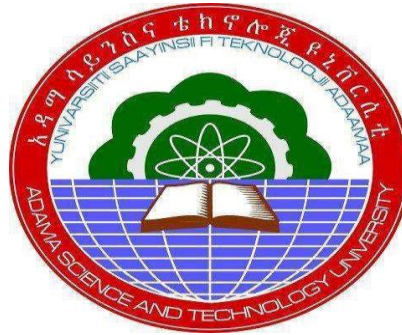


**REFRACTIVE INDEX METHOD FOR MEASUREMENT OF
ALCOHOLIC STRENGTH OF LOCAL ALCOHOLIC BEVERAGES
(AREKES) USING PRISM SPECTROMETER**

By

Sagana Bodde Heribaye



**A Thesis Submitted to
The Department of Applied Physics
School of Applied Natural Science
Presented in Partial Fulfillment of the Requirement for the
Degree of Master's in Physics
Office of Graduate Studies
Adama Science and Technology University**

**November 2017
Adama, Ethiopia**

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Declaration

I declare that this thesis is prepared for the partial fulfillment of the requirements for the degree of Masters of Science in Physics entitled, “**Refractive Index Method for Measurement of Alcoholic Strength of Local Alcoholic Beverage (*Areke*) Using Prism Spectrometer**” is my original work prepared by my own effort with the close advice and guidance of my advisor. I also declare that this thesis has not been presented in any university and all sources that I have used or quoted have been indicated and acknowledged by means of complete references.

Name: Sagana Bodde Heribaye

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This M.Sc. thesis has been submitted to for examination with my approval as University advisor.

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Place and date of submission:

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November 2017

APPROVAL SHEET
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APPLIED PHYSICS PROGRAM

This is to certify that the thesis prepared by Sagana Bodde entitled: “**Refractive Index Method for Measurement of Alcoholic Strength of Local Alcoholic Beverage (Areke) Using Prism Spectrometer**” submitted in partial fulfillment of the requirement for the Master of degree of science in physics complies with regulation of the university and meets the accepted standards with respect to originality.

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Final approval and acceptance of the research is contingent up on the submission of its final copy to the final of graduate studies (CGS) through the candidates department or school of graduate committee (DGS or SGC).

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Dedication

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List of Abbreviations

CoA:	Coenzyme
GC – FID:	Gas Chromatography with a Flame Ionization Detector
He-Ne:	Helium Neon
NADH:	Nicotinamide Adenine Dinucleotide
NIR:	Near – Infrared Reflectance
R.I:	Refractive index
%(v/v):	Percent by volume per volume
SNNPR:	Southern Nations, Nationalities, and People’s Region
USA:	United States of America
VLDLs:	Very Low Density Lipoproteins

Abstract

Several methods were developed to determine the refractive index of different solutions. In this research, the refractive index of various local beverages (Areke) and Alcoholic strength were determined using a prism spectrometer by measuring the angle of minimum deviation using laser light of different wavelengths. The refractive index of Dagim, Cereal Yegibto and Yetenaadam and Yenechshinkurt were in the range of $(1.3540 \pm 0.0001$ to $1.3635 \pm 0.0002)$. The minimum refractive index was recorded for cereal areke (1.3540 ± 0.0001) and the maximum for Dagim Areke (1.3635 ± 0.0002) using red diode laser (650nm). The alcoholic strength of areke collected from different parts of Ethiopia was analyzed by adopting the method of determination of reactive index of ethanol solution. The alcohol strength of 'areke' was determined by using regression equation using origin software. The alcoholic strength (%v/v) of the traditional alcoholic beverage (areke) was found to be 45.05 – 49.29, 30.02 – 42.03, 44.05 – 48.08 and 38.40 – 43.03 for Dagim, Cereal Yegibto and Yetenaadam and Yenechshinkurt, respectively.

CHAPTER ONE

1. Introduction

The alcohol content (strength) of alcoholic drink is an indication of the drinks' ethanol concentration as measured in percent ethanol per 100 mL. In other words, alcoholic strength is measured as volume per volume and expressed as % (v/v). Alcoholic drinks can be broadly classified into distilled and fermented liquors. Distilled liquors, typical examples of which are whisky, brandy, and shochu, are produced by distilling of fermented liquors, so they generally have higher alcohol content than the fermented liquors. On the other hand, the alcohol content of fermented liquors such as wine, beer and sake can be varied by the degree of fermentation of sugar by yeast. It also can be varied by material and maturation. Alcohol content is an extremely important numeric indicator for both manufacturers of liquors and their consumers.

Currently, there are many methods used to measure alcoholic strength (content) of alcoholic beverage including refractive index method [1], densitometry and pycnometry [1, 2], enzymatic method [3, 4] sequential injection analysis [5], beer analyzer [1, 7], liquid or gas chromatographic methods [7–11], biosensor [5, 7], potentiometry [7], oxidation of the distillate [1,2], dichromate oxidation [1,6,7] and near – infrared spectroscopy [11]. Due to complicated pretreatment procedures (e.g. sample distillation and accurate weighing process) and large sample volume required, the two most popular methods, pycnometry and densimetric analysis, are not applicable for samples with small amount. As for oxidation of the distillate and dichromate oxidation spectrophotometry, more than 5 mL of sample volume is required for analysis. Besides, the reagents used are highly toxic and classified. Low stability, low reproducibility and low accuracy are the disadvantages for enzymatic method, biosensor, and potentiometry [4]. High performance liquid chromatography and Gas chromatography, although they provide simultaneous determination of several alcohols, need long analysis time, require time consuming pretreatment process for obtaining reliable data, are expensive, demand skilled operator [10]. Recently developed NIR spectroscopy and beer analyzer are time consuming in establishing calibration curves and have low accuracy as they can be interfered by other alcohols in alcoholic beverages. This thesis is introducing inexpensive and quick refractive index method,

which used to determine alcoholic strength (content) in local alcoholic beverage are ke from calibration curve correlation between refractive index versus ethanol solution.

Refractive index is one of the most important physical properties of solutions. By measuring the refractive index of a solution, one can determine the composition of the solution [12]. The refractive index of a substance describes an important part of its interaction with electromagnetic radiation. The refractive index is a basic optical property of materials and its accurate measurement is often needed in many branches of physics and chemistry and it has several applications to many industries and materials. It is measured for many reasons. It is clearly important to know the refractive index of materials used for their clarity, such as glasses and solid plastics. In complex fluids such as drinks or foods, the refractive index is a measure of dissolved or sub micronic material [13, 14, 15].

There are numerous methods used for measuring the refractive index of a liquid solution that are well documented in text books [16], the most suitable and easiest method was reported [17, 18]. The diversity of methods is largely due to differences in applications, test objects and the required measurement accuracy. A separate class of devices for measuring the refractive index of transparent liquids consists of systems designed for examining liquids in cuvettes made of glass, quartz or other transparent materials. During the last decade, numerous practical devices based on the principle described above using various cuvettes have been constructed for determining the refractive index [19].

Previously some researchers (authors) have reported that the use of an equilateral hollow prism would allow for the measurement of the refractive index of most ordinary liquids. It opens the possibility of studying small variations in the refractive index of a solution with concentration. So measurements of refractive index are widely used in many industrial and research applications to determine the concentration of solutions. However, refractive index at the same time also varies with temperature, pressure, and wavelength [20, 21].

Previous studies [22 -24] provide more detailed discussion on the concentration mapping by the measurement of refractive index of liquids. Temperature coefficient of refractive index can also be used to calculate thermal expansion coefficient [25].

This thesis reports a relatively simple and effective technique, which can be used to measure the refractive index of ethanol solution at different concentration for different areke type at room temperature. We have utilized red and green diodes lasers and He-Ne laser with wavelength of 650 nm, 532 nm and 632.8 nm, respectively. Previously, refractive index of popular alcoholic drinks with different percentage of alcohol was determined by [26]. Abbe's Refractometer is an industrial made instrument used to determine the concentration of a solution by determining the optical refractive index of the solution. However, it is very expensive. So, in this research we used an equilateral hollow prism locally constructed from inexpensive and easily found materials to determine the refractive index of ethanol solution and the refractive index of local alcoholic beverage areke by applying Snell's law. A plot of refractive index versus ethanol concentration (percent by volume) was prepared and the alcoholic strength (content) of local alcoholic beverage areke was determined using the results of a liner regression analysis.

The thesis is organized as follows;the first chapter is the introductory chapter that includes the statement of the problem, the general and specific objectives, and the significance of the study. In Chapter two, the literatures related to ethanol, alcoholic beverage, areke and refractive index have been presented. The Ethiopian traditional alcoholic beverage, classification of areke, use of areke and effect of alcohol on man were presented as well as Snell's law, Angle of minimum deviation, Condition for minimum deviation, using a hollow prism to determine concentration of a solution, Concentration of the components of the liquids, its density and refractive index and Description of refractive index using electron oscillator model were presented.

In chapter three, the materials and methods used in this work are presented. Sampling and sample preparation and preparation of ethanol solution at different level of concentrations were presented. In the same chapter, the techniques of the measurement of ethanol solution and areke are presented.

In chapter four, results and discussion of the thesis are presented. The calculated values of refractive index of ethanol solutions at different concentrations and different local alcoholic beverage (areke) were presented. The alcoholic strength of local alcoholic beverage of different areke are presented. And the dependence of refractive index on the wavelength was presented.

Finally, the thesis windup by conclusions and recommendations.

1.1. Statement of the Problem

Several methods have been employed to measure alcoholic strength of distilled alcoholic beverage and most of these methods have been expensive and may not always be available locally. Therefore, there is a need to obtain alcoholic strength of local alcoholic beverage (*areke*) by refractive index method, using hollow prism spectrometer. Refractive index method is a simple and inexpensive method that can be used to measure alcoholic strength of local alcoholic beverage (*areke*) samples. To the best of our knowledge, no research was done on the measurement of alcoholic strength of local alcoholic beverage (*areke*) by refractive index method using hollow prism spectrometer in Ethiopia.

1.2. Objective of the Study

1.2.1. General Objective

- Determination of refractive index and alcoholic strength of local alcoholic beverage (*areke*) using prism spectrometer.

1.2.2. Specific Objective

The Specific objectives of this study are:

- To obtain regression equation using index of refraction and concentration using ethanol solution as standard solution.
- To examine wavelength effect on refractive index of ethanol solution.
- To determine the refractive index of each local alcoholic beverage (*areke*).
- To compare the refractive index of each local alcoholic beverage (*areke*).
- To compare alcoholic strength of each local alcoholic beverage (*areke*).

1.3. Significance of the Study

Determination of refractive index of different type of *areke* useful, to control quality and to determine alcohol content. Knowledge of alcoholic strength (i.e. ethanol level) in alcoholic beverages (*areke*) is an important task for economic reasons in relation to the taxes imposed on alcohol. The higher the ethanol contents in alcoholic beverages, the higher the tax. Other importance of knowing the alcoholic content of alcoholic beverage is it helps people to drink in moderation. Moderate alcohol consumption may provide some health benefits such as: reduce risk of heart disease, cancer, diabetes and trauma. And researchers, students, teachers, and academicians in the area can use the findings of the study as a reference material. The following may happen in future: enhancement of the research activities and facilities in contemporary optics and laser applications at the University of Adama. The research will provide motivation to continue advanced comprehensive research and develop new inexpensive local techniques for measuring refractive index of other alcoholic beverages.

CHAPTER TWO

2. Review of Related Literature

2.1. Ethanol

Ethanol ($\text{CH}_3\text{CH}_2\text{OH}$) has been described as one of the most exotic synthetic oxygen-containing organic chemicals because of its unique combination of properties as a solvent, a germicide, a beverage, antifreeze, a fuel, a depressant, and especially because of its versatility as a chemical intermediate for other organic chemicals. Ethanol, also known as ethyl alcohol or grain alcohol, is a clear, colorless, volatile, flammable chemical compound. In dilute aqueous solution, it has a somewhat sweet flavor, but in more concentrated solutions it has a burning taste. It is a monohydric primary alcohol that crystallizes at -114.1°C , boils at 78.5°C , and has a density of 0.789 g/mL at 20°C [27- 29]. Ethanol is slightly more refractive than water, having a refractive index of 1.36242 (at $\lambda = 589.3\text{nm}$ and 18.35°C or 65.03°F) [30]. The triple point for ethanol is 150K at a pressure of $4.3 \times 10^{-4}\text{Pa}$. It is miscible (i.e., mixes without separation) with water in all proportions [27-30]. Ethanol – water mixture have less volume than the sum of their individual components at the given fractions [30].

It is made from the fermentation or chemical breakdown of sugars by yeasts. All beverage ethanol and more than half of industrial ethanol is still made by this process. And also it is made from plants and grains such as corn, wheat, barley. During the fermentation process, the starches of the grains are turned into alcohol. The ethanol produced by fermentation ranges in concentration from a few percent up to about 14 percent. Above about 14 percent, ethanol destroys the zymase enzyme and fermentation stops. Ethanol is normally concentrated by distillation of aqueous solution, but the composition of the vapor from aqueous ethanol is 96 percent ethanol and 4 percent water. Therefore, pure ethanol cannot be obtained by distillation. Commercial ethanol contains 95 percent by volume of ethanol and 5 percent of water. Dehydrating agents can be used to remove the remaining water and produce absolute ethanol [27, 31].

Ethanol is commonly used in industrial applications (such as solvents, detergents, paints, printing inks, photo-chemical applications, latex processing, dyes, etc.), the beverage market, medicinal, pharmaceutical and food products and the sole manufacturer and supplier to the fuel market [27].

2.2. Alcoholic beverages

Alcoholic beverages are class of organic compounds, containing one or more hydroxyl group (OH), attached to a carbon atom [32]. Ethanol is the only type of alcohol that can be prepared and consumed at a commercial or household level. Alcoholic beverages can be produced by fermentation and/or distillation and distilled alcoholic spirit categories include two groups in itself: namely distilled spirits and liquors. Fermented drinks are prepared by the action of yeasts that metabolize sugars or other carbohydrates in grapes, other fruits, vegetables and grains by producing alcohol as a waste product whereas distilled spirits are produced by the distillation of fermented raw materials with the separation of alcohol from fermented liquid by increasing the alcohol content. Wine, beer, whisky, rum, vodka, brandy, gin and liquor are the most predominant types of commercially produced and consumed alcoholic beverages all around the world [31, 33, and 34].

In commercially produced alcoholic beverages, monitoring ethyl alcohol is crucial in the liquor industry beverages because it determines the quality of alcoholic beverages (wine, beer, liquor and spirit) since it is the main product of the alcoholic fermentation, and for some food preparations (packaging of “bakery products”) since it is often added as a conserving agent due to its antibacterial properties and its ability to limit staleness. Accurate alcoholic beverages analysis ensures excellent beverages quality [35]. Therefore this analysis is an essential part of the daily work in the labs in the breweries, vineyards and distilleries. Knowledge of the alcohol content is necessary to ensure that the beverage conforms to the label declaration of alcohol content and to establish the basis for the payment of tax.

Alcohol content of percentage by volume (% vol) is used to indicate the ethanol content of beverages; this is also called the French or Gay-Lussac system. The American proof system is double the percentage by volume; avodka which is 40% by volume is thus 80 proof in the USA [36]. The ethanol content in beer usually varies from 2.3% to over 10% vol, and is mostly 5–5.5% vol. In some countries, low-alcohol beer, i.e. below 2.3% vol, has obtained a considerable

share of the market. In general, beer refers to barley beer, although sorghum beer is consumed in large quantities in Africa. The ethanol content of wine usually varies from 8 to 15% vol, but light wines and even non-alcoholic wines also exist. The ethanol content of spirits is approximately 40% vol, but may be considerably higher in some national specialty spirits. Also within the spirits category are aperitifs, which contain around 20% vol of alcohol. Alcopops, flavoured alcoholic beverages or ready-to-drink beverages usually contain 4–7% vol of alcohol, and are often pre-mixed beverages that contain vodka or rum [37].

In addition to commercialized products, in many developing countries different types of home-made or locally produced alcoholic beverages such as sorghum beer, palm wine or sugarcane spirits are available and consumed [31]. Home – made or locally produced alcoholic beverages are produced through fermentation of seed, grains, fruit, vegetables or parts of palm trees, by a fairly simple production process. This kind of production seems especially common in many African countries, where a wide variety of different beverages can be found.

2.2.1. Ethiopian Traditional Alcoholic Beverages

Traditional alcoholic beverages, *tella*, *tej*, *keribo*, *birz*, *borde*, *korefe* and *areke* are indigenous to Ethiopia [38]. *Areke* is distilled whereas the rest are fermented alcoholic beverages [39–41]. *Tella* and *korefe* (a malt beverage like beer) are made from a mixture of *enkuro* (a dark brown toasted flour of barely, maize or sorghum), germinated wheat grain (*bikil*), *gesho* (*Rhamnus prenoide*s) and water [41]. *Tej* and *birz* (honey beer) are prepared from honey and water. But, *tej* has leaves or stem of *gesho* (*Rhamnus prenoide*s) as an additional ingredient [39, 41]. *Borde* and *keribo* are made from a mixture of roasted grain (barely, wheat, maize or sorghum) and their malts [39, 42].

2.2.1.1. Local Alcoholic Beverage (Areke)

Areke is a distilled beverage. It is a colorless (sometimes colorful), clear, traditional alcoholic beverage which is distilled from fermentation products prepared in almost the same way as *tella* except that the fermentation mass in this case is more concentrated (has high level of ethanol) [43,44]. It can have alcohol content in the range of 30 to almost 50 percent [45]. *Areke* is usually brewed in rural and semi-urban areas and is used more commonly by farmers and semi-urban dwellers than by people who live in the cities. In cities, those who drink *areke* are predominantly

lower class people or those who have become dependent on alcohol and cannot afford to buy industrially produced alcohol [31].

2.2.1.2. Classification of Areke

Traditionally *areke* is classified into two: *Terra-areke* and *Dagim-areke*. The term *dagim* in Amharic refers to ‘second time’ and, indicates that it is distilled second time, whereas the term *terra* in Amharic refers to ‘ordinary’ or single distilled type [41].

2.2.1.2.1. Terra –Areke

Terra-areke is a colorless (sometimes colorful), clear, local alcoholic beverage, which is single distilled type from a fermentation product known as *Yereke-tinsis* [44, 45]. *Terra areke* includes Yekosso (*Hagenia abyssinica*), Cereal, Yegibto (*Lupinus albu* or *white lupin*), Yemar (honey), Yetenaadam and Yenechskuit (*Ruta chalepensis* and *Allium sativum* or *garlic*). All those areke are made from powdered *Gesho* leaves (*Rhamnus prenois*) and different cereals such as corn, wheat, *teff* (*Eragrostis tef*), and *Dagussa* (*Elusine coracann*). All of them except cereal – based areke have additional ingredient as their names.

2.2.1.2.2. Dagim-Areke

Dagim-areke (*double distilled*) is a stronger type of *terra-areke*, which is prepared in the same way as *terra-areke*, except that the distillation process is allowed to proceed for a shorter period of time, or three volumes of *terra-areke* are redistilled to give about one volume of *dagim-areke* [45]. The redistilled *areke* will then have higher alcohol content.

2.2.1.3. Use of Areke

Traditional alcoholic beverage plays important roles in social function such as marriage, get together and any other ceremonies. They have also special place in traditional such as burials and settling disputes. Ethiopian traditional beverage consumed on wedding, holydays and other festive occasions in traditional environment [46]. Also traditionally, the Ethiopian alcoholic beverages like *areke* have been used as medicine [47]. The local community in North – western Ethiopia used ‘*gibto areke*’ as a locally made antihypertensive medicinal preparation [48]. *Kosso* areke provides medicinal properties, which used as an anthelmintic to combat tapeworm or control internal parasites [49].

2.3. Effects of Alcoholic Beverage on Man

The main active ingredient of local beverage *areke* is alcohol and therefore, the health effects of alcohol apply to *areke*. The moderate consumption of alcohol associated with a decreased risk of cardiac disease, stroke and cognitive decline [50]. The long-term effects of alcohol abuse, however, include the risk of developing alcoholism and alcoholic liver disease. High ethanol consumption over the years leads to liver damage. For healthy man, the limit is about 60g per day and for women 50g. However, these values are strongly dependent on body weight, health status and other factors. Ethanol related high levels of NADH+H⁺ and acetyl-coA in the liver leads to increase syntheses of neutral fats and cholesterol. However, since the exports of these in the form of VLDLs is reduced due to alcohol, storage of lipids occur (fatty liver). This increase in the fat content of the liver (from less than 5% to more than 50% of the dry weight) is initially reversible. However in chronic alcoholism, the hepatocytes are increasingly replaced by connective tissue. When liver cirrhosis occurs, the damage to the liver finally reaches an irreversible stage, characterized by progressive loss of liver functions [51].

2.4. Description of Refractive Index Using Snell's Law

Snell's law (also known as Descartes' law or the law of refraction), a formula used to describe the relationship between the angles of incidence and refraction, for light or electromagnetic waves, passing through a boundary between two different transparent medias, such as air and glass or others [52]. Snell's law states that the ratio of the Sine of the angles of incidence to the Sine of the angle of refraction is a constant that depends on the properties of the media [53]. When a light ray incident on an interface point of the two medias with different refractive indices at an angle of incident θ_i , which is the angle between the ray of incident and the normal line which is perpendicular to the surface, it becomes refract either towards or away from the normal depending on the optical density of the two medias at an angle of refraction θ_r , which is the angle formed between ray of refracted and the normal line as shown in Figure 2.1.

Suppose a light ray is incident on an interface separating two transparent media. The incident ray is then partially reflected to the first medium and partially transmitted (reflected) to the second medium. The refracted ray makes an angle θ_r , with the normal ray and the incident ray makes an

angle θ_i with the normal. If n_1 is the refractive index of the first medium and n_2 is that of the second medium, then Snell's law states that:

$\frac{\sin \theta_i}{\sin \theta_r} = \frac{n_2}{n_1}$ Moreover, if v_1 is speed of light in the first, medium and v_2 is speed in the second

medium, the Snell's law is expressed as: $\frac{\sin \theta_i}{\sin \theta_r} = \frac{v_1}{v_2}$.

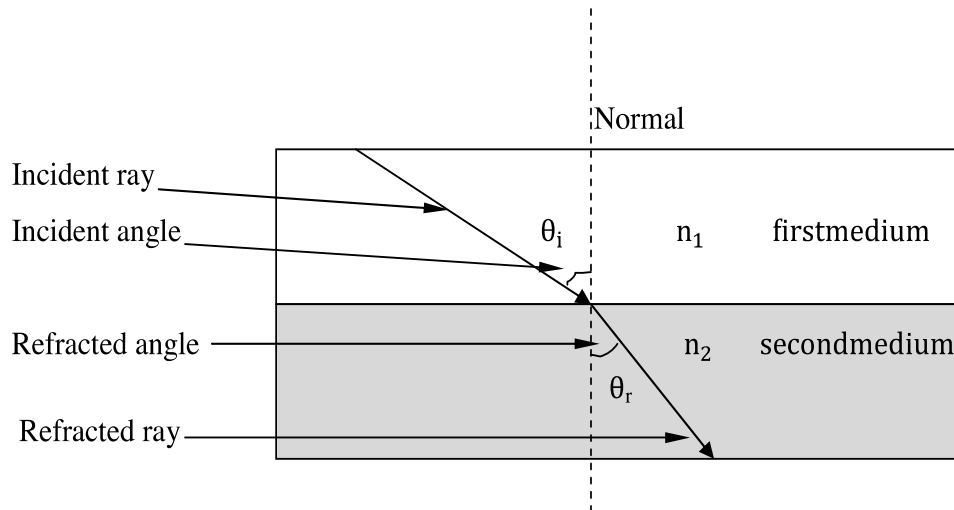


Figure: 2.1 Graphic of Snell's law

Mathematically Snell's law also can be rewired as:

$$n_1 \sin \theta_i = n_2 \sin \theta_r \quad (2.1)$$

$$\frac{\sin \theta_i}{\sin \theta_r} = \frac{n_2}{n_1} = \text{constant } t(n) \quad (2.2)$$

Where θ_i and θ_r are angle of incidence and angle of refraction respectively. n_1 and n_2 are the refractive indices of the first and second medium respectively and $n = \frac{n_2}{n_1}$ is the relative refractive index.

As light passes the border between the two medias, depending upon the relative refractive indices of the two medias, the light will either be refracted to a lesser angle or a greater one. These angles are measured with respect to the normal line, line perpendicular to the boundary at the point of incidence.

Snell's law used to determine the direction of light rays through refractive media with varying indices of refraction. The indices of refraction of the media, labeled n_1 and n_2 , are used to represent the factor by which light is "slowed down" within a refractive medium, such as glass or water, compared to its velocity in a vacuum.

2.4.1. Angle of Minimum Deviation

Angle of deviation is the angle through which an incident ray deviates. By measuring the angle of deviation from various angles of incident, we can determine the angle of minimum deviation and refractive index of the prism.

Figure 2.2 shows the set up to determine the angle of minimum deviation using a prism.

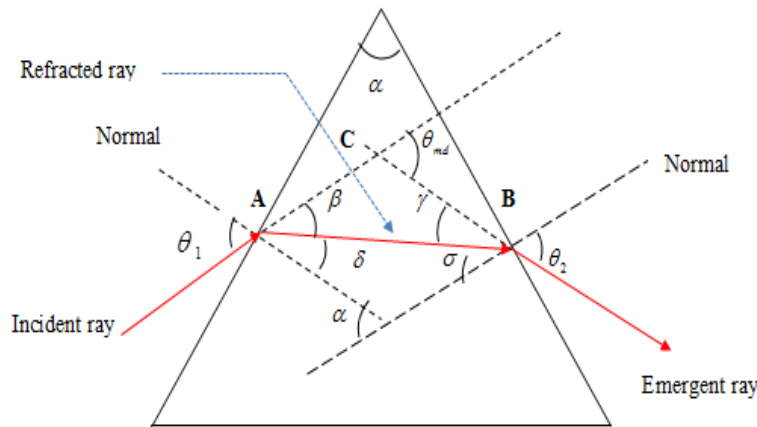


Figure: 2.2 Geometry of a light rays passing through a prism

Where: α is the apex angle; θ_1 and δ are the angles of incidence and angle of refraction on surface A, respectively. σ and θ_2 are the angle of incidence and angle of refraction on surface B, respectively. θ_{md} is the angle of minimum deviation. If light ray is incident on the first surface of the prism at an angle of θ_1 , then the refraction at both surfaces obeys Snell's law [54]. Then due to the refraction at surface A, we can write Snell's law as:

$$\frac{\sin\theta_1}{\sin\delta} = \frac{n_g}{n_a} \quad (2.3)$$

Where: n_g and n_a are refractive indices of the glass and air respectively. Application of Snell's

law at point B yields:

$$\frac{\sin \delta}{\sin \theta_2} = \frac{n_a}{n_g} \quad (2.4)$$

$$\text{or } \frac{\sin \theta_2}{\sin \delta} = \frac{n_g}{n_a} \quad (2.5)$$

Equates equation (2.3) and (2.5) yields:

$$\text{or } \frac{\sin \theta_1}{\sin \delta} = \frac{n_g}{n_a} = \frac{\sin \theta_2}{\sin \delta} \quad (2.6)$$

The angle of deviation produced by the first surface is:

$$\beta_1 = \theta_1 - \delta \quad (2.7)$$

And the angle of deviation produced by the second surface is:

$$\gamma_1 = \theta_2 - \delta \quad (2.8)$$

The total angle of minimum deviation is thus:

$$\theta = \beta + \gamma \quad (2.9)$$

Now substitute equation 2.7 and 2.8 into 2.9 and it yields:

$$\theta_{md} = \theta_1 + \theta_2 - (\delta + \sigma) \quad (2.10)$$

But, $\delta + \sigma = \alpha$

Therefore, equation (2.10) becomes: -

$$\theta_{md} = \theta_1 + \theta_2 - \alpha \quad (2.11)$$

2.4.2. Condition for Minimum Deviation

The angle of minimum deviation occurs at a particular angle of incidence where the refracted ray inside the prism makes equal angles with the two prism faces. This occurs when the path of the light inside the prism is parallel to the base of the prism. Therefore, it means that in Figure 2.2

$$\theta_1 = \theta_2, \quad \gamma = \beta, \quad \delta = \sigma$$

On account of equation (2.11) we can thus write the minimum angle of deviation as:

$$\theta_{md} = \theta_1 + \theta_2 - \alpha \quad \text{Or} \quad \theta_{md} = 2\theta_1 - \alpha \quad (2.12)$$

This implies that:

$$\theta_1 = \frac{1}{2}(\theta_{md} + \alpha) \quad \text{Or} \quad \theta_2 = \frac{1}{2}(\theta_{md} + \alpha) \quad (2.13)$$

And from equation (2.10) it follows that:

$$\delta = \frac{1}{2}\alpha \quad \text{Or} \quad \sigma = \frac{1}{2}\alpha \quad (2.14)$$

2.4.3. Determining Refractive Index of a Solution Using a Hollow Prism Spectrometer

A *spectrometer* is a spectroscope with a meter or detector so it can measure the *amount* of light (number of photons) at specific wavelengths. Light coming from a source is usually dispersed into its various constituent wavelengths by a dispersive element (prism or grating) and then the resulting spectrum is studied. As shown in Figure 2.3, a spectrometer consists of a collimator, a telescope, a circular prism table and a graduated circular vernier scale. The collimator holds an aperture at one end that limits the light coming from the source to a narrow rectangular slit. A lens at the other end focuses the image of the slit onto the face of the prism. The telescope magnifies the light dispersed by the prism (the dispersive element for your experiments) and focuses it onto the eyepiece. The angle between the collimator and telescope are read off by the circular scale [55, 56].

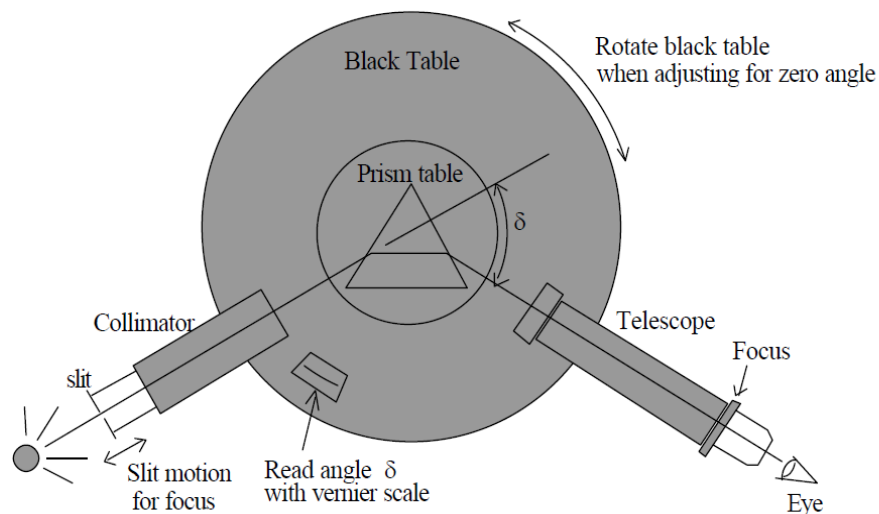


Figure: 2.3. Prism spectrometer

The hollow-prism spectrometer is suitable for measuring ray deviations and refractive indices. Sometimes a diffraction grating is used in place of the prism for studying optical spectra. A

prism refracts the light into a single spectrum, whereas the diffraction grating divides the available light into several spectra. Because of this, slit images formed using a prism are generally brighter than those formed using a grating. Spectral lines that are too dim to be seen with a grating can often be seen using a prism. Unfortunately, the increased brightness of the spectral lines is offset by a decreased resolution, since the prism doesn't separate the different lines as effectively as the grating. However, the brighter lines allow a narrow slit width to be used, which partially compensates for the reduced resolution [56].

With a prism, the angle of refraction is not directly proportional to the wave length of the light. Therefore, to measure wavelengths using a prism, a calibration graph of the angle of deviation versus wavelength must be constructed using a light source with a known spectrum. The wavelength of unknown spectral lines can then be interpolated from the graph. Once a calibration graph is created for the prism, future wavelength determinations are valid only if they are made with the prism aligned precisely as it was when the graph was produced. To ensure that this alignment can be reproduced, all measurements are made with the prism aligned so that the light is refracted at the angle of minimum deviation.

The theory of the prism spectrometer indicates that a spectrum of maximum definition is obtained when the angular deviation of a light ray passing through the prism is a minimum. Under such conditions it can be shown that the ray passes through the prism symmetrically. For a given wavelength of light traversing a given prism, there is a characteristic angle of incidence for which the angle of deviation is a minimum. This angle depends only on the index of refraction of the prism and the angle between the two sides of the prism traversed by the light. The mathematical relationship between these variables is derived as follows.

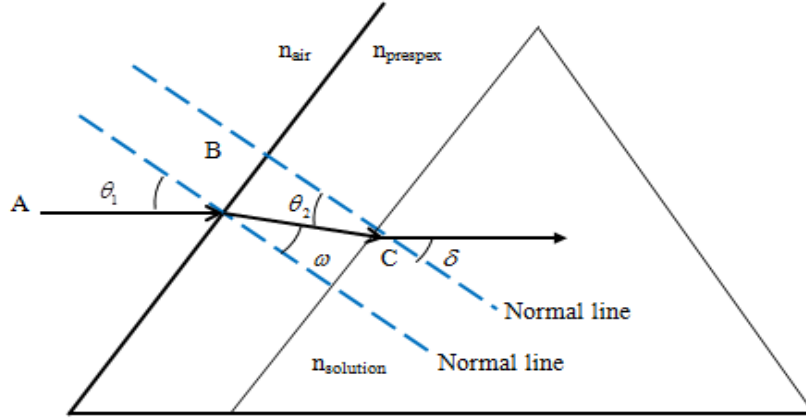


Figure: 2.4 Geometry of a light ray passing through a hollow prism

From A to B, light is travelling in air. It is refracted at B and travels in the Perspex up to C. At C it is refracted again and we assume it travels parallel to the base of the prism to achieve minimum deviation.

By applying Snell's law at B:

$$n_{air} \sin \theta_1 = n_{per} \sin \omega \quad (2.15)$$

Where: n_{air} and n_{per} are the refractive indices of air and Perspex respectively. The only unknown parameter in equation (2.15) is the angle of refraction, ω .

It can be obtained by rearranging Eq. (2.15)

$$\sin \omega = \frac{n_{air}}{n_{per}} \sin \theta_1 \quad (2.16)$$

Again by applying Snell's law at C,

$$n_{per} \sin \theta_2 = n_{sol} \sin \delta \quad (2.17)$$

Where, n_{sol} - refractive index of solution. Because $\theta_2 = \omega$, we see that

$$n_{per} \sin \omega = n_{sol} \sin \delta$$

$$\text{Or } \sin \omega = \frac{n_{sol}}{n_{per}} \sin \delta \quad (2.18)$$

Now, equates equation (2.16) and (2.18), we note that

$$\frac{n_{air}}{n_{per}} \sin \theta_1 = \frac{n_{sol}}{n_{per}} \sin \delta$$

$$n_{air} \sin \theta_1 = n_{sol} \sin \delta \quad (2.19)$$

Now substitution of equation (2.14) into (2.19) leads to:

$$n_{air} \sin \frac{1}{2}(\alpha + \theta_{md}) = n_{sol} \sin \left(\frac{1}{2} \alpha \right) \quad (2.20)$$

Since the refractive index of the air is 1, the refractive index of the solution can be written as:

$$n_{sol} = \frac{\sin \left(\frac{1}{2}(\alpha + \theta_{md}) \right)}{\sin \left(\frac{1}{2} \alpha \right)} \quad (2.21)$$

For this research the angle of apex $\alpha = 60^\circ$. Therefore, equation (2.21) becomes:

$$n_{sol} = \frac{\sin \left(\frac{1}{2}(\alpha + \theta_{md}) \right)}{\sin 30^\circ} \quad (2.22)$$

It then follows that:

$$n_{sol} = 2 \sin \left(\frac{1}{2} \theta_{md} + 30^\circ \right) \quad (2.23)$$

Where: θ_{md} is determined experimentally using a prism spectrometer.

2.5. Concentration of the Components of the Liquids, its Density and Refractive Index

Refractive index is an important characteristic of liquid substances for analyzing, monitoring, and identification of liquids in several industries now days. Accurate determination of the concentration of the solution by measuring the refractive index is widely used in chemical and physical studies, food technology, materials processing, medical diagnostics, monitoring systems, fuel cells directly measuring pollution, and many other fields.

In general, the measured refractive index of a multicomponent mixture is a function of its temperature T , concentration C , and the wavelength of the incident light λ [57]:

$$n = n(T, C, \lambda) \quad (2.24)$$

From eqn (2.24), the change of refractive index (Δn) of a multicomponent mixture is:

$$\Delta n \approx \frac{\partial n}{\partial T} \Delta T + \frac{\partial n}{\partial C} \Delta C + \frac{\partial n}{\partial \lambda} \Delta \lambda \quad (2.25)$$

For small changes of the temperature ΔT , concentration ΔC , and wavelength $\Delta \lambda$. Since laser light is highly monochromatic and wavelength-stable, $\Delta \lambda \approx 0$ and wave length effects can be neglected.

In this work, the concentration is defined on a mass basis for liquids A and B:

$$C_A = \frac{M_A}{M_A + M_B} \text{ and } C_B = \frac{M_B}{M_A + M_B} \quad (2.26)$$

Where, M – mass (kg)

Thus, by measuring the change in refractive index and the liquid temperature, the concentration change can be expressed using Eq. (2.25) as follows:

$$\Delta C \approx \left(\Delta n - \frac{\partial n}{\partial T} \Delta T \right) \left(\frac{\partial n}{\partial C} \right)^{-1} \quad (2.27)$$

Changes in the density of matter are always accompanied by some change of refractive index. Usually refractive index increases with increasing density. Theoretical study of the relationship between the density of matter and its refractive index, as well as the experimental data shows that there is a direct proportion between some refractive index function $f(n)$ and density d :

$$f(n) = r d \quad (2.28)$$

The constant factor r that is specific for this substance is called as specific refraction. As distinct from n and d the specific refraction does not depend on external conditions (temperature, pressure) and very slightly depends on the state of aggregation of matter. There are many attempts to derive a general formula for the function of all substances $f(n)$, but a universal function that is independent of the properties of matter does not exist.

The most widely used theoretical method for predicting the refractive index n_L of binary mixtures of liquids is the Lorentz–Lorenz relation [58]:

$$\frac{n_L^2 - 1}{n_L^2 + 2} = \Phi_1 \frac{n_{D1}^2 - 1}{n_{D1}^2 + 2} + \Phi_2 \frac{n_{D2}^2 - 1}{n_{D2}^2 + 2} \quad (2.29)$$

where n_L is the refractive index of the mixture according to the Lorentz–Lorenz formula; Φ_1 and Φ_2 are the volume fractions of alcohol and water, calculated from volume determinations based on the mass and density measurements, n_{D1} and n_{D2} are the refractive indices of the alcohol and water, respectively.

Empirical equations have also been developed to determine refractive indices of binary systems of liquids, including that proposed by Gladstone-Dale [59]:

$$\frac{n_G - 1}{\rho} = \omega_1 \left(\frac{n_{D1} - 1}{\rho_1} \right) + \omega_2 \left(\frac{n_{D2} - 1}{\rho_2} \right) \quad (2.30)$$

Where n_G is the refractive index of the mixture according to the Gladstone–Dale formula; ρ is the experimental density of the mixture; ω_1 and ω_2 are the mass fractions of alcohol and water; n_{D1} and n_{D2} are the refractive indices of the alcohol and water, respectively.

2.6. Classical Description of Refractive Index Using Electron Oscillator Model

From classical electron oscillator model the Lorentz force is given by [60]:

$$\vec{F} = e \left(\vec{E} + \vec{v} \times \vec{B} \right) \quad (2.31)$$

Where: \mathbf{F} is Lorentz force, \mathbf{v} is velocity, e is electron charge, \mathbf{E} is electric field and \mathbf{B} is magnetic field.

We assume that Eq. (2.31) applies to the individual protons and electrons in atoms. Although these particles and their interactions can be properly treated only using quantum theory, their interaction with light can be treated very accurately in most cases with classical laws and concepts.

If the electron has mass m_e and charge e and nucleus has mass m_n and charge $+e$. The nucleus exerts a binding force \mathbf{F}_{en} on the electron, depending on the relative separation $\mathbf{r}_{en} = \mathbf{r}_e - \mathbf{r}_n$. The electron also exerts a force \mathbf{F}_{ne} on the nucleus, and according to Newton's third law,

$$\vec{F}_{en}(\mathbf{r}_{en}) = -\vec{F}_{ne}(\mathbf{r}_{ne}) \quad (2.32)$$

The Newton equations of motion for the electron is therefore

$$m \frac{d^2 x}{dt^2} = eE(x, t) + F_{en}(x) \quad (2.33)$$

Assume x is the electron coordinate relative to the nucleus, $x = r_{en}$ and $m =$ mass of electron. In the absence of external force, the atom has a certain equilibrium position. Under the influence of an electromagnetic field, the electron experiences the Lorentz force Eq. (2.31) and is displaced from its equilibrium position; according to Lorentz “the displacement will immediately give rise to a new force by which the particle is pulled back towards its original position, and which we

may therefore appropriately distinguish by the name of elastic force” [61]. Lorentz’s assertion is equivalent to the replacement $\mathbf{F}_{en}(x)$ by $-k_s x$, where: k_s is the “spring constant” associated with the hypothetical elastic force. This leads to the equation:

$$\left. \begin{aligned} m \frac{d^2 x}{dt^2} &= eE(R,t) - K_s x \\ \left(\frac{d^2 x}{dt^2} + \omega_0^2 \right) x &= \frac{eE(R,t)}{m} \end{aligned} \right\} \quad (2.35)$$

Where: $\omega_0 = \sqrt{\frac{k_s}{m}}$ is the electron’s natural frequency of oscillator, R -for the position of the stationary nucleus.

If we take a frictional force for granted and explore its consequences, we simply amend the Newton force law Eq. (2.35) to be written as:

$$m \frac{d^2 x}{dt^2} = eE(R,t) - k_s x + F_{fric} \quad (2.36)$$

This frictional (drag) force can be given as:

$$F_{fric} = -bv = -b \frac{dx}{dt} \quad (2.37)$$

For an electron oscillator in a linearly polarized monochromatic plane wave Eq. (2.36) takes the form [60]:

$$\frac{d^2 x}{dt^2} + 2\beta \frac{dx}{dt} + \omega_0^2 x = \hat{e} \frac{e}{m} E_0 \text{Cos}(\omega t - kz) \quad (2.38)$$

Where: $\beta = \frac{b}{2m}$ the unit vector \hat{e} defines the polarization of the applied field. And then Eq. (2.36) for the electron oscillator with frictional damping is most easily solved by first writing it in complex form as:

$$\frac{d^2 x}{dt^2} + 2\beta \frac{dx}{dt} + \omega_0^2 x = \hat{e} \frac{e}{m} E_0 e^{-i(\omega t - kz)} \quad (2.39)$$

Where: we follow the convention of writing: $E_0 \text{Cos}(\omega t - kz)$ as $E_0 e^{-i(\omega t - kz)}$

$E_0 \text{Cos}(\omega t - kz)$ - is real part of $E_0 e^{-i(\omega t - kz)}$

This means that $x(t)$ in Eq. (2.39) is also regarded mathematically as a complex quantity in our calculations, but only its real part is physically meaningful. In other words, we may submit the process of taking the real part of (2.39) until after our calculations, at which point the real part of our solution for $x(t)$ is the (real) electron displacement. This approach is used frequently in solving linear equations such as Eq. (2.38). We solve Eq. (2.39) by writing;

$$X(t) = a e^{-i(\omega t - kz)} \quad (2.40)$$

and after inserting this into Eq. (2.39) we obtain

$$\left(-\omega^2 - 2i\beta\omega + \omega_0^2\right) a = \hat{e} \frac{e}{m} E_0 \quad (2.41)$$

Therefore, the assumed solution Eq. (2.40) satisfy Eq. (2.39)

$$a = \frac{\hat{e} \frac{e}{m} E_0}{\left(-\omega^2 - 2i\beta\omega + \omega_0^2\right)} \quad (2.42)$$

And the physically relevant solution is expressed as:

$$X(t) = \text{Re} \left[\frac{\hat{e} \left(\frac{e}{m}\right) E_0}{\left(\omega_0^2 - \omega^2 - 2i\beta\omega\right)} e^{-i(\omega t - kz)} \right] \quad (2.42)$$

Note that Eq. (2.42) actually gives only the steady-state solution of Eq. (2.38).

From equation (2.42) for classical oscillator, the electron displacement due to an applied electric field $\hat{e}E_0(\text{Cos}(\omega t - kz))$: is given as [61]:

$$X(t) = \hat{e} \frac{e}{m} E_0 \left[\frac{\omega_0^2 - \omega^2}{(\omega_0^2 + \omega^2)^2} e^{-i(\omega t - kz)} \text{Cos}(\omega t - kz) + \frac{2\beta\omega}{(\omega_0^2 + \omega^2)^2 + 4\beta^2\omega^2} \text{Sin}(\omega t - kz) \right] \quad (2.43)$$

The first term in brackets is in phase with the electric field, whereas the second term is “in quadrature,” that is, its phase differs is by $\frac{\pi}{2}$ from that of the field. The in-quadrature part of the induced electric dipole moment $d = e x$ is responsible for absorption (or stimulated emission) of light [60]. If the medium consists of N particles per unit volume, each of which has a dipole moment p, the polarization density is Np. We assume that the medium has no polarization in the

absence of a field, so that the only dipole moments \mathbf{p} in the medium are induced by the field, that is, the electric field causes charge displacements that result in an induced dipole moment given by the formula:

$$P(r, \omega) = \alpha(\omega) \varepsilon(r \cdot \omega) \quad (2.44)$$

Where the polarizability $\alpha(\omega)$ is defined by writing the in-phase component of \mathbf{d} as $\alpha \hat{e} E_0 (\cos(\omega t - kz))$ Thus, from Eq. (2.43),

$$\alpha(\omega) = \frac{e^2}{m} \left[\frac{\omega_0^2 - \omega^2}{((\omega_0^2 - \omega^2)^2 + 4\beta^2 \omega^2)} \right] \quad (2.45)$$

$$\text{and } n^2(\omega) - 1 = \frac{Ne^2}{m \epsilon_0} \left[\frac{\omega_0^2 - \omega^2}{((\omega_0^2 - \omega^2)^2 + 4\beta^2 \omega^2)} \right] \quad (2.46)$$

As in the case of spontaneous emission and absorption, this result of the classical oscillator model must be modified to include the oscillator strength f :

$$n^2(\omega) - 1 = \frac{Ne^2 f}{m \epsilon_0} \left[\frac{\omega_0^2 - \omega^2}{((\omega_0^2 - \omega^2)^2 + 4\beta^2 \omega^2)} \right] \quad (2.47)$$

Unlike absorption, the refractive index is usually attributable to non-resonant transitions, that is, transitions such that, $|\omega_0^2 - \omega^2| \gg \beta\omega$. In this case:

$$n^2(\omega) - 1 = \frac{Ne^2 f}{m \epsilon_0 (\omega_0^2 - \omega^2)} \quad (2.48)$$

In this no resonant situation, however, no one transition is necessarily dominate, and so we must add the contributions of all transitions connected to the ground state in which the atoms are presumed (for now) to reside. Thus, if the transitions from the ground state have oscillator strengths f_i and transition frequencies ω_j , the refractive index at the radiation frequency ω is given by the formula:

$$n^2(\omega) - 1 = \frac{Ne^2}{m \epsilon_0} \sum \frac{f_i}{\omega_j^2 - \omega^2} \quad (2.50)$$

This result applies when there is one type of atom or molecule in the medium; more generally we simply add the contributions of the different species. In a gas, furthermore, the density N is

generally sufficiently low that $n(\omega) \approx 1$ and therefore $n^2 - 1 = (n - 1)(n + 1) \approx 2(n - 1)$. Thus, for a gas consisting of a single type of atom or molecule with number density N , the formula for the refractive index is approximately:

$$n(\omega) = 1 + \frac{Ne^2}{m \epsilon_0} \sum \frac{f_i}{\omega_j^2 - \omega^2} \quad (2.51)$$

It is interesting to relate this result to a formula that is often used in tabulations of the refractive index of gases. For this purpose, we first rewrite Eq. (2.51) in terms of radiation wavelength ($\lambda = \frac{2\pi c}{\omega}$) and transition wavelengths ($\lambda_j = \frac{2\pi c}{\omega_j}$):

$$n(\omega) = 1 + \frac{Ne^2}{8\pi \epsilon_0 mc^2} \sum \frac{f_i \lambda^2}{1 - \frac{\lambda_j^2}{\lambda^2}} \quad (2.52)$$

As noted, electronic resonance in molecules (and in many atoms) tends to lie in the ultraviolet, in which case $\lambda_j \ll \lambda$ for optical wavelength λ . In this case we can approximate $\left(1 - \frac{\lambda_j^2}{\lambda^2}\right)^{-1}$

by the first two terms of its binomial series expansion, $1 - \frac{\lambda_j^2}{\lambda^2}$ then:

$$n(\lambda) - 1 \approx A_1 \left(1 + \frac{B_1}{\lambda^2}\right) \quad (2.53)$$

$$\text{Where } A_1 = \frac{Ne^2}{8\pi \epsilon_0 mc^2} \sum f_i \lambda_j^2 \quad (2.54)$$

$$B_1 = \sum \lambda_j^4 \quad (2.55)$$

An empirical relation of the form Eq. (2.53) was proposed by Cauchy in 1830, before the electromagnetic theory of light. Our derivation of Cauchy's formula gives explicit expressions for the coefficients A_1 and B_1 . Unfortunately, it is difficult to calculate the numerical values of A_1 and B_1 for a given atom or molecule because we require the transition wavelengths and the oscillator strengths of all transitions connected to the ground state, including transitions to "continuum" states in which the electrons are unbound, that is, in which an atom is ionized. For a gas at STP [$P = 760$ Torr, $T = 273$ K, and, $N = 2.69 \times 10^{25} / \text{m}^3$],

$$A_1 = 1.2 \times 10^{10} \sum f_i \lambda_j^2 \quad (2.56)$$

Cauchy's formula correctly accounts for the fact that most transparent materials we encounter daily (e.g., water, air, glass) have refractive indices greater than unity at visible wavelengths. According to our analysis, this is a consequence of these materials having resonance wavelengths λ_j that are small compared to optical wavelengths (which lie roughly between 400 and 700 nm).

It also follows from (2.48) that $\frac{dn}{d\lambda} < 0$, which is also a familiar feature of refractive indices in the visible: A glass prism, for instance, causes violet to be dispersed more than red when it separates white light into its spectral components. In fact, the increase of $n(\lambda)$ with decreasing λ ($\frac{dn}{d\lambda} < 0$) is sufficiently ubiquitous that it is called "normal dispersion." An example of normal dispersion appears in Fig. 2.5 [60].

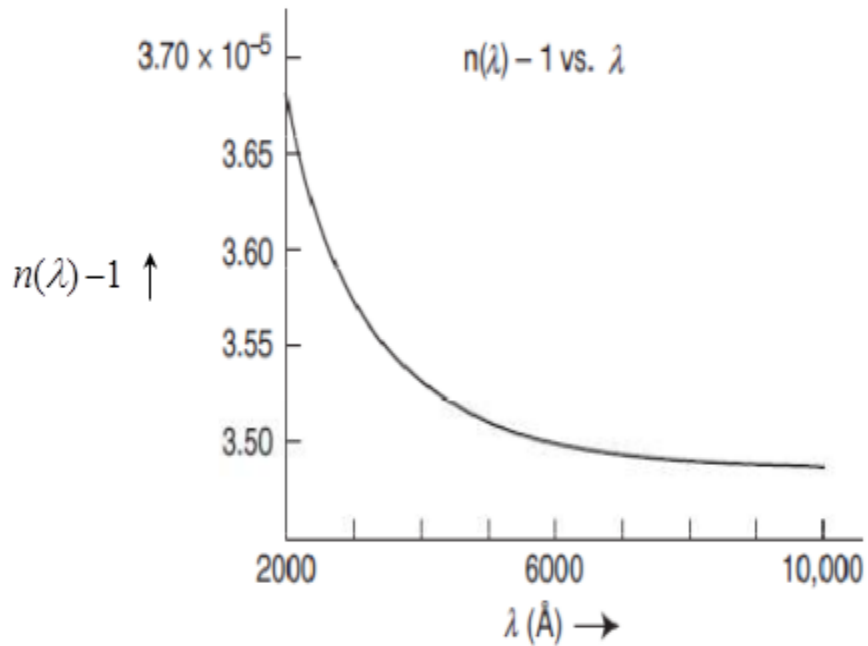


Figure: 2.5 Refractive index of helium at standard temperature and pressure [60].

It is also interesting to consider the case in which the frequency of the radiation is much greater than the resonance frequencies of the medium. The simplest example occurs for free electrons, for which there is no binding force. The resonance frequencies ω_j are then zero, and the dispersion formula (2.50) reduces to [see Eq. (2.59)].

$$N(\omega) = \left(1 - \frac{Ne^2}{\epsilon_0 m \omega^2}\right)^{\frac{1}{2}} = \left(1 - \frac{\omega_p^2}{\omega^2}\right)^{\frac{1}{2}} \quad (2.56)$$

Where N is now the density of free electrons

$$\omega_p = \left(\frac{Ne^2}{\epsilon_0 m}\right)^{\frac{1}{2}} \quad (2.57)$$

ω_p Is called the plasma frequency

If $\omega < \omega_p$ the refractive index (2.56) is a pure imaginary number. In this case the free-electron gas will not support a propagating electromagnetic wave, and an incident wave is instead reflected.

Consider the limit of Eq. (2.50) in which the field frequency ω is very large compared to any of the transition frequencies ω_j :

$$n^2(\omega) - 1 = \frac{Ne^2}{\epsilon_0 m \omega^2} \sum_j f_j \quad (2.58)$$

In this limit the transition frequencies ω_j are effectively zero, that is, the atom behaves as though its energy levels form a continuum, as is the case for unbound electrons. It is then plausible that in this limit the refractive index should be identical to that of N free electrons per unit volume, which is given by Eq. (2.50). This implies that the oscillator strengths f_j must obey the electric dipole sum rule:

$$\sum_j f_j = 1 \quad (2.59)$$

Which in fact may be derived using quantum mechanics? Our less rigorous “derivation” of this sum rule is based on the assumption that each of the N atoms per unit volume has one bound electron. In the case of Z electrons per atom Eq. (2.58) should reduce to $n^2(\omega) - 1$ for the case

$$\sum_j f_j = Z \quad (2.60)$$

Of Z free electrons per unit volume

We have assumed in our discussion of the refractive index that the N atoms per unit volume are all in the ground state with high probability, but it is straightforward to deal with the more general situation where there are N_i atoms per unit volume in energy level E_i . Equation (2.51), for instance, generalizes to:

$$n(\omega) = 1 + \frac{e^2}{2m\epsilon_0} \sum_i \sum_j \frac{N_i f_{ij}}{\omega_{ji}^2 - \omega^2} \quad (2.61)$$

Where $\omega_{ji} = (E_j - E_i)/\hbar$ and f_{ij} is the oscillator strength for the $i \rightarrow j$ transition. In particular, the contribution to the index from the $1 \rightarrow 2$ transition is:

$$n(\omega) = 1 + \frac{e^2}{2m\epsilon_0} \left(\frac{N_1 f_{12}}{\omega_{21}^2 - \omega^2} + \frac{N_2 f_{21}}{\omega_{21}^2 - \omega^2} \right) \quad (2.62)$$

$$n(\omega) = 1 + \frac{e^2}{2m\epsilon_0} \frac{N_1 f_{12}}{\omega_{21}^2 - \omega^2} \left(N_1 - \frac{g_1}{g_2} N_2 \right) \quad (2.63)$$

For any level of an atom. In other words, the sum over oscillator strengths in the electric dipole sum rule must, in the case of excited states, include both the oscillator strengths for absorption (positive) and for emission (negative). The sum rule for oscillator strengths played an important role in the formulation of quantum theory in the 1920s [62]. It was already known, based on the physical argument we have used in going from (2.58) to (2.59), before some of the most important features of quantum mechanics (e.g., before the Schrödinger equation).

The atoms or molecules of a medium do not form a continuum but have empty space between them. As a result, there is a difference between the “mean” field and the actual field acting on a

given atom. In many cases the only practical consequence of this difference is that the relation between the refractive index and the polarizability α becomes:

$$\frac{n^2(\omega) - 1}{n^2(\omega) + 2} = \frac{N\alpha(\omega)}{3\epsilon_0} \quad (2.64)$$

The origin of this ‘‘Lorentz – Lorenz relation’’ is discussed in many textbooks on electromagnetism. Note that when the refractive index is close to unity, so that $n^2(\omega) + 2 \approx 3$, the Lorentz – Lorenz relation reduces to the relation between n and α assumed in writing (2.61).

$$\frac{n^2(\omega) - 1}{n^2(\omega) + 2} = \frac{N\alpha(\omega)}{3\epsilon_0} \quad (2.65)$$

And this procedure has been taken as to the famous formula known as sellmeier equation which summarized below.

2.6.1. Sellmeier Equation

The derivation made in the previous topic is often represented in different way, called the sellmeier equation. This equation gives the refractive index as a function of frequency, but does not take the Lorentz local field in to account. Even if it’s lacking physical accuracy, it has found widespread use as a way to express the refractive index of optical materials. To derive it we start by combining:

$$P = \epsilon_0 \chi E \quad \text{and} \quad \epsilon = \frac{\epsilon_0 E + P}{\epsilon_0 E} = 1 + \chi$$

Giving an expression for the permittivity,

$$\epsilon = 1 + \frac{P}{\epsilon_0 E} \quad (2.66)$$

If we then combine the result with that of equation; $P = N_p = -N_{er} = \frac{Ne^2 E}{\epsilon_0 m(\omega_0^2 - \omega^2 - i\omega\gamma)}$ we get

$$\epsilon - 1 = \frac{Ne^2}{\epsilon_0 m(\omega_0^2 - \omega^2 - i\omega\gamma)} \quad (2.67)$$

Which also can be written in terms of the refractive index as

$$n^2 - 1 = \frac{Ne^2}{\varepsilon_0 m (\omega_0^2 - \omega^2 - i\omega\gamma)} \quad (2.68)$$

If we assume that the operating frequency ω is far away from the resonance frequency, the effect of the damping γ can be neglected and γ equals zero. By converting the angular frequency to wavelength by the relation $\omega = 2\pi c / \lambda$ we get

$$n^2 - 1 = \frac{\frac{N}{m\varepsilon_0} \lambda_0^2 \lambda^2}{(2\pi c)^2 (\lambda^2 - \lambda_0^2)} = \frac{A\lambda^2}{\lambda^2 - B} \quad (2.69)$$

Where, $A = \frac{Ne^2 \lambda_0^2}{(2\pi c)^2 m\varepsilon_0}$ is the resonance strength and $B = \lambda_0^2$ is the resonance wavelength. For

solid materials, A and B must be found empirically as they are depending on the Lorentz local field as well as the external field. Up until now it has been assumed that the electric field acting on the molecule in question only affects the vibration of the electrons. Depending on the substance, other modes of vibration may also be excited by an incident light beam. This means that equation 2.69 must be expanded to include all relevant vibrational modes [63, 64]

$$n = \sqrt{1 + \frac{A_i \lambda^2}{\lambda^2 - B_i}} \quad (2.70)$$

This is the Sellmeier Equation with Sellmeier constants A_i and B_i . The sum is often reduced to either two or three terms in the visible frequency range, as this proves to be sufficient to describe the refractive index accurately. The Sellmeier equation can describe the refractive index of all kinds of substances, as long as they have clearly defined resonance peaks.

The refractive index of a material has been shown to be dependent on the wavelength of the light and this dependence is referred to as dispersion. The Sellmeier equation from 2.70 is used to describe this relationship.

CHAPTER THREE

3. Materials and Methods

In this chapter, the materials and methods employed in the thesis are presented. The first section of this chapter describes the various materials used for the experiment; the second section describes sampling and sample preparation, the third section methods of the research; and in the last section the experimental setup was presented.

3.1. Materials

Prism spectrometer, red and green diode laser and He-Ne laser as a light source. In addition to these, different instruments such as syringes and measuring cylinder to measure the volume of ethanol solution (97% ethanol) and distilled water, the Mercury Thermometer to measure the temperature of the room, plastic bottles to keep different concentration of ethanol and magnetic stirrer were used.

3.2. Sampling and Sample Preparation

For this study traditional local alcoholic beverage (*areke*) samples were collected from Addis Ababa and other potential producing areas such as Arsi Negele (Ormiya Regional State), Butagira (SNNPR Regional State) and Dembecha (Amhara Regional State). The collected *areke* samples include *cereal (only cereal-based)*, *Dagim (double distilled)*, *Yetenaadam and Yenechshinkurt (Ruta chalepensis and Allium sativum or garlic)*, and *Yegibto (Lupinus albu or White lupin)*. For each type *areke*, 200-mL samples were collected in screw-capped plastic bottles (bottles of highland springs) from the three vending houses randomly from particular site. All the *areke* samples except cereal based have additional ingredient as their names indicate. The samples are kept at room temperature after collection and until the time of analysis. Finally 50 mL of *areke* (alcoholic beverage) was taken from each collected sample for experimental work.

3.3.Methods

3.3.1. Methods of Measuring Refractive Index of Ethanol Solution and Local Alcoholic Beverage (Areke) by Snell's Law

3.3.1.1. Preparation of Standard Solution of Ethanol Solution

For preparing calibration curves, solution of ethanol was prepared by volume – volume ratio at 0,10,20,30,40,50 and 60 % v/v in distilled water. (eg. To prepare 10 % of ethanol solution, from 97% ethanol from total of 50ml, 5.2 ml of ethanol was mixed with 44.8 ml of distilled water).



Figure: 3.1 Preparation of ethanol solution by different concentrations

3.3.1.2. Method of Measuring of Angle of Minimum Deviation

In order to measure the angle of minimum deviation, the set up were arranged as shown in Figure 3.2. The sample prepared for each concentration of ethanol solution was poured into the sample holder (Hollow prism) which is placed on the prism spectrometer. And for each ethanol solution, the angles of minimum deviation have been measured three times using red (650 nm) and green (532 nm) diodes and He-Ne (632.8 nm) laser lights and the averages were calculated. The refractive index of each solution was calculated from angle of minimum deviation using equation (2.23). For local alcoholic beverages (*areke*), we use the same procedures to determine angle of minimum deviation and refractive index using red (650 nm) diode.

3.3.2. Determination of Alcoholic Strength in Local Alcoholic Beverage (Areke) from the Calibration Curves

Linear curve fit was applied to find linear regression equations for the standard solutions of ethanol solution. The alcoholic strength of local alcoholic beverage (*areke*) was deduced from linearly fitted calibration curve of refractive index versus ethanol concentration graph. Alcoholic strength of local alcoholic beverage (*areke*) was generally expressed as % v/v, or percentage volume per volume.

3.4. Experimental Set up

Experimental set up for determination of angle of minimum deviation for ethanol solution and local alcoholic beverage (*areke*).

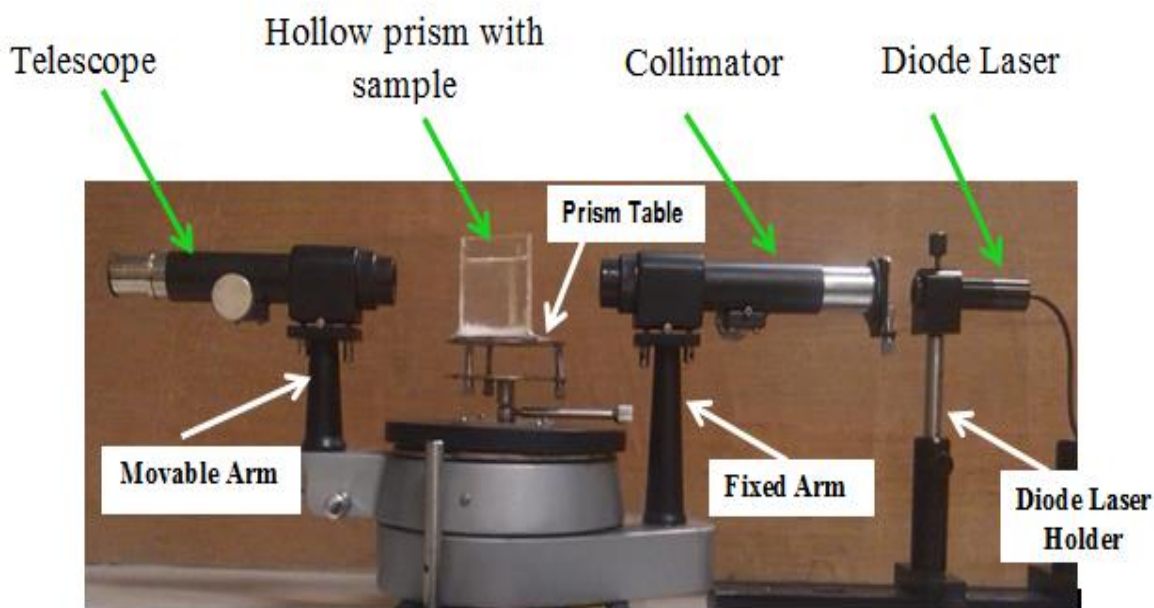


Figure 3.2: Experimental setup for determination of angle of minimum deviation

CHAPTER FOUR

4. Results and Discussion

In this chapter, the results and discussion of the thesis were presented. In the first sections of this chapter the results of refractive indices of ethanol solutions determined by the three lasers were presented; graph of refractive index versus ethanol concentration and graph of refractive index versus wavelength were presented. In the last section alcoholic strength of local alcoholic beverages (*areke*) (*Dagim*, *Cereal*, *Yegibto* and *Yetenaadem* and *Necishunkurt*) were presented.

4.1. Refractive Index of Ethanol Solution Using Red and Green Diode Lasers and He-Ne Laser

4.1.1. Concentration Dependent of Refractive Index of Ethanol Solutions

Table 4.1-4.3 show the refractive indices of ethanol solution measured at different concentrations using red diode laser ($\lambda = 650$ nm), green diode laser ($\lambda = 532$ nm) and He-Ne laser ($\lambda = 632.8$ nm). The results in the tables indicate the refractive index of the ethanol solutions in the mentioned concentration range (10%-60%) were found to be 1.3370 ± 0.0002 - 1.3605 ± 0.0003 , 1.3374 ± 0.0003 - 1.3617 ± 0.0002 and 1.3407 ± 0.0002 - 1.3645 ± 0.0002 using red diode, He-Ne lasers and green diode laser light respectively. The refractive index increases as the concentration of ethanol solution increases. In the mentioned concentration range, the minimum value of refractive index was 1.3370 ± 0.0002 obtained using red diode laser for 10% concentration and the maximum value was 1.3645 ± 0.0002 obtained using green diode laser for 60% concentration. The obtained results are in a good agreement with the reported results by [65, 66].

Table 4.1. Results of refractive index of ethanol concentration using red diode laser ($\lambda = 650nm$)

Ethanol concentration % (v/v)	Angle of minimum deviation (θ_{md})	Refractive index (n)
0	23.46±0.004	1.3313±0.0001
10	23.90±0.005	1.3370±0.0002
20	24.28±0.001	1.3420±0.0003
30	24.67±0.002	1.3470±0.0004
40	25.06±0.01	1.3520±0.0001
50	25.49±0.003	1.3575±0.0002
60	25.72±0.015	1.3605±0.0003

Table 4.2. Results of refractive index of ethanol concentration using He-Ne laser ($\lambda = 632.8nm$)

Ethanol concentration % (v/v)	Angle of minimum deviation (θ_{md})	Refractive index (n)
0	23.49±0.002	1.3317±0.0001
10	23.93±0.002	1.3374±0.0003
20	24.31±0.003	1.3424±0.0001
30	24.70±0.002	1.3474±0.0002
40	25.09±0.003	1.3524±0.0001
50	25.64±0.001	1.3595±0.0003
60	25.82±0.002	1.3617±0.0002

Table 4.3. Results of refractive index of ethanol concentration using green diode laser ($\lambda = 532nm$)

Ethanol concentration % (v/v)	Angle of minimum deviation (θ_{md})	Refractive index (n)
0	23.74±0.002	1.3350±0.0003
10	24.18±0.001	1.3407±0.0002
20	24.57±0.005	1.3457±0.0001
30	24.96±0.003	1.3507±0.0002
40	24.35±0.005	1.3557±0.0004
50	25.86±0.002	1.3623±0.0001
60	26.03±0.003	1.3645±0.0002

Figure 4.1 shows the plot of refractive index versus concentration of ethanol solution measured using red diode laser, green diode laser and He-Ne laser. As shown in the figure the experimental values are best linearly fitted ($R^2 = 0.99563, 0.99247, 0.99273$) for red diode, He-Ne laser and green diode laser respectively. The regression equations of the graph obtained using different

laser light are shown in Table 4.4. Using these regression equations we have calculated the alcoholic strength in local alcoholic beverage (*Areke*). Table 4.5 shows the refractive index calculated by our techniques using red diode, green diode lasers and He-Ne laser light. The obtained results are similar with the previous reported results using sodium lamp (589.8 nm) [65, 66].

The refractive index shows a non-linear dependence with the ethanol concentration in the samples, as described in the literature [67]. However; experiments were performed to assess the linearity range of refractive index values determined for ethanol–water mixtures at room temperature. A standard curve for ethanol–water mixtures was prepared. The results are presented in Figure 4.1. We found that, all three laser refractive index-concentration curves have similar shapes in that they increase linearly with increasing concentration up to about 40%, but for an ethanol content of >40% (v/v), a departure from linearity exists, and accordingly a standard curve could be drawn only between 0 and 40% (v/v). The correlation equation for ethanol–water mixtures between 0 and 40% (v/v) was $y = (4.95E-4)C + 1.3391$, with a best correlation coefficient $R = 0.99563$.

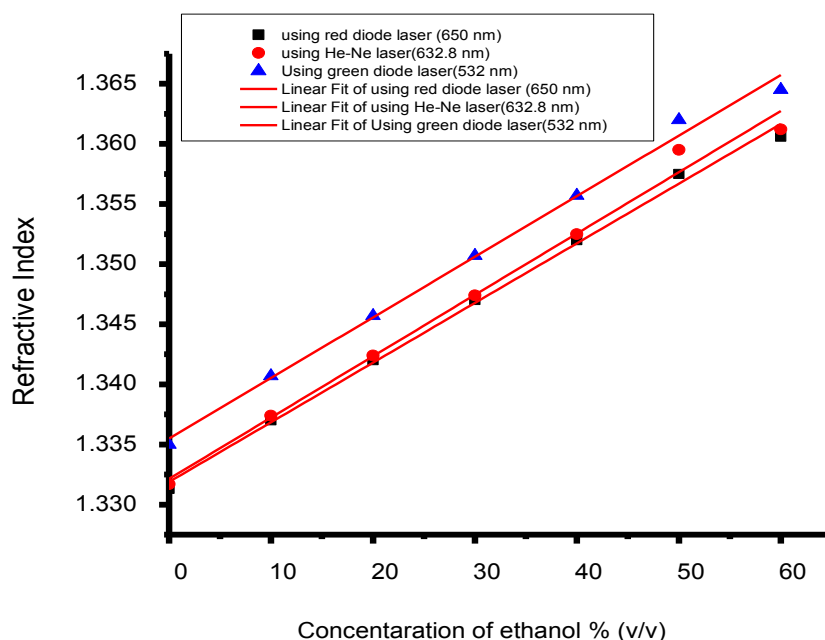


Figure: 4.1 Graph of refractive index versus ethanol concentration using red, green diode and He-Ne Laser

Table: 4.4 Results of slopes, intercept and regression equation for refractive index versus ethanol concentration for the three lasers (red diode, He-Ne and green diode) laser

Source of light	Slopes	Intercept	R ²	Regression equation
Red diode laser	4.95E-4	1.33191	0.99563	$n = (4.95E - 4)C + 1.33191$
He-Ne laser	5.15E-4	1.33205	0.99247	$n = (5.15E - 4)C + 1.33205$
Green diode laser	5.06E-4	1.33547	0.99273	$n = (5.06E - 4)C + 1.33547$

Where c is concentration of ethanol solution.

Table: 4.5 values of refractive indices experimentally determined and the literatures values

Ethanol concentration % (v/v)	Experimental values Using red diode ($\lambda = 650\text{nm}$)	Experimental values Using He-Ne laser ($\lambda = 632.8\text{nm}$)	Experimental values Using green diode ($\lambda = 532\text{nm}$)	Literatures value ($\lambda = 589.29\text{nm}$) [63,64]
0	1.3313	1.3317	1.3350	1.3329
10	1.3370	1.3374	1.3407	1.3370
20	1.3420	1.3424	1.3457	1.3420
30	1.3470	1.3474	1.3507	1.3480
40	1.3520	1.3524	1.3557	1.3530
50	1.3575	1.3595	1.3623	1.3560
60	1.3605	1.3617	1.3645	1.3590

4.2. Determination of Alcoholic Strength in Local Alcoholic Beverage (Areke)

Determination of the concentration of the components of a mixture can often be done utilizing indirect methods. This experiment was performed for the purpose of determining the alcohol content /alcoholic strength / (percentage by volume) of local alcoholic beverage by measuring its refractive index. Table 4.6 and 4.7 shows the alcoholic strength in local *areke* calculated using the regression equations shown in table 4.4 for the standard concentration of ethanol solutions. As shown in table 4.7, the determined values of ethanol contents in local alcoholic beverage

(*areke*) using red diode laser are found in the range (30.02 -49.29) %v/v which is comparable with the reported values, by other literatures; that is in the range of (30.1-56.0) % v/v [68].

The results obtained indicated that the amount of alcoholic ranges from 30.02 to 49.29 % (v/v). The alcoholic strength of *Yetenaadam and Yenechshinkurt* (38.4 – 43.03), *Yegibto* (44.05 – 48.08), *Dagim* (45.05 – 49.29), and *Cereal Arekes* (30.02 – 42.03) were with the mean values of 40.74, 46.06, 47.58, and 37.21 % (v/v), respectively. *Dagim Arekes* have an alcoholic content higher than the other *Arekes* since they are *double distilled*. For cereal and *Dagim Arekes*, the mean values obtained are 37.21 % and 47.58 % (v/v) whereas the reported values are 37.2 and 48.0 % (v/v), respectively [46]. On the other hand, when the alcoholic content of *Areke* is compared with other distilled beverages, it is in the range of liqueurs' alcoholic content, 20 – 60 % (v/v) [69].

It is important to consider the many sources of error that were in the experimental procedure. The first to note is that refractive index is a function of temperature and temperature recordings were not made during this procedure. Thus it is the best to obtain refractive index of ethanol solution at same temperature. However, prism spectrometer that used in this study was not equipped with a temperature regulating system, therefore, to avoid evaporation of ethanol during refractive index measurement, the ethanol solution and local alcoholic beverage *areke* were measured at room temperature but room temperature varies, this would affect refractive index to some extent so, this show some small fluctuations that would contribute to indeterminate error. There was also indeterminate error present due to the use of volumetric pipets. In particular, the use of the graduated volumetric pipet in the preparation of the known solutions was a challenging experimental technique.

There is also a flaw in the basic premise of this experiment. That is, local alcoholic beverage *arekes* with a large number of “impurities” were compared to mixtures of pure ethanol and water. Surely the other substances present in local alcoholic beverage *arekes* would affect its refractive index to some extent and therefore also affect the percent by volume determination.

Table: 4.6. Results of refractive index of local alcoholic beverage *areke* (*Dagim, Cereal, Yegibto and Yetenaadam and Yenechshinkurt*) using red diode laser

Types of Areke	Sample Site	Sample No.	Average θ_{md}	Refractive index (n)	Result of alcoholic strength
Dagim	Addis Ababa	1	25.89 ± 0.0003	1.3626± 0.0002	47.50
		2	25.80 ± 0.006	1.3614± 0.0003	45.05
		3	25.92± 0.002	1.3629± 0.0004	48.08
	Dembecha	1	25.96 ± 0.005	1.3635± 0.0002	49.29
		2	25.93± 0.0001	1.3631± 0.0007	48.50
		3	25.88± 0.0001	1.3624± 0.0003	47.07
Cereal	Addis Ababa	1	25.37± 0.0002	1.3559± 0.0006	34.06
		2	25.52± 0.0003	1.3579± 0.0004	38.07
		3	25.48± 0.0003	1.3574± 0.0002	37.04
	Demecha	1	25.56± 0.0004	1.3584± 0.0008	39.07
		2	25.60± 0.0001	1.3589± 0.0008	40.03
		3	25.44± 0.0003	1.3569± 0.0006	36.08
	Arsi Negele	1	25.60± 0.0004	1.3589± 0.0002	40.05
		2	25.52± 0.0002	1.3579± 0.0002	38.03
		3	25.68± 0.0002	1.3599± 0.0006	42.03
	Butagera	1	25.22± 0.0006	1.3540± 0.0001	30.02
		2	25.48± 0.0004	1.3574± 0.0007	37.06
		3	25.41± 0.0002	1.3564± 0.0003	35.02
Yegibto	Addis Ababa	1	25.80± 0.0007	1.3614± 0.0004	45.05
		2	25.84± 0.0001	1.3619± 0.0008	46.06
		3	25.88± 0.0005	1.3624± 0.0007	47.07
	Dembecha	1	25.84± 0.0001	1.3619± 0.0004	46.06
		2	25.92± 0.0007	1.3629± 0.0006	48.08
		3	25.76± 0.0004	1.3609± 0.0003	44.05
<i>Yetenaadam and Yenechshinkurt</i>	Arsi Negele	1	25.54± 0.0001	1.3581± 0.0006	38.40
		2	25.68± 0.0003	1.3599± 0.0008	42.02
		3	25.59± 0.0006	1.3587± 0.0003	39.60
	Dembecha	1	25.62± 0.0004	1.3591± 0.0004	40.40
		2	25.64± 0.0006	1.3594± 0.0001	41.01
		3	25.72± 0.001	1.3604± 0.0003	43.03

Table: 4.7 The result of alcoholic strength of local alcoholic beverage (Areke) experimentally determined and the literatures values

Types of Areke	Alcoholic Strength % (v/v) in this thesis	Literature Value of alcoholic Strength % (v/v) [65]
Dagim Areke	45.05 – 49.29	51.1-56.0
Cereal	30.02 – 42.03	30.1-48.2
Yegibto	44.05 – 48.08	42.8-50.3
Yetenaadam and Yenechshinkurt	38.40 – 43.03	43.0

4.3.Wavelength Dependence of the Refractive Index of Ethanol Solution

Figure 4.2 shows the refractive index of ethanol solution versus wavelength graph for different lasers. The graph indicates the refractive index of ethanol solution decreases as the wavelength of the laser light increases. The results are in a good agreement with the theoretical description expressed in Eq. (2.53). In Eq. (2.53), the refractive index decreases exponentially as the wavelength of laser light increases as shown in Figure 2.5. From this graph the values of Cauchy’s coefficients A and B were determined using non-linear curve fitting. The value of A and B are 0.030305 ± 0.00106 , 32236 ± 1341.4 respectively with $R^2 = 0.96369$. A and B are the parameters that depend on the nature of the medium in which the light is propagating.

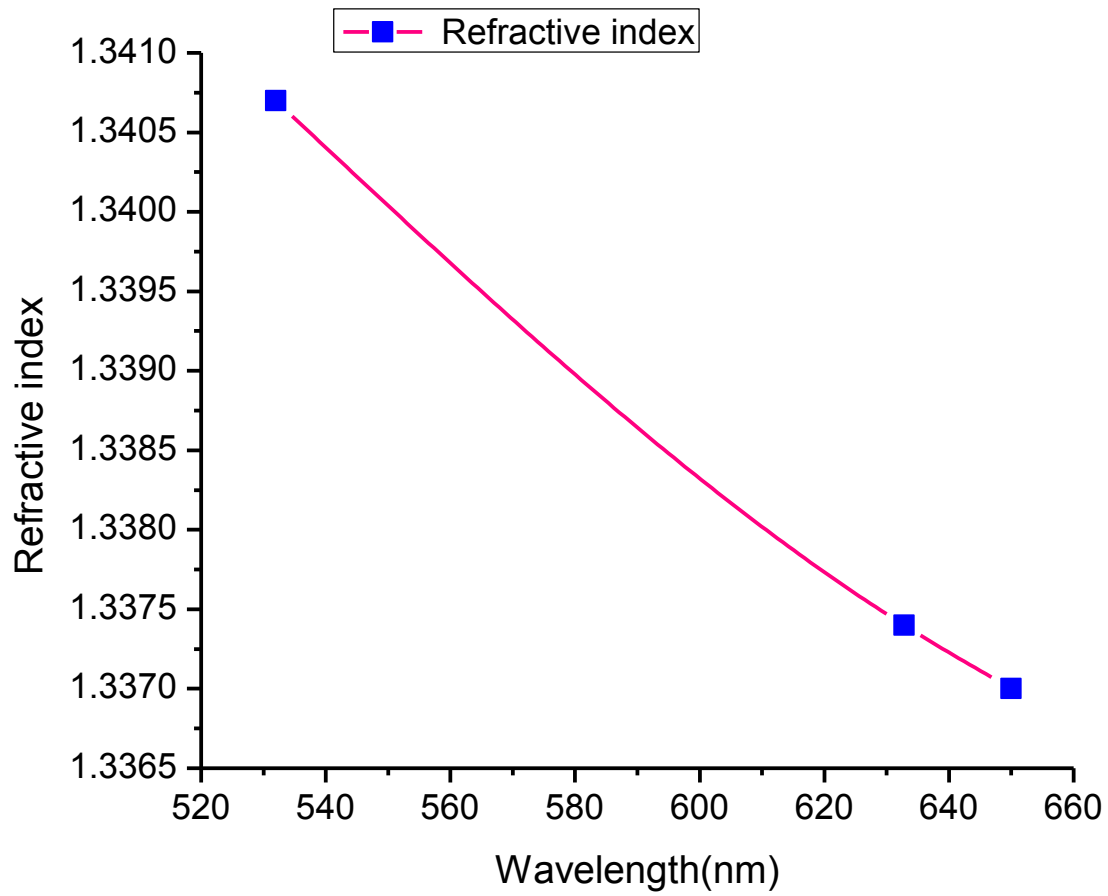


Figure 4.2 Graph of refractive index versus wavelength (532nm, 632.8nm & 650nm)

5. Conclusions and Recommendations

5.1. Conclusions

A simple method for measuring the alcoholic strength of local alcoholic beverage (*areke*) has been presented. The refractive index allow the indirect determination of alcoholic strength in local alcoholic beverage (*areke*). We have successfully determined the refractive index of ethanol solution using Red diode laser (650nm), Green diode laser (532nm) and He-Ne laser (632.8nm) and the refractive of *Dagim, Cereal, Yegibto and Yetenaadam and yenechshunkurt Areke* by using Red diode laser (650nm) by the minimum angle of deviation method using a hollow prism shaped cell. The experimental results showed that this method could be safely employed to study the dependence of refractive index of local *areke* on their alcoholic strength. The red diode laser light was proven to be powerful and accurate to determine refractive index of local *areke*. A correlation study was undertaken to find possible relationships between the refractive index and the alcoholic strength . Values derived from measurements (Table 4.4) with the reference ethanol solution were subjected to regression analysis. The regression equation found for measurement of refractive index values at room temperature by using transformation tables (4.4) was $n = (4.95E-4) C + 1.3391$; $R^2 = 0.99563$. Compared with previously reported procedures involving infrared measurements, the recommended techniques provide good analytical qualities, which are comparable with those offered by other published methods. In addition, the techniques are in good agreement with the results obtained from GC-FID measurements, and they are rapid, low cost, and reliable for the determination of alcoholic strength in locally produced *areke*. Refractive indices of all samples of *areke* for given composition of ethanol-water mixture are higher than corresponding ethanol-water binary mixture. Local alcoholic beverage arekes with a large number of “impurities” were compared to mixtures of pure ethanol and water. Surely the other substances present in local alcoholic beverage *arekes* would affect its refractive index to some extent and therefore also affect the percent by volume determination and also the alcoholic content variability in local alcoholic beverages could be attributed to the spontaneous fermentation and distillation differences that the producers build.

5.2.Recommendations

The developed methods can also be applied for other types of solutions. On top of this, in a future work, the hollow prism made by expert or industrially made and prism spectrometer that equipped with a temperature regulating system would need to provide better predictive results. In Ethiopia the government has no control over the production and quality of traditional beverages. There is, however, wide spreading and serious alcohol- related problems. Thus, control in the production and supervision with the development of comprehensive national alcohol policy is recommended.

It is not difficult to imagine that large number of population in the most parts of the country consumes the local beverages. As the process of fermentation is spontaneous, high amount and variability of toxic substances is inevitable. Hence from the point of view of public health, investigation on the mechanism of production and means to avoid predominance of harmful contents are necessary.

Further researches need to be conducted toward the development of standardized ways of production. Estimation of the amount of the harmful components of the traditional beverages, in relation to human health, is required. It is also important to deal with these harmful components so as to assess whether they are found in the beverages above or below their maximum allowable concentrations.

Developed methods have wider economic and a scientific application, even for other mediums, RI method is the most preferable for determination of alcoholic strength in alcoholic beverages. Further research may improve it, if necessary, for other mediums. Thus it is recommended for quality inspection centers and alcoholic beverage exporters to use RI techniques for their quality control.

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