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THE COMPOSITION OF THE GUM EXUDATES FROM SOME *Combretum* SPECIES; THE BOTANICAL NOMENCLATURE AND SYSTEMATICS OF THE *Combretaceae**†

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ABSTRACT

An analytical study has been made of gum specimens from *Combretum collinum*, *C. collinum* subsp. *hypopilinum*, *C. erythrophyllum*, *C. nigricans*, *C. fragrans*, and *C. glutinosum*. In comparison with the genus *Acacia*, both the botanical classification and synonymy, and the chemistry, are more complex. Glucuronic acid, galacturonic acid, 4-*O*-methylglucuronic acid, galactose, arabinose, and rhamnose are present in all the specimens studied. In addition, mannose and xylose are present in the gum from *C. collinum*, which is therefore similar to *C. hartmannianum*. The data now available suggest that, as a group, the genus *Combretum* produces gum exudates that are more viscous, of higher molecular weight, and more acidic than those of the *Acacia* group.

INTRODUCTION

The Sahelian droughts of 1972–1974 devastated the natural and cultivated stands of *Acacias* which provided the bulk of the world's commercial supplies of gum arabic. The *Combretum* forests of Africa were not affected to such a great extent, and consequently there has been a great deal of industrial interest in the possible utilisation of *Combretum* gum.

Botanically, the genus *Combretum* is complex: much of the botanical data is contained in remote journals, and therefore an Appendix to this paper summarises the essential details of the botanical relationship between the 20 different genera of the family COMBRETACEAE R. Br., as given in the revision by Exell and Stace[2]; this revision involves an organisation into 2 sub-families, 2 tribes, and 3 sub-tribes, which are further sub-divided extensively.

In comparison with the *Acacia* genus, where considerable work has been carried out in the field of chemical taxonomy, relatively little is known about gums from the *Combretaceae*, apart from the following species: *Anogeissus latifolia*[3–7]; *Anogeissus schimperi* Hochst. (syn. *A. leiocarpus* (DC.) Guill. et Perr.)[8–12]; *Com-*

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bretum verticillatum[13] (syn. *C. collinum* Fresen. subsp. *hypopilinum* (Diels) Okafor); *Combretum leonense* Engl. and Diels[14,15] (syn. *C. glutinosum* Perr. ex DC.); *Terminalia tomentosa*[16]; *Terminalia sericea* and *T. superba*[17]; and *Combretum hartmannianum*[1].

Anogeissus latifolia is one of the main components of gum ghatti, which is of considerable commercial importance, and as *T. sericea*[17], *T. superba*[17], and *C. harmannianum*[1] gums have colloidal properties of possible industrial interest, analytical studies of gum specimens from the *Combretum* species listed below have been made.

EXPERIMENTAL AND RESULTS

Origin of gum specimens. — (*a*) *C. fragrans* F. Hoffm., syns. *C. kilossanum* Engl. and Diels, *C. ghasalense* Engl. and Diels, *C. ternifolium* Engl. and Diels, and *C. tetraphyllum* Diels (Sect. GLABRIPETALA). Sample *A*, small, dark nodules collected in September 1965 by Mr. J. H. Dick, Regional Forest Officer, at Isenegaza Public Lands, Tabora, Tanzania. Sample *B*, dark, large nodules collected in May 1970 by Mr. A. G. Sief-el-Din, Gum Research Officer, at Rashad, Republic of the Sudan.

(*b*) *C. nigricans Lepr. ex Guill. et Perr. var. elliottii (Engl. and Diels) Aubrèv.* (Sect. CILIATIPETALA). Large, very dark nodules collected in 1955 by Mr. Oseni, Botanist at Western Region Department of Forestry, Ibadan, and placed at our disposal by (the late) Professor R. J. McIlroy.

(*c*) *C. glutinosum* Perr. ex DC., syn. *C. leonense* Engl. and Diels (Sect. GLABRIPETALA). Small, dark brown or yellow nodules collected in April 1947 at Bauchi, Northern Nigeria, for Mr. A. G. Kenyon, Tropical Products Institute, London.

(*d*) *C. erythrophyllum (Burch.) Sond.*, syns. *C. glomeruliflorum* Sond., *C. riparium* Sond., *C. sonderi* Gerr. ex Sond., *C. ligustrifolium* Engl. and Diels ex Bak. f., *C. lydenburgianum* Engl. and Diels, and *C. salicifolium* sensu Monro. (Sect. ANGUSTIMARGINATA). Collected in October 1970 by Mr. T. Gordon at Audley End Farm, Darwendale, nr. Salisbury, Rhodesia.

(*e*) *C. collinum Fresen.*, syns. *C. binderanum* Kotschy, *C. mechowianum* O. Hoffm., *C. laeteviride* Engl. and Gilg., *C. cognatum* Diels, *C. bajonense* Sim, *C. gazense* Swynnerton and Bak. f., *C. junodii* Dummer, *C. album* De Wild., *C. angustilanceolatum* Engl., *C. griseiflorum* S. Moore, *C. Milleranum* Burtt, *C. tophamii* Exell ex Burtt Davy and Hoyle, *C. abercomense* Exell, *C burtii* Exell, and *C. eylesii* Exell (there are other less well-known synonyms, and also 11 subspecies) (Sect. METALLICUM). Medium-size, dark nodules collected by Mr. J. H. Dick, Regional Forest Officer, in September 1965 at Isenegaza Public Lands, Tabora, Tanzania.

(*f*) *C. collinum Fresen. subsp. hypopilinum (Diels) Okafor*, syns. *C. verticillatum* Engl., *C. hypopilinum* Diels, *C. kattoense* Exell, and *C. flaviflorum* Exell (Sect. METALLICUM). Collected in 1955 for (the late) Professor R. J. McIlroy by Mr. Oseni, Botanist at Western Region Department of Forestry, Ibadan. A preliminary

TABLE I

ANALYTICAL DATA FOR *Combretum* SPECIES

	C. fragrans *Sample* A	C. fragrans *Sample* B	C. glutinosum	C. nigricans	C. erythro-phyllum	C. collinum	C. collinum *subsp.* hypopilinum	C. hartman-nianum
Data for crude gum								
Moisture, %	13.5	13.1	13.0	10.6	12.0	9.8	15.7	11.9
Ash, %[a]	7.0	7.7	3.5	3.0	3.6	4.1	13.5	3.8
Nitrogen, %[a]	0.27	0.27	0.25	0.35	0.12	0.11	0.09	0.64
Hence, protein, % ($N \times 6.25$)[a]	1.7	1.7	1.6	2.2	0.8	0.7	0.6	4.0
Acetyl, %[b]	4.4	2.5	4.2	2.5	1.7	0.3	0.5	0.4
Data for purified gum								
Recovery from crude gum, %	79	65	n.d.	80	78	73	87	83
Moisture, %	12.5	12.0	11.9	9.1	10.6	8.9	13.8	8.3
Ash, %[a]	7.7	8.3	3.3	3.1	5.7	3.3	11.7	3.7
Nitrogen, %[a]	0.17	0.18	0.07	0.10	0.11	0.13	0.10	0.61
Hence, protein, % ($N \times 6.25$)[a]	1.1	1.1	0.4	0.6	0.7	0.8	0.6	3.8
Methoxyl, %[b]	0.85	1.07	0.58	0.24	0.33	0.45	1.46	0.25
$[\alpha]_D$ in water, degrees[b]	+35	+41	−9	−43	−54	−81	+53	−35
Intrinsic viscosity, $[\eta]$, $ml.g^{-1}$[a]	162	170	75	35	110	312	60	63
Molecular weight, $M_w \times 10^5$[a]	17	49	5.3	4.8	14	116	7.3	6.4
Equivalent weight[b]	505	487	1073	1244	745	1405	398	1173
Hence, uronic anhydride[b,c]	35.0	36.2	16.4	14.0	23.6	12.5	44.3	15.0
Sugar composition[b] *after hydrolysis*								
4-*O*-Methylglucuronic acid[d]	5.1	6.4	3.5	1.4	2.0	2.7	8.8	1.5
Glucuronic acid	19.7	19.8	8.9	7.7	13.9	6.7	23.9	7.4
Galacturonic acid	10.2	10.0	4.0	4.9	7.7	3.1	11.6	6.1
Galactose	34	34	40	30	27	22	36	22
Arabinose	14	16	31	41	33	47	5	43
Rhamnose	15	14	13	15	16	8	13	4
Mannose	2	—	—	—	—	9	1	10
Xylose	trace	trace	sl. trace	trace	trace	3	1	6

[a]Corrected for moisture content. [b]Corrected for moisture and protein content. [c]If all acidity arises from uronic acid. [d]If all methoxyl groups located in this acid.

examination of this gum was published[13] by Professor McIlroy, who generously placed the original Fraction *A* at our disposal.

Purification of gum samples. — The gum nodules were crushed lightly and ground finely. Samples (3 g) from each species were treated with cold water (300 ml); after 48 h, the gums had dissolved completely, except for those from *C. collinum*, *C. fragrans* (sample *B*), and *C. nigricans*, which required the addition[18] of sodium borohydride (0.5 g) to facilitate dissolution. The solutions given by *C. fragrans* and *C. collinum* gums were so viscous that dilution had to be effected to assist complete dissolution. The solutions (all at pH ~4.5) were filtered through muslin to remove bark and insoluble debris, and then through Whatman Nos. 41, 1, and 544 papers. After dialysis against running tap-water for 48 h (96 h for the borohydride-treated samples) and distilled water for 24 h, the gum samples were obtained as the freeze-dried products (yields recorded in Table I).

Analytical methods. — The standard analytical methods described[19,20] previously were used for the determination of moisture, ash, nitrogen, methoxyl, acetyl, equivalent weight, specific rotation, limiting viscosity number, and molecular weight. The chromatographic systems and the hydrolysis conditions used to determine the relative proportions of both neutral and acidic sugars were as described[1] recently for the study of *C. hartmannianum* gum.

The analytical data obtained are shown in Table I; the data for *C. hartmannianum* gum are included to facilitate comparisons between the species yielding mannose and xylose in addition to galactose, arabinose, and rhamnose.

DISCUSSION

The two specimens of gum from *C. fragrans*, from widely differing geographical locations, are closely similar, although the Sudanese sample is of considerably higher molecular weight. In contrast, the gum from *C. collinum* differs extensively from that of its subspecies *hypopilinum*. Although few data are available to date, recognised botanical subspecies can be very distinct, both in terms of their external morphological characters and the chemical composition of their gum exudates[21]. The specific rotation for *C. collinum* subsp. *hypopilinum* agrees with the value published by McIlroy[13], although he reported that the gum contained only galactose, arabinose, and glucuronic acid. The data for *C. glutinosum* agree well with the average of the values reported for several nodules of gum identified by the synonym *C. leonense*[14]. Botanically, *C. glutinosum* is a very variable species with many subspecies, varieties, and forms[22]. The extensive synonymy noted above indicates that the classical botanists had difficulty in recognising good new species, and the remarks of experts quoted in the Appendix indicate that *Combretum* gums must be expected to originate from complex, heterogeneous populations. The botanical sections METALLICUM, GLABRIPETALA, CILIATIPETALA, and ANGUSTIMARGINATA are all placed within the sub-genus *Combretum* of the genus *Combretum* Leofl.

The analytical data show many interesting features. The nitrogen contents are low, with the exception of *C. hartmannianum*. Methoxyl contents are variable, but sufficiently high for 4-*O*-methylglucuronic acid to be readily detectable. Optical rotations range from $-81°$ to $+53°$; the former value, for *C. collinum*, appears to be the highest negative rotation recorded for a gum exudate. The uronic acid content also varies over a wide range, from values that occur commonly within the *Acacia* group (12–16%) to very high values (36.2, 44.3%) that are comparable to those recorded for *Khaya*[23] and *Sterculia*[24] spp. There appears to be a reasonable correlation between the ash content of the purified gums and their uronic acid contents, as is typical in the genus *Acacia*.

Perhaps the most interesting feature, however, concerns the variable, but high, values of limiting viscosity number shown by the gums from these species. The value for the least viscous gum, *C. nigricans*, is much higher than typical values for *Acacia* gums ($\eta = 4\text{–}24\ \text{ml.g}^{-1}$); the exceptional viscosity given by *C. fragrans* and *C. collinum* resembled that of a gum karaya or gum tragacanth. These high viscosities are reflected in high molecular weights; the molecular weight of *C. collinum* gum (11.6×10^6) is the highest observed in this laboratory to date for a gum exudate.

Of the species studied, only the gums from *C. hartmannianum*, *C. collinum*, and its subspecies *hypopilinum* contain significant, if small, quantities of mannose and xylose. *C. hartmannianum* is placed, together with *C. fragrans* and *C. glutinosum*, in section GLABRIPETALA; *C. collinum* and its subspecies *hypopilinum* are placed by Exell and Stace[3] in section METALLICUM, although this is a synonym, *pro parte*, of GLABRIPETALA. It will therefore be of interest in future studies of other *Combretum* species to ascertain whether the presence or absence of xylose and mannose, and other structural features of the gums, have any correlation with the present botanical classifications. Attempts to establish the main structural features of the gums from *C. erythrophyllum* and *C. nigricans* are in progress.

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APPENDIX

Botanical nomenclature of the genus Combretum

The genus *Combretum* Loefl. is the largest in the Family COMBRETACEAE (order, MYRTALES); it is cosmopolitan in the tropics and sub-tropics, although absent[1] from Australia and the Pacific Islands. The naming of species in *Combretum* has long been recognised[2] as a difficult problem, especially in the absence of flowers. Over 600 specific names are now known[1] to have been used to represent some 250 actual species; consequently, the synonymy is often unusually extensive (see origin of samples described above). The greatest range of structure and most of the difficult taxonomic problems are found in Africa, where ~180 of the known species occur[1]; there are ~30 Asian species[3].

There were several attempts at taxonomic classification in the nineteenth century. Engler and Diels[4] divided the species on a world-wide basis into 55 sections in 1899; their classification still forms the basis for modern revisions, such as Exell's account[2] of the American species. A revision by Exell and Stace[5] in 1966 grouped Engler and Diel's sections, and 10 sections added by other authors, into subgenera, the major of which are subgenus *Combretum*, subgenus *Cacoucia*, and subgenus *Apetalanthum* (Table II). In 1969, Exell and Stace[6] described 3 new sections (all African species) of the sub-genus *Combretum*; this brought the total number of sections in the genus to 68, of which 22 in subgenus *Combretum* and 5 in subgenus *Cacoucia* are recognised in Africa.

These taxonomic difficulties have arisen because the genus *Combretum* is a complex, heterogeneous population, in which there appears to be continuous re-shuffling of genes[2]; a number of characters are found in nearly every combination. All that can be done is to give the "complexes" or "aggregates" of species the earliest legitimate name available; with new material continually being collected and ideas constantly changing, there is no way[7] of avoiding the inconvenience of changing the preferred name for species and the extensive use of synonyms. Exell[2] has pointed out that there are three types of synonym: (*a*) nomenclatural synonyms indissolubly linked with the accepted name, (*b*) names given to plants that appear to be identical to the type, and (*c*) names given to plants that differ from the type in certain characteristics, each of which represents one of the ways in which a number of characters can combine in a heterogeneous population.

TABLE II

TAXONOMIC CLASSIFICATION OF THE FAMILY COMBRETACEAE

FAMILY → SUB-FAMILIES

COMBRETACEAE
(ORDER, MYRTALES)

(*a*) STREPHONEMATOIDEAE Engl. and Diels
[1 genus: *Strephonema* Hook. f. (7 spp.)]

(*b*) COMBRETOIDEAE (10 genera)

TRIBE

COMBRETEAE (16 genera)

TRIBE

LAGUNCULARIEAE Engl. and Diels
[3 genera: *Laguncularia* Gaertn. f. (1–2 spp.)
Lumnitzera Willd. (2 spp.)
Macropteranthes F. Muell (4 spp.)]

SUB-TRIBE

COMBRETINAE (7 genera)

[*Thiloa* Eichl. (3 spp.)
Quisqualis L. (16 spp.)
Calopyxis Tul. (22 spp.)
Meiostemon Exell and Stace (2 spp.)
Guiera Juss. (1 spp.)
Calycopteris Lam. (1 spp.)
Combretum Loefl. (approx. 200 spp.)]

SUB-TRIBE

PTELEOPSIDINAE
(1 genus)

[*Pteleopsis*
Engl. (10 spp.)]

SUB-TRIBE

TERMINALIINAE (8 genera)

[*Terminalia* L. (approx. 150 spp.)
Ramatuella Kunth (6 spp.)
Terminaliopsis Danguy (1 spp.)
Bucida L., (9 spp.)
Buchenavia Eichl. (22 spp.)
Anogeissus Wall. (14 spp.)
Finetia Gagnop. (1 spp.)
Conocarpus L., (2 spp.)]

SUB-GENUS *Combretum*

SUB-GENUS *Cacoucia*

SUB-GENUS *Apetalanthum*

(further sub-divided into a total of 68 sections)

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