,m}(\vec{r}, \mathrm{t})\mathrm{Y}_{1,\mathrm{m}}(\hat{\mathrm{s}}) = \frac{3\mathrm{J}(\vec{r},\mathrm{t})}{4\pi}. \hat{\mathrm{S}} \ldots\ldots\ldots 2.7.3.27$$

Hence we can approximate radiance by the sum of both isotropic and anisotropic terms as [23]

$$\mathrm{L}(\vec{r}, \hat{\mathrm{s}}, t) = \frac{\Phi(\vec{r},\mathrm{t})}{4\pi} + \frac{3(\vec{r},\mathrm{t})}{4\pi} \quad \ldots\ldots\ldots 2.7.3.28$$

Substituting equation (2.7.3.28) for radiance in RTE equation (2.7.3.25), integrating the scattering term of the RTE over the complete (4π) solid angle and multiplying the RTE (2.8.3.25) for the vector form in the direction ŝ before evaluation, the proper RTE can be respectively rewritten in scalar and vector forms as follows [16]:

$$\frac{\partial\Phi(\vec{r},t)}{V\partial t} + \mu_a\Phi(\vec{r},t) + \nabla.\vec{J}(\vec{r},t) = S(\vec{r},t) \text{.......................................} 2.7.3.29$$

$$\frac{\partial\Phi(\vec{r},t)}{V\partial t} + (\mu_a + \acute{\mu}_s)\vec{J}(\vec{r},t) + \frac{1}{3}\nabla\Phi(\vec{r},t) = 0 \text{.......................................} 2.7.3.30$$

From the second assumption of diffusion theory above, we note that the fractional change in current density $\vec{J}(\vec{r},t)$ over one transport mean free path ($1/\acute{\mu}_s$) is negligible. The vector representation of the diffusion theory RTE equation (2.8.2.30) reduces to Fick's law [16, 17]:

$$\vec{j}(\vec{r},t) = \frac{-\nabla\Phi(\vec{r},t)}{3(\mu_a + \acute{\mu}_s)} \text{...} 2.7.2.31$$

This defines current density in terms of the gradient of fluence rate. And Substituting Fick's law equation (2.7.2.31) into the scalar representation of the RTE equation (2.7.2.29) gives the diffusion equation [16]:

$$\frac{\partial\Phi(\vec{r},t)}{c\partial t} + \mu_a\Phi(\vec{r},t) - \nabla.[D\nabla\Phi((\vec{r},t)] = S(\vec{r},t), \text{..............................} 2.7.2.32$$

Where $D = \frac{1}{(\mu_a + \acute{\mu}_s)}$ is the diffusion coefficient and $\acute{\mu}_s = (1-g)\mu_s$ the reduced scattering coefficient.

2.7.4 Solutions of the diffusion equation with point sources in an infinite homogeneous media

For simple case, using a short-pulsed point source in an infinite homogeneous medium, the diffusion equation can be solved for fluence rate. The source term in the diffusion equation (2.7.2.32) above becomes S $(\vec{r}, t, \vec{\acute{r}}, \acute{t}) = \delta(\vec{r} - \vec{\acute{r}})\,\delta(t - \acute{t})$, where $\vec{\acute{r}}$ and $\acute{t}$ are the positions at which fluence rate is measured and the position of the source respectively and the pulse peaks at time t'. Thus the solution can be presented as of the following [16];

$$\Phi(\vec{r}, t, \vec{\acute{r}}, \acute{t}) = \frac{v}{4\pi D(\vec{r} - \vec{r}')^{\frac{3}{2}}} \exp\left[-\frac{\left|\vec{r} - \vec{\acute{r}}\right|^2}{rDv(t - \acute{t})}\right] \exp\left[\mu_a v(t - \acute{t})\right] \text{..................} 2.7.2.33$$

The exponential decay in fluence rate due to absorption in accordance with Beer's law is expressed in the equation (2.7.3.33) above with the term exp [$\mu_a v$(t $-\acute{t}$)] and the other terms represent broadening due to scattering. Taking time variation out of the diffusion equation gives the following for the time-independent point source S ($\vec{r}$) = $\delta(\vec{r})$

$$\Phi(\vec{r}) = \frac{1}{4\pi Dr} exp^{-\mu_{eff} r} \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots 2.7.2.34$$

Where $\mu_{eff} = \sqrt{\frac{\mu_a}{D}} \sqrt{3\mu_a(\mu_a + \mu_s)}$ is the effective attenuation coefficient and indicates the rate of spatial decay influence.

3. MATERIALS AND EXPERIMENTAL PROCEDURES

3.1 Introduction

In this study different methods and techniques are used to make it easy and clear. The theories and laws used to analyze optical properties of diffusing medium (coffee solution) have been also reported. Different model of light propagation in turbid medium such as radiative transfer equation, diffusion approximation and solutions of the diffusion equation with point source in an infinite homogeneous media are presented to determine the effective attenuation coefficient.

3.2 Materials

The materials used in this experiment for standard sample preparation are as follows. Filter paper, distilled water, volumetric flasks, beakers , measuring cylinders, magnetic stirrer , funnel, separatory funnel, cylindrical glass cuvette with diameter 15mm and rectangular glass cuvette (15mm, 15mm, 20mm). Unwashed Nekemte Arabica coffee sample prepared as a sample. The laboratory apparatus and instruments used for this work are the following He-Ne laser at 62.8nm wavelength, chopper, chopper controller, collimator, photo diode detector, lock-in amplifier, and computer (PC).

3.3 Lock-in Amplifier

In its most basic form a lock-in amplifier is an instrument with dual capability. It can recover signals in the presence of an overwhelming noise background or, alternatively, it can provide high resolution measurements of relatively clean signals over several orders of magnitude and frequency. However, modern instruments offer far more than these two basic functions and this increased capability has led to their acceptance, in many scientific disciplines, as units which can provide the optimum solution to a large range of measurement problems. For example, the modern lock-in amplifier will function as: - an AC Signal Recovery Instrument, a Vector Voltmeter, a Phase Meter, a Spectrum Analyzer, a Noise Measurement Unit,…..and much more. It is this versatility, available in a single compact unit, which makes the lock-in amplifier an invaluable addition to any laboratory. This Technical Note describes the basic “building blocks” of the lock-in amplifier so that the user and potential user may better understand how the instruments work and how the choices made in their design affect their performance.

3.4 Methods

3.4.1 Coffee sample preparation

The preparation of sample of coffee solution for this experimental work was carried out as follows. A rectangular and cylindrical cuvette were thoroughly cleaned, rinsed with distilled water and dried before used. An accurately weighed amount of grinded green coffee beans (7.5g) was dissolved in distilled water and made the solution 240ml. The solution was heated and stirred for about 25 minutes using magnetic stirrer with hot plate. In addition the solution was filtered using a filter paper to get rid of particles from the solution. And then by weighing the filtered residue after drying, it was necessary to subtract the amount of residue from the first dissolved amount of coffee (7.5g) to know the amount of solute concentration in water solvent. This is our concentrated stock solution. Then after that by taking a small amount of sample (2ml first, with gradually equal increase till 8ml) solution from the prepared stock solution. The proportions of concentration are expressed using table and figure as follows:

(a) Green coffee beans (b) Green coffee powder (c) sample solutions

Figure 3.4.1.1: proportions of prepared solutions with gradually increased concentrations.

Solution	1	2	3	4
Volume stock soln.(ml)	2	4	6	8
Final dilute soln.(ml)	40	40	40	40
Concentration(%)	5	10	15	20

3.4.2 Liquid-Liquid Caffeine Extraction

Liquid-Liquid extraction is a process of separation of components of a solution in which a solute is distributed between two immiscible solvents and this process can be implemented by chloroform and dichloromethane solutions [46]. Here dichloromethane solution was used for extraction of caffeine from coffee.

The extraction process as used by Abebe [46] is used. The first step was preparing Sample and accurately weighed amount of grinded green coffee beans (200mg) was dissolved in 100ml of distilled water. The solution was stirred for about 25minutes using magnetic stirrer with hot plate. In addition the solution was filtered through filter paper to get rid of particles from the solution. Then this prepared solution was mixed with dichloromethane by volume ratio (1:1). This mixture was stirred for 15 minutes, and then by separatory funnel caffeine was extracted by dichloromethane from the coffee solution. The extraction was repeated for 4 times by 100ml dichloromethane at each round. Then the extracted caffeine by dichloromethane and the solution in which the caffeine was extracted from it was stored in volumetric flasks. Then finally the attenuation coefficient μ_t and particle size of the caffeine extracted coffee solution were measured and compared with the normal solutions before the caffeine was extracted from it.

3.5 Optical measurement techniques

The study was focused on the measurement and calibration of the optical properties of the diffusing medium such absorption, scattering, reduced scattering effective attenuation coefficients and an isotropic factor. In measuring the optical properties and particle size of a diffusing medium three phases of measurement were considered. These are collimated transmission measurement, fluence rate measurement and angular distributions of intensity measurement.

3.5.1 Collimated transmitted measurement

The collimated transmitted light that passes through the sample must be passed through the collimator. The aim of passing the collimated transmitted light through collimator is that to avoid the scattered light from entering the detector. Figure 3.5.1.2 shows how it works.

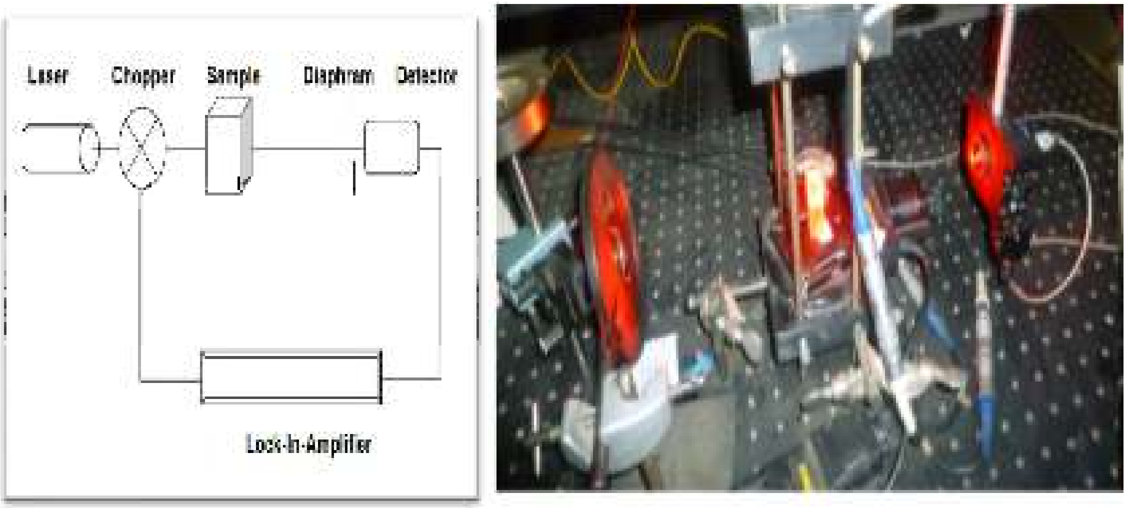

Figure 3.5.1.2 Schematic diagram of the experimental setup for transmission measurement. The transmitted (unscattered) light through the coffee sample was detected with photo diode detector connected to the lock-in amplifier. The lock-in amplifier again connected to the chopper, which is fixed for all phase measurements, to purify the signal from other interfering frequency noise signals. The photo-diode detector used had a sensitivity curve in visible range of wavelengths. The initial intensity of light I_O in which it was first passed though the sample cell containing only distilled water was recorded. This was done to compensate for refractive index mismatches between the external medium (air) and the surface of the cell. Then the corresponding intensities of light I, passing through different concentrations of the sample in the light path was measured. This was done by taking a small sample solution from diluted sample prepared first with gradually increased concentrations by 2ml (5%) until 8ml (20%) values were reached. The collimated transmission method is used to calculate the total extinction coefficient of the diffusing medium ε_t. It was also used to investigate the optical properties of the sample and this would be done by applying Beer Lambert Law.

$$I = I_o \exp(-\mu_t L) \qquad \text{3.5.1}$$

Where $\mu_t(\mu_t = c\varepsilon_t)$ is the total attenuation coefficient, ε_t is the total extinction coefficient, c is the concentration of the sample and L is thickness for total path length of light through the sample in the Cavite.

3.5.2 Fluence Rate Measurements

In this method (phase) the multi angular distance measurement fluence rate or emittance in which the rate of total amount of photons crossing over surface of unit cross section, inside the highly diffusing medium was performed. Figure 4.5.4 shows the set up. He-Ne laser beam was incident on a cylindrical sample cell of diameter 15mm containing distilled water only to measure the initial light intensityI_o. The cylindrical sample cuvette used here was preferable because it's smooth and curved surface was necessary to get equal thickness around the sample cuvette for photo diode detector.

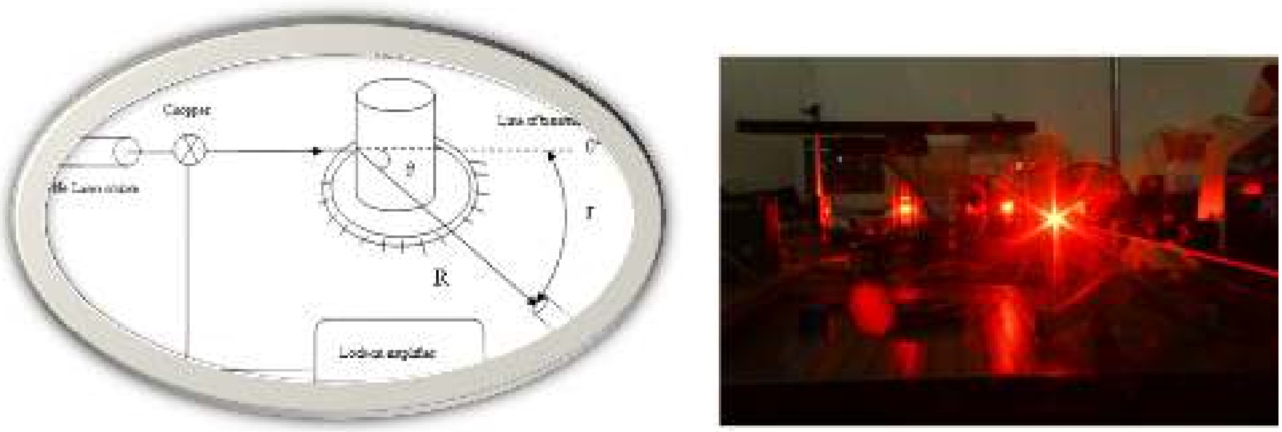

Figure 3.5.2.3: Schematic diagram of multi-angular distance measurements set up.

The corresponding intensities of laser light I for a series of gradually increased variable concentrations were measured. The concentrations of the samples used in these measurements were taken from the first prepared solutions with gradually increased concentration from 2.3% 9.2%.For a variably concentrated coffee samples of the added amount concentration, the fluence rate was measured by varying the angle from 18 °to 45° in steps of9°. The angular measurement was taken from the line of transmission in both clockwise and counterclockwise directions. The source to detector distance 17cm was kept constant.

The curve distance from the line of transmission to the detector at any angle θ is r=Rθ which is measured using spectrometer scale. The fluence rate as a function of the curve distance between the line of transmission 0^o and the detector (r) as it mentioned in the radiative transfer equation in chapter two is defined as

$$\phi(r)=\frac{1}{4\pi D}\exp(-\mu_{eff}r) \quad \text{3.5.2}$$

Where D is the diffusion coefficient defined by $D = \frac{1}{3\mu_s^{'}}$ for the medium having high albedo (α) and by rearranging the above fluence rate equation it can be written as follows:

$$\ln(\phi(r)r) = -\mu_{eff}r + \ln\frac{3\mu_s^{'}}{4\pi} \quad \text{3.5.3}$$

After the effective attenuation coefficient was found, it was possible to find the reduced scattering coefficients $\mu_s^{'}$. Using the equation $\mu_{eff}^2 = 3\mu_a\,(\mu_a+\mu_s^{'}) \approx \; 3\mu_a\mu_s^{'} \rightarrow$ for $\mu_a \ll \mu_s^{'}$ the anisotropy factor (g) can be calculated using the relations $\mu_s^{'} = (1\text{-g})\mu_s$ for a specific concentration.

3.5.3 Angular Distribution of Intensity Measurements

In this phase we took two types of measurements as of the first phase that used for determination of the particle size of coffee sample solutions with and without caffeine. These measurement has great significant to compare whether or not caffeine particles participate for scattering phenomena in the solution. Both these measurements have the same method.

The only difference for caffeine extracted coffee sample measurements was taking sample from the stock solution itself directly without dilution with water. This was done because the concentration of the extracted caffeine cannot measure directly from dichloromethane.

The first step in this phase should be measuring the incident He-Ne laser beam intensity on a sample cell containing only distilled water. A 4ml coffee stock sample solution was diluted by distilled water to make a 5% by volume 40ml solution. And then by taking 16.7ml sample from these 40ml solution the intensity measurements were carried out. The intensity of light was detected and measured four times by varying the angle around the sample from the line of transmission 0^oclockwise and counterclockwise using the photo diode detector of the spectrometer. The Airy disk (or a diffuse circular disk) would be formed from the distribution of detected light intensity rather than forming a bright dot as image when light from He-Ne laser source passes through spherical scattering particle [9]. The diameter of the Airy disc corresponds to the diameter of the scattering particle. The size of the particle can be calculated from the angle measurement and using the diffraction equation

$$d = \frac{1.22\lambda_o}{\theta} \quad \ldots\ldots 3.5.4$$

The spherical scattering particle size (diameter) can be computed using the value of d. θ in the above expression represent the angle from the transmission line 0° to where the first minima occur. The particle size parameter can then be calculated

$$x = \frac{d\pi}{\lambda_o} n_{med} \quad \ldots\ldots 3.5.5$$

where n_{med} is the refractive index of the surrounding medium at room temperature and λ_o is the wavelength of the incident He-Ne laser beam. A Mie scattering theory of particle size calculation again yields the scattering efficiency Q_s of the scatterer in the solution to know the amount of efficient cross-sectional area for scattering phenomena as follows.

$$Q_s = \frac{\mu_s}{\rho_s A} \quad \ldots\ldots 3.5.6$$

where A is the true geometrical area of the scatterer and $\rho_s$is the particle number density which relates the scattering cross sectional area σ_s to the true geometrical area $A = 4\pi r^2$ of the spherical particle by the equation

$$\sigma_s = \frac{\mu_s}{\rho_s} \quad \ldots\ldots 3.5.7$$

After the determination of the optical properties of the diffusing medium (coffee) such as the absorption, scattering, reduced scattering, effective attenuation coefficients and anisotropy factor it was tried to relate with the quality of coffee.

4. RESULTS AND DISCUSSIONS

In this chapter the results found from the three phases of experiments are presented independently. The analysis was performed by using Beer-Lamberts Law and RTE in the diffusion approximation theory. Graphs and some algebraic calculations are used in the discussion to show the relationship between various quantities. The relationship between concentration & absorbance was used to calculate the extinction coefficient of the given coffee sample medium. Curve distance & fluency rate graph is used to determine scattering, reduced scattering and absorption coefficients and anisotropy factor. The diffraction equation was used to calculate the particle size and scattering parameter of the sample. Finally comparisons of particle sizes of the caffeine extracted coffee sample with the sample solution without caffeine is extracted would be performed.

4.1 Total Extinction Coefficients.

The collimated transmission measurement phase of the experiment, section (3.5.1) phase I is used to calculate the total extinction coefficients of the prepared sample coffee medium. In the diluted samples, the concentration of coffee was varied from 5% to 20% with 5% variation by volume. From the total sample 2ml Sample having a concentration increment of 0.34% (from 0.34% to 1.36%) were taken by a rectangular cuvette for the measurement. The initial incident intensity I_O was measured with the sample cuvette containing only the solvent, which was distilled water and found to be I_O = 118.1a.u. Then the corresponding intensities of light I, passing through different concentrations of the sample (coffee) in the light path was measured. The experimental results for the sample with different concentrations of prepared stock sample solutions were shown in Table 4.1

Using Beer-Lamberts law equation (3.5.1), the relationship was plotted for $\frac{1}{L}\ln\left(\frac{I_O}{I}\right)$ against concentration, Figure (4.1.1). The slope of the straight line represents the total extinction coefficient ε_t, which is the attenuation of the intensity of light due to absorption and scattering in the sample. Thus we obtain ε_t=4.5x10^{-3} from the graph.

Table 4.1.1: Transmission intensity measurements for different concentrations of coffee type samples

Coffee sample	**Concentration**	**Intensity**	$\frac{1}{L}\ln\left(\frac{I_O}{I}\right)$
Unwashed Nekemte	0	118.1	0
	0.34	116.2	0.0011
	0.68	114.4	0.0022
	1.02	111.9	0.0037
	1.36	107.8	0.0061

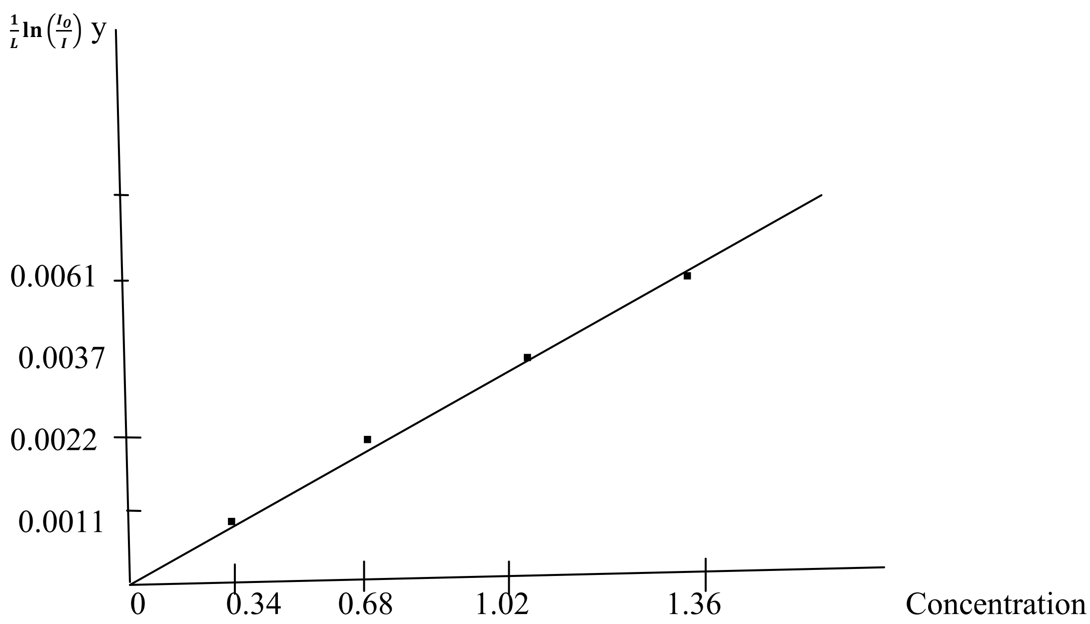

Figure 4.1.1: Graphs of $\frac{1}{L}\ln\left(\frac{I_O}{I}\right)$ versus concentration C of coffee samples used in this experiment.

The total attenuation coefficient μ_t of caffeine extracted coffee sample were calculated using equation (3.5.1). In this step of experiment we use the sample which caffeine was extracted and the corresponding intensity for the given sample was detected as I = 107.8a.u. And the result of calculation from the experiment gives us the total attenuation coefficient μ_t=6.08x10^{-3}mm^{-1}.

4.2 Absorption, Scattering and Reduced Scattering Coefficients and Anisotropy Factors

Multi angular distance or fluence rate measurement method was carried out to investigate absorption, scattering and reduced scattering coefficients and anisotropy factors of used coffee sample mediums. This phase measurement results were analyzed based on the solution of the radiation transport equation (RTE) for a point source in an infinite medium with diffusion equation (DE) approximations. The concentrations of coffee were varied from 2.11%to 8.44% in volume ratio for which the fluence rate intensity ϕ(r) was attenuated. The raw data of the measurements were presented in Table (4.2.2) and their corresponding graphs for ln(ϕ(r)r) versus r are shown as figure (4.2.2).

Table 4.2.2: Fluence rate measurement raw data of coffee samples used in the experiment

Coffee sample	**Angle θ°**	**Curve distance r= θR(mm)**	**Φ(r) ΔC=2.11%**	**Φ(r) ΔC=4.22%**	**Φ(r) ΔC=6.33%**	**Φ(r) ΔC=8.44%**
Un washed Nekemte	18	56.34	13.1	16.98	21.43	26.81
	27	84.51	8.3	9.4	12.3	16.1
	36	112.68	4.2	6.2	7.2	11.2
	45	140.85	2.5	4.1	4.6	7.2

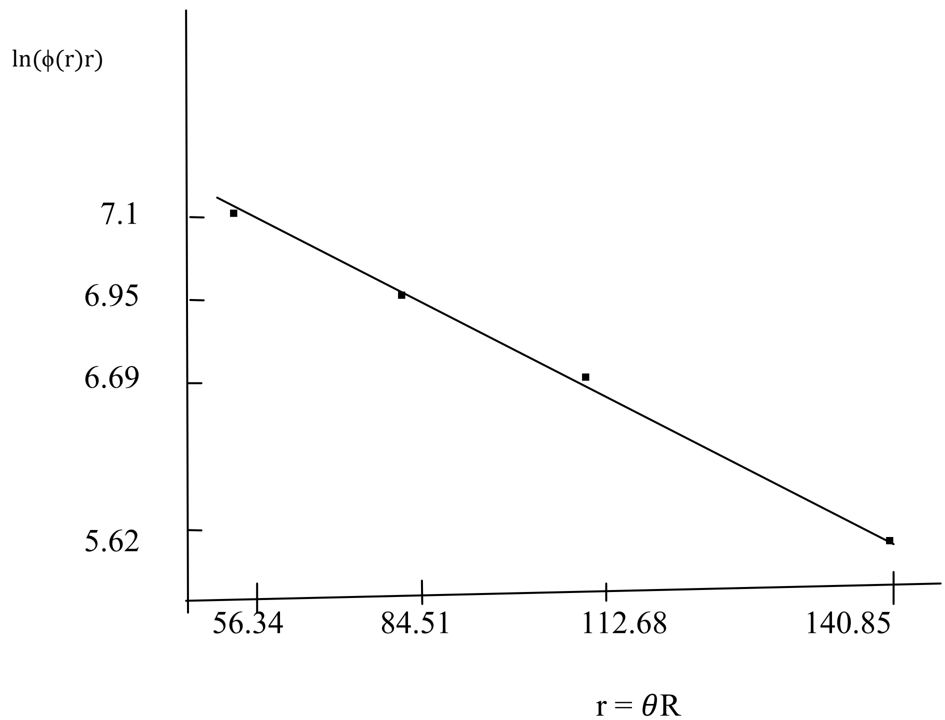

Figure 4.2.2: Graphs for fluence rate measurements (ln(ϕ(r)r)versus r) for coffee samples used in the experiment.

From the graph it can be observed that the fluence rate increases as the concentration of the medium (coffee) increases. The slopes of these curves in the graph are increasing according to the increase in concentration of coffee samples. This indicates that the samples of coffee used in this experiment have high albedo constant or it has no absorption coefficient (μ_a =0) for this given wavelength value of light, in which 633nm He-Ne laser. This shows that the results are in good agreement with the diffusion equation approximations in the literature in which high albedo medium is necessary to use those approximations. To calculate the effective attenuation coefficient μ_{eff} and reduced scattering coefficient $\mu_s^{'}$ from the slope of the graphs and equation (3.5.3) respectively, a curve with specific concentration of the scatterer 6.33% by volume is selected from samples of coffee graphs.

The evaluation of the slope of (ln(ϕ(r)r) as a function of distance, r, gives us the effective attenuation Coefficient μ_{eff}^2=3$\mu_a\mu_s^{'}$ which intern is used to find the reduced scattering coefficient.

μ_{eff}^2= 3$\mu_a\mu_s^{'}$, the $\mu_s^{'}$ is simply proportional to the concentration of scatterers, where as the absorption coefficient in the scattering medium is due to only the absorption of the distilled water used for dilution which has an absorption coefficient of $2.6\text{x}10^{-4}$ at 633nm wavelength[25,26].

Since μ_a =μ_{as} +μ_{aw} and μ_{as}=0, where μ_{as} and μ_{aw} are the absorption due to both scatterer and pure water respectively, the reduced scattering coefficient $\mu_s^{'}$ of coffee samples are presented in table (4.2.3) by using $\mu_s^{'} = \frac{\mu_{eff}^2}{3\mu_{aw}}$

Considering the loss in intensity due to reflection is minimum, the total extinction coefficient is as a result of absorption and scattering. And since the absorption coefficient of scatterer is zero (μ_a= 0), the total extinction coefficient is equal to the extinction coefficient due to scattering only (ε_t = ε_s). Hence multiplyingε_s , which is obtained using phase-I measurement results of the experiment, with concentration c results in the scattering coefficient μ_s of the scatterer. Using this value and the value obtained for the reduced scattering coefficient, the magnitude of the anisotropy factor g for coffee sample was computed. All the results of necessary parameter calculations are summarized in Table (4.2.3) as follows:

Table 4.2.3: Summery for calculation values of different optical properties of coffee samples used in the experiment

Coffee sample	C(%)	$\mu_{eff}(mm^{-1})$	$\mu_s'(mm^{-1})$	$\mu_s(mm^{-1})$	g-value
Unwashed Nekemt	6.33	$1.8\text{x}10^{-2}$	$0.42x10^{-2}$	$0.28\text{x}10^{-1}$	0.85

4.3 Particle Size Measurement Results

Determination of diameter (or particle size) of the scattering particle and particle size parameter x can be performed using the results of the 3rd phase measurements of the experiment, angular distribution of intensity measurements. Coffee stock samples used in this phase experiment were diluted to have a concentration of 5% by volume in a 40ml solution. And then a 16.7ml (concentration by volume) of sample was taken by cylindrical cuvette from these diluted 40ml solution to get a volume fraction of f_v =0.0083 And this is used to calculate the particle number density ρ_s of the scatterer in the sample solution when it is divided by the true geometrical volume of the scatterer. The particle number density ρ_s intern is used to calculate the scattering efficiency Q_s and the scattering cross-sectional area σ_s using equations (3.5.6 & and 3.5.7).

In this experiment, the intensity distribution is symmetric and centered at $0°$ and the airy pattern and airy disk is formed around this center. Using equations (3.5.6 & 3.5.7), the particle size (or diameter 'd' of the particle), particle size parameter X and other necessary parameters were calculated precisely and the results were presented in table (4.3).

Table 4.3.4: calculation summery of different parameters related to scattering of light by coffee samples used in the experiment.

Coffee sample	$\theta(°)$	d(μm)	X	A(μm^2)	$\rho_s(\mu m^{-3})$	Q_s	$\sigma_s(\mu m^2)$
Unwashed Nekemte	0.549	1.726	9.3	1.5512	4.767×10^{-2}	5.55×10^{-3}	5.4541×10^{-3}

The particle size of Caffeine extracted Unwashed Nekemte coffee is measured and calculated in similar procedure. The scattering angle obtained from the measurement is 0.69° and the result of calculation gives d=1.119 μm, and its particle size parameter x = 7.38 with geometrical area 0.9829μm^2. From these result of calculation it can be conclude that caffeine in the coffee has its own effect on the scattering of light, and also it affects the quality of coffee. In general in this experimental measurement it may be mentioned that there is an average error of $\pm 0.043\mu$m in particle size determinations. Results of particle size parameter X, tells us that our results agreed with Mie scattering theory in which it has value x> 1.42

5. CONCLUSION, RECOMMENDATION AND FUTURE PLAN

5.1 Conclusion

Particle size measurement and the determination of particle size distribution are essential to any one dealing with particulate system. Particles have different shapes and are involved in different process. For quality control or system property description purposes, the need to determine the particle size distribution of food powders becomes apparent, including processing and functionality of many food powders. Incorrect particle size and particle size distribution, particle shape, surface area, and density can lead to poor coffee quality. The method for investigation of Particle size dependency of quality of coffee and it's optical properties, (μ_t, μ_a, μ_s, μ_s' and g), have been developed. The scattering coefficient has direct relation with the particle size as well as the quality; the larger the scattering coefficient is the finest the particle size and the highest the quality. The particle size of ground coffee is an extremely important parameter in determining the final characteristics of the brewed product, thus care needs to be taken during grinding to ensure that the resulting particle size matches the required taste profile for the product being made.

5.2 Recommendation and future plan

Particle size is one of the most important parameters in materials science and technology as well as many other branches of science and technology, from medicine, pharmacology and biology to ecology, energy technology and the geosciences. The characterization of particulate system is usually made through the analysis of particle size and its distribution. Particle size measurement is most often required a simple quality control technique, a method used to investigate changes in research and design process, and or as a requirement in patent or product specification application. As the study indicates the laser light scattering method for investigation coffee quality is simple, fast and valid; more over it is highly sensitive and more suitable for on line monitoring with equipments mostly available in common laboratories. Since the developed methods have wider economic and scientific applications, even for other turbid mediums like milk chocolate, tea and etc, light scattering method is the most preferable for classification of coffee quality standards. Thus it is recommended for quality inspection centers and coffee exporters to use light scattering techniques for their quality standard classifications. Further research may improve it, if necessary, for other mediums.

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