Chapter 8:

Chemotaxonomical studies on some members of the Punicaceae, Onagraceae, Lecythidaceae and Alangiaceae

These are small families grouped with Myrtales in one or other classifications. Since one or two members of each family were available for studies, all these four families are treated together in one chapter.

The **Punicaceae** are a unigeneric family of *Punica* which is having two species, *P. granatum* and *P. protopunica*; the former common in Balkan region and India and the latter, a native to Island of Socotra. The plants are small trees or shrubs with quadrangular wings and simple opposite leaves with vestigial stipules. Flowers are large, solitary or in clusters, perfect, regular, hypogynous with the colored hypanthium prolonged well beyond the ovary. Sepals 5-8 appearing as lobes of hypanthium. Petals 5-8, crumpled in bud. Stamens many produced on the hypanthial tube in centrifugal sequence. Gynoecium of several carpels, undergoing differential growth to become superposed in 2 to 3 layers, the lower layer with axile placentation and the upper appearing parietal because of asymmetric growth. Style slender, stigma capitate. The fruit with a leathery rind and crowned by the persistant sepals. Seeds embedded in a pulpy mass representing the proliferated sarcotestas. Seed fats yield a rare fatty acid, the punicic acid.

Traditionally this family is considered closer to Myrtaceae by almost all taxonomists. But Bridgewater and Baas (1978) considered that wood anatomy of *Punica* supported is close relationship with the Lythraceae. Therefore Thorne (2000) included this taxon as a subfamily Punicoideae within the Lythraceae. *Punica granatum* yields the edible fruit Pomegranate. The fruit rind and bark of fruit and stem yield pyridine alkaloids such as pelletierine, pseudopelletierine etc which are used as anthelmintics and febrifuges.

51

The **Onagraceae** consist of mostly aquatic herbs or shrubs (18/650) of cosmopolitan distribution. The plants are herbs or arborescent shrubs with unicellular hairs. Leaves are of various types but basically opposite, simple, at times lobed. Flowers borne solitary in the axils or in leafy bracteate or naked spikes/racemes, sometimes unisexual. Sepals and petals 4-5 each. Stamens 2-8, bicyclic generally attached within the hypanthium surrounding an epigynous disc. Carpels 4-5, syncarpous or the partitions sometimes imperfect. The placentation axile or parietal. Style with a 4-lobed stigma. Ovules many in a locule. Fruit a loculicidal capsule or berry.

This family is somewhat isolated in the order Myrtales, being well set of in embryological characters such as 4-nucleated embryo sac and diploid endosperm. Thorne groups this family along with Lythraceae and Combretaceae in the suborder Lythrineae in Myrtales. Many plants of this family like *Oenothera, Fuchsia, Epilobium* and *Clarkia* are well known ornamental plants.

The Lecythidaceae are a tropical family of 20 genera (285 sp.) containing trees or shrubs. The plants produce triterpenoid saponins and proanthocyanins and stem with mucilage and stratified phloem. The leaves are alternate, commonly crowded at the tips of branches, simple and epipetalous. Flowers, typically pollinated by bats/insects, are large in axils of leaves, or cauliflorous. Flowers are showy, perfect, regular or irregular and epigynous. Calyx of 4-6 sepals, valvate or connate and calyptrate. Petals are the same as sepals, imbricate or wanting. Stamens many, centrifugal, filaments connate below and distinct above, produced on one side into a flat ligule that is curved over the ovary as a hood, some of the stamens reduced and staminodial. Intrastaminal nectary disc well developed. Gynoecium of 2-6 carpels united to form an inferior or seldom half-inferior ovary with a terminal style and capitate stigma. Placentation axile with

half-inferior ovary with a terminal style and capitate stigma. Placentation axile with many ovules in each locule. Ovules bitegmic and tenuicellular. Fruit capsular with a distal operculum, often very large or drupaceous or baccate. Seeds with a funicular aril.

The Lecythidaceae have traditionally been referred to the order Myrtales because of their combination of separate petals, numerous stamens and syncarpous inferior ovary with axile placentation. They differ from the characteristic members of the Myrtales, however, in their alternate leaves, bitegmic tenuicellular ovules, lack of internal phloem and a series of embryological features that have been elucidated by Mauritzon (1939). Cronquist (1981) considered these differences too formidable to ignore and since no other order could accommodate the Lecythidaceae with undue strain, he recognized an order Lecythidales. Once removed from the Myrtales, the placement of Lecythidales was a problem and the Rosidae and Dilleniidae were the two subclasses which could accommodate this order.

The Lecythidales were found to be more at home in the Dilleniidae than in the Rosidae. The bitegmic, tenuinucellar ovules of this order are seen in orders like Theales, Primulales and Ebenales, but are rare and scattered elsewhere. Although centrifugal stamens have been demonstrated in the Myrtales, this kind of androecium is much more common in the Dilleniidae. Stratified phloem of the Lecythidales is especially common in the Malvales and to a lesser extent in the Theales. Wedge-shaped phloem are likewise more common in the Malvales and Theales. Barringtonia in the Lecythidaceae produces 3-sambubiosides, similar to those of Hibiscus. In addition, the Lecythidales resemble characteristic members of the Malvales in their valvate calyx, connate filaments and mucilage (Lecythis), but differ in the absence of stellate pubescence and cyclopropenoid fatty acids. The Lecythidales resemble several families of the Theales in their bitegmic, tenuinucellar ovules and they resemble the Ochnaceae in having cortical vascular bundles. The pollen structure also is readily comparable with that of the Theales and Malvales. With all these reasons Cronquist (1981) derived Lecythidales and Malvales as cases of parallel evolution with divergent specializations from a common ancestry in the Theales. Thorne (2000) also supported this contention.

The members of the Lecythidaceae show striking differences in androecium and therefore some authors recognized several segregate families like Asteracanthaceae, Barringtoniaceae, Foetidiaceae, Napoleonaceae, etc. But Kubitzki (1990) recognized four families within this family. They are (1) Lecythidoideae, (2) Napoleonaeoideae, (3) Foetidioideae and (4) Planchonioideae (Barringtoniaceae).

The seeds of *Bertholletia excelsa* yield Brazil nuts of commerce. Bark of *Barringtonia acutangula* contains dicarboxylic acid, barringtonic acid and barringtogenol E. Leaves contain acutangulic and barringtogenic acids and stigmasterol.

A single genus *Alangium* (20 spp.) native to tropics forms the members of the family **Alangiaceae**. The plants here are trees or less often shrubs or wines, often with stellate or peltate hairs and containing saponins and alkaloids of emetine type. The leaves are simple estipulate. Flowers in axillary cymes, perfect or rarely unisexual and epigynous. Calyx lobes 4-10 or absent. Petals 4-10 linear, valvate, hairy within, sometimes reflexed after anthesis. Stamens as many or twice as many as petals, in a single rind around a prominent, pulvinate epigynous disc. Filaments hairy within, sometimes adnate to the petals at base. Gynoecium of two carpels united to form a compound bilocular ovary that lacks axile vascularisation or more often pseudomonomerous with a single locule. Style short with two elongate ventrally stigmatic branches. Ovules solitary and pendulous in each locule. Fruit a drupe crowned by the sepals and disc containing a single uni/bilocular stone, each locule with one seed. Seeds large with foliaceous cotyledons.

This family is distinct from all the other families included in Myrtales in having latex, isoquinoline alkaloids and unitegmic crassinucellar ovules. Therefore almost all recent taxonomists group this family with the Cornales.

Alangium salvifolium is a well-known medicinal plant in India.

Previous Chemical Reports

Fruits of *Punica granatum* were rich in flavonoids. Bark of stem and root contained alkaloids of pyridine group like methylpelletierine ($C_9H_{17}ON$), and pseudopelletierine

(granatonine) (C₉H₁₅ON), and isopelletierine (Anonymous, 1995). Nine triterpene saponins, acutangulosides A-F and acutangulosides D-F methyl esters and a single triterpene aglycone were isolated from water extract of the bark of *Barringtonia acutangula* (Mills *et al.*, 2005). The roots, rootbark, stembark and leaves of *Alangium salvifolium* contained isoquinoline alkaloids like alangine A and B, cephaeline, emetine, tubulosine, etc. (Anonymous, 1985).

In the present work three members of the Lecythidaceae and one member each of the Onagraceae, Alangiaceae and Punicaceae have been studied for their chemical constituents.

Materials and Methods:-

All the plants except *Careya* were collected from in and around Baroda. *Careya* arborea was obtained from Kerala. Voucher specimens of all these plants have been deposited in the Herbarium, Department of Botany, The Maharaja Sayajirao University of Baroda (BARO), Vadodara. Standard procedures are described in chapter 2 were followed for the extraction, isolation and identification of the phytochemicals.

Results:-

The results of screening of 1 member each of Punicaceae, Onagraceae and Alangiaceae as well as three members of Lecythidaceae for the flavonoids, alkaloids and quinones are presented in Table 6.

The leaves of *Punica granatum* contained flavones only and no flavonols. The flavones present were acacetin (4'-OMe apigenin), luteolin and 3'-OMe luteolin. The stem contained flavonols only (no flavones) such as gossypetin, 3'OMe gossypetin and 3', 4'-diOMe quercetin. Besides vanillic, syringic and gallic acids which were common in both stem and leaves, leaves contained melilotic acid. The large amount of gallic acid in leaves is noteworthy. Alkaloids were located in both stem and leaves. Tannins were also abundant in this family.

Ludwigia octovalvis, the only plant screened in Onagraceae is found to contain quercetin in stem and leaves. Gallic acid was found in large amounts in leaves which

also contained vanillic and syringic acids. The stem contained *p*-hydroxy benzoic acid also in addition. Quines were located in the stem of this plant. Alkaloids, saponins and tannins were found to be absent in the plants screened.

All the plants of Lecythidaceae screened, *Careya, Couropita* and *Barringtonia* were found to contain various flavonoids. *Careya arborea* contained kaempferol and 3' OMe quercetin in leaves and luteolin, a flavone, in stem. *Couropita* contained kaempferol and quercetin in leaves and gossypetin and quercetin in stem. *Barringtonia* is found to contain gossypetin, 3' OMe gossypetin and 3', 4'-diOMe quercetin in leaves, the former two compounds in young stem and myricetin and gossypetin in bark. The phenolic acids in *Couropita* were sinapic acid in leaves and *p*-coumaric acid in stem besides vanillic and syringic acids. Gallic and *p*-coumaric acids were located in both leaves and stems of *Careya* while the leaves contained *p*-hydroxy benzoic acid also. In the case of *Barringtonia*, the stem and bark contained vanillic and syringic acids only whereas the leaves contained gallic, melilotic and *p*-coumaric acids also. Quinones were present in *Couropita* and *Barringtonia*. Alkaloids were found to be absent from all the plants screened.

Alangium, the only one plant screened in Alangiaceae contained kaempferol,

4'-OMe kaempferol and traces of quercetin in leaves. The bark contained only traces of flavonoids alongwith vanillic, syringic, *o*-coumaric and protocatechuic acids. Stem possessed only quercetin and phenolic acids such as vanillic, syringic, melilotic and sinapic acids. Quinones were present in all the parts of the plant screened

Discussion:-

Among the four small families screened, the Alangiaceae are very distinct in possessing *o*-coumaric and protocatechuic acid. This family does not contain gossypetin, the flavonol characteristic to the Myrtales. Instead, it contains isoquinoline alkaloids which are not seen in Myrtales. All these chemical differences support the shifting of this family to another order Cornales by all recent taxonomists.

The family Lecythidaceae is very similar to the rest of the Myrtales in containing gossypetin, its derivatives and other flavonols. Therefore the exclusion of this family from the Myrtales does not get any chemical support. Morphological characters such as the presence of inferior ovary and large number of stamens and clustering of leaves at the tip of stems (as in the Combretaceae) support this contention.

The Lecythidaceae with their clear disc, calyptrate calyx, inferior ovary, peculiar fruits and arillate seeds are entirely different from the Theales or Malvales. The triterpenoids saponins of the Lecythidaceae also are not common in these orders. Therefore the shifting of Lecythidaceae (or Lecythidales) near to Theales and Malvales cannot be justified.

But the differences in other characters such as lack of internal phloem and in having alternate leaves and bitegmic tenuinucellar ovules indicate that this family possesses distinct identity and therefore it may be treated as a suborder Lecythidineae as practiced by Takhtajan (1980). The presence of gossypetin, a typical flavonol of the Malvaceae, and the characters such as stratified phloem and connate filaments, by the strength of which this family was shifted to sub class Dilleniidae near Malvales and Theales, may be treated as cases of parallel evolution. Incidentally some of the members of the Myrtaceae also possess stratified phloem. *Barringtonia* is chemically similar to *Couropita* and *Careya* and thus establishing a separate family status to this plant as Barringtoniaceae is not chemically supported. Due to the differences in morphology, at best, it may be treated as a subfamily with in the Lecythidaceae.

The **Punicaceae** are distinct in having flavones in leaves and flavonols in the stem. Presence of alkaloids of pyridine type in both stems and leaves is another distinctive feature of this family. These chemical characters do not support including Punicaceae in Lythraceae as a subfamily Punicoideae as practised by Thorne (2000). Lythraceae are characterized by quinones and the alkaloids of *Punica* are not reported in any member of Lythraceae. The Punicaceae can be considered as an advanced family of the Myrtales.

Onagraceae with quercetin and gallic acid, the latter in large amounts, fits very well with the other families of the Myrtales.