# SCIENTIFIC SECTION

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# COMMON AND ORIENTAL CARDAMOMS.\*

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## INTRODUCTION.

While considerable information has been collected concerning the official cardamom (*Elettaria Cardamomum* Maton), fruits belonging to the family of *Zingiberaceæ*, no critical survey has thus far been made to establish the exact relationship and the comparative value of several other varieties which are recognized articles of trade in other countries, especially those of the Orient. Certain varieties, widely cultivated, in China, for instance, and possessing a distinct flavor might equally deserve an official recognition. Although the study of products in laboratories, located far removed from the countries of origin and growth, always renders difficult final decisions as to the natural grouping of those varieties, some light, nevertheless, can be thrown upon the relationship and actual value of these forms, now classified both as Elettaria or Amomum species.

This difficult subject has not had the attention of the pharmacognosists until the time of Dr. Pereira, to whom we are indebted for his important observations, particularly on the pharmacognostical history of various species of Amomum, growing on the West Coast of Africa. Shortly afterward Guibourt of Paris has carefully conducted researches on certain species of cardamoms, which, although rarely met with in America or Europe, are important articles of trade among the Oriental countries.

To these Oriental Cardamonum species, esteemed highly by the majority of the Chinese physicians, Daniel Hanbury has favorably alluded in papers on some rare kinds of cardamom. He stated: "It is my hope that Europeans residing in the countries in question, who take an interest in natural history, may be stimulated to some exertion to discover the botanical origin and obtain further accounts regarding the culture, the collection and the uses of these productions, which, apart from their interest to the pharmacologist, are derived from plants, many of which, remarkable for a splendid inflorescence, would become valuable additions to the horticultural collections of this country (1)."

We shall record here the results of our investigation correlated with the data in the literature. In view of the official recognition of several species of cinnamon, ginger, etc., in the U. S. Pharmacopœia, there are good reasons to consider for adoption as official also some other important varieties of cardamom. Hence, critical evaluation of some of the Chinese varieties in the proper light is the object of the following pages.

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## AMERICAN PHARMACEUTICAL ASSOCIATION

# PART I.

## HISTORY OF THE DRUGS AND SPICES.

1. Origin, Use and Extent of Growth.—Although a drug of so striking a flavor as cardamom can hardly have been unknown to the Ancient classical writers, it cannot with certainty be identified in their writings. There has been considerable doubt in the origin of the name; whether the word cardamom is of Greek, Arabian or Indian derivation, still remains a matter of great obscurity. Elettari is the Malayan or native name of the plant in Malabar, while Cardamomum, Kapsauwuor is the name of some Indian spice used in classical times. However, the Greek nomenclature is apparently suggestive of "Delicious Spice."

According to Susruta, the author of "Ayurvedas," an old Sanskrit medical work in which a large number of Oriental drugs are mentioned, cardamoms, Elā, have been used in India from a remote period (2). The drug, being thought to have reached Europe prior to the Christian Era, is found to agree (though not exactly), in description with the Kapsauwuor of Theophrastus and Dioscorides, and Amomum or Amomis of Pliny.

In the curious list of Eastern Drugs and Spices, liable to taxation at the Roman Customhouse in Alexandria about 176–180 A.D., Amonum as well as Cardamonum are mentioned (3). It has been incidentally mentioned by St. Jerome in common with musk as perfumes used among the voluptuous Eccleciastics of the 4th century (4).

Hanbury reports in this Pharmacographia that cardamoms are named by Edrisi about A.D. 1154 as a production of Ceylon and also as an article of trade from China to Aden. In the same century, among the highly esteemed spices, cardamom is mentioned together with cinnamon and cloves in the long and interesting article by a pharmacist of Cairo, named Abul Mena.

Though the drug has found its way into Europe as early as the 12th century, only in the time of Garcia de Orta, 19th century, cardamom was brought to the careful attention of the world and became much valued in the European market.

Of the place of growth of cardamom the first distinct notice seems to be that of the Portuguese navigator, Barbosa, A.D. 1514, who, while searching for spices, discovered for the first time the cardamom as a product of the Malabar coast.

The early history of cardamoms renders it somewhat likely that the writers of the classical times have erroneously used the name for many other fruits of the family of Zingiberaceæ. It is scarcely possible to determine what products they actually referred to, as their notices of it are so brief and indefinite, though many believe it to have been the variety which we call *Elettaria Cardamomum*.

2. Brief Historical Discussion of the Rarer Cardamons.—For the purpose of comparison we feel it necessary while discussing the history of cardamom in general, to briefly review facts concerning a few of those varieties which have some importance in pharmacy and medicine.

(a) Amomum Cardamomum, L.—The Cluster or Round Siam cardamom. Being considered of great rarity and obtained from a native plant of East Indies, the fruits reached Europe in the early part of the 17th century. Drug dealers at that time considered it as the only product to be used when Amomum was ordered. The chief source of supply is reported to have been Bangkok, Siam, where large shipments were made to England, China and Japan. In 1857, several batches of Bangkok cardamom were offered for sale in London and figures at 1s. 6d. per pound, cost price. The amount of drug exported to Singapore and China in 1871 was estimated to be 4678 piculs (623,733 pounds) (2). Hence it thoroughly testifies to the fact that the drug is an article of considerable traffic in Eastern Asia. The Round cardamom occurs in small compact bunches, possessing a strong camphoraceous and aromatic flavor, and having a general resemblance to that of the Malabar Cardamom (Elettaria Cardamomum). Round cardamoms are rarely employed in this country, but they were formerly official in the French codex and Dublin Pharmacopœia. According to Hanbury, they are also mentioned in the celebrated Dispensatorium of Valerius Cordus.

The export of cardamoms from Siam is classified under two headings, "Best Cardamoms" and "Bastard Cardamoms." Bastard cardamoms are the product of one or more species of Amomum growing wild. This class comes almost entirely from Eastern Siam (5).

"Best Cardamoms" are probably derived entirely from cultivated plants; the species that yields them being Amomum Krervanh. This plant is chiefly grown in the provinces of Chantaburi and Krat. Plantations are made in the forests often in mountain valleys. In making these plantations the forest trees are not felled, but some clearing of the undergrowth is done.

In the five years ending March 31, 1930, the average annual export of "Best Cardamoms" was 1118 piculs, valued at Tcs. 234,417. In the same period the average annual export of "Bastard Cardamoms" was 4061 piculs, valued at Tcs. 205,418. Hongkong takes most of the export of both qualities.

(b) Amomum Melegueta.---This is the so-called Amomum grana-Paradise which furnishes the "Grains of Paradise" or guinea grains of commerce. The plant is native of Africa and has been transplanted in the West Indies. So far as known, it has never been found in Asia. Some writers have erroneously referred it to Amomum xanthioides which is now understood to be the "Bastard Cardamom." It is possible that the seeds of the latter variety have occasionally been used as a substitute for those of Amomum Melegueta, but they are nevertheless not the true "Grains of Paradise." The name Melegueta refers to the ancient empire of Melle of the upper Niger region of Africa. And again, Melegueta or Malaguelta is also said by Hanbury to be the African name of the seeds, though it is more probably of Spanish origin. But in modern Spanish the word is applied to various small pungent spices. Though the early history of grains of Paradise falls short of unauthentic record, there is an incidental allusion to its use as a spice, among the aborigines of West Africa. There are many references showing the fruits to have been employed in medicine in the 13th century. Again among important spices, the "Grains of Paradise" were used and mentioned by John, King of France, 1359-1360. Even Christopher Columbus, while voyaging to the coast of Guinea, has referred to it in connection with Costa Di Maniguetta, which later became a monopoly of the Kings of Portugal.

Grains of Paradise, as seen in commerce, agree in many respects with the seeds of the official cardamom, having a round, ovoid or somewhat wedge-shaped outline and shining golden, brown appearance. Their surface is slightly rough from the presence of small longitudinal striations, and internally they are white, when crushed and rubbed between the fingers, a characteristic aromatic odor is developed. They are highly esteemed by the native African as the most wholesome of spice; in Great Britain and the United States, it is primarily employed in the preparation of veterinary medicines, and to impart pungency to cordials. "Hippocrus," the famous spiced wine of the 14th century, was heavily flavored with grains of Paradise, cinnamon and ginger.

(c) Ovoid China Cardamom.—In the hands of modern pharmacognosists, the plant yielding Ovoid China cardamom has been referred to Amomum medium of Loureiro. It is described in the "Chinese Herbal" together with Alpinia globosum, from which it is with difficulty distinguished. It is a product of Southern China, and abundant in the drug shops of the latter. This cardamom is characterized by its large, hard, angular seeds, which alone seems sufficient to prove its distinctness from the other varieties of cardamom. The drug is employed by the Chinese in much the same cases as the Amomum globosum, to which it is preferred in the treatment of various gastric disorders. It should also be mentioned that the seeds have been formerly much used as a condiment or spice.

(d) Amomum Villosum.—This form is described by Loureiro to be a Cochin-Chinese species of Amomum, which has early been naturalized in China and is largely grown in the province of Kwan-Tung. Some writers have designated it the "Hairy China Cardamom." It is rather unfortunate that the Chinese have erroneously regarded it as identical with Amomum xanthioides (Bastard cardamom); and they are usually found on the market admixed with the seeds of the latter. The "Hairy China Cardamom" is sometimes sold with the stalk attached, sometimes removed from it. The stalk or scape, when perfect, measures 3-4 inches, carrying ten or twelve capsules. It is the scape that is beset with hairs and not the capsules. The same tonic and stomach properties are ascribed to the seeds of this plant as to those of cardamoms in general.

(e) Amonum Amarum.—"I-Chih-Tse" is the correct Chinese name for this special variety: the Bitter-Seeded Cardomom; the origin of which is still very little understood. The botanical origin is first mentioned by Guibourt who has (doubtfully) referred it to Zingiber nigrum of Contract a classifier the

of Gaertner, a plant identical with *Alpinia Allughas* of Roscoe, which is considered by Pereira and Hanbury to be a totally different species. The "Pun-Tsao-Kungmuh," a celebrated Chinese herbal, says that the fruits formerly occurred in Thibet and Cochin China, but are now chiefly produced in the Province Kwang-Tung.



Fig. 1.—1/2 × Official Malabar cardamom.

(f) Amomum Globosum of Loureiro.—(Tou-Kow or Tsao-Kow, Chinese.) The plant has been referred to Alpinia globosa by Stuart (6). It is actually the Amomum globosum Loureiro, described by Hanbury as "The Large Round Chinese Cardamom." The actual plant, furnishing



Fig. 2.— $1/2 \times$  Long Ceylon cardamom.

the well-known Chinese cardamom in commerce, is still uncertain and is yet to be carefully studied by botanists. It is found to be early naturalized in Southern China, namely, the provinces of Kwang-Tung, Kwang-Si-Yunnan and Fukian. In commerce there is the so-called "Small Round China Cardamom," which Hanbury and Perrot consider simply a variety of the preceding. Both cardamoms and the flowers are used in Chinese medicine and reputed to counteract the effects of wine in the system.

An interesting botanical discussion of Alpinia, Amonum and Elettaria species, as possible or assured sources of cardamoms, has recently been given by Brandt and Wasicky (7); see also Tschirch (8).

For illustration of some of these cardamoms, official and non-official, including Oriental, see Figs. 1, 2, 3, 4, 5—all 1/2 natural size.



Fig. 3.— $1/2 \times$  Korarima (African) cardamom.

# Part II.

#### CULTIVATION, PRODUCTION AND COMMERCE.

1. The plant of the *Elettaria Cardamonum* grows abundantly, both wild and under cultivation, in the mountainous districts of Indo-China, Ceylon and Southern India, at an elevation of 2500-5000 feet above the sea. It is also reported that the place where cardamoms grow best has an average temperature of  $22^{\circ}$  C. and an average rainfall of 121 inches. The fruits are largely obtained from cultivated plants; and the gathering of the fruits before maturity commences in October and continues during dry weather for two or three months. However, the methods of cultivation vary considerably in the different localities.

In some moist shady mountain forests cultivators, shortly before the rainy season commences, usually locate a spot where some cardamom plants are growing. Not infrequently they root out the surrounding weeds that are decidedly injurious to the plants and at the same time



Fig. 4.—1/2 × Chinese Round cardamom.

a certain amount of light is admitted for the proper growth of the plant. The plant, having attained 2 to 3 feet in height, is well protected and left to itself for a year. At the end of the second year plants begin to flower, and bear fruits a year later. Plants thus remain productive for quite a few years.

The cardamom plants in Western Mysore are cultivated in the betel-nut regions and thus are raised between the palms, from which they derive the necessary amount of shade. They commence to bear fruits in their third year and these fruits are gathered from October to late in December. Either the entire spike may be cut or only ripe fruits may be plucked from the spikes. But these fruits are dried in different ways; thus in some cases, the white scape of fruits is gathered at once and dried; or the fruits, when collected, are carried to the houses and partially dried on mats for a few days. They are then stripped from their scapes and dried by a gentle fire-heat. Occasionally they have to be bleached by exposure to dew and sunlight or sulfurous acid and



Fig. 5.— $1/2 \times$  Bitter cardamom.

steam. The seeds are best kept in their pericarps, in which condition they are imported in chests, but when required for medicinal use the seeds should be separated from them and the pericarps discarded. Those seeds which have been removed from capsules become inferior in time through loss of volatile oil, responsible for the flavor, as compared to those that have been kept enclosed.

Most of the drug goes through Bombay or Madras to the various parts of the world. The cardamom fruits are esteemed in proportion to their plumpness and heaviness and the sound and mature condition of the seeds they contain. In commerce they are found in the two forms of "Shorts" and "Shortlongs." The former variety is usually broad and plump, while the latter is finer ribbed and lighter than the "Shorts." The Malabar cardamoms are of the highest commercial value and consist of both varieties. The Mysore cardamom is considered to be the next best grade, consisting mostly of "Shorts," but being less pungent.

Beside the Malabar or official cardamoms, above described, cardamoms of other species, rather generally believed to have an inferior flavor, have been and are now widely employed in the Oriental countries. The forms mainly considered are the Amomum Globosum of Loureiro (Large Round Chinese cardamom) and the Chinese Bitter-Seeded cardamom.

2. The Round Chinese cardamom, as we have previously stated, is indigenous to Southern China (6). This evergreen plant, said to resemble the Myristica in appearance, now grows best both wild and under cultivation in the Kwang-Tung province. The flower has not yet been de-

scribed, but the other parts in many respects are similar to those of Elettaria Cardamonum. The plant as a rule thrives best in a light, moderately rich and well-drained soil which has been carefully prepared before planting. Though the fruits, furnishing the Round cardamoms of commerce are largely obtained from the cultivated variety, it is not at all uncommon to find that the latter is often admixed with that of the wild growing species. The seeds, from which the cultivated plant grows, should not be more than 2 years old and are sown dispersedly, covered one to two inches deep. Usually at the end of the second year fruits can be gathered in a ripened condition. Since the plants develop very slowly it is therefore not advantageous to sow the seeds in weedy soil.

3. The Bitter-Seeded cardamom grows wild in the shady forests, where slight admission of light helps the plant to develop in abundance. It is believed to grow better in moist soil, and growers do not seem to have much difficulty in germinating the seeds in the place where the climate is warm and damp. However, the writers have not succeeded in growing the seeds in the greenhouse and it is very likely that the seeds were not viable. But the plant, which produces this variety of bitter seeds is still not identified. The writers have also failed to obtain the true plant from the place of growth, namely, the province of Kwang-Tung. Hanbury states "Received from R. Swinhole, a specimen which he says is from Hainan, where the aborigines use it as tea. He adds that there is good reason to believe it to be the fruit of the Alpinia which yields the galanga root of commerce" (9).

In considering the commercial aspects of the two Chinese varieties, the market demand for these drugs is naturally the chief factor in determining the prospects for their commercial production under cultivation in the foreign countries. So far as studied, they are cultivated with benefit in China, where they are grown under conditions of soil and climate resembling those of many localities in the United States. Often the most suitable plants for a particular locality cannot be foretold, especially in those situations where no attempts have yet been made to grow them in the places other than the native. In such cases, it is therefore well to try and select for cultivation the plants like cardamoms which may thrive in the new situation under conditions closely resembling their old habitat. As a number of native medicinal plants, which in their wild state are restricted to certain localities, have been successfully cultivated in situations far beyond their natural range, there are good reasons to believe that Chinese cardamoms will thrive in sections where they are not now grown.

Annually, fairly large sums of money are expended for the official cardamoms (*Elletaria Cardamomum*), imported from Ceylon and India, where they are cultivated under conditions similar to that of many sections in the United States. As a means of guaranteeing the future supply of the spice in question and of alleviating the necessity of depending on specific importations, attention might well be directed to the possible cultivation of the various cardamom plants in this country. There is now a wide use and remunerative production of Chinese cardamom in China.

# PART III.

# BOTANICAL CHARACTERISTICS.

1. Plants. (a) General Morphology.—Large Round China Cardamom (Tou-Kow or Tsao-Kow).

The plant under consideration produces the large Round and small Round Chinese cardamom of commerce. It is attributed by M. Guibourt to the *Amomum* globosum of Loureiro. This species has been described and illustrated by Hanbury (10) "On Some Rare Kinds of Cardamom." But identity of the plant is still far from being conclusive. It is a native of South China and Cochin China, and is now widely cultivated in all parts of Kwang-Tung and Kwang Si, as well as in parts of Yunnan and Fukien.

Stuart describes the plant as follows: "The plant is said to resemble the Myristica in appearance, and bears a red flower, changing to yellow, in the axils of the leaves, which has some likeness to the Hibiscus. The leaves resemble those

of the Wild ginger, and are sometimes gathered in the miniature state in a similar manner to tea buds (6).

The plant of the wild growing variety, often confounded with those under cultivation, is much more difficult to propagate from the seeds than the cultivated species, commonly grown in previously prepared moist soil. Hence these species for cultivation in this country are not easily propagated but require special conditions if good results are to be realized.

#### THE BITTER-SEEDED CARDAMOM-AMOMUM.

The plant believed to be that of Zingiber nigrum, which is identical with Alpinia Allughas, a Cochin Chinese species, the origin of which is still somewhat doubtful. The Chinese term is also applied to Nephelium longa, but recent writers restrict it to the Bitter-Seeded cardamom. According to R. Swinhoe it is probably a fruit of the Alpinia which furnishes the galanga root of commerce.

This evergreen herbaceous perennial is said to resemble Amonum Malegueta (Grains of Paradise) in having a long, slender, branched rhizome and an erect stem. The flowering stem is usually 4-6 feet in height, bearing pinkish flowers of moderate size. Numerous green and leathery leaves are described by some of the native herbals.

(b) Differentiation of Cardamomum Plants from Amomum Species.—Cardamomum, which is mentioned in the Sanscrit writings, apparently derives its name from "KAPSAUWUOR," a designation for some Indian spice in classical times. Likewise Amomum, "AUWOUR" is accepted as the classical name of some undetermined Eastern spice-bearing plant. Being derived from a common origin, the terms are indiscriminately used by the botanists for the various fruits of the family Zingiberaceæ, grouped under the name of Cardamoms.

From a close examination of the characters by which they are distinguished and of the chief points of interest attached to them, the writers so far as they studied the taxonomy of the fruits in question have failed to agree with the general opinion of placing them under separate genera. From the standpoint of classification, in Elettaria the connective tissue of the flower shows no extension and combines the halves of anthers in their entire length, while there is either no extension or an insignificant extension of the connective in Amonum (Sect. Geanthus) and broad extension of connective in case of Amonum Sect. Euamonum (11). Apparently the points of differentiation of Amonum from Cardamonum are far from being conclusive.

2. Seeds. (a) General Morphology of Forms Mainly Concerned.—The fruit of the Amomum globosum of Loureiro constitutes the large and small Round Chinese cardamom of commerce. It is a round or globular capsule having an average diameter and thickness of 15 mm., weighs about 475 mg. The fruit is trivalved and loculicidally dehiscent, a breaking away of the valves from the septa or partitions.

Capsule:

Color: Externally—Pale yellow. Internally—Very light shade of yellow. Shape: Round or globular. Surface Marking: Longitudinally streaked. Inner Surface: Indistinct network markings. Extremities: Pointed at both ends, surrounded by unicellular non-glandular hairs. Texture: Thin and easily torn. Taste: Slightly aromatic, practically tasteless. Oct. 1937

Seeds:

Shape: Pyramidal or wedge-shaped.
Number: Average of 24 seeds in each pod, forming 3 coherent
masses which are separated by a thin whitish membrane.
Size: Thickness-average 2.65 mm., maximum 3 mm., mini-
mum 2.05 mm. Length: Average 3.5 mm., maximum
4 mm., minimum 3.25 mm.
Color: Brownish yellow.
Weight: Average 20 mg., maximum 24 mg., minimum 17 mg.

TABLE I. OBNERAL CHARACIERISTICS OF THE OFFICIAL AND THE CHINESE CARDAM
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Official Cardamom.		Large Round China Cardamom.	Bitter-Seeded Cardamom.	
Shape of fruit	Ellipsoidal or ovoid	Round or globular	Oval or ovate oblong	
Color	Light buff	Yellow	Dusky brown	
Surface markings ) of the capsule	Longitudinally streaked	Interrupted longitudinal ridges	Lines of irregularly interrupted ridges	
Texture of the capsule	Thin and papery	Thin and easily torn	Hard and coriaceous	
Extremities	Round at base, contracted at apex, devoid of hair	Tapering at both extremi- ties covered with numerous long non- glandular hairs	Pointed at both ends, having a few short hairs	
Number of seeds ( in a fruit	15-30 seeds	Average about 24 seeds	25–30 seeds	
Description of seeds	Brown, red or grey, irregu- larly angular trans- versely rugose and a de- pressed hilum, longitu- dinally grooved on one side	Seeds are brownish yel- low, pyramidal or wedge - shaped with deep furrow on one side	Dark brown in color, obtusely angular but smaller than that of the round variety. They ad- here firmly together. Each seed weighs about 14 mg.	
Size of Seeds:	4 mm. in length. 3 mm. in diameter	Same as Elettaria	3.5 mm. in length. 2 mm. in diameter	
Taste of Seeds }	Aromatic and pungent	Distinctly aromatic in taste and smell	Bitter and myrrh-like	

(b) Anatomy-Histology.—Microscopical Characteristics—Transverse sections of the Round Chinese cardamom seed show the following tissues from periphery toward the center.

TABLE II.--HISTOLOGICAL CHARACTERISTICS OF THE ROUND CHINESE CARDAMOM.

- 1. Arillus, a whitish transparent membrane of elongated cells loosely attached to the outside of the testa.
- 2. Spermoderm consisting of:
  - (a) Epidermis—A single layer of big oblong cells with thickened walls. Width,  $20\mu$ ; length,  $50\mu$ .
  - (b) Cross-Cell—A very narrow layer of nearly isodiametric cells occasionally undifferentiated from the epidermal layer. Tannin is present.
  - (c) Oil Cell—Big oil cells, usually devoid of oil globules. Length of the cells exceeds the width. It is usually a one-celled layer.
  - (d) Parenchyma—An indistinct layer of tangentially elongated parenchyma cells with traces of brownish tannin contents.
  - (e) Sclerenchyma—A layer of brownish stone cells with thickened walls, each having a distinct lumen, which is 12μ in diameter. Considerable amount

of tannin is found in these stone cells. Also the presence of silicic acid is detected by macerating the seeds in concentrated sulfuric acid. These cells are usually  $16\mu$  wide,  $24\mu$  long.

- 3. Perisperm, containing numerous starch grains and few aleurone grains. Calcium oxalate crystals found.
- 4. Endosperm, a narrow zone surrounding the embryo. Here crystals of calcium oxalate are also noticed.

(c) Histology of the Bitter-Seeded Cardamon.—Cross sections of the Bitter Seeds upon examination present the following microscopical characteristics.

TABLE III.—HISTOLOGICAL CHARACTERISTICS OF THE BITTER CHINESE CARDAMOM.

- 1. Arillus, Is usually detached.
- 2. Spermoderm:
  - (a) Epidermis—A layer of thick-walled epidermal cells, which in cross section appear rectangular, while in surface view they are both polygonal and rectangular. Average cell measures 58μ in length and 24μ in width.
  - (b) Cross-Cell—A single layer of pigment cells with brownish tannin contents and resin-like bodies.
  - (c) Oil Cell—One-celled layer of rectangular oil cells, whose walls are slightly thicker than that of the Round China cardamom. The length of the cell is about half as broad as the width.
  - (d) Parenchyma—Occasionally indistinguishable.
  - (e) Sclerenchyma—These stone cells appear in much darker shade, dark brown; walls are excessively thickened, producing lumina of 8 to 9μ in diameter. Tannin and silicic acid are always present.
- 3. Perisperm-Consists largely of starch grains.
- 4. Endosperm—Enveloping the embryo, which is stained red with tincture of alkanna showing the presence of oil.

(d) Histochemistry or Microchemistry. 1. Presence and Location of Silicic Acid.— Silicic acid in the cardamoms is chiefly found in the sclerenchyma layer of the spermoderm. With respect to the identification of silica in cardamom as well as in other plant structures, stress should be laid upon the structure of the elements after ashing and the formation of characteristic crystals of sodium or potassium fluorsilicate upon treatment with hydrofluoric acid. However, the presence of silica in the sclerenchyma cells can also be readily detected by macerating the whole seed with concentrated sulfuric acid and after a short time treating with chromic acid solution and then washing with water and alcohol. Characteristic skeletons produced as a result of the reaction are conclusive for the test.

It should also be mentioned that both varieties of the Chinese cardamom under consideration render satisfactory results with all the tests just described; and the amount of silica found in the Bitter-Seeded is relatively greater than that of the Round variety.

2. Pigments, Tannins, etc.—Tannin occurs as a constituent of the cell-sap, and the cells containing it are the cross-cells, parenchyma cells, sclerenchyma cells of the spermoderm of the cardamom seeds. Its presence is frequently noted in making sections of the seeds with a razor; the liberated cell-sap is colored dark blue. It may also be determined by use of dilute solutions of methylene blue as proposed by Pfeffer, which colors the cell-sap blue, afterward precipitating the tannin. This reagent has the advantage that when used in very dilute solution (1-500,000) it does not injure the protoplasm of the living cells (12). Ferric chloride, ferric acetate and copper acetate are the few good reagents very frequently employed for the precipitation of tannin.

In comparing the amount of tannin present in cell structures of the Chinese cardamoms, especially in the sclerenchyma cells, the writers have found that there is also heavier precipitation of tannin in the Bitter variety.

Not infrequently, brownish resin-like bodies occur in the form of a cell-content, being present in the pigment layer (cross-cells) of the Bitter-Seeded cardamoms. Like the fixed oils, they are colored brownish or brownish black with osmic acid and are intensely colored with alkannin. 3. Volatile Oil or Flavoring Principle.—Volatile aromatic principles are found in the large clear cells of the seeds of both varieties which arise either as a metamorphosis of the cell-wall described by Tschirch as a "resinogenous layer" or merely as an indirect product connected with photosynthesis.

The volatile oil present in the Bitter cardamom is distinguished by containing the bitter substances which are characteristic of the oil as well as the seed.

The oils are colored a carmine red with very dilute solutions of fuchsin. They also readily respond to osmic acid, alkannin, sudan III and cyanin.

4. Calcium Oxalate.—In the localization of the crystals of calcium oxalate in the cardamom seeds, the polarization-microscope is distinctly of value. These crystals are chiefly found in the perisperm tissue and occasionally in the endosperm. Aside from the optical method employed, resorcin-sulfuric acid for identifying calcium oxalate gives an intense blue color with oxalic acid or calcium oxalate. The test is brought about by adding concentrated sulfuric acid to the section and then resorcin in amounts sufficient to effect the reaction (13).

5. Starch, Fixed Oil and Aleurone Grains.—The perisperm is loaded with starch grains which, when treated with chlorzinc iodine solution, is colored blue, while embryo and endosperm are unaffected by the same reagent.

The presence of fatty oil in the embryo is indicated by the fact that the latter is readily stained pink with tincture of alkanna.

Aleurone grains, mainly found in the endosperm tissue, may be detected by the use of eosin-picric acid. Take the section (oils are previously removed by alcohol, or alcohol and cther) and leave it for 15 minutes in a saturated aqueous solution of picric acid. Transfer it to alcohol. Next treat the section with rather dark aqueous eosin solution for a few minutes, followed by alcohol and then xylol. The aleurone grains are either colored red with eosin or yellow with picric acid.

### PART IV.

## CHEMICAL CHARACTERISTICS.

(a) Isolation of Oil-Steam Distillation.—The most valuable constituent of cardamom is the essential oil, of which the seeds of the Round variety yield 4 to 6 per cent and the Bitter-Seeded 2 per cent.

The oil of cardamom was first distilled in 1540 by Valerius Cordus. An oil prepared "by extraction from the seed," as suggested by the critic, would be far from satisfactory. His sample made as described, "by taking the cardamom seeds, grinding them and extracting with a solvent and evaporating the solvent," was probably not more than one-half to one-third pure essential oil, because fixed oil in this seed is more abundant than the volatile. The ordinary solvents would extract this and leave it in the residue on evaporation (14).

Steam distillation was first recommended in 1826 by H. Zeise and especially for volatile oils by Van Dijk in Utrecht who thereby materially aided in its introduction. He demonstrated that the volatile oils which were obtained by steam alone from the vegetable material, distinguished themselves from those obtained by distillation over open fire, by a lighter color and purer odor.

By the distillation of the seeds of the Chinese cardamoms with steam the essential oils in question are obtained. The exact procedure employed in isolating the oil of cardamom in the laboratory is as follows:

The seed used for analysis should remain in the husks, and always be freshly ground, as the exposed or powdered material is apt to lose strength through volatility (15). One hundred grams of the seed in the form of moderately fine powder are placed in a liter flask; 500 cc. of water are added. The mixture is macerated for 15-20 minutes and then distilled with steam long enough (3 hours) to insure complete extraction. A Florentine receiver is frequently employed to collect

# TABLE IV.—COMPARATIVE STUDY

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	Elettaria Cardamomum.	Elettaria Cardamomum B.	Chinese Round Cardamom.	Bitter-Sweet Cardamomum.	Amomum Cardamomum L.
Specific Gravity	0.923-0.944	0.895-0.906	0.965-0.975	0.985-0.987	0.905
Optical Rotation	$A_D + 24^{\circ} \text{ to } + 41^{\circ}$	+12° to +15°			+38° 4'
Congealing Point	Fraction of 150°- 164° C. (17). So- lidifies in a freez- iny mixture		Does not con- geal at -5° C.	9° C. A portion does not congeal at 3° C.	Semi-solid at ordinary tem- perature
Melting Point	M. p. cineol 112° -113°. M. p. d - a - terpineol (18) 35°-37°	A solid crystal line (20) sub- stance melts at 60°-61° — dis- tillation residue		Camphor - like crystals melt at 110°-112° C.	42° (Oil)
Refractive Index	N <sub>D20</sub> ° 1.462- 1.467		1.462	N <sub>D30</sub> ° 1.471	
Solubility	Soluble in 2-5 vols. 70% alcohol	1-2 vols. 80% al- cohol	4.5 vols. 80% alcohol	5 volumes 80% alcohol	1.2 vols. of 80% alcohol
Acid Figure	A. V. up to 4.0	25-75	1.5-3	7.5	
Saponification Figure	132	25 to 70	25-35	81-87	18.8
Ester Figure	94-150			60-62	
Percentage Vield	3%; 5%; 7% Peri- carp-0.2% (19)	4%-6%; Seed 4%; Pericarp 0.2%	5%	2%-3%	2.4% (17)
Color	Light yellow	Light yellow	Straw Yel.	Yellowish brown	
Odor	Penetrating but agreeable	Strong aromatic	Distinctly aro- matic	Pungent	Distinct odor of camphor and borneol
Taste	Camphoraceous burning taste	Pleasant cooling taste	C a m p h o r a- ceous and cool- ing	Camphoraceous and bitter	
Constituents iso- lated; their be- havior toward certain reagents	The acid constitu- ent of the ester is acetic acid form- ing. No formic, no higher fatty a c i d s present. High saponifica- tion value due to terpineol acetate from saponified oil fractionation in vacuum yielded crystallizable d-a terpineol. M. p. $35^{\circ}-37^{\circ}; AD + 81^{\circ}$ 37' in the super- cooled state. The first or lowest frac- tion cineol iden- tified by iodol compound melting at 112°-113°	Fractional por- tion between $165^{\circ}-167^{\circ} + HCl$ yield terpinene dihydrochloride melting at 52°. Terpinene nitro- site of fraction of 178° to 182° melting at 155°. Fraction of 205°- 220° + HCl a hy- drochloride melt- ing at 52° and by shaking it with conc. Hi, a hy- driodide melting at 76°-terpineol -4° (Wallach) (18)	Oil boils at 170° C.	Oil boils at 200° C. with partial decomposition acquiring a dark brown color	Saponification value after acetylization raised to 7.2 and contained alcohols 22.5% borneol. From 800 Cm. of the oil 100 Cm. of the solid com- pound were ob- tained by cen- trifugal action. They consist of equal parts of <i>d</i> -borneol and <i>d</i> -camphor (21)

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OF THE OILS OF CARDAMOM.

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Amomum Melegueta. 0.894 (20) 0.825 (2)	Amo- mum Xan- thoides.	Amo- mum Villo- sum.	Kame- roon Car- damom 0.907	Amo- mum Aromati- cum. 0.920	Galanga Carda- mom.	Ovoid China Carda- mom.	Amo- mum Augusti- folium. 0.9017	Carda- mom Root of Indo- China. 0.9066
-4° -3°58′			-20°34' -23.5° Hansel	-13°			16°50′	- 32° 57′
			1.4675				1.46911	1.48151
10 vols. of 70% alcohol			7-8 vols. 80% al- cohol 50, 107 af- ter acetyli- zation		 		6 vols. 80% al- cohol 0.4	0.5 vol. 95% alcohol 3.7
							4.2	87.9 96.7 (after a cetyliza- tion)
0.3% (2) 0.75% (17)			2.33% 1.60%	1.12%			4.5%	
Faint yellow- ish			1.60% (18)				Colorless	Lemon - yel- low
Agreeable and spicy but not characteristic		Aromatic tar-like	Odor of cineol	H i g hl y aromatic	Aroma resem- bles za- lange root	Terchin- thinate odor	Res. caju- put oil	Peculiar spicy odor
Aromatic, not acrid taste	Distin- guishable from the peculiar aromatic taste of the oil of Malabar Card a- mom (1)	Warm and bit- ter and tar-like (5)		Cam - phora- ceous	Pungent burning	Woody and ter- chin- thinate		
The oil begins to boil at 236°, the larger p or tion passing over between 257° and 258°. Analysis yielded results in agreement with the for- mula C <sub>20</sub> H <sub>HO</sub> . The oil dis- solves liodine without ex- plosion			The pe- culiar odor pre- vents its use as substi- tute for Ceylon carda- mom	Large quanti- ties of cineol				Oil contains cineol, bi- saboline and paraffin ow- ing to the presence of paraffin the residue left upon distil- lation solidi- fied at 15° C.

the distillate, and by this means the separation of the light oily layer from the accompanying water is effected. Then the distillate is saturated with salt and extracted with ether. However, in the course of operation, the cooling surface effected by the condenser causes the deposition of whitish camphor-like bodies, accompanying the distillate. They are also rendered soluble in ether and removed by the use of the same reagent.

(b) The Isolation of Oil-Extraction with Solvents.—The problem of extracting the oil from the aqueous distillate, obtained through steam distillation, requires the selection of suitable solvents. The solvent most commonly used is ether which will extract the oil very readily, without producing any turbidity. The ethereal solution is preferably dried over anhydrous sodium sulfate and the greater part of the ether is distilled off, the remaining portions being allowed to evaporate spontaneously.

Less frequently petroleum ether is used since it caused incomplete solubility, the turbidity noticed when the distillate comes in contact with petroleum ether.

Likewise, carbon disulfide is not serviceable and moreover the extract always retained some of the disagreeable odor of the solvent. Neither has carbon tetrachloride proved satisfactory.

(c) Composition of the Oil (Volatile).—The analysis of the volatile oil of cardamom is always a difficult subject because most of the constituents are liquid in character and can, therefore, be separated only by fractional distillation. But this operation is also unsatisfactory because certain constituents of the oil are not volatile without decomposition. For this and other reasons it is best to subject the oil to a preliminary examination, the results of which simplify the examination considerably. It consists primarily in the determination of the physical properties of the oil; and also in the study of the behavior of the oil toward certain group reagents, whereby the presence or absence of certain constituents can be ascertained.

Of the physical constants, the specific gravity, the optical activity, boiling point and the congealing point help one to draw conclusions as to its chemical composition.

Results of the comparative physical and chemical study of the oils of cardamom with the chief points of differentiation are tabulated in Table IV.

The oil of the Round cardamoms is light yellow in color, somewhat viscid, has a strong odor of cardamom and a characteristic pleasant cooling taste. The specific gravity lies between 0.965 and 0.975. This may indicate a mixture of compounds which in the main are suggestive of terpenes and camphor. The oil is soluble in 4-5 vols. of 80 per cent alcohol.

At a temperature somewhat below the ordinary temperature the oil of the Bitter-Seeded cardamoms (which is deeper in color than the preceeding and has a bitter taste) forms a semi-solid substance with a strong camphoraceous odor. In order to dissolve the separated crystals, the oil has to be heated to about  $40^{\circ}$  C.; at which temperature the specific gravity is 0.985 and refractive index is 1.471 (30° C.). Saponification value 87.03; acid value 7.5; soluble in 5 volumes of 80 per cent alcohol. The oil boiling at 200° C. acquires a dark brown color with partial decomposition.

The camphor-like crystalline bodies accompanying the distillate in the distillation of both varieties with steam are found to be present in much less amount in the oil of Round cardamoms than that of the Bitter-Seeded cardamoms. These crystals upon repeated purification with ether or benzol tend to melt at 110–112° C. with the aid of so-called "micromelting-point apparatus," which was used in connection with the microsublimation of plant products in the Bureau of Chemistry, Washington, D. C. (16).

#### PART V.

#### PREPARATIONS AND USES.

Cardamoms owe their properties essentially to the presence of the volatile oil, the effects of which are in the main agreeably aromatic, usually devoid of acridity. For this and other reasons, the uses of various cardamoms will more or less be collectively discussed. Cardamoms are used either on account of their flavor or for their carminative and stimulant properties. The seed or the oil is, however,

rarely prescribed alone, but frequently employed as adjuvants or correctives of cordial, tonic and purgative medicines.

In China, tonic, stomachic, cordial, pectoral and astringent properties are ascribed to the fruits of the Bitter-Seeded cardamoms, and the occasional use to which they are applied at the present time is in the treatment of urinary troubles and chronic coughs. The seeds alone are used, and are given in the form of a decoction for affections of the stomach, or as a tincture in ague, malarial or intermittent fever, catarrh, or other systemic diseases (22). But the fruits of the Round Chinese cardamom are believed to be of value in cases of "heartburn" (pyrosis), vomiting, flatulent dyspepsia. They are also reputed to be serviceable in ague and disorders arising from drunken dissipation. They are said to have been formerly much used as a condiment or spice.

Cardamoms are but little used in the United States and Great Britain, being only employed in medicine, and to a very limited extent as an ingredient in the preparation of the condiment known as Curry Powder. Hence the consumption is small in comparison with what it is in Russia, Sweden, Norway and parts of Germany, where they are constantly employed as a spice for the flavoring of cakes (23). In these countries Ceylon cardamoms are also used, but exclusively for the manufacture of liquers. But in the East Indies, besides their medicinal use, they are largely consumed as a condiment and for chewing with betel, as described under "Areca Catechu," by Bentley and Trimen.

#### U. S. P., N. F., B. P., PREPARATIONS, ETC.

Though cardamoms enter into a considerable number of pharmaceutical preparations, only two derive their names from these fruits, namely, Tinctura Cardamomi Composita and Oleum Cardamomi. Cardamom, however, constitutes an important ingredient in Pulvis Aromaticus of N. F. and Pulvis Cinnamomi Compositus of B. P.

Owing to the presence of the fixed oil, the seeds are very difficult to powder alone. Hence the practice, in preparing compound powders containing cardamom, of mixing the other ingredients with it, so that they may absorb the oil.

Among the important preparations of cardamom, the compound tincture of cardamom deserves individual consideration especially in reference to their incompatibilities. Dunning mentions the possibility of tannin in compound tincture of cardamom, causing precipitation of alkaloid strychnine (24). Likewise, McCutcheon and Alexander discuss the incompatibility of the preparation with sodium bromide, with bismuth mixtures and with alkaloidal salts (25).

#### OFFICIAL PREPARATIONS WITH CARDAMOM.

Ι.	From	the Seed	Official in		
	1.	Tinctura Cardamomi Composita	U. S. P. XI		
	2.	Tinctura Gentiana Composita	U. S. P. XI		
	3.	Pulvis Aromaticus	N. F. VI		
	4.	Pulvis Cretæ Aromaticus	N. F. VI		
	5.	Extractum Colocynthidis Compositum	N. F. VI		
	6.	Pilulæ Aloes et Myrrhæ (from Pulvis Aromaticum)	N. F. VI		
	7.	Pilulæ Hydrargyri Chloridi Mitis Compositæ (Containing Compound			
		Extract of Colocynthidis)	N. F. VI		
	8.	Oleum Cardamom	N. F. VI		
II.	Containing Volatile Oil				
	9.	Spiritus Cardamomi Compositus	N. F. VI		
	10.	Spiritus Vanillini Compositus	N. F. VI		
III.	Cont	aining Compound Spirit of Cardamom			
	11.	Elixir Cardamomi Compositum	N. F. VI		
	<b>12</b> .	Elixir Glycyrophosphatum Compositum	N. F. VI		
IV.	Cont	aining Compound Spirit of Vanillin			
	13.	Elixir Vanillin Compositum	N. F. VI		
	14.	Elixir of Barbital	N. F. VI		

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V. Containing the Compound Tincture of Cardamom

15.	Elixir Euphorbíæ Compositum	N. F. VI
16.	Elixir Gentianæ	N. F. VI
17.	Elixir Gentianæ et Ferri	N. F. VI
18.	Elixir Gentianæ Glycerinatum	N. F. VI
19.	Elixir Glycerophosphatum Compositum	N. F. VI
<b>2</b> 0.	Elixir Taraxaci Compositum	N. F. VI
21.	Elixir Viburni Opuli Compositum	N. F. VI
22.	Elixir Viburni Prunifolia	N. F. VI

#### SUMMARY.

1. The close relationship of fruits obtained from Elettaria Cardamomum, yielding the official Malabar cardamoms, and those from several Amomum species appears well established as a result of the survey, based on literary, morphological and chemical evidence.

2. The character and percentage yield of volatile oil obtained from the Round Chinese cardamom suggest that the fruit might well be used as a substitute of, if not an equal to, the fruit of the official Malabar cardamom.

3. The Bitter Chinese cardamom, while chemically closely related to the other cardamoms, contains a bitter crystalline camphor, melting at 110-112° C.

The anatomical and chemical data obtained should greatly facilitate the identification of cardamom fruits, whether obtained from Elettaria or Amomum species.

5. The various differences observed between Elettaria Cardamomum and Amomum species appear not marked enough to justify separation in different genera.

6. Attention is called to new sources of supplies, both for condimental and medicinal uses; these supplies can be readily obtained through more extensive importations or possibly through domestic cultivation.

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## ASSAY OF SPIRIT OF CAMPHOR.\*

#### BY SAMUEL W. GOLDSTEIN AND WILLIAM F. REINDOLLAR.<sup>1</sup>

The assay of Spirit of Camphor official in the United States Pharmacopœia XI has been found very unsatisfactory in this Laboratory. The first supplement to the United States Pharmacopœia XI provides for the use of a 5-cc. sample, and it has been suggested that a further reduction of the amount of sample to 2 cc. is advisable (1). The official method involves treatment of the sample with solution of dinitrophenylhydrazine, refluxing on a water-bath for four hours, diluting and setting aside for twenty-four hours, collecting, drying and weighing the precipitate, and multiplying the resulting weight by a factor. We have found that the above reaction is not stoichiometric, the reagent used is exceedingly unstable, the procedure requires at least twenty-nine hours, and the results are unreliable.

The method recommended by Randall (2) is based upon precipitation of the camphor by an acidified aqueous calcium chloride solution in a Babcock bottle, dissolving the camphor in 1 cc. of benzin and, after centrifuging, noting the increase in volume, which when multiplied by a factor gives the per cent w/v of camphor in the spirit. This method, in the hands of one of us, gave unsatisfactory results, owing principally to the volatility of the benzin; hence a modified procedure has been developed.

#### EXPERIMENTAL.

Two samples of Spirit of Camphor were prepared by dissolving 9.7905 Gm. of natural camphor (A) and 9.8359 Gm. of synthetic camphor (B) in enough alcohol to make 100 cc. Results obtained following the U. S. P. XI procedure are given in Table I.

TABLE I.—U. S. P. XI PROCEDURE.					
Sample.	Amt. Used in Cc.	Gm. Camphor per 100 Cc.	Results Calcd. to 10% Spirit		
Α	5	5.84	5,9 <b>2</b>		
В	5	7.20	7.32		
В	5	5.58	5.67		
В	2	9.30	9.46		
В	2	9.30	9.46		
В	2	9.50	9.65		
В	2	10.05	10.21		
В	2	9.00	9.15		
в	2	9.15	9.30		
В	2	9.20	9.45		

\* Scientific Section, A. PH. A., New York meeting, 1937.

<sup>1</sup> From the laboratories of the Bureau of Chemistry of the State of Maryland Department of Health.