# *PART-I*

## *THERMOPHYSICAL PROPERTIES OF SOME MEDICINAL PLANT LEAVES AND FRUITS*

# CHAPTER-1

## INTORDUCTION

### 1.1. DETAILS OF THE MEDICINAL PLANTS SELECTED

For the present study, four medicinal plant leaves and two fruits sample have been selected for thermo-physical properties, as they are often used in the treatment of various ailments in the traditional as well as modern system of medicine.

**A: Nephafu** (*Clerodendron colebrookianum* Walp, Verbenaceae)**:** This tree (figure1.1) is distributed widely in south and south-East Asia[1]. In India, it mainly grows in the North-Eastern region up to an altitude of 1700 $m^2$. In India traditional medicine, the leaves of C. colebrookianum (Locally known as "Nephafu") are mostly used in the treatment of hypertension[2-3] The roots are used as an anthelmintic[4], antibacterial[5] and to cure bronchitis, asthama, fever, stomach troubles, syphilis and gonorrhoea[6]. In China it is used to induce diuresis[7]. The leaves and roots of *Clerodendron colebrookianum* contains many chemical compounds such as flavones and

their glycosides[8-9] which may have significant antioxidant activity. Excessive production of reactive oxygen species plays an important role in the pathogenesis and progression of various diseases involving different organs[10-11]. Lipid peroxides, produced from unsaturated fatty acids via free radicals, cause toxic effects and promote the formation of additional free radicals in a chain reaction.[12-13].

Since a very long time the tribal people of the North- East region of India have used the leaves of the plant of *Clerodendron colebrookianum* to keep away many degenerative diseases[14]. The scientific basis of such a beneficial effect of plant leaves is

**Figure 1.1:** Nephafu (*Clerodendron colebrookianum*)

**Figure 1.2**: Mahaneem (*Azadirachta indica*)

not clear. It is possible that the compounds present in *Clerodendron colebrookianum* might contribute to its overall protective effect.

**B: Mahaneem** (*Azadirachta indica*, Meliaceae):The tree(figure1.2) *Azadirachta indica*[15-16] a large evergreen tree, 40-50 feet in height, common through out the greater part of India, Burma and some parts of south Asia. The plant has been imported to have wide spectrum of bio-activities. It is frequently planted as homestead or avenue tree as it is believed to purify air. Almost every part of this tree is used for medicinal purpose in India. It is commonly known as margosa or the Indian lilac and less commonly as neem, khomba, verpu, mamba or suder[17]as originally located in southern Asia and is now widespread over tropical and subtropical Africa, America and Australia. There are an estimated 14 million trees in India alone[18] .For centuries, Neem has been held in high esteem by Indian folk because of its medicinal and insecticidal properties. Neem preparations have been used since ancient times in unani medicine for the treatment of a wide range of human ailments due to their anti-inflammatory, concoctive, fever, thirst, nausea, vomiting, blood purifying, antiarthritic, anti pyretic, anti microbial and anti helmintic actions. The leaves of neem are used to protect grain and cloths from insects[19]nd the seed oil used as an insecticide[20]nd medicine of treatment of leprosy, skin disease and maleria[21]

The bark exudes a clean bright amber coloured gum, which is collected in small tears or fragments. It is considerably esteemed medicinally as stimulant. As an external application to ulcers and skin diseases, neem leaves are used in variety of forms such as poultices, ointments and liniments. The fruits are described as purgative and emollient and are useful in the treatment of intestinal worms, urinary diseases, piles etc. the dried flowers are also used as tonic after fever, under the name of 'Panchaaurita' a medicine is prepared which contains the flowers, fruits, leaves bark and root of the tree[15].

**C: Tulsi** (*Ocimum sanctum*, Lamiaceae)

*Ocimum sanctum*[14,22] is a tropical much branched annual herb, up to 18 inches tall and grows into a low bush in figure 1.3. The tulsi or holy basil, considered sacred by the Hindus, has small leaves with a strong smell and purple flowers. There are two verities: a red- and a green one. The variety with green leaves is called locally, Shri tulsi and one with reddish leaves is called Krishna tulsi. The natural habitat of tulsi varies from sea level to an altitude of 2000m. Its leaves are nearly round and up to 5cm long with the margin being entire or toothed. These are aromatic because of the presence of a kind of scented oil in them. Tulsi flowers are small having purple to reddish colour present in small compact clusters on cylindrical spikes. The fruits are small and the seeds yellow to reddish in color. Red Holy basil has a stronger smell. This plant, originally from India, grows abundantly in Suriname.

Apart from the religious significance it also has substantial medicinal meaning and is used in Ayurvedic treatment. Tulsi has antimicrobial properties since it is also anti-inflammatory due to the Eugenal oil present in the leaves. It is useful in respiratory tract infection. The ursolic acid present in tulsi has anti allergic properties. The plant can play a role in the management of immunological disorders such as allergies and asthma. The juice of the leaves is used against fever and as an antidote for snake and scorpion bites. It's antispasmodic properties, relives abdominal pains and helps in lowering the blood sugar level [22] Tulsi leaves contain a bright yellow volatile oil, which is useful against insects and bacteria. The principal constituents of this oil are Eugenol, eugenol methyl ether and carvacrol. The oil is reported to possess anti-bacterial properties and acts as an insecticide. It inhibits the in vitro growth of Mycobacterium tuberculosis and micrococcus pyogenes voroureus. It has marked insecticidal activity against mosquitoes. The juice of leaves and or a concoction, called jushanda a kind of tea gives relief in

common cold, fever bronchitis, cough, digestive complaints etc. when applied locally. It helps in eradicating ringworms and other skins diseases. Tulsi oil is also used as eardrops in case of pain. The seeds are used in curing urinary problems. Aphrodisiac virtue has been attributed to it, where powered tulsi root with clarified butter (ghee) is prescribed.

**D: Nayantora** (*Vinca rosea*, Linn, Apocynaceae): It is know as Nayatora, Sadabahar, sadaphul etc. (figure 1.4) in various Indian languages. In botanical nomenclature it is known as *Vinca rosea*[23-24] Linn belonging to Apocynaceae botanical family. Normally it is an annual plant sometime it was also noticed that it lived for years. Occasional pruning is needed to ensure continues and sufficient flowering. The plant grows maximum up to a meter. Leaves appear in pair, smooth and oval. Flowers have fine petals small. There are pink, light pink and white flowers. Flowers appear round the year, so also its beans. Each bean has a number of seeds. It can be propagated either by cutting process or through seeds. It proper care is not taken the plant may die of varus attack. Entire plant is bitter hence cattle do not relish for which the plant is often grown as fences. It is also poisonous to cattle's

Two alkaloids pusiline and pusiline besides tree sterols has been isolated in V. Pusilla. Both the alkaloid are heart depressants. Leaves of Vinca rosea contain two glycosidal principles and ursolic acid. Alkaloids like leurosine, isoleurosine perivine, mitraphylline, lochnerine, perosine sitsirikine, perividine and convincine etc. two alcohols (lochnerrol and lochnerollol), tennin, carotenoids, sterols, a flavona derivative, and oleoresin and sasquiterpenes. Whole plant contains ursolic acid, about forty four alkaloids (isoleurosine, lochne ridsoe, sitsirikine, Vincamicone, catharins, Vindo licone, Vindorosine, lichnarinine, reserpine, akuammine, tetrahydroalstonins, vincaleucoblastine, vondoline, virocine, catharanthine, Lichnarinine etc.) so far twenty four alkaloids isolated from its roots, such as ajmalicine (5-Yohimbine), vinosidine lochnerivine, leirosivine,

cavincine etc. root bark contains alkaloids (Serpentine, ajmalicina, lochnaricine, alstonice), a phenolic resin 2%, d-camphor 3%, a neutral substance , Rose-purple flower contains anthocynine. No reference has been found about this plant either in vedic literature or samhitas nor even in later Nighuntos. It is probable that this useful herb was introduced in India much later. However traditional herbal physicians could derive its virtues later by trial and error process.

**Figure 1.3:** Tulsi (*Ocimum sanctum*)

**Figure 1.4:** Nayantora (*Vinca rosea*)

Entire plant is used in Queensland of Australia and Natal province of South Africa for treating diabetes. It is also used for same purpose in India and Sri Lanka. Juice of the plant is also useful for treating blood secretion and diabetes. Infusion of leaves for either in hot or cold water is useful for expressive blood secretion and vaginal discharge of blood. Juice of fresh leaves is applied externally for scorpion stinge or insect bite. Entire plant with root-reduces hypertension, sedative, reduces nervous excitement and causes sound sleep. Root is poisonous but digestive. In entire plant about seventy alkaloids found, out of which Vinchristine and Vinblastine are found to be useful for treating for blood cancer. Some alkaloids have capacity to reduce urine flow. Four alkaloids possessing antibacterial activity are found in leaves. Some alkaloids are in *Raulfia serpentina* has their hypotensive and hypoglycemic action. It also yields anticancer alkaloids.

Traditional experienced herbal physicians apply it for various diseases with utmost care since over does often produce major toxicity. It is advisable not to use it as medicine without proper professional consultations. Some of the applications are discussed here.

Leukemia is a dreadod disease whose cure seems to be impossible even during modern days. It is reported this herb could cure some patients. In case of diabetes mellitus, it acts miraculously. But due care should be taken and blood sugar be monitored since excess use may cause different problems. In case of irregular menstruation or irregular blood secretion in vagina, it acts as miracle. It can reduce hypertension but drug be applied with proper monitoring the blood pressure since excess dose may be fatal. In case of joint pains, the extract of plant be cooked in till oil (Thoiding oil) and applied externally only on consultation of physicians. Juice or paste of leaves of applied relives pain and cure insect bites, bees' sting or even scorpion stinge. It is powerful herbs and

has to be applied with due care. It is grown everywhere but virtues are not everywhere. It applied appropriately it can be effective yet cheapest drug.

**E: Bandordima(*Chisocheton paniculatus* Hiern. Meliaceae)**

The plant *Chisocheton paniculatus* Hiern belongs to the family Meliaceae[25]. In Assam this tree is locally known as "Bandordima". It is a small tree scarcely exceeding 40 ft. in height and 2.5 ft in girth, with ascending branches and a narrow crown; young shoots and inflorescence brown hirsute; bark dark brown outside, plain but warty, exfoliating in distant round flakes, often with white blotches; inside light dun brown oxidizing to dirty puple on exposure. Leaves 2-4 ft long, dark green; rachis hairy; leaflets 7-33, opposite or sub opposite, 3-10 by 1-4 in, oblong; ovate oblong or lanceolate, acuminate, entire, chartaceous, glabrescent above, thinly pubescent especially along the nerves beneath; lateral nerves 7-13 on either half, obliquely arenate, ends parallel to the leaf margin, prominent beneath; petiolules short. Panicles axillary, 2-3 ft long, dropping with elongated lax branches. Flowers polygamous, pale yellow, 0.5-0.7 in long, faintly scented, 4-merous; pedicels 0.1-0.5 inch long, articulated below the calyx; buds 0.5-0.6 inch long, club shaped. Calyx small, cup shaped, sub truncate. Petals starp shaped, imbricate, minutely puburulous. Staminal tube white, as long as the petals, with 6-8 obscure bifid lobes, more or less pubescent; anthers 8, sessile, linear, included. Disk short, fleshy. Ovary 4-celled, each cell usually with one ovule; style slender, pubescent below, slightly exceeding the anthers. Fruit a capsule, 1.5-3 inch across, globose with a pyriform base, smooth and shining, dark orange or red; seeds black, shinning with a white or yellow aril' endocarp fleshy. This plant is available in the forests of the Northeastern India. The fruits are generally matured in the month of April and May[25] in figure 1.5.

**F: Saru-moin** (*Cudrania javanensis*, Trecul, Moraceae)

*Cudrania javanensis*[25] locally known as "Saru moin" (figure 1.6) a large rambling or subscandent evergreen spinous shrub or a small tree; young shoots pubescent. Bark smooth. lenticellate, yellowish-brown or cinerous, exfoliating in thin flakes; thorns axillary ,straight or some what curved; latex profuse, pale yellow, leaves 1.3-5 by 7-2 in, variable, elliptic-obovate or oblong-elliptic, mucronate, rounded, acute or obtusely acuminate, entire, coriaceous, glabrous, dark-green above, palar beneath; lateral nerves 6-12 on either half, slender, pairs forming intramarginal loops; base usually acute, sometimes sub-obtuse; petiole ·1- ·3 in, long (local specimen); stipules minute. Flower heads ·2-·3 in. across, greenish or yellowish. Male flowers yellow; tepals usually 5, obovate. inflexed at the tips. Female flower heads ·3 in. across; tepals 4 obovate-oblong, thickened and softly tomentose at the tips. Fruit globose, 1-1·2 in. across, velvety and orange when ripe.

This plant is common throughout the province. The ripe fruits are eaten and the heart wood gives a yellow dye. The flowering season of the Plant starts from April to June and the fruits are seen from July to November of the year.

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**Figure 1.5:** Fruits of Bandordima (*Chisocheton paniculatus*) with leaves

**Figure 1.6:** Plant of Saru moin F(*Cudrania javanensis*)with fruits

## 1.3 REVIEW OF THE PREVIOUS WORK

### 1.3.1 XRD STUDIES

The crystallinity of plant fibre has a very important role in describing the thermophysical properties. Reports have been found about the studies on the crystallinity in plant fibres[26-33].Some worker studied the crystallinity of silk and plant fibres by X-ray diffraction method[34-36].

Guinier[37],Kratky[38] and other co-workers[39-40] developed a method for the determination of the size, shape and distribution of the crystallites from the study of the continuous X-ray scattering at very low angles near the incident beam. By using the method Heyn[41]estimated the average diameter of the crystallites of raw and chemically treated plant fibres. Herzog and Jancke[42]shows that cellulose crystalline structure of rame, jute, cotton etc. are same. The crystalline orientation of cellulose fibre is investigated by some workers[43-46].Orientation factor of ramie have been investigated by Sen Woods[47].Hermans[48].studied the crystallinity by X-ray method in native state and mercerized state.Warwicker[49]observed a decrease in the crystalline orientation of the fibre when treated with Caustic soda.But,Biswanathan[50] found no change in crystallinity in native and mercerized states of the fibre.Kulshrestha[51]reported the evidence of paracrystallinity in the ramie fibre, during his course of investigation on the axial order of the fibre.

The optical constants of native and mercerized ramie are determined by a number of researcher[52-57].Iyer et al.[58]esteblished a correlation between crystalline and optical orientation of ramie fibre.

The X-ray diffraction studies of silk have first observed by Herzog and Jancke[59].X-ray investigation of silk of various types is carried out by Kratky and Kuriyama[60].Brill[61]drew the attention to the similarity between the X-ray diagrams of silk

fibroin and silk and synthetic polyamides. Trongs and Hess[62] put forward the theory of two crystalline phases for silk as a possible explanation of their observation that certain X-ray reflections are the same for all silks whereas others varied with the type of silk. Interpretation of the X-ray diagram of fibrion in terms of two crystalline phases is also suggested by other worker[63-64].Corelations of the structural characteristics and textile properties of silk fibre is reported by Padhi *et.al*[65].Conformations of silk in the natural fibre and in regenerated silk are investigated by several workers[66-67].Some researchers carried out studies on the conformations of silk fibroin film prepared from the solution obtained from the silk gland[68-70].Bardoloi[71] *et.al.* study the crystallographic structure of some plant and silk fibre. From X-ray diffractogram the degree of crystallinity (non-irradiated and irradiated ), interplanner spacing ($d_{hkl}$) and relative intensities RI % (raw and degummed) of muga, pineapple ramie and lady's finger fibres are found under different thermal condition[72].

### 1.3.2 XRF STUDIES

The use of X-ray fluorescence (XRF) spectroscopy has open new idea to study the role of the trace elements on structural setup of materials under different conditions. The chemical analysis of various elements using XRF spectroscopy has been used by various workers as a rapid and accurate analysis method[73-74]. Multi elements analysis of geological and fibre samples by XRF technique has been found extensively[75].

The XRF studies on plant and silk fibres has been done by Bora and Bardoloi[76]. They found that among the major elements present in the plant fibres, the relative abundance of Ca is found to be higher in silk fibres compared to plant fibres; which indicates, remarkable significance between silk and plant fibres.

### 1.3.3 TG, DTG, DTA & DSC STUDIES

The study of thermal behaviors of cellulose materials has been made by many investigators[77-80]. The Differential Thermal Analysis (DTA), Thermogravimetry (TG) derivative Thermogravimetry (DTG) and Differential scanning calorimetry (DSC) techniques are becoming important tools for investigation of thermo physical properties of various materials Tang and Bacon[81] have studied the pyrolysis of cellulose up to 773K using Infrared spectroscopy, static Thermogravimetric analysis, gas evolution and physical properties data. The determination of kinetic data from thermogravimetry (TG) curves has been widly reviewed by many workers [82-85] again TG,DTG and DTA and TG studies are silk fibres (Muga , Eri and Pat) and plant fibres (Jute, Ramie and cotton) under different chemical conditions have been done Talukdar. C. Murphy[86] investigated the effect of physically observed water on cellulose degradation. Kilzer and Broido[87-88] have offered a reasonable explanation of the Thermogravimetric Analysis (TGA) and Differential Thermal Analysis (DTA) profiles from pure cellulose. The thermal decomposition of cellulose power in inert oxidizing atmospheres is studied using TG by Fairbridge et al[89]. The pyrolysis of cellulose resulted in the formation of carbonyl and carboxyl containing compounds is investigated by Clifford and Fargher[90] Breger and Whitehead[91] using the vacuum DTA apparatus studied the thermal properties of cellulose. Schwanker and Beck[92] studied by DTA technique, the thermal degradation of polymeric materials in air and nitrogen atmosphere. They observed that the thermal degradation mechanisms are different for the air and Nitrogen atmospheres. Solution[93] has developed an electronic digital computer programme for the calculation of reaction kinetics by the Freeman and Carrol[94] method. The programmed involves computation of temperature and weight and rate of reaction from the values of D.C. mili volt signals originating from the thermo balance. The thermal decomposition of organic fibres using

TG curves been studied and activation energies at different temperatures range are calculated by Venger *et al* [95].

Investigation on the reaction kinetics of organic complexes with the technique differential scanning calorimetry (DSC) has been made by many workers [96-102]. Evaluation of kinetic data from these techniques has also been made by Lengyel et al[103-105] Ishikawa[106] has studied the reaction kinetics of silk fibroin with the DSC thermo grams.

But no such studies on medicinal plants sample A (*Clerodendron colebrookianum*), B (*Azadirachta indica*), C (*Ocimum sanctum*), D (*Vinca rosea*), E (*Chisocheton paniculatus*) and F (*Cudrania javanensis*) under different thermal condition have been noticed till now.

## 1.4 THE PRESENT WORK

In the present investigation, attempts have been made to study the thermophysical, crystalline properties under different thermal and gaseous conditions of the medicinal plants leaves sample A(*Clerodendron colebrookinum*), B(*Azadirachta indica*), C(*Ocimum sanctum*) and D(*Vinca rosea*);and also fruits sample E(*Chisocheton paniculatus*) & F(*Cudrania javanensis*) readily available in North Eastern region of India. Efforts have been made to investigate the feasibility of these medicinal plant leaves to be used in modern health treatments.

### 1.4.1 XRD STUDIES

The crystalline nature of these medicinal plant leaves and fruits sample: A, B, C, D, E, and F at different thermal condition has been investigated by x-ray diffractogram (XRD).

### 1.4.2 XRF STUDIES

The major element present in the unheated and heated medicinal plant leaves sample (A, B, C & D) and fruits sample (E & F) except C, H, O are evaluated by x-ray fluorescence (XRF) technique.

### 1.4.3 TG, DTG, DTA & DSC STUDIES

Attempts have been made to study the various thermodynamical properties of the medicinal plant leaves sample (A, B, C & D) and fruits sample (E & F) abundantly found in the North Eastern Region (specially Assam) have been investigated by Thermogravimetry (TG), Derivative Thermo-gravimetry (DTG) Differential Thermal Analysis (DTA) methods in air, oxygen and nitrogen atmospheres in the temperature range (300-743) K and Differential Scanning Calorimetry (DSC) method air atmosphere.

From weight loss (%), time and temperature, the activation energies of these medicinal plants have been computed.

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