palm fiber oil and expression palm oil are quite similar so the carotene content and its biological value may be assumed to be the same. The carotene of palm oil contains about 60-65% of beta-carotene and 35-40% of alpha-carotene, besides a variable quantity of lycopene and gamma-carotene.

Table I shows the carotene content of the various sources including palm fiber oil.

Palm fibers represent important tonnages. In the oil mills of French Africa 800 to 1,000 tons of fibers remain after the production of 1,000 tons of expression oil. These contain 15% of water and 10% of oil. The new mills, in the Ivory Coast, Dahomey, and the Cameroons alone, have a potential yearly output of nearly 5 tons of carotene pigment. If the total available palm fibers were processed, the carotene thus produced would be an important asset to the food and pharmaceutical industries.

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Tung Oil Review, 1951-1952

RALPH W. PLANCK, Southern Regional Research Laboratory,[1] New Orleans, Louisiana

DURING 1951 and 1952 more than 250 books, articles, and patents dealing with the chemistry and technology of tung oil and other tung products appeared in the technical and trade literature. The pertinent information culled from these numerous sources comprises this review.

General

The first comprehensive book on tung products, Fonrobert's "Das Holzöl" (70), was published in 1951. It covers the source, extraction, trade, properties, chemistry, testing, and uses of tung oil. A comprehensive "Abstract Bibliography of the Chemistry and Technology of Tung Products, 1875-1950" (160), which appeared in 1952, contains nearly 3,000 abstracts of articles and patents dealing with all phases of the tung industry.

Botany

Aleurites fordii, the tung oil tree grown in the United States, has been compared with *A. montana* and other related species grown abroad in regard to morphology of its parts, growth characteristics, and distribution (42, 54).

Culture of the Tung Tree

Recommended procedures for use in solving the numerous problems involved in tung culture are presented in Farmers' Bulletin No. 2031, "Tung Production" (168) by Potter and Crane, and in the new book "Successful Tung Farming" (27) by Beebe and Greer. For the farmer interested in establishing a tung orchard there appeared articles on land selection and preparation (59, 166), selection of planting stock (42, 141-2, 165), transplanting seedlings (5, 108, 149), and studies on plant growth (80, 150, 181), and diseases (32, 175). Numerous reports were published on fertilizing with balanced proportions of N, P, and K (58, 162, 183), with liquid ammonia (8, 73, 109, 118), with lime (56), and with the minor elements (57-8, 78, 127, 143, 179-80). Related to fertilizing are the articles published on leaf analysis (58, 148) and the practice of planting cover crops (117, 151, 167). Good farm management practices have been discussed (46, 59, 103, 117, 151, 164, 167) and measures for minimizing frost damage described (21, 155, 166, 183). Potter (163) reviewed 13 years of research on tung culture by the U. S. Department of Agriculture. Other writers have described tung production in Argentina (11, 12, 136), Brazil (9, 11, 136), Paraguay (136-7), Nyasaland (91, 107), Madagascar (33, 66, 82), Portuguese Africa (46), and still other countries (28, 36, 42, 172).

Tung Fruit

A mechanical harvester (picker) has been improved by Jezek (6, 26). Tung fruit have been analyzed, using a modified Hamilton-Gilbert method wherein the oil is extracted from ground fruit in a Waring Blendor (79). Fruit grown in the United States in 1951 was found to be abnormally low in oil content by 1-2% (169). This phenomenon was attributed to unfavorable weather conditions. For the determination of moisture in tung fruit, kernels, and hulls, the Seed and Meal Analysis Committee (134) of the American Oil Chemists' Society recommended that the forced-draft oven method (Ad 2-48) (17) be adopted as an official method. However Holmes *et al.* (100) considered drying in a vacuum oven to be the most accurate of the six methods they compared for determining moisture in tung fruit and seed. Moisture contents determined in the vacuum oven were found to be 0.4-0.8 percentage units higher than those measured in a forced-draft oven. The Seed and Meal Analysis Committee also recommended that present tentative methods (17) for sampling tung fruit and for determining oil in whole fruit and in kernels be made official methods. Equilibrium moisture contents at 25°C. were determined on tung fruit and its components at various relative humidities (99). Cutting (50) found that efficient drying of tung seed is a prerequisite for efficient oil expression. Detailed analyses for organic and inorganic constituents were made on tung fruit and press cake (101). McKinney (133) has reviewed work done at the U. S. Tung Oil Laboratories at Bogalusa, Louisiana (active) and at Gainesville, Florida, (closed) on drying, analysis, storage, pressing, and solvent extraction of tung fruit and on the composition and utilization of tung oil and its by-products, hulls and press cake. Medical reports have been published giving symptoms and treatment of people poisoned by eating tung fruit (22-4, 74). Hexane extraction of kernels to give shell-free meal for toxicity and detoxification studies was reported (145).

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[1] One of the laboratories of the Bureau of Agricultural and Industrial Chemistry, Agricultural Research Administration, U. S. Department of Agriculture.

Oil

Two German books, one by Fonrobert (70) and one by Fritz (72), were published in 1951 on the chemistry, processing, and uses of tung oil. Less detailed information on tung oil is found in Jordan's "Oils for the Paint Industry" (106) and in Garoglio's Spanish oil treatise (75).

Test Methods. Several improvements have been made in the testing of tung oil. The total unsaturation present in the oil can be measured by catalytic hydrogenation in acetic acid at atmospheric pressure (156). Dupin (60) obtained an iodine value of 230 for a Chinese tung oil, using the method of Klee and Benham (122), but Sacco Neto (176) got low values (164-171), using Wijs reagent with mercuric acetate. Reaction of tung oil with p-benzoquinone (130-31) gave diene values slightly higher than did reaction with maleic anhydride. Spectrophotometric methods for determining polyunsaturated fatty acids have been reviewed (140). A new method (94) for detecting adulteration in tung oil obtained from either *Aleurites fordii* or *A. montana* depends on crystallizing the oil under test from petroleum ether at —70°C. and determining the eleostearic acid content of the fatty acids in the glycerides in the filtrate. Another specific test (123) for adulteration of edible oils with tung oil is based on the gelling produced by treatment with a mixture of sulfuric acid, nitric acid, and ferric chloride. Paper chromatography (111-2) and displacement analysis techniques (97-8) have been improved to permit the detection and determination of eleostearic acid in the presence of mixtures containing other fatty acids. In one technique (111) the acids on the paper were reacted with inorganic salts and then with organic dyes. In another technique (112) conjugated acids (*e.g.*, eleostearic acids) were separated and distinguished from non-conjugated ones by reaction with maleic anhydride followed by development of the paper chromatogram with petroleum ether. Treatment of methyl esters of tung oil acids with aqueous solutions of urea yielded crystalline clathrate compounds of the less unsaturated esters and a solution of esters having a higher iodine value than the starting material (152).

Composition. Disagreements still exist as to the fatty acid composition of tung oils. For the first time lauric acid (0.2%) and myristic acid (0.5%) have been reported in a tung oil (Formosan) which also was said to contain: eleostearic acid, 70%; oleic acid, 15.5%; stearic acid, 2.2%; palmitic acid, 3.0% (43). A Brazilian tung oil was reported to contain: eleostearic acid, 90.6%; linoleic acid, 0.6%; oleic acid, 4.0%; saturated acids, 4.6% (9). The composition of typical *Aleurites fordii* oils from Madagascar was given as linoleic acid, 0-10%; oleic acid, 1-5%; palmitic acid, 5% (84). These values may be compared with previous compositions reported by Hilditch and Riley (95), namely: eleostearic, 82%; linoleic acid, 8.5%; oleic acid, 4.0%; palmitic acid 5.5%. Proportions of mono-, di-, and trieleostearic glycerides in tung oil were reported also (93).

Wheeler (201) concluded from infrared measurements that alpha-eleostearic acid is 9-cis, 11-trans, 13-trans, or 9-trans, 11-trans, 13-cis-octadecatrienoic acid and that beta-eleostearic acid is 9-trans, 11-trans, 13-trans-octadecatrienoic acid.

Properties. Beta (isomerized) tung oil was found (198) to exist in three crystalline forms, depending upon the rate of cooling. These melted at 52.8, 44, and 28°C., respectively. Heats of fusion, specific heats, entropies, and heat contents were reported for both alpha (normal) and beta tung oils. Another study (182) gave expansibilities and melting dilation for solid alpha tung oil and for the three forms of solid beta tung oil, also expansibilities and absolute densities for the liquid oils. Isomerization to the beta form caused a volume decrease of 4%. Cutting (51) has found that free fatty acids produce a greater reduction of gelation time with *A. montana* than with *A. fordii* oil. It has been suggested (51, 93, 107) that gelation is controlled by the content of trieleostearin and of free fatty acids, the effect of the two being opposed.

Reactions. The older work of Böeseken and others on the hydrogenation of tung oil was reviewed by Waterman (199) in "The Hydrogenation of Fatty Oils." Thompson (189) followed the changes in composition that occurred when a mixture of tung and linseed oils was progressively hydrogenated. It is now established (90) that vaccenic acid is one of 5 or more positional isomers formed when tung oil is two-thirds hydrogenated. Biochemical reduction of eleostearic acid was demonstrated by feeding tung oil to laying hens, thereby producing deposition of both alpha-eleostearic acid and a newly synthesized dienoic acid in both the neutral fat and in the phosphatides of eggs laid soon after ingestion of the oil (170). Styrene has been shown to react with tung oil and with eleostearic acids simultaneously by Diels-Alder addition and by free-radical copolymerization to give a mixture of products (19, 37, 158). Styrenated tung oil dries to clearer films than does styrenated isomerized tung oil indicating an influence of cis-trans configuration on the rate of copolymerization of the oil with styrene (65). Ricinus lipase splits tung oil into glycerine and fatty acids (102).

Oxidation, Polymerization, Film Formation. When air is blown over the surface of heated tung oil-linseed oil mixtures, oxygen is absorbed, and this is said to promote cyclization and polymerization (177). The oxidation of tung oil is reportedly accelerated by exposing the oil to air containing ketene vapors (116). Studies have been made on the bulk oxidation of tung and other oils (92), on the oxygen absorption by methyl eleostearate (15), and on the differences in the oxidation process occurring with conjugated and isolated double bond systems (191). Both autoxidation and thermal polymerization were reviewed by Jordan (107). The drying time of tung oil is reduced from 8 to 1.5 hours by refining the oil with aluminum oxide and to 2.5 hours with silica gel (114). A straight-line relationship exists between the time tung oil is bodied at constant temperature and the logarithm of the viscosity of the oil (104). Tung oil is said to become bodied by exposure to high-frequency electric fields (69). Vacuum distillation removed free fatty acids and glycerides of unchanged monomeric fatty acids from stand oils, but no significant improvement in the quality of the stand oil was found (129). Free radicals like triphenyl methyl cause tung oil to dry rapidly (115). Kaufmann and Strüber (113) discussed the factors that cause films to dry to a wrinkled finish. Gas-proofing to prevent wrinkling can be accomplished by heating tung oil with disulfides (47) such as those naturally found in kerosene or by polymerizing the oil with styrene in the presence of certain terpenes (196). The addition of peroxides to core-bonding materials containing tung oil increases the rate of curing of molds containing those materials (147). The gelation of tung oil by heat (124) and by ferric chloride (125) has been studied. Gels formed by ferric chloride can be made soluble by fusion with metallic salts (126).

The Federation of Paint and Varnish Production Clubs inaugurated a program for the study in several laboratories of the formation, properties, and deterioration of films formed by pure compounds. One phase (44) of this program revealed that eleostearin and pentaerythritol eleostearate in thin layers rapidly absorb oxygen, part of which is used to build up the peroxide content to a maximum of 0.3 moles per mole of eleostearic acid. Another report (40) showed that films of trieleostearin absorbed more oxygen than trilinolenin films, acquiring an oxygen content of about 30% in 1-30 days depending on film thickness. The shrinkage (17-18%) of trieleostearin films during drying was comparable to that of trilinolenin films (39). Stress strain measurements (63) showed that there was less change of distensibility on aging for trieleostearin films than for trilinolenin films, and less for pentaerythritol eleostearate films than for the corresponding linolenic ester. Hardness, abrasion resistance, and accelerated weathering tests (110) were made on trieleostearin, pentaerythritol eleostearate, and glycerol-eleostearate alkyds. Moore (146) correlated the properties of alkyd films with the composition of oils used to modify the resins and has shown that tung oil contributes both yellowing and hardness.

Uses and New Compositions. The use of tung oil with soybean oil is discussed in Markley's "Soybeans and Soybean Products" (135), and its use with rubberseed oil is described in Jordan's book (106). Results have been published on the use of tung oil for the modification (83, 186) of alkyd resins and the manufacture of oleoresinous varnishes (41, 62, 86). Flexible wrinkle finishes (197) are obtained by tung oil modification of alkyd resins made with non- or semi-drying oils. Another wrinkle coating is made with carbic anhydride (adduct of maleic anhydride and cyclopentadiene), tung or another drying oil, and enough glycerin and diethylene glycol to give a 10-25% excess of hydroxyl groups (144). A varnish base has been made by reacting tung oil acids with a reaction product of soybean oil acids, pentaerythritol, maleic anhydride, and chlorostyrene (174). Interpolymerization of acylic mono- and di-olefines with tung oil in the presence of Friedel-Craft catalysts yields a synthetic drying oil of good quality (195).

Cobodying tung oil with a hydrocarbon drying oil and condensing the product with aldehydes or ketones yields resinous or elastomeric materials (76). Unusual products are obtained by reacting tung oil with haloölefine and then replacing the halogen with other polar groups (178). Thiophene-aldehyde resins modified with tung oil also make coating compositions (25). Tung oil (or its acids or their esters) will condense with acrolein on treatment with 0.01-5.0% of aluminum isopropoxide (202). When drying oils, such as tung oil, are heated with the reaction product of furfural and mesityl oxide, the products are useful in brake linings, coating compositions, etc. (89). Laminating resins are produced by condensing tung oil and phenol with formaldehyde in aqueous solutions (157). An unusual film-forming material results from condensing tung oil with phenol and then treating the product with a polyepoxide formed by reaction of glycerine with epichlorohydrin (85). Combinations of styrenated alkyd resins (which may contain tung oil) with vinyl esters of tall oil acids yield clear and durable films (139). Another new product is copolymer of styrene with polyesters of tung oil acids esterified with glyceryl polyethers of dihydric phenols (188). New also are copolymers of styrene with acrylic esters of unsaturated alcohols produced by sodium-alcohol reduction of drying oil (*e.g.*, tung oil) acids (88).

Procedures have been given for the copolymerization of styrene with tung oil alone (35, 87, 154), or with modifying agents such as diallyl succinate (161) and polyallyl ethers (171). Tung oil has been reacted successively with polystyrene (or a related polymer), a polyhydric alcohol and a polybasic acid (31). Difficulties due to the incompatability of styrenated oils with other varnish constituents are overcome by using styrenated oils with styrene oxide-phenolic resin-tung oil solutions (138). A heat-bodiable drying resin is obtained by interpolymerizing butadiene and isobutylene with $AlCl_3$ in propane and then transferring the polymer into a drying oil like tung oil (190). Tung oil has been copolymerized also with a cyclic dimer of butadiene or piperylene (81), with ethylene (30), with cyclopentadiene (77, 154), and with vinyl type monomers (77, 154). The hydrogenation of tung oil in the vapor phase was patented, and the apparatus needed for the purpose was described (96). New esters were formed when ethylene was copolymerized with vinyl acetate or carbon monoxide and the polymer first hydrolyzed and then esterified with tung and other drying oil acids (200). Polyvinyl esters of tung oil acids are prepared by interesterifying polyvinyl alcohol with the methyl esters of the acids while distilling off the methanol formed (61). Excellent drying properties are claimed for the ester of polyallyl alcohol and eleostearic acid (14) and for eleostearyl chloride (128). Partial esters of eleostearic acids can be prepared by treating tung oil with glycerine and fractionating the products with a mixture of polar and non-polar solvents (205). Polyamide resins of unusual solubility characteristics result from condensing diamines with dimerized tung oil acids (34).

Dispersion of a tung oil varnish in a solution of a thermosetting resin (*e.g.*, phenolic resin) gives a new type of wood filler (38). Tung oil with a partially hydrolyzed vinyl acetate-chloride resin is said to be useful in the manufacture of linoleum (187). Tung oil can be modified by the addition of alkenyl ethers, such as allyl sucrose (68), or used to modify tall oil-sorbitol varnishes (71). Coe (45) claimed the use of tung oil for shrinkproofing wool. Danzig (52) has patented the use of eleostearic acid as a short-stop after the polymerization of vinyl chloride. Tung oil has now entered the cosmetic and pharmaceutical field with the marketing of a tung oil skin lotion for which remarkable healing properties are reported (53).

Substitutes. Kamala oil, obtained from the seeds of *Mallotus philippinensis* in India, has been called a threat to the supreme position now held by tung oil because it is reported (13) to be comparable to tung oil in its speed of drying and to gel in 9.5 minutes in the Browne heat test. Other tung oil substitutes (64) of current interest are dehydrated castor oil (105) and catalytically conjugated linseed oil (29).

Economics

Tung oil has been scarce during 1951 and 1952 because the military conflict in Korea caused the shipment of the oil from China to the United States to be embargoed in December, 1950, both by Communist China (2, 3) and by the United States (2). As a result, prices of the oil (drums, carlots, f.o.b. New York) rose from 26.4 cents per pound in November, 1950, the last "normal" month, to a high of 43.4 in February, 1952, and then dropped again to 34.2 cents in December, 1952. In the same period consumption in the United States (192) dropped from 7.7 million pounds in November, 1950, to as little as 3.6 million pounds in December, 1951, and July, 1952, and was about five million pounds in December, 1952. Production of tung oil in the United States was low (11 million pounds) in 1951 due to frost damage (48, 162) but reached an all-time high (est. 30 million pounds) in 1952 (193). This high domestic production is desired from a national security stand point (18, 48).

Economic studies have analyzed costs and practices involved in orchard establishment (132), fruit production (132), oil extraction and marketing (120, 194), and pointed out the competition existing between tung and other oils (119, 121). Three tung oil cooperatives in the U. S. mill tung fruit (67). Government aid to the tung industry has included research programs (7, 16, 55, 133, 159, 163), price supports (1, 2, 49, 120), production loans, and publication of statistics (7, 203). Plans for crop insurance have been discussed (185). Tariffs (10) and import quotas (153) also have been requested by tung growers.

Foreign Production and Imports into the United States

Production of tung oil has continued to increase significantly in Argentina (12, 120), Brazil (9, 11), Paraguay (11, 136-7), Nyasaland (91), and Madagascar (33, 66, 82) and to a lesser extent in other countries (204). Since the embargo terminated imports from China, all major incoming shipments of tung oil have come from South America (12).

The culture of the *Aleurites montana* tree has achieved commercial importance in Portugese East Africa (47), where, as in Nyasaland (91, 107), the climate is not satisfactory for raising the *A. fordii* species grown in the Americas. Chavancy (42) described the botanical characteristics and the culture of *A. montana*. Jordan (107) and Chavancy (42) compared the compositions, properties, and relative technical values of the two kinds of tung oil. Cutting (50) has described the harvesting and extraction procedures practiced on *A. montana* fruit in Nyasaland.

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