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Studies on Some Local Legumes

II. Cyanogenic Glucosides

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(*Paper accepted : 12 October* 1977)

Abstract : The seeds of nearly 50 selected legumes (mainly those commonly found in Sri Lanka) were screened for the presence of cyanide and cyanogenic glucosides. Only four of these contained significant amounts of cyanogenic glucosides ; all were varieties of *Phaseolus lunatus* (sieva bean or lima bean). Methods of processing to eliminate the cyanogenic glucoside was investigated with success. Local strains of *Vigna sinensis*, *Phaseolus vulgaris* and *Pisum sativum* were found to contain less cyanogenic glucoside than previously reported for strains grown elsewhere.

1. Introduction

The presence of cyanogenic glucosides have been reported in some plants of Family Leguminoseae. Its occurence in lima bean (*Phaseolus lunatus*) and sieva bean (*Phaseolus lunatus* L.) has been well documented.[1,5,7,8,9,13] The glucoside was identified as linamarin as early as 1907,[5] and since then there have been many studies on its distribution in the plant[10] and some on the effect of processing.[3,12] The only studies in Sri Lanka have been those of Charvanapavan[3] who in 1944 gave some data on the analysis of a locally available variety of lima bean but the account bears no description of either the variety or the method of analysis used.

Very little work has been conducted on cyanogenic glucosides of other legume seeds although Jaffe[6] and Montgomery[10] have reported that varieties of *Phaseolus vulgaris* (kidney or navy bean) *Pisum sativum* (garden pea) and *Vigna sinensis* (black-eye pea) contain cyanide in the order of 20 p.p.m. Cyanogenic glucosides have also been reported in *Cicer arietinum* and *Lathyrus sativus*.[9]

As in the case of the cyanogenic glucoside content of manioc, the methods of assay previously reported relied upon were either autolysis[2] or acid hydrolysis. The former assay is probably valid for most fresh samples but cannot be applied to processed material. The latter assay, as in the case of manioc, gives low results and is therefore not valid. In addition to the methods mentioned, another assay based on the use of sweet almond extract[2] (a source of glucosidase to decompose the amygdalin type of cyanogenic glucoside) has been recommended, but this would be ineffective on the linamarin type of cyanogenic glucoside which is generally present in the Phaseolae ; cyanogenic glucosides can be classified into four groups according to the chemical structure of the aglycone.[4]

Our studies have been mainly based on the application of the enzymic assay worked out for the cyanogenic glucosides of manioc, viz. the use of exogenous linamarase to hydrolyse bound cyanide. The assay has been found to work successfully on lima bean. In studies on screening, our work has been based on the reasoning that all cyanogenic glucosides except the linamarin type are decomposed by acid to give high yields of cyanide. Due to this, an initial screening was done by acid

hydrolysis and further tests were done only on those giving at least a slight positive reaction or those previously suspected for containing linamarin. The studies have shown that : (1) nearly all the types tested contained little (less than 5 p.p.m) or no cyanide, (2) varieties of lima bean could contain up to 800 p.p.m. total cyanide, (3) it is possible to process the lima bean so that only a small fraction of the total cyanide is present in the cooked material.

2. Experimental

2.1. Plant Material

2.1.1. *General*

Seed material was obtained from a variety of sources. Varieties of *Dolichos*, *Psophocarpus* (introduced) and *Phaseolus lunatus* (white) were obtained from the Agriculture Department Dry Zone Research Station (Mahailluppallama). Seeds of cover crops were obtained from Mr. Roy Bandaranaike, while common varieties of grain legumes were purchased in the open market or from Agriculture Department sales outlets. The remaining seeds were collected during field trips or from home gardens. The last category included *Canavalia gladiatus* (awara) *Cajanus cajan* (tur dhal) *Phaseolus lunatus* (lima bean-black varieties) and *Psophocarpus tetragonolobus* (local variety).

2.1.2. *Lima bean* (black variety).

The seeds tested were obtained from plants grown in home gardens. The plant is a climber with leaves and flowers typical of the Phaseolae. Pods are 6 to 7cm long and green when tender and generally bear 3 black seeds (due to a dark purple pigment in the testa). The seeds are flat with an average weight of 0.33g (at 13.5% moisture) bearing the typical markings of *Phaseolus lunatus* varieties.

2.2. Preliminary screening of samples for cyanogenic glucoside

The seeds (20g, 10% to 15% moisture) were coarsely ground and hydrolysed with 4N H_2SO_4 during steam distillation (45 min). The distillate (175 ml) was collected in 50 ml of 6.25% Na_2CO_3 and tested for cyanide by the picric acid test.[14]

2.3. Tests with manioc linamarase

The samples were finely ground in an edge runner mill (350μ particle size) and known weights (20, 10 or 5g) suspended in 100ml of 0.02M citrate buffer (pH 5.4) was incubated with manioc linamarase (100 units) for 4 to 8h and total cyanide determined as described previously.[11] Autolysis experiments were carried out in the absence of manioc linamarase. Free cyanide was determined as described previously.[11] Manioc linamarase was prepared by the method of Wood.[15]

3. Results and Discussion

3.1. Screening tests for cyanide

Screening tests for cyanide in the legumes using acid hydrolysis gave negative results (sensitivity 10 p.p.m. for the linamarin type and 5 p.p.m. for other types of bound glycosidic cyanide) for most of the seeds tested (Table 1). Possible trace quantities were detected in a *Mimosa* and *Crotolaria brownei*, while definite cyanide positive reactions were observed with 4 varieties of *Phaseolus lunatus* (Table 1).

TABLE I—Screening of legumes for bound cyanide

Code	Plant Species	Cyanide (Acid hydrolysable)
PB1a	Atylosia	—
PB2a	Cajanus cajan	—
PD1a	Crotalaria anagyroides	—
PD1b	Crotalaria brownei	+
PD1c	Crotalaria usaramoensis	—
PD2a	Trigonella foenum graecum	—
PE1a	Centrosema pubescens	—
PE2a	Clitoria ternatea	—
PE3a	Glycine max	—
PF1a	Arachis hypogea	—
PF2a	Desmodium gyroides	—
PF2b	Desmodium incenatum	—
PF2c	Desmodium ovalifolium	—
PG1a	Calpagoniam mucunoides	—
PG2a	Canavalia gladiatus	—
PG3a	Dolichos bi florus	—
PG3b	Dolichos lablab (yellow)	—
PG3c	Dolichos lablab (brown)	—
PG4a	Erythrina	—
PG5b	Phaseolus lathyroides (Pueraria javanica)	—
PG5c	Phaseolus lunatus (black)	++++
PG5d	Phaseolus lunatus (white, small)	++
PG5dl	Phaseolus lunatus (white, small)	+++
PG5e	Phaseolus lunatus (white, large)	++
PG5f	Phaseolus mungo	—
PG5g	Phaseolus mungo (var. radiatus)	—
PG5h	Phaseolus vulgaris (butter bean)	—
PG5i	Phaseolus vulgaris (navy bean)	—
PG5j	Phaseolus vulgaris (bush bean)	—
PG5k	Phaseolus vulgaris (wal bonchi)	—
PG6c	Psophocarpus tetragonalobus (light brown)	—
PG7a	Pueraria kudzu	—
PG7b	Pueraria phaseoloides	—
PG8a	Stizolobium nivea (mottled)	—
PG8b	Stizolobium nivea (grey)	—
PG8c	Stizolobium nivea (black)	—
PG8d	Stizolobium nivea (black, large)	—
PG9a	Vigna sinensis (black)	—
PG9b	Vigna sinensis (brown)	—
PG9c	Vigna sinensis (yellow)	—
PG9d	Vigna sinensis (panduru me)	—
PH1a	Tephrosia candida	—
PH1b	Tephrosia candida belga	—
PH1c	Tephrosia candida belga S	—
PH1d	Tephrosia vogelii	—
PI1a	Pisum sativum	—
MB2a	Mimosa I	+
CA1a	Cassia species	—

+, Trace 5—10 p.p.m. cyanide
++, 20—50 p.p.m. cyanide
+++, 50—100 p.p.m. cyanide
++++, > 100 p.p.m. cyanide

Studies using linamarase as the cyanide releasing agent on those seeds giving cyanide positive reactions and also those legumes previously reported[6,10] to contain glucosidic cyanide showed that the mimosa variety contained 14 p.p.m. total cyanide and *C.brownei* less than 5 p.p.m. Contrary to reports in the literature (which have quoted values of the order of 20 p.p.m.) the total cyanide levels in the edible legumes, *Pisum sativum*, *Vigna sinensis* (4 varieties: black, brown and yellow medium sized seed varieties and 'panduru mc') and *Phaseolus vulgaris* (4 varieties : Navy bean, butter bean, bush bean and 'wal bonchi', all sold by Agriculture Department sales outlets) were very low, being present only in trace quantities (less than 5 p.p.m.)

Cyanide levels of *Phaseolus lunatus* varieties are given in Table 2. All results quoted refer to one batch of seeds. These studies showed agreement with the literature on the point that smaller, darker seed types contain more bound cyanide.

TABLE 2—Cyanogenic glucoside content of some varieties of lima bean

	Variety	Source	Total cyanide (p.p.m.)
1.	Black small PG5c	Kegalle	460
2.	Black small PG5c	Grown in Colombo	500
3.	Black small PG5cl	Matara	792
4.	White small PG5d	Mahailluppallama	93
5.	White small (Henderson Bush) PG5dl	Mahailluppallama (Nigerian)	110
6.	White large (Burpees Bush) PG5e	Mahailluppallama (Nigerian)	46

Assays done using autolysis and exogenous linamarase. For experimental details see 2.3. Each experimental point refers to one batch of seeds.

3.2. Conditions of assay for total cyanide of *P. Lunatus* (PG5c).

Before initiating detailed studies on the effect of processing and cooking on the cyanogenic glucosides of the seeds, it was decided to compare the efficiency of the various methods of analysis available. It was found that the best results were obtained either by homogenising the fresh seed followed by autolysis or by use of exogenous linamarase on an extract of the powdered seed (Table 3). For the latter purpose the seed must be dried to less than 8% moisture at a temperature below 70°C before powdering in an edge runner mill. It was found that drying at 110°C caused some lowering of the total cyanide value possibly due to quick autolysis of the tissue and the resulting loss of free cyanide formed. Drying also reduces linamarase activity rendering autolysis ineffective (Table 4). However, the results of most of the methods used in Table 2 are comparable considering that sampling of seeds for assay results in an almost 10% standard deviation (90% probability) when random samples of 20 seeds are used for the assay (done six times in parallel). This was no doubt, due to the variation of total cyanide content from seed to seed and this deviation could be reduced by vastly increasing sample size. However, it was decided to slightly increase sample size to 30 seeds for the experiments described below, due to : (1) the limited quantity of seed material available (2) the fact that the effects studied showed relatively large changes (much larger than the standard deviations observed).

TABLE 3—Assay of total cyanide in lima bean by various methods

Method of Assay	Temp. of drying of seed (°C)	Total cyanide (p.p.m.)
1. Acid hydrolysis	—	158
2. Autolysis (4h)	—	460
3. Autolysis (15 min) + Exogenous linamarase (5h)	70	473
4. Autolysis (1½h) + Exogenous linamarase (3h)	70	464
5. Autolysis + Exogenous linamarase (4h)	70	492
6. Autolysis (4h)	70	423
7. Autolysis + Exogenous linamarase (overnight)	110	396
8. Autolysis + Exogenous linamarase (4h)	110	418

In 3 and 4 autolysis and exogenous linamarase hydrolysis were carried out in a two step process with a distillation step inbetween. See Table 3 for autolysis values. In other cases (5.7 and 8.) the reaction was stimultaneous.

For further experimental details see 2.2, 2.3. One batch of such was used of the above set of results.

TABLE 4—Effect of temperature of drying on the autolysis of cyanogenic glucosides

Time (h)	Temperature (°C)	Cyanide released (p.p.m.)	
0.25	70	90	
0.75	70	150	
1.5	70	279	
3	110		48
4	70	423	
4	110		66
5	110		83

For experimental details see 2.3. Results refer to one batch of seeds.

The results of Table 2 show that use of manioc linamarase is a valid method of assaying linamarin in lima bean. This was important as this method has to be used for assaying processed lima bean where autolysis cannot be used and acid hydrolysis results in abnormally low values for bound cyanide content.

3.3. Location of cyanogenic glucosides

In many cyanogenic tissues the outer layers of the tissue contain larger quantities of glucoside. Separation of the testa (13.5% by weight) from the rest of the seed showed that the reverse was true in this case ; the testa contained only 90 p.p.m. total cyanide compared to 582 p.p.m. for the rest of the seed. In other words, only 2.5% of the total cyanide was present in the testa. This variation of cyanogenic glucoside content of the different regions of the seed might account (in part) for the deviations observed in the assay.

3.4. Effect of soaking and boiling

There are several methods of cooking lima bean (in Sri Lanka). Most of these methods involve a soaking stage prior to boiling. A number of variations were tried out and it was noticed that swelling up of the seed markedly affected the final total cyanide content of the boiled seed (Table 5). Whereas soaking in water or dilute Na_2CO_3 resulted in only about half the seeds swelling and considerable residual total cyanide, induced swelling caused by slightly damaging the seeds or by removal of the testa (of undried fresh seed) resulted in considerably greater loss of total cyanide. However, the most promising results were obtained by boiling germinated seeds ; here only 3% of the original total cyanide remained.

All the above observations were true for seeds that were freshly harvested (within one month). However old seeds (15 months after harvest) are much easier to detoxify (Table 6). These seeds which are 80% to 90% non-viable swell up abnormally.

TABLE 5—Effect of soaking and boiling on total cyanide content of lima bean seeds

Treatment	Seeds swollen (%)	Residual total cyanide (p.p.m.)	Total cyanide remaining (%)
1. S — B — R — B	47	348	70
2. D — S — B — R — B	100	87	17
3. S* — B — R — B	47	306	61
4. R — S — B — B	100	196	25
5. G — R — B — B	100	15	3

S, Soak, (24h) ;
B, Boil 15 min ;
R, Remove testa ;
D, Damage ;
G, Germinate (5 days moistening)
S*, Soak in Na_2CO_3 (24h)

For details see 3.4: Results refer to one batch of seeds.

TABLE 6—Effect of soaking and boiling on total cyanide content of old lima bean seeds

Treatment	Residual total cyanide (p.p.m.)	% Remaining
1. S — B — B (over swollen)	<5	<1
2. S — R — B — B (over swollen)	<5	<1
3. S — B — B (selected unswollen)	160	35
4. B — R — B	40	09
5. B — B	127	28

See Table 4 for abbreviations.
For details see 3.4. Results refer to one batch of seeds.

3.5. Fate of cyanide in seed on soaking

Studies on the effect of soaking showed that there was very little free cyanide in the soak water. Next, it was decided to find out at which stage the cyanide was lost. In order to determine this, two sets of experiments were designed.

Experiment 1.

The seeds were soaked in a closed system (a cyanide determination steam distillation apparatus). After the soaking period (3 days) the seeds were boiled (during the steam distillation process). The boiled seeds were then separated from the aqueous medium and both medium and seed tested for bound cyanide. The results (Table 7) showed that although more than 50% of the original total cyanide was present in the soak-boil water in the form of bound cyanide, very little free cyanide was present. Further, a cyanide balance calculation showed that about 20% of the original total cyanide was unaccounted for. This suggested the possibility that some cyanide (free ?) was converted to other products, probably by a natural detoxification mechanism in the seed. The next set of experiments resulted in a similar conclusion.

TABLE 7—Fate of bound cyanide in lima bean on soaking and boiling

		Cyanide	
		p.p.m.	%
A			
1.	Free cyanide released on soaking and boiling	13	2.6
2.	Total cyanide in seed	120	24
3.	Bound cyanide released on soaking and boiling	265	53
B			
1.	Free cyanide in soak water	0	0
2.	Bound cyanide in soak water	0	0
3.	Total cyanide in seed	154	31
4.	Bound cyanide in boil water	169	33

For details see 3.5. Results refer to the batch of seeds.

Experiment 2.

In this experiment, free and bound cyanide in the soak water and bound cyanide in the boil water was determined. As no free cyainde is formed during boiling stage (Table 7A), the results (Table 7B) again showed some of the original cyanide was unaccounted for. The results also showed that release of bound cyanide takes place not during the soaking stage but in the boiling stage.

3.6. Detoxified flours

An attempt was made to prepare detoxified lima bean flour using the data accumulated above. The seeds were dried at 60°C and then ground into a fine flour. Water was added until a thick paste was formed and the mixture incubated for 4h after

which the paste was dried first at 60°C (to 15% moisture) and then at 100°C for 2h. The total cyanide content of the resulting flour was found to be 85 p.p.m. (17% of original total cyanide). Although it was considered possible to remove a larger fraction of the cyanide by (1) increasing amount of water added to flour and (2) by increasing incubation time (for autolysis), further work was not carried out as the flour obtained had a highly undesirable purple colouration arising from the pigment of the testa and because the removal of the testa was an operation which is unlikely to be accomplished easily on a large scale.

4. Conclusions

Most edible legumes contain little or no free and bound cyanide. The same is true also for legumes that are not usually eaten. The only exception appears to be the lima bean which can contain dangerous levels of bound cyanide. However these studies show that proper cooking procedures can be used to reduce the total cyanide content to relatively safe levels. The availability of safe methods of processing may provide sufficient impetus for the cultivation of this bean, which is a high yielder and whose cyanogenic character protects it from insect attack.

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J. Natn. Sci. Coun. Sri Lanka 1978 6(1) : 11—14

Bellows Effect of Sarong and Trousers

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Abstract : The bellows effect of two garments—sarong and trousers—upon the ventilation of the skin of the lower limb was measured by determining the loss in weight of pieces of a volatile substance, paradichlorobenzene, placed inside wire cages which were attached to the leg and thigh during 30 minutes of walking. The weight loss was of the order of 100 mg and there was no significant difference between sarong and trousers, or leg and thigh, with regard to the weight loss. The bellows effect is due to a wind created *in situ* under the garment rather than to a wind entering the garment from below.

1. Introduction

Garments have a bellows effect, that is, the movement of garments gives rise to the pumping of air over the skin which is covered by the garments. This gives rise to convective exchange of heat between the body and the air. In warm climates, the bellows effect is helpful in promoting heat loss from the clothed body during movement if the air is cooler than the skin.

The sarong and trousers are the commonest garments worn by men in contemporary Sri Lanka. Both hang down from the waist and cover the lower part of the trunk and the lower limbs. The sarong is a simple untailored tube of thin cloth, whereas trousers are tailored to fit the crotch and each lower limb separately with cloth of variable thickness. Many people have the custom of wearing trousers to work, and sarong during relaxation or sleep.

The present investigation sought to estimate the bellows effect of sarong *versus* trousers upon the lower limb.

2. Experimental

The principle of the method used in this investigation is to attach a sublimable substance to the lower limb between the garments and the skin ; the loss of substance during movement is assumed to be an index of the bellows effect.

If the skin is painted with an alcoholic extract of asafoetida, the bellows effect removes the asafoetida ; and the observer's nose can detect the decrease or disappearance of the odour of asafoetida from the skin. This method is, of course, subjective and non-quantitative.

Nishi and Gagge used naphthalene held near the skin to measure convectional loss of heat from the skin. Naphthalene balls, 3 cm in diameter, were held 3 cm off the skin at various points in the body. The ventilation of the skin was held at 30 feet per minute. Activities tested were sitting, walking and cycling.

The volatile substance paradichlorobenzene (PDB) which is commonly used, like naphthalene, as an insecticide for domestic articles such as textiles, is more sublimable than naphthalene. Whereas the weight loss of a piece of napthalene held in a wire cage on the leg under a sarong or trousers was of the order of 10 mg at the end of a half hour's walk, that of a similar piece of PDB was of the order of 100 mg. The weight loss of a 1.7 g piece of PDB if it were simply kept in a wire cage on a laboratory table for half an hour was found to be about 25 mg ; if a fan blew air on it with a wind speed of about 280 metres per minute the weight loss increased to about 60 mg. If it were kept outside the wire cage the weight loss was about 50 mg, or about 90 mg with a fan blowing on it.

The PDB was cut into roughly square blocks about 1.0 to 1.5 cm long. It is assumed that small differences between the shape and size of the blocks used in the present series did not have any important effect on the loss of weight of the blocks. A block of PDB was put into a wire cage. The shape of the cage was cylindrical with a length of about 5 cm and a diameter of about 1 cm. The wire was thin (gauge 0.2 mm) and of close mesh (2 mm square). The wire cage was weighed and then attached to the middle of the leg, or the junction of the lower and middle thirds of the thigh on its medial side, by means of a wire handle and rubber bands. One cage was attached to the left limb and another to the right. The subject then walked outside the laboratory for 30 minutes. He took a stopwatch with him so as to time the walking. At the end of the walking, the cages were detached and weighed to determine the loss in weight of the PDB inside the cage.

3. Results

The results are shown in the accompanying table.

None of the differences between the mean values for subject ERMR (sarong *vs* trousers, or left *vs* right, or leg *vs* thigh) was statistically significant, except possibly for the difference between leg and thigh with trousers (mean value for leg 101, thigh 84, t value 2.1 ; $0.05 > P > 0.02$).

4. Discussion

There is a very appreciable loss in weight in the block of paradichlorobenzene attached to the leg or thigh during walking. It is of the same order of magnitude as that produced in a similar piece of the substance by blowing air over it at nearly 300 m/min.

TABLE I. Weight loss of a piece of paradichlorobenzene placed in a wire cage attached to the leg or thigh during ½ hour of walking in sarong or trousers. Mean ± standard deviation.

	Number of determinations	Weight loss of the wire cage (mg)		
		Left side	Right side	Both sides
A. Wire cage attached to the *leg*				
Subject ERMR				
Resting (sitting) with sarong tucked up to expose the legs	1	20	25	23
Walking for ½ h wearing—				
Sarong	8	96 ± 30	90 ± 41	93 ± 35
Sarong tucked up to expose the legs	1	90	70	80
Trousers	8	89 ± 26	78 ± 24	84 ± 22
Subject RP				
Walking for ½ h wearing —				
Sarong	1	97	60	79
Trousers	1	92	80	86
B. Wire cage attached to the *thigh*				
Walking for ½ h				
Subject ERMR wearing —				
Sarong	8	112 ± 35	111 ± 18	111 ± 27
Sarong tucked up to expose the legs	1	63	60	62
Trousers	8	93 ± 23	109 ± 28	101 ± 26
Subject RP wearing —				
Sarong	1	58	60	59
Trousers	1	156	170	163
Subject BMPB wearing				
Sarong	1	102	114	108
Trousers	1	148	127	138

Possible causes of the loss in weight of the block of PDB are (i) sublimation of paradichlorobenzene and (ii) dropping off of pieces of paradichlorobenzene as powder or flakes. The latter is not likely to be important. In experiments done with the wire cage enclosed in a cloth bag there were no flakes or powder to be seen in the bag at the end of the experiment and there was no increase in the weight of the bag alone. The paradichlorobenzene still lost weight but the weight loss was halved, presumably because sublimation was hindered by the cloth bag.

The similarity in the order of magnitude of the weight loss of PDB in the leg and thigh sheds some light on the mechanics of the bellows effect. Let us consider two hypotheses : (*a*) the bellows effect is due to a current of air entering and leaving through the lower open end of the garment, and (*b*) it is due to a mixing and a blowing about of the air which already exists between the garment and the skin. If hypothesis (*a*) were correct we would expect the loss in weight of PDB to be greater on the leg than on the thigh. Since in fact there was no significant difference

between leg and thigh, hypothesis (*b*) seems more likely to be correct than (*a*). This conclusion was further supported by the finding that the application of a ligature on the trousers above the ankle or knee had little or no effect on the weight loss of the PDB.

5. Conclusions

The weight loss of the paradichlorobenzene (PDB) placed in a wire cage attached to the leg or thigh during 30 minutes of walking was of the order of 100 mg. There was no significant difference whether the garment worn was sarong or long trousers. The weight loss of the PDB is attributed to a "bellows effect" i.e. a wind created under the garment during movement. This wind seems to be created *in situ* under the garment rather than one which enters the garment from below.

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Absolute Convergence Factors for Cesàro Means

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Abstract : The problem considered in this paper is an extension of the problem (A) : What conditions on the convergence factor $g(x)$ are necessary and sufficient in order that the product $f(x)\,g(x)$ is absolutely Cesàro limitable of order r whenever $f(x)$ is absolutely Cesàro limitable of order k ? We find necessary and sufficient conditions in order that, for some l' ; $f(x)\,g(x) \sim l'\,x^{p+q} \mid C, r \mid$ whenever $f(x) \sim lx^p \mid C, k \mid$ for some l, where $p > -1$, $p+q > -1$, $r, k \in N$, $r \geqq k$, and f, g satisfy suitable local conditions. When $p = q = 0$, the conditions we obtain in this case are precisely those required in problem (A).

1. Introduction

We have considered[6] the problem of finding conditions which are necessary and sufficient in order that $f(x)\,g(x) = 0(x^{p+q}) \mid C, r \mid$ whenever $f(x) = 0(x^p) \mid C, k \mid$, where $r, k \in Z^+$, $r \geqq k$, $p, q \in \mathbb{R}$, $p + q \leqq -1$, f is locally Lebesgue integrable and g^{k-1} is locally absolutely continuous. We showed that in this case, the convergence factor g belongs to a restricted class, viz., a subclass of the class of all polynomials.

In this paper, we consider the case $p + q > -1$ and the problem in a slightly more general form. We find conditions necessary and sufficient in order that for some l', $f(x)\,g(x) \sim l'\,x^{p+q} \mid C, r \mid$ whenever $f(x) \sim lx^p \mid C, k \mid$ for some l, where $p > -1, p + q > -1, r, k \in Z^+, r \geqq k, g$ is bounded and measurable locally when $k = 0$, and g^{k-1} is locally absolutely continuous when $k \geqq 1$. The sequence analogue of this problem for $p = q = 0$ has been considered in (5).

The following are some preliminary definitions :

If $f \in L_{loc}$, $k \in \mathbb{N}$, define $I_k f(x) = f_k(x) = \int_1^x \frac{(x-t)^{k-1}}{(k-1)!} f(t)\,dt$, and $f_0(x) = f(x)$.

If $p > -1$, we write $f(x) \sim lx^p \mid C, k \mid$ if

$(p+1)\,(p+2) \ldots (p+k)\,x^{-p-k} f_k(x) \to l$ as $x \to \infty$ and $x^{-p-k} f_k(x) \in BV(1, \infty)$.

In particular, $f(x)$ is absolutely Cesàro limitable of order k to l

if $k\,!\,x^{-k} f_k(x) \to l$ as $x \to \infty$ and $x^{-k} f_k(x) \in BV(1, \infty)$.

2. Statement of the Theorems

We assume throughout that $p > -1$, $p + q > -1$ and the local conditions stated above hold.

Theorem 1. (*a*) If $r = k = 0$, a necessary and sufficient condition that $f(x)\, g(x) \sim lx^{p+q} \;\; C, 0\,|$ whenever $f(x) \sim lx^{p}\,|\,C, O\,|$ is that

$$x^{-q}\, g(x) \in BV(1, \infty).$$

(*b*) If $k = 0$, $r = 1$, a n.a.s.c. that $f(x)\, g(x) \sim l'\, x^{p+q}\,|\,C, 1\,|$ whenever $f(x) \sim lx^{p}\,|\,C, 0\,|$ is $(i)_b : x^{-p-q-1}\, Ix^{p}\, g(x) \in BV(1, \infty)$.

(*c*) If $k = 0$, $r > 1$, then conditions n.a.s. in order that $f(x)\, g(x) \sim l'\, x^{p+q}\,|\,C, r\,|$ whenever $f(x) \sim lx^{p}\,|\,C, 0\,|$ are :

$(i)_c$ $\displaystyle\int_1^x t^{-q}\, g(t)\, dt = 0(x)$ as $x \to \infty$,

$(ii)_c$ $x^{-p-q-r}\, I_r\, x^{p}\, g(x) \in BV(1, \infty)$.

Theorem 2. If $r \geq k \geqq 1$, conditions n.a.s. that $f(x)\, g(x) \sim l'\, x^{p+q}\,|\,C, r\,|$ whenever $f(x) \sim lx^{p}\,|\,C, k$ are :

(i) $g(x) = 0(x^{q})$ as $x \to \infty$

(ii) $g^{k-1}(x) = 0(x^{q+1-k})$ as $x \to \infty$

(iii) $x^{-p-q-r}\, I_r\, x^{p}\, g(x) \in BV(1, \infty)$.

3. Auxiliary Results

In this section we give some results which will be used in the proofs of the theorems.

Lemma 1 If $d_{ij} = \dfrac{\theta^{r+1-j}}{(r+i-j)!}$, $i, j = 1, 2, \ldots r-1$, $r \in \mathbb{N}$, then

$D = det\,(d_{ij})_{(r-1)\times(r-1)} = K\, \theta^{r(r+1)}$ where $k \neq 0$, and D_{ij}, the cofactor of d_{ij} in D is given by $D_{ij} = K_{ij}\, \theta^{(r-2)\,r+j-1}$, where K_{ij} is independent of θ.

This result is easily proved, by induction with respect to r.

Lemma 2 If $n \in \mathbb{N}$, $\phi^{n} \in AC_{loc}$, $\phi(x) = 0(x^{q})$ as $x \to \infty$, and $\phi^{n}(x) = 0(x^{q-n})$ as $x \to \infty$, then $\phi^{j}(x) = 0(x^{q-j})$ for $j = 0, 1, \ldots n$.

See (1), page 309 and (7), Lemma 1(c).

Lemma 3 Let $a(x,t) \in L(1,x)$ for $1 \leqq t \leqq x$, $a(x,t) = 0$ for $t > x$, and $v(x) = \int_1^x a(x,t)\, s(t)\, dt$. Then a n.a.s.c. that $v \in BV(1,\infty)$ whenever $s \in BV(1,\infty)$ is that there exists H independent of t such that

$$\int_t^\infty |\, d_x A(x,t)\,| \leqq H \text{ for all } t \geqq 1, \text{ where } A(x,t) = \int_t^x a(x,u)\, du.$$

This result follows from (4) Theorem 3 after an integration by parts. Cf. Also (3) — VIII. Note that $A(t,t) = A(t+,t) = 0$ in this case.

Lemma 4 Let $a(x,t)$, $A(x,t)$, $v(x)$ be as in Lemma 3. Let $n \in Z^+$ and s^n be the class of all real functions on $[1,\infty)$ such that $s^n(t)$ is absolutely continuous, locally and $s \in BV(1,\infty)$. Then, if $V(t) = \int_t^\infty |\, d_x A(x,t)\,| \in L(1,T)$ for every $T > 1$, a necessary and sufficient condition that $v \in BV(1,\infty)$ whenever $s \in S^n$ is that there exists H independent of t, and a constant T such that

$$V(t) \leqq H \text{ for almost all } t \geqq T.$$

This result follows from the theorem proved in (9) after an integration by parts. Cf. also (8), Theorem 4.

Lemma 5 If $p + k > -1$, $k' > k$ and if $g(x) \sim lx^p\,|\,C,k\,|$, then

$$g(x) \sim lx^p\,|\,C,k'\,|.$$

'$\sim lx^p$' may be replaced by '$= o(x^p)$' or '$= 0(x^p)$' here. Cf. (2), Lemma 3.

Lemma 6 If $p + k > -1$, $p + q > -1$ and $g(x) \sim lx^p\,|\,C,k\,|$, then

$$x^q g(x) \sim lx^{p+q}\,|\,C,k\,|.$$

'$\sim lx^p$' may be replaced by '$= o(x^p)$' or '$= 0(x^p)$' here. Cf. (2), Lemma 4.

4. Proofs of the Theorems

Theorem 1(a) is well known and its proof is omitted. We note at the outset that conditions (i)$_b$, (ii)$_c$ and (iii) are necessarily satisfied, since we may take $f(x) = lx^p$, $l \neq 0$, in particular. Hence we assume that $x^{-p-q-r} I_r x^p g(x) \in BV(1,\infty)$ in what follows. (1)

Case I Let $r > k$.

By repeated partial integration we have, when $k \geqq 1$,

$$I_r f(x)\, g(x) = \int_1^x \frac{(x-t)^{r-1}}{(r-1)!} f(t)\, g(t)\, dt = (-1)^k \int_1^x f_k(t)\, D_t^k G_r(x,t)\, dt \qquad (2)$$

where $D_t = \frac{\partial}{\partial t}$ and $G_r(x, t) = \frac{(x-t)^{r-1}}{(r-1)!} g(t)$.

This formula holds for $k = 0$ too.

Define $s(t) = t^{-p-k} f_k(t)$, $a(x, t) = (-1)^k t^{p+k} x^{-p-q-r} D_t^k G_r(x, t)$ and

$$A(x, t) = \int_t^x a(x, u)\, du.$$

Then, $x^{-p-q-r} I_r f(x) g(x) = v(x) = \int_1^x a(x, t) s(t)\, dt,$ (3)

and we want n.a.s.c. in order that $v \in BV(1, \infty)$ whenever $s \in BV(1, \infty)$ when $k = 0$, and whenever $s \in S^{k-1}$ when $k \geq 1$.

Now, $A(x, t) = \int_1^x x^{-p-q-r} u^{p+k} D_u^k G_r(x, u)\, du - \int_1^t x^{-p-q-r} u^{p+k} D_u^k G^r(x, u)\, du$

$= (-1)^k x^{-p-q-r} (p+1) \ldots (p+k) I_r x^p g(x) - x^{-p-q-r} \sum_{m=0}^{r-1} c_m x^{r-m-1} \int_1^t u^{p+k} D^k u^m g(u)\, du$ (4)

where $c_m = \frac{(-1)^m}{(r-1)!} \binom{r-1}{m}$, by partial integration and the binomial theorem.

By (1) and (4) it follows that $V(t)$ is bounded and hence Lebesgue integrable locally.

Thus, applying Lemma 3 when $k = 0$ and Lemma 4 when $k \geq 1$, we get the n.a.s.c. $\int_t^\infty |d_x A(x, t)| = 0(1)$, which, by (1) and (4) reduces to

$$\int_t^\infty |d_x (x^{-p-q-r} \int_1^t u^{p+k} D_u^k G_r(x, u)\, du)| = 0(1) \quad (5)$$

When $k = 0$, $r = 1$, it immediately follows that $(i)_b$ implies (5), and hence $(i)_b$ is both necessary and sufficient.

When $k = 0$, $r > 1$ repeated partial integration gives

$$\int_1^t \frac{(x-u)^{r-1}}{(r-1)!} u^p g(u)\, du = \sum_{m=1}^{r} \frac{(x-t)^{r-m}}{(r-m)!} I_m t^p g(t) \quad (6)$$

Since (1) holds, $\int_t^\infty | d_x (x^{-p-q-r} I_r t^p g(t)) | = | I_r t^p g(t) | t^{-p-q-r} = 0(1)$, and hence, by (6), condition (5) reduces to

$$\int_t^\infty | d_x \left(x^{-p-q-r} \sum_{m=1}^{r-1} \frac{(x-t)^{r-m}}{(r-m)!} I_m t^p g(t) \right) | = 0(1) \qquad (7)$$

If $(i)_c$ holds, then by Lemmas 4 and 5, we get

$$t^p g(t) = 0 (t^{p+q}) (C, m) \text{ for } m = 0, 1, \ldots r-1.$$

Hence $\int_t^\infty | d_x (x^{-p-q-r} (x-t)^{r-m} I_m t^p g(t)) | = | I_m t^p g(t) | \int_t^\infty | d_x (x^{-p-q-r} (x-t)^{r-m} | = 0 (t^{p+q+m})$. $A_1 t^{-p-q-m} = 0(1)$ for every m, showing that (7) holds.

Thus $(i)_c$ and $(ii)_c$ are sufficient in this case, and only the necessity of $(i)_c$ need be established.

By Lemma 5, since $f(x) g(x) \sim 1' x^{p+q} | C, r+1 |$ for $i = 0, 1, \ldots$, (7) holds with r replaced by $r + i$.

Thus $\int_t^\infty | d_x \left\{ x^{-p-q-r-i} \left(\sum_{j=1}^{r-1} + \sum_{j=r}^{r+i-1} \right) \left[\frac{(x-t)^{r+i-j}}{(r+i-j)!} I_j t^p g(t) \right] \right\} | = 0(1)$,

which reduces to

$$\int_t^\infty | d_x \left(x^{-p-q-r-i} \sum_{j=1}^{r-1} \frac{(x-t)^{r+i-j}}{(r+i-j)!} I_j t^p g(t) \right) | = 0(1), \qquad (8)$$

since $\int_t^\infty | d_x (x^{-p-q-r-i} (x-t)^{r+i-j} I_j t^p g(t)) | = 0 (t^{p+q+j})$. $A_2 t^{-p-q-j} = 0(1)$ for $j = r, \ldots\ldots r+i-1$.

(8) gives $\int_t^\infty | d_x \phi_i (x, t) | = 0(1)$, (9)

where $x^{p+q+r+i} \phi_i (x, t) = \sum_{j=1}^{r-1} \frac{(x-t)^{r+i-j}}{(r+i-j)!} I_j t^p g(t)$, $i = 1, \ldots r-1$.

Solving this system of linear equations for $I_j t^p g(t)$, we get

$I_j t^p g(t) = D^{-1} \sum_{i=1}^{r-1} D_{ij} x^{p+q+r+i} \phi_i (x, t)$ for $j = 1, \ldots r-1$, where by

Lemma 1, $D = K(x-t)^{r(r-1)}$, $K \neq 0$, $D_{ij} = K_{ij}(x-t)^{(r-2)r+j-1}$, K_{ij} being independent of x and t.

Hence $It^p g(t) = K^{-1}(x-t)^{-r(r-1)} \sum_{i=1}^{r-1} D_{i1} x^{p+q+r+1} \phi_i(x,t)$ giving

$$\frac{K(x-t)^{r(r-1)} It^p g(t)}{x^{rt-r+1+p+q}} = \sum_{i=1}^{r-1} K_{i1} \phi_i(x,t) \left[\frac{(x-t)}{x}\right]^{(r-1)^2-i} \tag{10}$$

Now (9) implies that $\int_t^\infty | d_x \left\{ \frac{(x-t)^s}{x^s} \phi_i(x,t) \right\} | = 0(1)$ for any $s \in \mathbb{N}$.

Putting $s = (r-1)^2 - i$, we get $\int_t^\infty | d_x \left\{ \left[\frac{(x-t)}{x}\right]^{(r-1)^2-i} \phi_i(x,t) \right\} | = 0(1)$,

and thus (10) gives $\int_t^\infty | d_x \left\{ \frac{(x-t)^{r(r-1)}}{x^{r^2-r+1+p+q}} It^p g(t) \right\} | = 0(1)$.

i.e. $| It^p g(t) |. A_3 t^{-p-q-1} = 0(1)$, which gives $(i)_c$ by Lemma 6, and thus Theorem 1 is complete.

Proof of Theorem 2 *Sufficiency*. We see that (i) and (ii) imply

$$g^j(t) = 0(t^{p-j}). j = 0, 1, \ldots k-1, \text{ by Lemma 2.} \tag{11}$$

Now, (5) may be written

$$\int_t^\infty | d_x \left\{ x^{-p-q-r} \int_1^t u^{p+k} du \left[\sum_{m=0}^{r-1} a_m x^{-r-m-1} \sum_{n=0}^{k} b_{mn} u^{m-n} g^{kn}(u) \right] \right\} | = 0(1) \tag{12}$$

where $a_m = (-1)^m \binom{r-1}{m}$ and $b_{mn} = \binom{k}{n} m(m-1)\ldots(m-n+1)$.

Now $\int_t^\infty | d_x \left\{ x^{-p-q-m-1} \int_1^t u^{p+k} u^{mn} g^{k-n}(u)\, du \right\} |$

$= \int_t^\infty | d_x\, x^{-p-q-m-1} || 0(u^{p+q+m}) | = 0(1)$ for $m = 0, 1 \ldots$ and $n = 1 \ldots k$ by (11) and

$$\int_t^\infty | d_x \{ x^{-pq-m-1} \int_1^t u^{p+k+m} g^k (u)\, du \} |$$

$$= \int_t^\infty | d_x \{ x^{-p-q-m-1} ([u^{p+k+m} g^{k-1} (u)]_1^t - \int_1^t (p + k + m)\, u^{p+k+m+1} g^{k-1} (u)\, du) \} |$$

$= 0\,(1)$ for $m = 0\ \ 1 \ldots$ by (ii).

Thus (12) holds proving the sufficiency of (i) (ii) and (iii).

Necessity We show that (5) implies (i) and (ii).

The proof is by induction. Assume that $(5)_{k = k_1}$ implies (i) and $(ii)_{k = k_1}$.

Suppose $k = k_1 + 1$. Then by lemma 5 (iii) and $(5)_{k = k_1 + 1}$ are necessary, and by the inductive assumption (i) and (ii) $k = k_1$ are necessary.

Also, $\int_1^t u^{p+k_1} D_u^{k_1} G_r (x, u)\, du + \int_1^t \frac{u^{p+k_1+1}}{p + k_1 + 1} D_u^{k_1} G_r (x, u)\, du$

$= \frac{t^{p+k_1+1}}{p + k_1 + 1} D_t^{k_1} G_r (x, t) + \sum_{m=0}^{r-1} a_m x^{r-m-1}$, where a_m is constant, and hence,

since $(ii)_{k = k_1}$ and $(ii)_{k = k_1 + 1}$ hold, it is necessary that

$$\int_t^\infty | d_x \{ x^{-p-q-r} t^{p+k_1+1} D_t^k G_r (x, t) \} |$$

$$= \int_t^\infty | d_x \left\{ x^{-p-q-r} t^{p+k_1+1} \sum_{k=0}^{k_1} (-1)^j \binom{k_1}{j} \frac{(x-t)^{r-j-1}}{(r-j-1)!} g^{k_1-j} (t) \right\} | = 0\,(1) \quad (13)$$

Now $\int_t^\infty | d_x \{ x^{-p-q-r} t^{p+k_1+1} (x-t)^{r-j-1} g^{k_1-j} (t) \} |$

$= t^{p+k_1+1} .0\ (t^{q+j-k_1}). A_4\, t^{-p-q-j-1} = 0\ \ (1)$ for $j = 1, 2, \ldots k_1$ by (i) and $(ii)_{k_1}$ (14)

(13) and (14) give $\int_t^\infty | d_x \{ x^{-p-q-r} t^{p+k_1+1} (x-t)^{r-1} g^{k_1} (t) \} | = 0\,(1)$ and hence $g^{k_1} (t) = 0\,(t^{q-k_1})$, which is (ii) $k = k_1 + 1$, and by induction, we get the necessity.

Case II $r = k$. Since (i) and (ii) are independent of r and are necessary when $r > k$, by Lemma 5. (i) and (ii) are *a fortiori* also necessary when $r = k$. We thus have to consider only the sufficiency. In this case we have

$$x^{-p-q-k} I_k f(x) g(x) = \int_1^x a(x\ t)\, s(t)\, dt + (-1)^k x^{-q} s(x) g(x) \tag{15}$$

and $$\int_1^x x^{-p-q-k} u^{p+k} D^k_u G_k(x\ u)\, du = (-1)^k x^{-p-q-k} (p+1) \ldots (p+k) I_k x^p g(x) - x^{-q} g(x) \tag{16}$$

by partial integration, instead of (3) and (4) respectively.

Now define $B(x, t) = A(x, t) + (-1)^k x^{-q} g(x)$.

Then, for fixed x, $-B(x, t)$ is still an indefinite integral of $a(x, t)$, and since

$$A(x, x) = 0, \quad \text{we have } B(x, x) = (-1)^k x^{-q} g(x).$$

Hence partial integration of (15) gives $x^{-p-q-k} I_k f(x) g(x) = \int_1^x B(x, t)\, ds(t)$,

and from this point onwards the proof of the sufficiency is exactly as in case I.

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The Set of Numbers { 1, 5, 10 }

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Abstract : The set of numbers { 1, 5, 10 } has the property that, the product of any two decreased by 1, is a square. It is shown that there exists no positive integer c such that the set { 1, 5, 10, c } possesses the same property.

1. Introduction

The set of numbers [1, 3, 8, 120] has the property that, the product of any two increased by 1, is a square. Baker and Davenport[1] have proved that the property does not hold, if 120 is replaced by any other positive integer. The proof is based on Baker's Theorem in Diophantine Approximation. Kanagasabapathy and Ponnudurai[2] have described another method using nothing deeper than quadratic reciprocity, by which the result may be obtained. In this paper, we deal with the numbers 1, 5 and 10 which have the property that the product of any two decreased by 1, is a square. We prove the following theorem concerning these numbers :

2. Statement of the Theorem

Theorem : There exists no positive integer c such that the product of every pair of numbers of the set [1, 5, 10, c] decreased by one, is a perfect square.

Proof :

To establish our result, it is sufficient to show that the equations

$$c - 1 = x^2$$

and

$$5c - 1 = y^2$$

$$10c - 1 = z^2$$

cannot hold simultaneously for any positive integral values of x, y, z and c.

These equations lead to

$$y^2 - 5x^2 = 4 \qquad (1)$$

and

$$z^2 - 2y^2 = 1 \qquad (2)$$

All the non-negative integral solutions of the Pell Equation (2) are obtained from the following formula :

$$z_n + \sqrt{2}y_n = (1 + \sqrt{2})^{2n} \tag{3}$$

where n is a positive integer or zero.

Using (3) we obtain easily the following relations :

$$z_{m+n} = z_m z_n + 2y_m y_n$$

$$y_{m+n} = y_m z_n + y_n z_m$$

$$z_{-n} = z_n,\ y_{-n} = -y_n$$

$$z_{2n} = z_n^2 + 2y_n^2 = 2z_n^2 - 1 = 1 + 4y_n^2$$

$$y_{2n} = 2y_n z_n.$$

The following congruence holds :

$$y_{n+2r} \equiv -y_n \pmod{z_r} \tag{4}$$

We need the following results which can be easily established by induction :

(i) z_n is odd.

(ii) If $k = 2^t$, where t is an integer, then

$$z_{4k} \equiv 1 \pmod 8, \text{ for } t \geqslant 0 \tag{5}$$

$$z_{4k} \equiv 2 \pmod 5, \text{ for } t \geqslant 0 \tag{6}$$

$$z_{4k} \equiv -1 \pmod{17}, \text{ for } t = 0 \tag{7}$$

$$z_{4k} \equiv 1 \pmod{17}, \text{ for } t \geqslant 1 \tag{8}$$

We require the following table of values :

TABLE 1

n	y_n	z_n
0	0	1
1	2	3
2	12	17
3	70	99

From (1) we obtain

$$(5x_n)^2 = 5(y_n^2 - 4). \tag{9}$$

The proof is now accomplished in three stages :

(*a*) (9) is impossible if $n \equiv 0 \pmod 4$,
For, using (4) we obtain

$$y_n \equiv 0 \pmod{17}$$

Thus, we find that

$$(5x_n)^2 \equiv -20 \pmod{17}$$

and since $(-20 \mid 17) = -1$, (9) is impossible.

(*b*) (9) is impossible if $n \equiv 2 \pmod 4$.
For, using (4) we obtain

$$y_n \equiv \pm 12 \pmod{17}$$

Thus, we find that

$$(5x_n)^2 \equiv 20 \pmod{17}$$

and since $(20 \mid 17) = -1$, (9) is impossible.

(*c*) (9) is impossible if $n \equiv \pm 1 \pmod 4$, $n \neq \pm 1$, that is $n = \pm 1 + 4k + 8rk$, where r is an integer and $k = 2^t$ with t an integer $\geqslant 0$.

For, using (4) we obtain

$$\begin{aligned} y_n &\equiv \pm y_{4k+1} \pmod{z_{4k}} \\ &\equiv \pm 3y_{4k} \pmod{z_{4k}} \end{aligned}$$

Which implies

$$\begin{aligned} 2y_n^2 &\equiv 9(z_{4k}^2 - 1) \\ &\equiv -9 \pmod{z_{4k}} \end{aligned}$$

Thus we find that

$$\begin{aligned} (10x_n)^2 &= 10(2y_n^2 - 8) \\ &\equiv 10(-17) \pmod{z_{4k}} \\ &\equiv -2.5.17 \pmod{z_{4k}} \end{aligned}$$

and since

$$\begin{aligned} \left(\frac{-2.5.17}{z_{4k}}\right) &= \left(\frac{-1}{z_{4k}}\right)\left(\frac{2}{z_{4k}}\right)\left(\frac{5}{z_{4k}}\right)\left(\frac{17}{z_{4k}}\right) \\ &= \left(\frac{2}{5}\right)\left(\frac{\pm 1}{17}\right) \quad \text{using the congruences (5) to (8)} \\ &= -1, \end{aligned}$$

and so (9) is impossible.

Hence there exist no integral values x, y, z which satisfy equations (1) and (2) simultaneously. The theorem now follows.

Acknowledgement

The author is deeply grateful to the late Professor P. Kanagasabapathy for suggesting the problem and for the kind help he gave in the preparation of this paper.

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Some Studies on the Corrosion of Copper and its Alloys in Calcium Hypochlorite

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Abstract : Corrosion behaviour of copper and eleven of its alloys has been studied in 20% calcium hypochlorite liquor containing 60% available chlorine at 35°C. Potential time curves, galvanostatic anodic and cathodic polarization behaviour and conventional weight loss data were utilised in the study. All the alloys studied were found to be self passivating under the experimental conditions used. The time taken for the formation of the protective film was less than an hour. The corrosion potentials of the alloys were in the range 0.650 to 0.680 V (vs SCE). Anodic polarization revealed that passivity broke down, leading to pitting in some alloys and general corrosion in certain other alloys. The latter behaviour was due to the formation of a soluble product. The Tafel slopes ranged from 0.120 to 0.254 mV in the transpassive region. Cathodic polarization curves exhibited two Tafel regions. Weight loss data under total immersion conditions indicate that nickel tin bronze (high nickel) is the most resistant. The corrosion rate of this alloy was three times higher under intermittent immersion conditions and eleven times higher under partial immersion conditions.

1. Introduction

Studies on the corrosion of copper and its alloys have been reviewed by Leidheiser.[7] Only a few workers have studied the corrosion of copper in bleaching liquor. Fitzerald Lee has reported the corrosion rates of six copper alloys in saturated $Ca(OCl)_2$ solution at 16°C.[6] Other workers have reported the corrosion of brass in NaOCl solution containing 9% available chlorine.[11] All these studies were based on weight loss data. No fundamental electrochemical studies of copper corrosion in hypochlorite has been reported.

This paper describes the study of the corrosion behaviour of copper and a number of its alloys in calcium hypochlorite liquor containing about 60% available chlorine (High Test Hypochlorite). Hypochlorite solutions are used for bleaching in the textile industries of Sri Lanka. Electrochemical and gravimetric methods were utilised in this study. The experimental work involved determination of the variation of open circuit electrode potential with time and the measurement of anodic and cathodic galvanostatic polarization data. In addition, weight loss data were also obtained.

2. Experimental

Only a brief outline of the experimental procedure is given below as full details have been given elsewhere.[1]

2.1 Materials and specimen preparation

The alloys were cast and analysed quantitatively by standard methods. The percentage composition of the alloys are reported in column 1 of Table 1. The surface was prepared by hand abrading with emery paper of successively finer grades from 120 to 500, then washing thoroughly with distilled water and finally by degreasing with acetone.

A concentrated solution (20%) of High Test Hypochlorite (DCM Chemicals New Delhi) in distilled water was used in all studies. This solution had a pH of 11.5 and an available chlorine content of 60.3%. All experiments were conducted at 35 ± 0.2°C.

2.2. Electrochemical studies

The polarization cell had two compartments separated by a sintered disc. The cell had a capacity of 100 ml. One compartment contained a cylindrical platinum counter electrode and the other contained the working electrode. The reference electrode (saturated calomel electrode) was kept in a cell of saturated KCl solution and connected to the working electrode compartment by a Luggin bridge designed after Greene.[4]

The working electrodes were cut from 0.86 cm diameter rods to 5 cm length. The working electrodes were mounted on a Teflon electrode holder so that only the polished surface (of area 0.586 cm^2) was exposed to the solution. The distance between the exposed electrode surface and the Luggin capillary tip was about 2 mm.

The variation of open circuit electrode potential with time was recorded, for about an hour after immersion, on a potentiometric recorder. The counter electrode was disconnected from the circuit during these measurements.

Galvanostatic polarization experiments were started after the electrode potential reached the steady state corrosion potential. Anodic polarization was first performed followed by cathodic polarization. Current densities ranging from 0.0025 to 15.5 mA cm^{-2} were applied. Potentials were measured five minutes after applying current.

TABLE 1. Corrosion Characteristics of Copper Base Alloys in Calcium Hypochlorite

Alloy	Corros Pot. (volts vs. SCE)	Tafel slope, b (volts per decade)	corros. current (mA per cm^2)	corros. rate (mm per year)
1. Copper	0.686	0.196	0.055	0.058
2. Silicon bronze (Si 1.5)	0.683	0.191	0.063	0.102
3. Silicon bronze (Si 3.0)	0.665	0.222	0.115	0.102
4. Brass (Zn 9.7)	0.688	0.251	0.123	0.284
5. Tin bronze (Sn 9.4)	0.650	0.168	0.148	0.130
6. Nickel tin bronze (Sn 5.8, Ni 2.1)	0.692	0.165	0.046	0.063
7. Nickel tin bronze (Sn 1.2 Ni 5.0)	0.648	0.161	0.239	0.038
8. Cupronickel (Ni 5.0, Fe 5.5)	0.655	0.195	0.380	0.185
9. Aluminium brass (Zn 17.7, Al 2.0)	0.650	0.254	0.234	0.358
10. High leaded tin bronze (Pb 14.8, Sn 5.9)	0.670	0.167	0.048	0.112
11. Gunmetal (Sn 8.4, Pb 3.8, Zn 3.1)	0.680	0.236	0.095	0.119
12. Leaded naval brass (Zn 36.7, Pb 2.1, Sn 1.9)	0.680	0.120	0.151	0.345

2.3. Gravimetric Methods

The cells used were cylindrical glass vessels 80 mm high and 40 mm diameter. A glass hook was fixed to the stopper to support the corrosion coupon. The spherical coupons had a diameter 12.5 mm and thickness 3 mm. A hole, 3 mm diameter, was drilled near the edge to support the specimen. The solutions were neither agitated nor aerated.

The corroded coupons were treated with 10% sulphuric acid to remove the corrosion products, then washed thoroughly with distilled water and finally rinsed with acetone. The coupons were then dried and weighed. An uncorroded specimen lost less than 0.0001 g during the above treatment.

All the twelve copper alloys studied were subjected to total immersion tests of 24 hour duration. On the basis of their corrosion rates the best alloy was selected to undergo the planned interval tests recommended by Wachter and Treseder[12] which provides an excellent procedure for evaluating the effect of time on the corrodibility of the metal and the corrosiveness of the environment. The maximum duration of these tests was 96 hours. The effect of changing the immersion conditions to partial and total immersion was also studied.

3. Results and Discussion

3.1. Potential-time curves

Typical potential-time curves are given in Figure 1 and the steady state corrosion potentials are given in column 2 of Table 1. The curves show that the electrode potentials of copper base alloys rise sharply to a noble steady state value of about 0.650 to 0.680 V (vs SCE). Such behaviour indicates the formation of a protective film on the metal surface. Consequently, here is a rare instance of copper exhibiting passivity. This may be due to the high oxidising power of the corrosive. The corrosion potentials recorded for copper in the present work are considerably higher than those reported in literature. The highest corrosion potential of copper, 0.45 V (vs SCE), was obtained in 0.05M chromic acid solution.[8]

The nature of the protective film formed is not certain. The colour of the film, which was brownish black, indicated that it is probably CuO. Potential-pH diagrams[10] and the recent work by Faita *et al.* who worked with aerated 0.5M NaCl solutions[3] indicate that the protective film is CuO. On the other hand, Hoar *et al.* have indicated that the black film covering brass in chloride solutions is Cu_2O.[5] North and Pryor support this view.[9] In the present work, polarization behaviour (see below) indicates the possibility that the film consists mainly of Cu_2O.

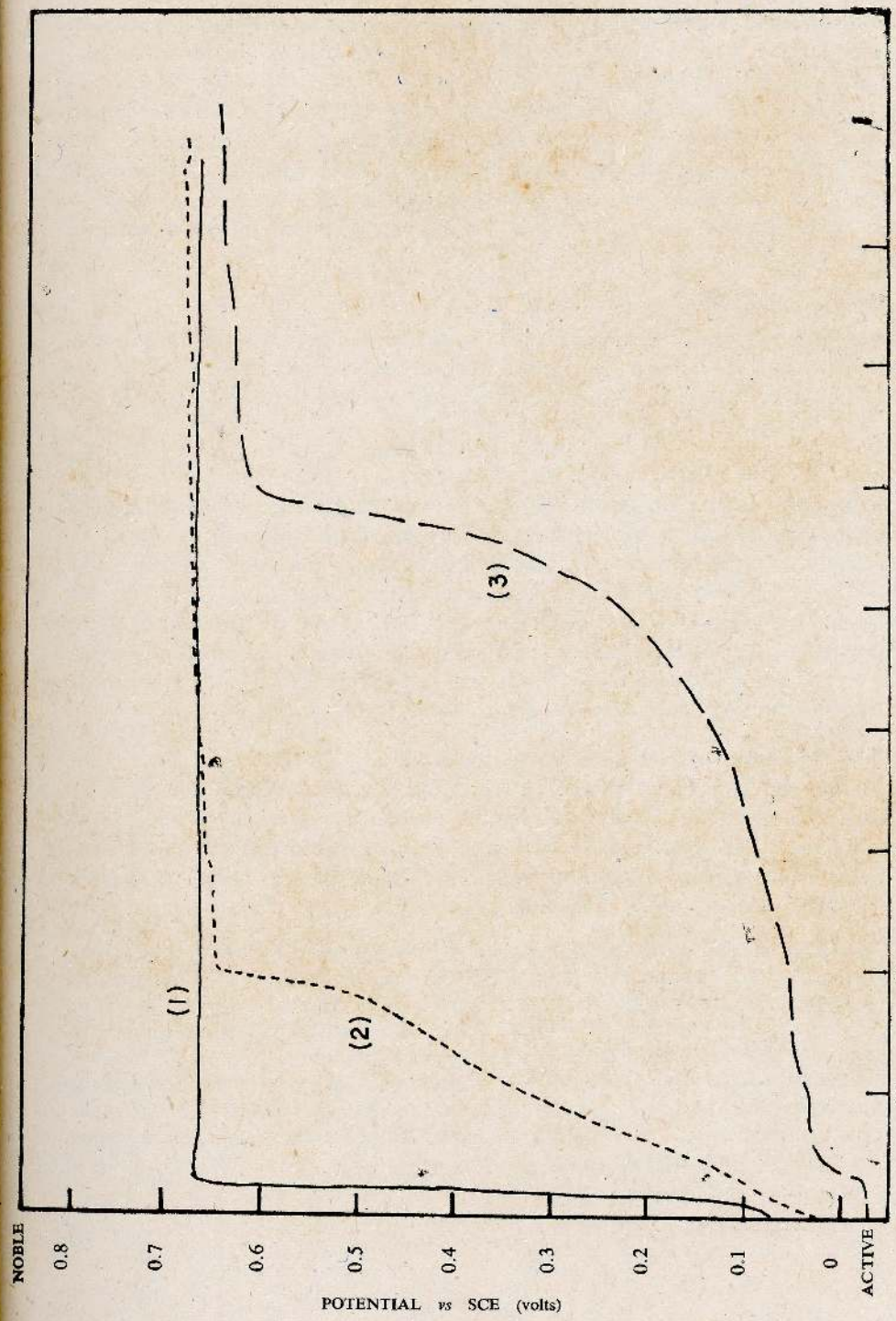

Figure 1. Potential—time curves

The time taken for the formation of the protective film varies with the alloy studied. Formation of the protective film was very rapid in the case of unalloyed copper, 90 : 10 brass, aluminium brass and the three nickel alloys. This behaviour is exemplified by the potential-time curve of nickel tin bronze-high tin (Curve 1). Film formation was slower with the addition of silicon (Curve 2, high silicon bronze) to copper and with all the three alloys containing lead. It seems that the addition of silicon and lead delays film formation while zinc, nickel, aluminium and iron have no effect on the rate of film formation. The longest time of one hour was observed with tin bronze (Curve 3).

3.2. Anodic polarization

Since copper appears to be self passivating in hypochlorite liquor, the anodic polarization curve is in the transpassive region. Typical anodic polarization curves are shown in Figures 2 and 3. The curves are seen to obey the Tafel equation

$$\eta = \frac{2.303\ RT}{\alpha\, nF} \log i_o + \frac{2.303\ RT}{\alpha\, nF} \log i \qquad (1)$$

at overvoltages greater than about 0.050V. In this equation η is the overvoltage, i the current density, i_o the exchange current density, R the universal gas constant, T the absolute temperature, α the transfer coefficient, n the number of electrons involved in the process and F the Faraday constant. The Tafel slopes for the transpassive region obtained in the present work range from 0.120 to 0.254 V for the different alloys and are reported in column 3 of Table 1. Figure 2 shows the anodic behaviour of brass and nickel tin bronze (high Ni). The other alloys exhibit an intermediate behaviour with respect to their Tafel slopes.

The anodic curve was extrapolated to the corrosion potential to calculate the corrosion current density. Corrosion current density (i_{corr}) values so calculated are given in column 4 of Table 1. Figure 3 shows two extreme behaviour with respect to i_{corr}. The anodic curve of copper ($i_{corr} = 0.055$ mA cm^{-2}) and that of Cu.Ni.Fe alloy ($i_{corr} = 0.380$ mA cm^{-2}) are shown in the figure. The other alloys exhibit an intermediate behaviour.

A few alloys (leaded naval brass, high leaded tin bronze, tin bronze, low silicon bronze and unalloyed copper) exhibit a second Tafel region at higher current densities with slopes ranging from 0.058 to 0.118V (Figure 3). As the values of the Tafel slopes in this case is approximately half as that in the early transpassive region, it is probable that the second anodic reaction involves double the number of electrons as that in the first reaction (see Equation 1). Hence it is reasonable to assume that the first reaction involves an one electron transfer and the second reaction, a two electron transfer. Consequently, it follows that the simplest anodic reactions are the formation of cuprous and cupric ions,

$$Cu \longrightarrow Cu^{+} + e^{-} \qquad (2)$$

$$Cu \longrightarrow Cu^{2+} + 2\,e^{-} \qquad (3)$$

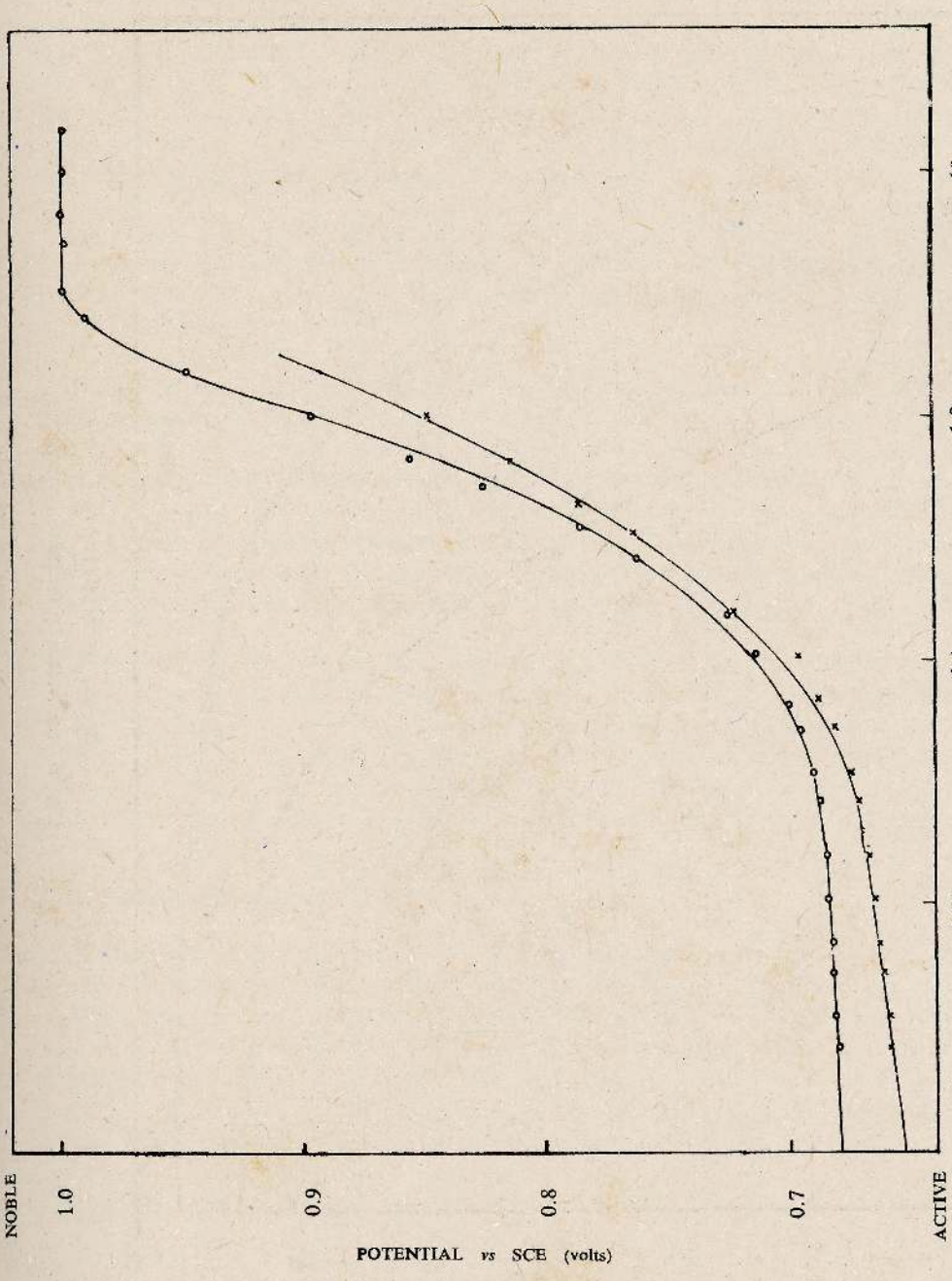

Figure 2. Anodic polarization curves
o-o-o brass
x-x-x-x nickel tin bronze (high Ni)

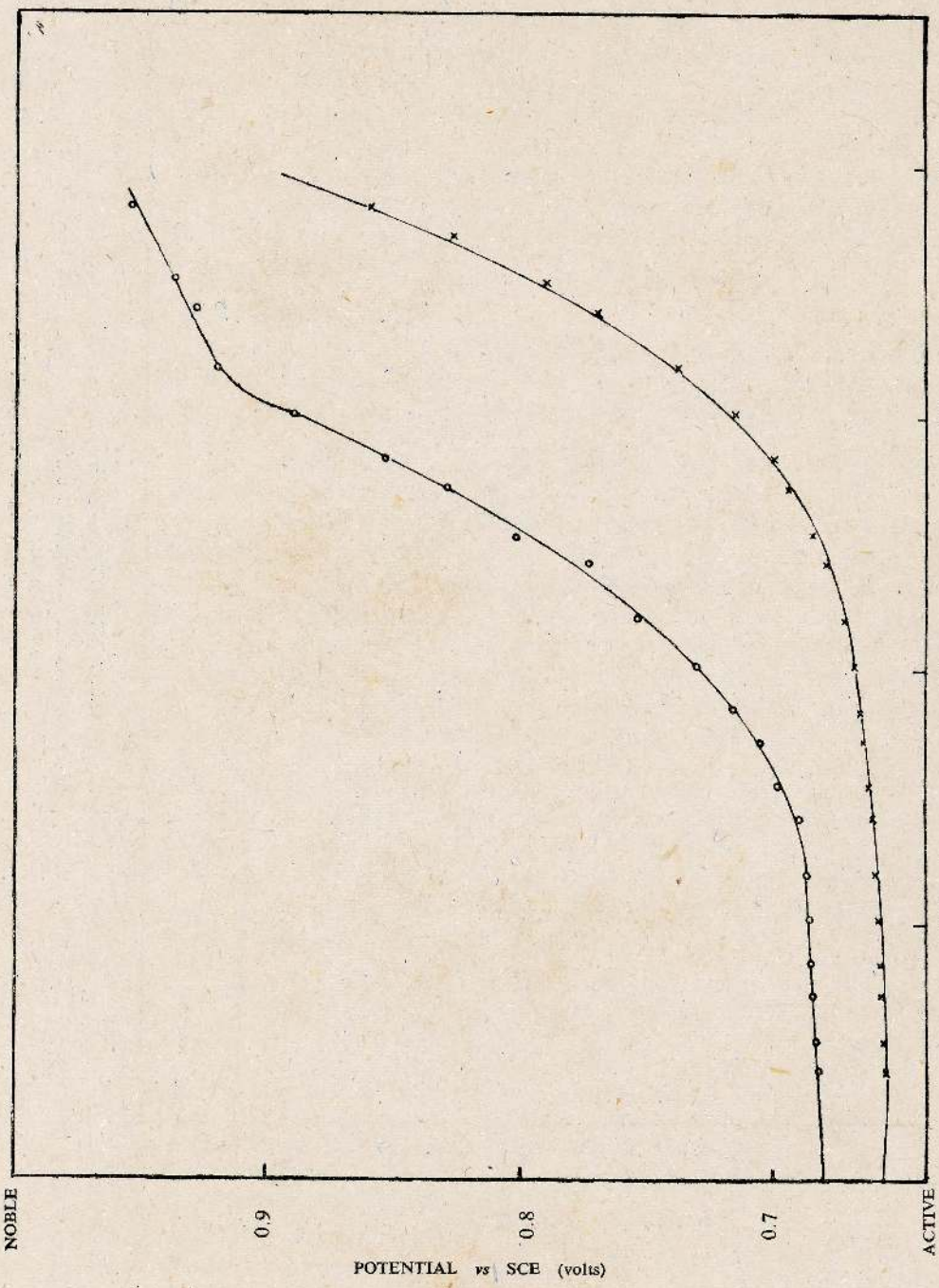

Figure 3. Anodic polarization curves
o-o-o-o copper
x-x-x-x Cu. Ni. Fe alloy

The Tafel slope values can be used to calculate the transfer coefficient for the two reactions (Equation 1). The transfer coefficient of the first reaction rnages from 0.24 to 0.51 and for the second reaction from 0.26 to 0.53. Myer[13] has indicated that low values for α experimentally obtained (as in the present work) indicate that the reaction process involves charge transfer through the film in addition to the usual charge transfer process at the interface.

The anodic polarization curve of gunmetal and brass exhibited a region of constant potential at high current densities (Figure 2). This can be interpreted as due to pitting. The breakdown potentials, E_b, for gunmetal and brass were 0.960 and 1.000 V respectively (vs SCE).

As for the nature of the corrosion product formed, reference has to be made to previous work. Many workers have indicated that the corrosion product covering copper in aerated, acid and alkaline solutions, is Cu_2O.[3,5,8,9] Hence, it is probable that the electrochemical reaction involving one-electron transfer leads to the formation of Cu_2O. Assuming that this reaction serves to plug any defects in the protective film, it can be said that the protective layer is likely to be made up of Cu_2O. The black colour of the protective layer observed also indicates this possibility as Hoar *et al.* have observed that the black layer covering brass in chloride solutions is Cu_2O.[5]

As for the two-electron reaction, the first impulse is to say that cupric oxide it formed. This is in agreement with the work of Faita *et al.*[3] who have shown that, at high potentials, the corrosion product covering copper in alkaline solutions is CuO. But in the present work a green product was observed in the vicinity of the electrodes. This leads to the assumption that CuO and $CuCl_2$ are formed initially and then the green complex $CuO.CuCl_2$, which is insoluble in water, is formed.

3.3 Cathodic polarization

Cathodic polarization behaviour of two alloys are shown in Figure 4. From the curves it can be noted that each alloy exhibits four regions. The first region is a slight curve where the overvoltage is not sufficient enough to produce polarization. The second region shows a sharp fall in potential indicating that the cathodic reaction is diffusion controlled. The third region is a Tafel slope showing that a second reduction reaction occurs in this region. The fourth region is again a steep fall in potential even beyond the range of the potentiometer (— 1.8 V vs SCE).

The first cathodic reaction is under diffusion control and obeys the equation

$$\eta_c = \frac{2.303\ RT}{nF} \log \frac{i_L - i}{i_L} \qquad (4)$$

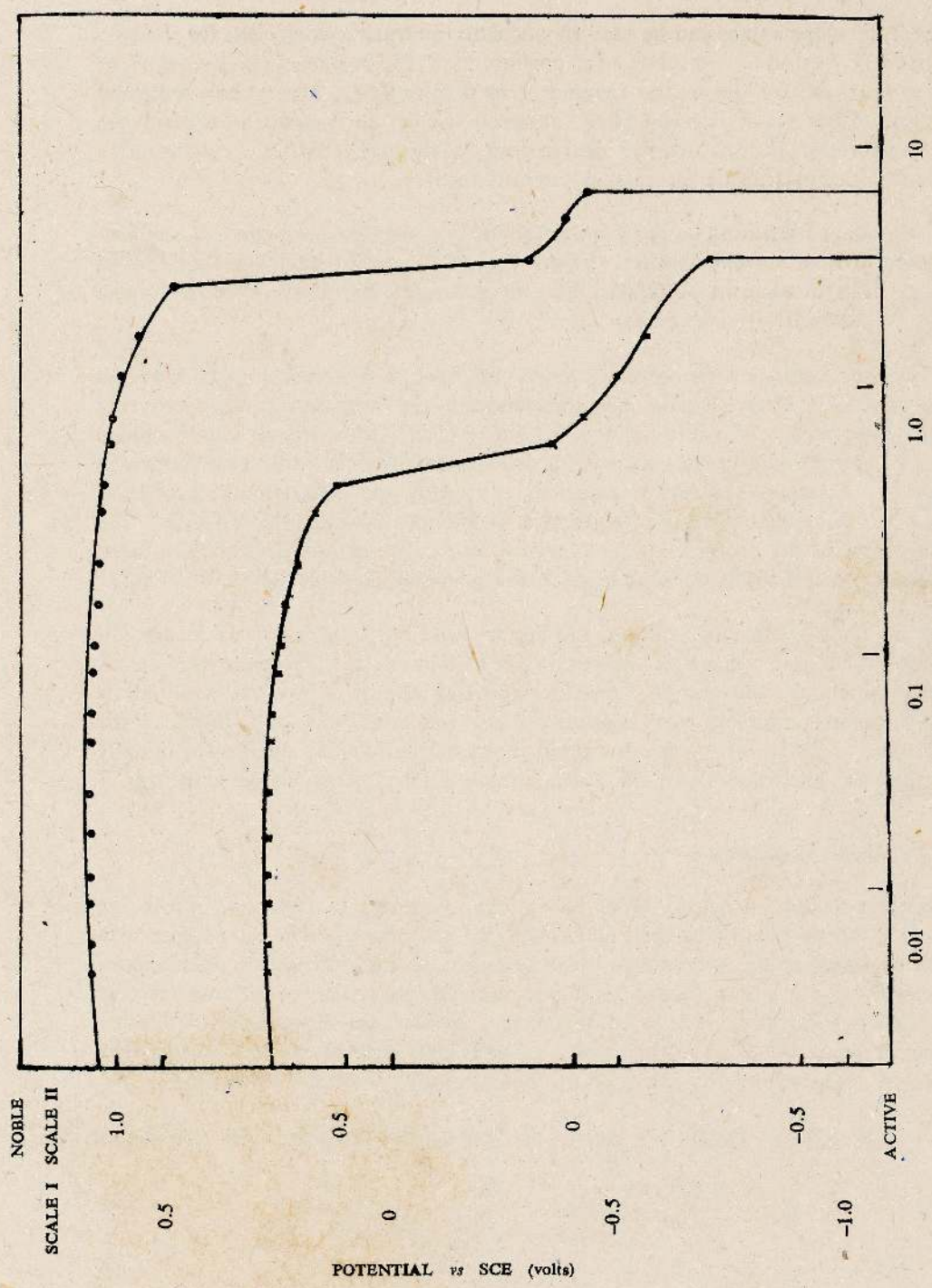

Figure 4. Cathodic polarization curves
scale I o-o-o-o tin bronze
scale II x-x-x-x High leaded tin bronze

where i_L is the limiting diffusion current density. When the applied current density approaches the limiting diffusion current density ($i \rightarrow i_L$) the overvoltage tends to infinity. That is, the potential falls sharply. In the present case, the limiting diffusion current density ranges from 0.34 to 3.4 mA cm^{-2}. Figure 4 shows the two extreme cases and the other alloys exhibit intermediate behaviour.

As for the nature of this reaction, it cannot be hydrogen evolution as the corrosion potential is very positive (around + 0.9V vs SHE) and hydrogen evolution occurs at negative potentials. Besides, gas evolution was not observed. This reaction cannot be the reduction of hypochlorite ion as the concentration of this ion in the solution is too high for diffusion control. Assuming the diffusion layer of thickness, δ, in an unstirred solution to be 0.05 cm and the diffusion coefficient, D, to be 8×10^{-6} cm^2 sec^{-1} the limiting diffusion current density has been calculated to be 62.5 mA cm^{-2} by using the equation

$$i_L = \frac{Dn\ F\ a}{\delta} \tag{5}$$

where a, the activity of the hypochlorite ion is 2.23 gm ions per litre and n the number of electrons involved in the reaction

$$ClO^- + 2\ H_2O + 2e^- \rightarrow Cl^- + 2\ OH^- \tag{6}$$

s 2. This value of 62.5 mA cm^{-2} is much higher than the experimentally obtained value of 0.34 to 3.4 mA cm^{-2}. Hence the first cathodic reaction is not the reduction of hypochlorite.

The only possibility, then, is that the first reaction is the oxygen reduction reaction, more so, as the solution was not deaerated.

$$O_2 + 2H_2O + 4e \rightarrow 4OH^- \tag{7}$$

The nature of the second cathodic reaction is not known.

3.4. Weight loss methods

The corrosion rate (in mm per year) for a duration of 24 hours is given in column 5 of Table 1. Of all the alloys tested, nickel tin bronze (high nickel) alloy is the most resistant. Pitting was not observed in any alloy. This may be because the test duration was very short.

The nickel-tin-bronze alloy (the most resistant alloy) was subjected to special tests. The planned interval tests showed that while the corrosiveness of the solution remained constant with time, the corrodibility of the alloy decreased with time. This can be explained as due to the formation of the protective film. Corrosion rate under

intermittent immersion conditions was about thrice as fast compared with that under total immersion conditions for the same duration (0.102 vs 0.036 mm per year). The most severe corrosion was observed in partial immersion tests, where the corrosion loss was 4.42 mm per year compared to 0.30 mm per year in total immersion. This severity is probably due to the availability of atmospheric oxygen at the water line which acts as an efficient depolariser of the cathodic oxygen reduction reaction.

4. Conclusions

Copper and its alloys were found to be self passivating in strong calcium hypochlorite solution. The passivity broke down in some alloys leading to pitting. In certain other alloys there was a region of active dissolution after transpassivity. Of the remaining alloys, from corrosion current and weight loss data, the nickel tin bronze seem to be the most corrosion resistant. However, this alloy cannot be used under partial immersion conditions as corrosion is very severe. Corrosion under intermittent immersion conditions is only slightly more severe than under total immersion conditions and the nickel tin bronze alloy can be used.

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Alkaloids of Some Plants of Sri Lanka—Chemistry and Pharmacology*

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Abstract : Studies on alkaloids in some plants of Sri Lanka are reviewed with special reference to chemical and biosynthetic aspects. Plant species investigated are grouped into families, a short description of the alkaloid types encountered in a family is given followed by a brief botanical description of the plant(s) studied. Where uses of these plants in indigenous and/or western medicine have been noted an attempt is made to relate these uses to the known pharmacology of their constituent alkaloids.

CONTENTS

*Based on a lecture delivered at a Workshop on Phytochemical, Pharmacological and Microbiological screening of local plants held as a prelude to the 3rd Asian Symposium on Medicinal Plants and Spices, Sri Lanka, Jan.—Feb. 1977.

1. General Introduction

Thanks to its geographical situation and climatic conditions, Sri Lanka has an abundant flora. From a total of 3368 flowering plant species, 830 are reported to be endemic to the country[1,2,3,4,11,164] and of this total around 750 species are claimed to have uses in the indigenous system of medicine.[2,3] A good number of local plants are also used as sources of drugs in Western medical practice. Distribution of these medicinal and/or drug-producing plants of Sri Lanka among the pteridophytes, gymnosperms and angiosperms has been presented by Abeywickrema[3] and is summarised in Table 1. Some of these plants owe their medicinal activity to the alkaloids contained in them.

TABLE 1. Distribution of Medicinal and/or Drug-producing Plants among Pteridophytes, Gymnosperms and Angiosperms in Sri Lanka.

		No. of species used as medicinal drugs			
		Total	Indigenous	Introduced	Endemic
Pteridophytes		5	5	0	0
Gymnosperms		1	1	0	0
Angiosperms	Monocots	125	100	22	3
	Dicots	619	564	44	11
TOTAL :		750	670	66	14

Amongst the natural products, alkaloids comprise the largest single class of secondary plant metabolites. Up-to-date, a total of about 6,500 alkaloids are known and a new alkaloid is discovered at the rate of about one per day. Since they display dramatic physiological activities, alkaloids find wide applications in medicine; some alkaloids are toxic to man and animals. There is no completely satisfactory definition of the term alkaloid. However, according to Harbourne,[77] alkaloids generally include "those basic substances which contain one or more nitrogen atoms, usually in combination as part of a cyclic system".

Alkaloids are widely distributed in the plant kingdom. Hegnauer's estimate of 15 to 20% for the distribution of alkaloids in vascular plants[79] seems to be somewhat high. Recent phytochemical screening programmes have however, suggested 10% to be a more logical estimate representing alkaloid-yielding plant species.[45] Alkaloids occur mostly in higher plants belonging to angiosperm families and are absent or infrequent in the gymnosperms, ferns, mosses and lower plants. However, even in the angiosperm families, alkaloid distribution is very uneven and certain families are characteristically devoid of them. Angiosperm families of Sri Lanka which are particularly rich in alkaloids are presented in Table 2, which also shows the number of genera and species of each of these families found here in Sri Lanka.

In addition, Table 2 gives the number of these species endemic to the country and the number employed in indigenous medicine.[3] Distribution of the endemic plant species in various districts of Sri Lanka has been listed by Sultanbawa and Weerasekara.[156]

TABLE 2. Important alkaloid bearing and medicinally useful plant families of Sri Lanka.

Family	No. reported from Sri Lanka			No. of species used in medicine	
	Genera	Species	Endemic	Non-endemic	Endemic
Amaryllidaceae*,†	4	10	—	5	—
Annonaceae†	17	45	18	2	—
Apocynaceae†	23	31	8	13	2
Compositae	60	115	20	21	1
Convolvulaceae	16	57	3	17	1
Cucurbitaceae	18	33	2	22	—
Euphorbiaceae	46	149	47	39	—
Flacourtiaceae	10	16	10	1	3‡
Graminae*	122	295	21	35	—
Lauraceae†	10	33	23	6	—
Leguminosae	85	283	12	88	—
Liliaceae*	14	18	2	5	—
Loganiaceae†	5	18	10	3	1
Malvaceae	13	44	2	21	—
Menispermaceae†	11	13	—	12	—
Moraceae	11	34	5	20	—
Rubiaceae†	50	158	74	23	1
Rutaceae†	18	40	4	14	—
Solanaceae†	9	29	—	11	—
Verbenaceae	16	36	3	14	—
Zingiberaceae*	12	38	17	13	—

*Monocotyledons †Major alkaloid bearing families

‡According to Attygalle[67] one endemic species, *viz*, *Litsea longifolia* (Nees) Alston is used in native medicine.

Research work carried out on the plants of Sri Lanka in the field of alkaloids, their chemistry and pharmacology are found scattered in the literature. The purpose of this review article is, therefore, to attempt to bring these together and provide relevant information to the researcher exploring into these fields. The information presented here is limited only to the work on those plant species collected from Sri Lanka. The alkaloids occurring in the plant species found in Sri Lanka (not necessarily collected from Sri Lanka) and their pharmacology would, however, be the subject of a future presentation.

In this article, the plant species investigated for alkaloids are grouped under their families and their applications in indigenous medicine, their chemistry, pharmacology and where relevant, biosynthesis, have been considered. An attempt has also been made to relate the known pharmacology of these alkaloid bearing plants and/or their contained alkaloids to the use of these plants or the drugs derived from them in indigenous and/or western medical practices.

2. Surveys for Alkaloids

2.1. Introduction

Prior to undertaking any detailed investigations on a particular class of compounds, it is logical for the natural products investigator to make his selection of plants with the help of preliminary screening or survey programmes. Survey of plants for the occurrence of alkaloids has been made easy as tests for this class of compounds in plant extracts are simple and could be conducted rapidly by reasonably reliable methods.[50]

Two systematic surveys of Sri Lanka plants for alkaloids have been reported.

2.2. A Survey of the Endemic Annonaceae for Alkaloids

In 1973, Sultanbawa, Wannigama and their co-workers reported a general survey of the endemic Annonaceae for alkaloids.[99] In this study, crude basic fractions from leaves and twigs were subjected to Mayer's test[119] and *TLC* examination. The endemic species investigated included *Alphonsea coriacea* (Thwaites) Finet and Gagnep, *Desmos elegans* (Thwaites) Safford, *Enicosanthum acuminatum* (Thwaites) Airy-Shaw, *Sageraea thwaitesii* Hook f. and Thoms., *Xylopia championii* Hook f. and Thoms., and *X. nigricans* Hook f. and Thoms. The following non-endemic species were also tested ; *Cyathocalyx zeylanicum* Champ. ex Hook f. and Thoms., *Miliusa indica* Leschen ex A.DC. and *Uvaria semecarpifolia* Hook. f. and Thoms. All these 9 species had shown the presence of alkaloids and authors have undertaken a fuller investigation into the alkaloids present in the endemic Annonaceae.[99]

2.3. A Screening of 464 Sri Lanka Plants for Alkaloids

An extensive survey of Sri Lanka plants for the occurrence of alkaloids has recently been reported.[157,158] In this study, 464 plant species including 170 endemic species have been investigated. Extracts obtained by 3 different procedures were tested for the presence of alkaloids by Mayer's reagent[119] and the approximate number of alkaloids in each extract determined by *TLC* analyses. The results obtained in this survey are summarised in Table 3. In this study, 128 new plant species including 59 endemics (distributed in 25 families) containing alkaloids have been uncovered.

TABLE 3. Summary of test results from the screening of 464 Sri Lanka plant species for alkaloids.

BOTANICAL COLLECTIONS			TEST RESULTS		
No. of families	Total	96	Meyer's test	No. tested	464
	Tropical	49		No. positive	91
No. of genera		314	Dragendorff (*TLC* method)	No. tested	417
				No. positive*	137
No. of species	Total	464	Iodoplatinate (*TLC* method)	No. tested	214
	Endemics	170		No. positive*	89
			No. positive†	Total	201
				Endemics	59
				New spp.	128

*Excludes doubtfully positive species

†By either Meyer's, Dragendorff or Iodoplatinate tests.

3. Alkaloids of Ancistrocladaceae

3.1. Introduction

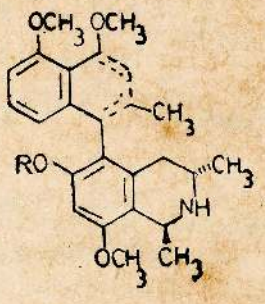
The genus *Ancistrocladus* of the family Ancistrocladaceae has been recently investigated for alkaloids, and some unique isoquinoline alkaloids have been reported.[62,63,64,65,66] Almost all the alkaloids isolated contained a 1,2,3,4-tetrahydroisoquinoline moiety coupled to an oxygenated naphthalene at varying positions of the aromatic ring of the former. Unlike other isoquinoline alkaloids, *Ancistrocladus* alkaloids have a polyketide biogenesis.[63]

In Sri Lanka, the family Ancistrocladaceae is represented by a single genus containing only one species, *Ancistrocladus hamatus* (Cahl) Gilg.[1], which is endemic to the country.[11] It is a very strong creeper and in rural areas it is used to tie cattle, hence the Sinhala name, gona-wal.[12]

3.2. Alkaloids of *Ancistrocladus hamatus*

Govindachari's group in the course of their studies on isoquinoline alkaloids in plants belonging to the genus *Ancistrocladus*,[62,63,64,65,66] have investigated roots of *A. hamatus* obtained from Sri Lanka.[66] In addition to ancistrocladine (I) which has already been isolated from several members of this genus, a new alkaloid named hamatine (III) was also isolated from the Sri Lankan species. The two alkaloids were present to the extent of 0.50 and 0.06% respectively. It has been shown that the product derived from *O*-methylhamatine (IV) by dehydrogenation is enantiomeric with the isoquinoline made from *O*-methylancistrocladine (II) by the same process. This finding coupled with the *NMR* and *CD* (Circular Dichroism) data suggested ancistrocladine and hamatine to be isomeric with each other, the only difference being the relative orientation of the substituted naphthalene ring.

(I) : R = H
(II) ; R = CH_3

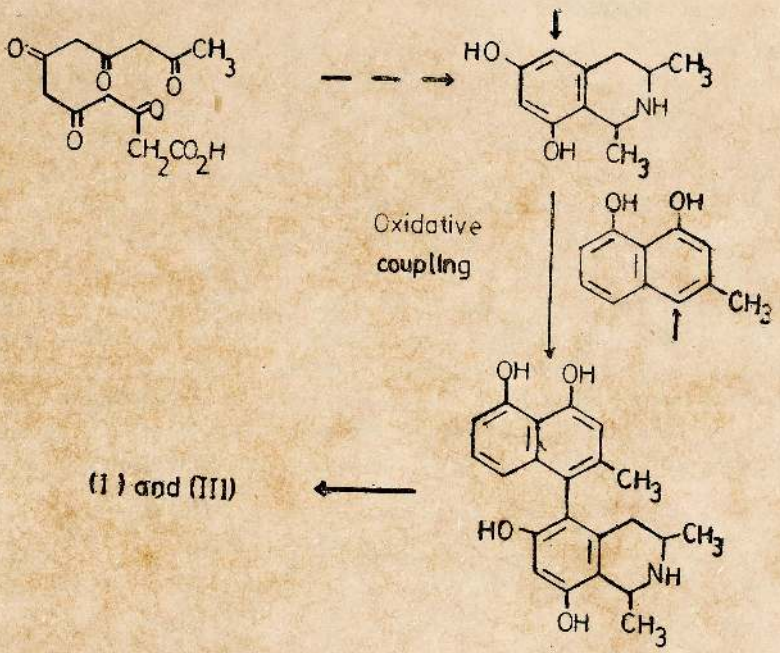


(III) ; R = H
(IV) ; R = CH_3

Although the 2 alkaloids from *A. hamatus* have not been subjected to any pharmacological evaluation, a related alkaloid ancistrocladidine from the Indian species *A. heyneanus* Wall. had shown spasmolytic activity on the isolated guinea pig ileum at a concentration of 5μg/ml, comparable with that of papaverine.

3.3. Biosynthesis of Hamatine and Ancistrocladine

Isoquinoline alkaloids are biogenetically derived from aromatic amino acids such as phenylalanine or tyrosine.[13] However, the rare oxygenation pattern in *Ancistrocladus* alkaloids suggested that the tetrahydroisoquinoline moiety in these has a polyketide origin.[63] Subsequent oxidative coupling of the tetrahydroisoquinoline with an oxygenated naphthalene accounts for the formation of all known *Ancistrocladus* alkaloids. Biogenetic origin of these alkaloids is given in Scheme 1. This polyketide biogenetic hypothesis is supported by the isolation of alkaloids coupled at other positions of the benzene ring in the tetrahydroisoquinoline moiety.[65]



Scheme 1. Biosynthetic origin of Hamatine and Ancistrocladine.

4. Alkaloids of Apocynaceae

4.1. Introduction

Botanically, the Apocynaceae or the dog-bane family is divided into 3 sub-families: Cerberoideae, Echitoideae (Apocynoideae) and Plumericideae. The plants of this family contain well over 250 alkaloids, majority having an isoprenoid origin. For example, the first two sub-families produce steroidal alkaloids, whereas the sub-family Plumerioideae contains indole alkaloids derived from the amino-acid tryptophan and a terpenoid moiety. The steroidal alkaloids are of the types aminosteroids and aminoglycosteroids. However, the genus *Holarrhena* in the sub-family Plumerioideae is an exception as the plants of this genus bear only steroidal alkaloids and as they do not contain indole alkaloids of this sub-family.[167]

The family Apocynaceae is well known for skynanthine-type monoterpene alkaloids, pregnane-type steroidal alkaloids and complex indole alkaloids. A number of these alkaloids bear pharmacological activity and have become indispensable drugs in western medicine ; e.g. complex indole alkaloids of *Rauwolfia* have anti-hypertensive action[139] and the *bis*-indole alkaloids of *Catharanthus roseus* are unique in their antileukemic activity[118] (see below).

4.2. *Holarrhena* Alkaloids

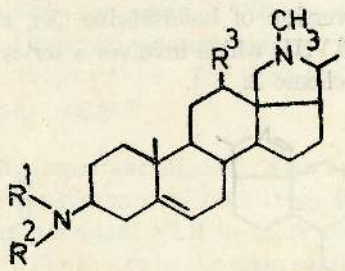
The genus *Holarrhena* in Sri Lanka contains a single species *H. mitis* (Vahl) R. Br. ex Roen and Schult. (*Sinh.*— Kiriwalla, Kirimawaran ; *Tam.*—Vellupalai) which is endemic to this country.[1,11] The bark juice of this plant under the name of Kalindu is reputed in native medicine as a remedy for dysentery and fevers.[95] Alkaloids of *H. mitis* had been the subject of a series of papers by Wannigama, Goutarel and their co-workers.[8,19,27,89,101,102] Alkaloids isolated from various parts of this plant by these workers are presented in Table 4. In addition to the isolation of conessine (V) from the bark of *H. mitis* during their very early investigations, Wannigama and Bhavanandan obtained evidence for the occurrence of *N*-demethylated conessines.[19] Further investigations had confirmed the occurrence of a large number of steroidal alkaloids in the bark extracts of this plant. The seeds of *H. mitis* have been shown to be devoid of mitiphylline (XV). Isolation of triacanthine (XIV) from these seeds marks the first occurrence of this base in any seed. The alkaloidal extract of the fruit pericarp was shown to constitute only of triacanthine.

TABLE 4. Alkaloids of *H. mitis*.

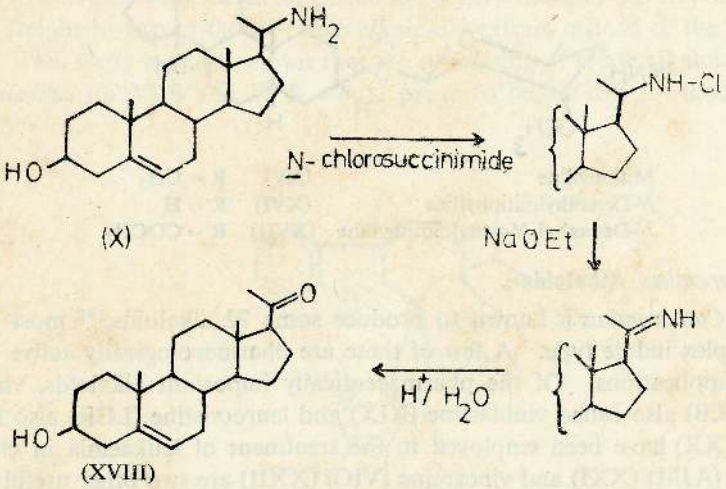
Plant part	Alkaloid(s) isolated	Yield (%)	Ref.
Bark	conessine (V)	—	19
Bark	conessine (V)	0.098	27
	iso-conessimine (VI)	0.098	27
	holadienmine (IX)	0.013	27
	conkurchine (XIII)	0.020	27
	holarrhenine (VII)	0.026	27
	holafebrine (X)	0.098	27
	holarrhimine (XI)	0.098	27
	N-3-methyl-holarrhimine (XII)	0.098	
Leaves	mitiphylline (XV)	[40(65]*†	89, 102
	N-desmethylmitiphylline (XVI)	[9(10]*†	89, 102
	triacanthine (XIV)	[45(20]*†	89, 102
Seeds	triacanthine (XIV)	0.080	101
	conamine (VIII)	0.360	101
	conessine (V)	0.160	101
	iso-conessimine (VI)	0.016	101
Fruits (pericarp)	triacanthine (XIV)	0.400	101

*Ref. 89,
†Represented as percentage of the total bases.

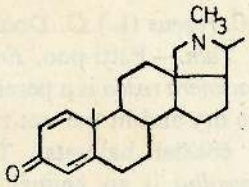
TABLE 5. Some pregnane type steroidal alkaloids of *H. mitis*.



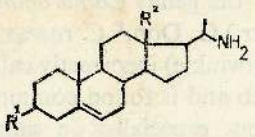
		R^1	R^2	R^3
Conessine	(V)	CH_3	CH_3	H
Iso-conessimine	(VI)	CH_3	H	H
Holarrhenine	(VII)	CH_3	CH_3	OH
Conamine	(VIII)	H	H	H



Scheme 2. Chemical transformation of Holafebrine to Pregnenolone.

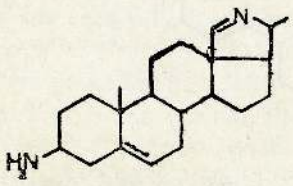


(IX) Holadienine

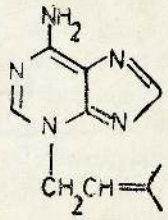


		R^1	R^2
(X)	Holafebrine	OH	CH_3
(XI)	Holarrhimine	NH_2	CH_2OH
(XII)	*N*-3-Methyl-holarrhimine	$NHCH_3$	CH_2OH

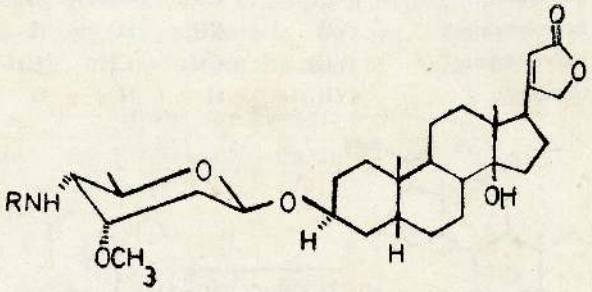
Much attention has been focussed on the steroidal alkaloids of *Holarrhena* spp. with the intention of economic exploitation of these in the synthesis of valuable steroidal hormones.[61] Conversion of holarfebrine (X), an alkaloid of Sri Lankan *H. mitis*, to pregnenolone (XVIII) which involves a series of simple chemical transformations is depicted in Scheme 2.



(XIII) Conkurchine



(XIV) Triacanthine



Mitiphylline	(XV)	R = CH_3
N-Demethylmitiphylline	(XVI)	R = H
N-Demethyl-*N*-acetylmitiphylline	(XVII)	R = $COCH_3$

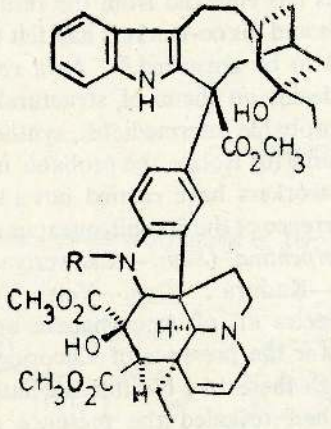
4.3. *Catharanthus* Alkaloids

The genus *Catharanthus* is known to produce some 72 alkaloids,[162] most of which are of complex indole type. A few of these are pharmacologically active and have medicinal applications. Of the pharmaceutically important alkaloids, vincaleukoblastine (VLB) also called vinblastine (XIX) and leurocristine (LCR) also known as vincristine (XX) have been employed in the treatment of leukaemia in children.[161] Ajmalicine (AJM) (XXI) and vincamine (VIC) (XXII) are two other useful alkaloids reported from *Catharanthus* species.[46]

In Sri Lanka, the genus *Catharanthus* has two species, *C. roseus* (L.) G. Don and *C. pusillus* (Murr.) G. Don.[1] *C. roseus* (*Sinh.*—Mini-mal; *Tam.*—Patti-poo, *Eng.*—Madagascar Periwinkle) incorrectly called *Vinca rosea* or *Lochnera rosea* is a perennial herb or subshrub and is found commonly as a weed in the dry and in the wet zones at low elevations, especially on sandy soil and in the coastal habitats. Three different forms of *C. roseus* have been reported.[85] *C. pusillus* is an annual herb indigenous to Sri Lanka and India. In Sri Lanka, it is a rather rare weed on cultivated land and has been recorded exclusively from Batticaloa and Jaffna districts.[107]

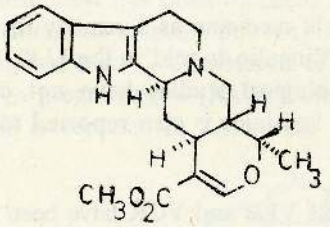
C. roseus, in the form of a tea, is used in folk medicine as a remedy for diabetes and an infusion of leaves under the name of Vinculin is sold in the U.K. as a cure for the same ailment.[35] However, pharmacological studies have not confirmed this alleged hypoglycaemic activity.[47,162] The leaf-juice is also reported to be used as an application for wasp stings.[35]

The two pharmaceutically important alkaloids VLB and VCR have been obtained from *C. roseus*. Although the synthesis of both these alkaloids[130] and the chemical transformation of the more abundant VLB to more potent antileukaemic alkaloid VCR is known, *C. roseus* still remains to be the main source of this valuable alkaloid.[46] Therefore, this plant is at present uprooted and exported by local agents to drug manufacturers in Europe for the extraction of these alkaloids.[9] Realisation of the disadvantages of this practice had prompted investigation into the feasibility of extracting these alkaloids from *C. roseus* (and *C. pusillus*) with locally available solvents and facilities with the hope of obtaining better returns to the country from this non-traditional export.[70,152] This work has led to 2 important findings, *viz.* the possibility of harvesting the leaves for extraction at periodic intervals and the reduction in cost of freight by exporting the crude alkaloidal extract instead of the dried plant material. This study has also shown that the processing of crude alkaloidal mixture from *C. pusillus* for VLB and VCR would prove to be less difficult than that from *C. roseus*.

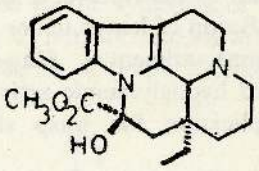


(XIX) Vincaleukoblastine (VLB) R = CH_3

(XX) Leurocristine (LCR) R = CHO



(XXI) Ajmalicine (AJM)



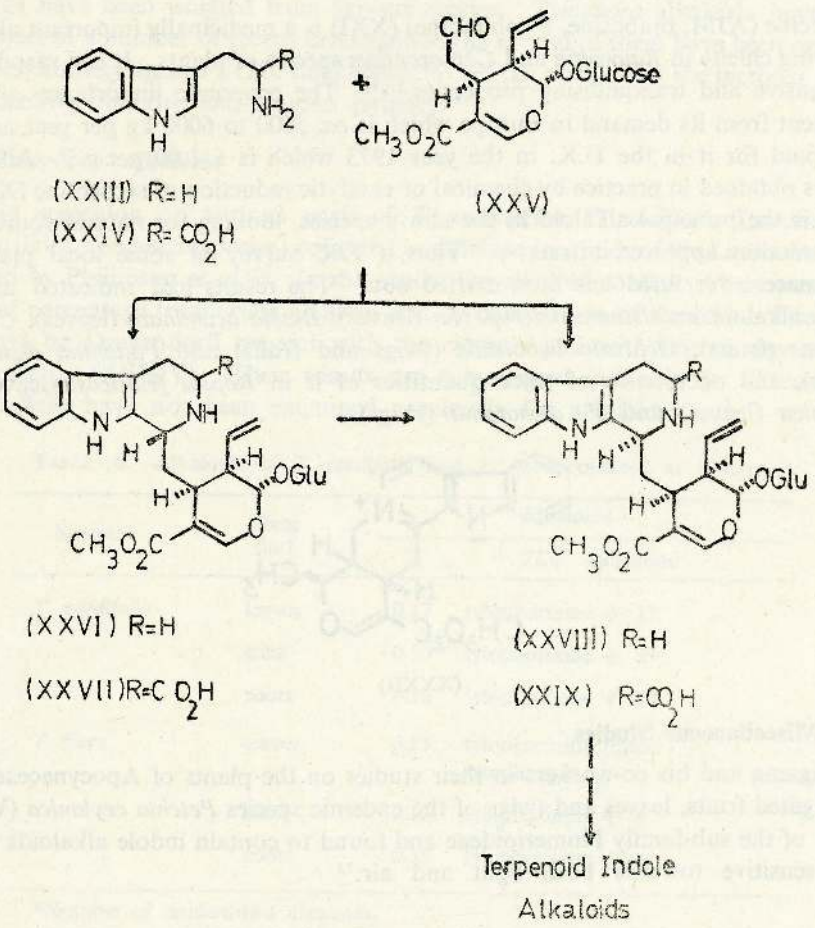
(XXII) Vincamine (VIC)

4.4. Pharmacology of *C. roseus* Alkaloids

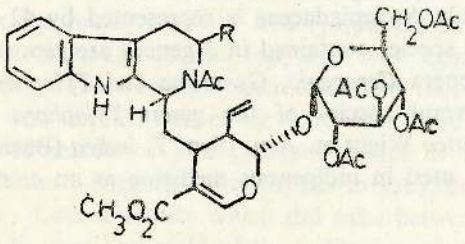
Two important drugs, Oncovin and Velban, produced by Eli-Lilly Co. in the U.S. contain the sulphates of VCR and VLB respectively.[114] Oncovin arrests mitosis in metaphase and is known to be effective in the treatment of leukaemia in children, lymphosarcoma, reticulum sarcoma, neuroblastena, Wilm's tumour and tumours of the breast, brain and lungs. Velban is a cytotoxic drug arresting cell growth in the metaphase and is effective in the treatment of Hodgkin's disease and other lymphomas and choriocarcinomas.

4.5. Screening for Biosynthetic Intermediates

The first step in the biosynthesis of terpenoid indole alkaloids, the major group of alkaloids in Apocynaceae (and Rubiaceae) involves condensation of tryptophan (XXIV) or its decarboxylation product, tryptamine (XXIII) with secologanin (XXV) having monoterpene origin (Scheme 3). Although a satisfactory biogenetic theory for terpenoid indole alkaloids has emerged from the results of extensive radioactive labelling experiments, de Silva and his co-workers had felt that a number of important questions in this theory had to be answered.[37] As a result they have approached the problem in order to (*a*) decide on chemical, structural and biosynthetic grounds, the compounds that can be probable intermediates, synthesise them and test them as precursors of the alkaloids, and (*b*) isolate the probable intermediates and test them as precursors. Thus, these workers have carried out a screening of indole alkaloid producing plants for the occurrence of the first nitrogenous monoterpenoid precursor.[36] In this survey, *Rauwolfia serpentina* (*Sinh.*— Ekaweriya, *Tam.*— Sorannamilbori), *Strychnos nux-vomica* (*Sinh.*—Kaduru ; *Tam.*—Yetti, Yettie-kottai) and a number of *Mitragyna* and *Vinca* species all of Apocynaceae and *Cinchona ledgeriana* of Rubiaceae have been tested for the presence of vincoside (XXIV) and 5α-carboxy-vincoside (XXVII). Although these two bio-intermediates have not been detected, macro-isolation techniques had revealed the presence of 5α-carboxystrictosidine (XXIX), an isomer of (XXVII), in all the plants tested and strictosidine (XXVIII), an isomer of (XXVI), only in *Rauwolfia*, *Vinca* and *Strychnos* species. Strictosidine (XXVIII) and its 5α-carboxy derivative (XXIX) have been isolated from *R. serpentina* and *S. nux-vomica* and were characterised as the pentaacetate (XXX) and the methoxycarbonyl pentaacetate (XXXI) respectively.



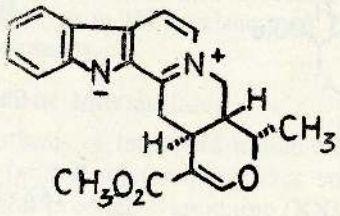
Scheme 3. Early stages in the Biosynthesis of Terpenoid Indole Alkaloids.



(XXX) R = H
(XXXI) R = CO_2CH_3

4.6. Screening for Ajmalicine

Ajmalicine (AJM, raubasine, δ-yohimbine) (XXI) is a medicinally important alkaloid occurring chiefly in *Rauwolfia* and *Catharanthus* species of plants. It has vasodilator, hypotensive and tranquillising properties.[21,35] The economic importance of AJM is evident from its demand in Europe which is *ca.* 5000 to 6000 kg per year and the price paid for it in the U.K. in the year 1973 which is £ 1.00 per g.[21] Although AJM is obtained in practice by chemical or catalytic reduction of serpentine (XXXII) which is the principal alkaloid in the above species, looking for natural sources for its extraction appeared attractive. Thus, a *TLC* survey of some local plants of Apocynaceae for AJM has been carried out.[7] The results had indicated absence of this alkaloid in *Alstonia macrophylla* (leaves), *Bassia acuminata* (leaves), *Cerbera manghas* (fruits), *Ochrosia bobornica* (twigs and fruits) and *Pajiantha dichotoma* (leaves), and occurrence of trace quantities of it in *Kopsia fruiticosa* (leaves), *O. bobornica* (leaves) and *P. dichotoma* (twigs).



(XXXII)

4.7. Miscellaneous Studies

Wannigama and his co-workers in their studies on the plants of Apocynaceae have investigated fruits, leaves and twigs of the endemic species *Petchia ceylanica* (Wight) Livera of the sub-family Plumerioideae and found to contain indole alkaloids which were sensitive towards both light and air.[73]

5. Alkaloids of Asclepiadaceae

5.1. Introduction

In Sri Lanka, the family Asclepiadaceae is represented by 42 species belonging to 24 genera,[1] of which 6 species contained in 3 genera are reported to be endemic to the country.[11] The genera *Ceropegia*, *Gymnema* and *Tylophora* each contains two endemic species.* Several species of the genus *Tylophora* have folk-medicinal reputation. *T. asthmatica* Wight et. Arn. (Syn. *T. indica* (Burm.) Merr.), known as Indian Ipecacuanha is used in indigenous medicine as an emetic, expectorant and anti-dysenteric.[112,133]

*However, according to a recent revision[86], Sri Lanka has 37 species in 21 genera of which 3 species are endemic.

The genus *Tylophora* comprises some 50 species and phenanthrene-indolizidine alkaloids have been isolated from several species. *Tylophora* alkaloids have been the subject of a number of recent investigations as several of them have been reported to possess activity against l-1210 Leukaemia,[40,54,132] the position of the methoxy group been decisive for the anti-tumour potency.[141]

5.2. *Tylophora* alkaloids

Alkaloid screening on various parts of *T. asthmatica*, *T. cordifolia* Thwaites and *T. flava* Trim. (endemic species) collected in different localities of Sri Lanka has been reported by Phillipson *et al.*[126] In this study, the alkaloid contents were assessed in terms of percentage total crude alkaloid and by comparison of the colour intensities produced by Dragendorff reagent with the extracts and with known amounts of tylophorinine (XXXIV). Their results are summarised in Table 6. *T. cordifolia* and *T. flava* have not been examined previously for alkaloids.

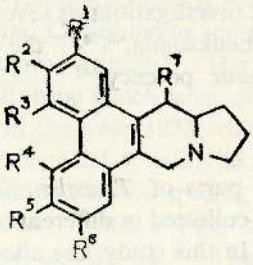
TABLE 6. Alkaloids of *T. cordifolia* and *T. flava* collected at Matara.

Species	Plant part	Alkaloids	
		%	*TLC* indication
T. cordifolia	leaves	0.17	tylophorinine + 1*
	stem	0.10	tylophorinine + 2*
	roots	0.16	tylophorinine + 3*
T. flava	leaves	0.15	tylophorinine (major) + tylophorine
	stem	0.12	tylophorinine + 4*
	roots	0.15	tylophorinine + 4*

*Number of unidentified alkaloids.

Eight samples of *T. asthmatica* collected from different localities were also screened by the same group of workers for the content of alkaloids.[126] *TLC* has indicated that in most samples the major alkaloid was tylophorinine (XXXIV) in at least one part of the plant, although in one leaf and two stem samples tylophorine (XXXIII) predominated. Their results contrasted with those previously reported for Indian plants[67,133] and implied that either there were some variations in alkaloid content from season to season or that different strains of *T. asthmatica* existed. However, these authors have suggested that further investigations would be required to verify these points. One similarity which did exist between the Indian and Sri Lankan materials was the very low yield of the antileukaemic alkaloid tylophorinine (XXXIV).[116]

TABLE 7. Alkaloids of *Tylophora* species.



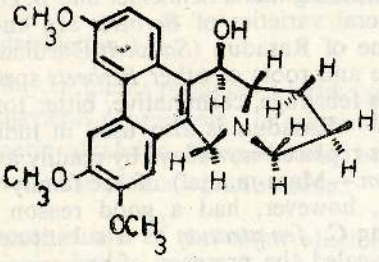
		R^1	R^2	R^3	R^4	R^5	R^6	R^7
Tylophorine	(XXXIII)	OCH_3	OCH_3	H	H	OCH_3	OCH_3	H
Tylophorinine	(XXXIV)	H	OCH_3	H	H	OCH_3	OCH_3	OH
Tylophorinidine	(XXXV)	H	OCH_3	H	H	OH	OCH_3	OH
Tylocrebine	(XXXVI)	OCH_3	OCH_3	H	OCH_3	OCH_3	H	H
Isotylocrebine	(XXXVII)	H	OCH_3	OCH_3	H	OCH_3	OCH_3	H

A detailed isolation work of a large batch of *T. asthmatica* was also carried out by Phillipson's group. Their results are given in Table 8 along with the alkaloids isolated from the Indian species by previous workers. A detailed analysis of the *NMR* spectrum of tylophorinine (XXXIV) has aided in the postulation of the preferred conformation of this alkaloid as given in (XXXVIII).

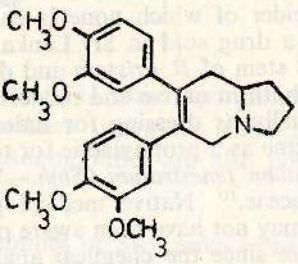
TABLE 8. Alkaloids of *T. asthmatica* from India and Sri Lanka.

Origin	Alkaloids isolated	Ref.
India	tylophorine (XXXIII), tylophorinine (XXXIV), tylophorinidine (XXXV), isotylocrebine (XXXVII), septicine (XXXIX), alkaloids A*, B* and C*	67, 68, 138
Sri Lanka	tylophorine (XXXIII), tylophorinine (XXXIV), tylophorinidine (XXXV)	126

*Unidentified alkaloids



(XXXVIII)



(XXXIX)

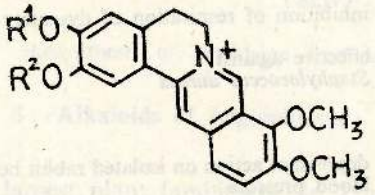
5.3. Pharmacology of *Tylophora* species

In addition to the above mentioned uses, *T. asthmatica* has been employed widely in folk medicine to cure asthma, one leaf per day being taken during a period of 6 days. Recent clinical trials by Shivpuri's group while proving the validity of its use in treating asthma has also shown it to be active against allergic rhinitis.[151] Preliminary clinical studies have also indicated that tylophorine (XXXIII) could be the active constituent in this plant.

6. Alkaloids of Berberidaceae

6.1. Introduction

The family Berberidaceae has yielded a total of more than 50 alkaloids, many of which are from the genera *Berberis*, *Leontice* and *Nandina*.[129] These alkaloids are of the types : aporphine, bisbenzylisoquinoline, oxoaporphine, protoberberinium and tetrahydroisoquinoline. The pharmacologically important protoberberinium alkaloid, berberine (XL) and its close relatives palmatine (XLI) and jatrorrhizine (XLII) are known to occur in *Berberis*, *Mahonia* and *Mandina* species.[129] These three alkaloids have a wide distribution in the plant kingdom and have been reported from Annonaceae (1 genus), Menispermaceae (8 genera), Rananculaceae (5 genera), Papaveraceae (8 genera) and Rutaceae (3 genera).[80]



		R^1	R^2
Berberine	(XL)	$-CH_2-$	
Palmatine	(XLI)	CH_3	CH_3
Jatrorrhizine	(XLII)	H	CH_3

In Sri Lanka, the family Berberidaceae is represented by a single genus *Berberis* which has three species,[1] *B. tinctoria* Leschen, *B. wightiana* Schneider and *B. ceylanica* Schneider of which none is endemic. Several varieties of *Berberis* are known to yield a drug sold in Sri Lanka by the name of Rasadun (*Sanskrit*-Daruharidra).[10] Dried stem of *B. aristata* and dried rhizome and roots of other *Berberis* species are used both in native and modern medicine as febrifuge, carminative, bitter tonic and externally as dressing for indolent ulcers.[113] Rasadun is also used in indigenous medicine as a prophylactic for tetanus and is replaced nowadays by readily available *Coscinium fenestratum* (*Sinh.*—Veniwal, *Tam.*—Mara-manjal) of the family Menispermaceae.[10] Native medical practitioners, however, had a good reason though they may not have been aware of it, for using *C. fenestratum* as a substitute for *B. aristata* since the chemical analysis had revealed the presence of berberine (XL)[163] which is known to be the active principle in both species.

6.2. Alkaloids of *Berberis tinctoria*

B. tinctoria collected from Nuwara Eliya has recently been analysed by Shamma and his co-workers at the Pennsylvania State University. Their work has shown the presence of 12 alkaloids in the basic fraction of which two were new.[144] The alkaloids identified included simple tetrahydroisoquinolines, aporphines, oxoaporphines, protoberberinium salts and bisbenzylisoquinolines. A new protoberberinium having an unusual substitution pattern has been isolated and named lankamine.

6.3. Pharmacology of Berberine and *Berberis* species

The known pharmacology of *Berberis* species could be related to the presence of berberine in them. Berberine (XL) has been used internally as an anti-malarial, febrifuge, carminative and externally as dressing for indolent ulcers.[113] Pharmacological evaluation of several *Berberis* species, berberine and other plants known to contain this alkaloid and related alkaloids have been reported and an extensive review on alkaloids of the genus *Berberis* and their pharmacological action has recently appeared.[42] In addition to the above, some other properties are also observed with either *Berberis* species or berberine and a summary of these are presented in Table 9.

TABLE 9. Pharmacological action of *Berberis* spp. and berberine.

Sample tested	Pharmacological action	Ref.
B. amurensis roots	depression effects in cats and vasoconstriction in isolated rabbit ears	41
Berberis spp. and berberine	inhibition of respiration of dysentery bacteria	120
Berberine, palmatine and jatrorrhizine and extracts of *Coptis japonica* (Ranunculaceae).	effective against *Staphylococcus aureus*	75
B. lysinum roots	depressant action on isolated rabbit heart, acute fall in blood pressure	128
Berberine sulphate from *B. vulgaris* rhizome cortex	bactericidal on *Staphylococci*	98

7. Lauraceae Alkaloids

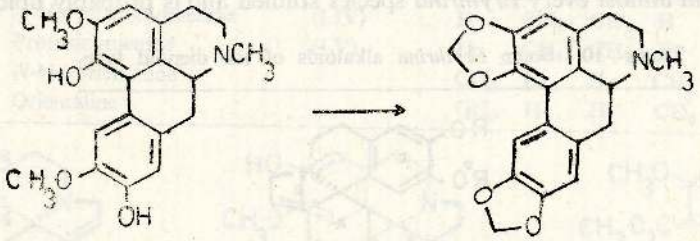
7.1. Introduction

This family of plants produces 5 basic types of alkaloids,[129] of which aporphines and their biogenetic progenitors, tetrahydrobenzylisoquinolines occupy a chief place. The dimeric alkaloids, bisaporphines and bisbenzylisoquinolines are also not uncommon in the plants of Lauraceae.

Lauraceae is one of the major alkaloid bearing families of Sri Lanka which has 23 endemic species out of the total of 33 species available.[11] In addition to 6 medicinally useful plants, this family contains the economically important cinnamon (*Cinnamomum zeylanicum* Bl.) plant.[170]

7.2. Alkaloids of *Neolitsea fuscata*

The genus *Neolitsea* of the family Lauraceae has 2 species in Sri Lanka,[1] *N. fuscata* (Thwaites) Alston and *N. Cassia* (L) Kosterman (= *N. involucrata* Alston) of which the former is endemic to the country.[11] Although *N. fuscata* has no reported medicinal uses, some other species of this genus have reputed medicinal activity.[96] A number of aporphine alkaloids have been isolated from plants of the genus *Neolitsea*.[87,117] Investigation of *N. fuscata* stem bark collected at Hakgala has shown the presence of aporphine base, isoboldine (XLIII) as the major alkaloid,[72] which represented the first report of its occurrence in this genus. Presence of isoboldine in this genus is significant since it could be regarded as the biosynthetic precursor[14] of neolitsine (XLIV), an aporphine base isolated from *N. pulchella*[87] (see Scheme 4).



(XLIII) (XLIV)

Scheme 4. Biosynthesis of Neolitsine from Isoboldine.

8 Alkaloids of Leguminosae

8.1. Introduction

Leguminosae, one of the largest plant families of Sri Lanka contains 283 species distributed in 85 genera,[1] of which 12 species are endemic to the island,[11] and 88 of these species find applications in indigenous medicine.[3] Although Leguminosae has no alkaloids characteristic of the family *Crotalaria* and *Erythrina*, two of the

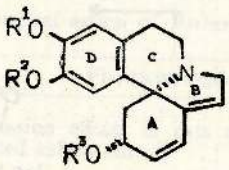
genera found in Sri Lanka contain alkaloids characteristic of these genera. The former genus bears alkaloids of pyrrolizidine type and the latter a spiroamine type (see below). The indole base, hypaphorine (LX) is recorded from a number of *Erythrina* and *Acacia* species.

8.2. *Erythrina* alkaloids

Four species of *Erythrina*, *viz. E. variegata* (= *E. indica*), *E. fusca*, *E. lithosperma* (= *E. subumbrans*), and *E. suberosa* have been reported from Sri Lanka.[164] Of these, various parts of *E. variegata* L. (*Sinh.*—era-badu, era-mudu ; *Tam.*—mullu murukku) are known to have an array of medicinal applications; the bark is used as an astringent and a febrifuge, in liver trouble and in epilepsy, as a nervine sedative an anti-asthmatic and as a collyrium in ophthalmia ; the leaves are used as a stomachic and diuretic and for relieving pain in joints[30] the juice is applied to syphilitic buboes. *E. variegata* is also used both as a lactogogue and an emmenagogue and is an ingredient in many decoctions.[10]

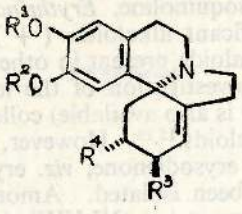
Up-to-date over 30 *Erythrina* alkaloids are known.[78] These are conveniently classified into 2 main structural types, those containing a conjugated diene in the A and B rings (Table 10) and those containing a $\Delta^{1,6}$ double bond in the ring A (Table11). A third group of alkaloids (LIV to LX) with various structural features includes α—and β—erythroidines and erythroculine (LIX), an *Erythrina* alkaloid isolated from *Coculus laurifolious* of the family Menispermaceae.[88] A number of other alkaloids not belonging to the *Erythrina* group have also been isolated from various *Erythrina* species. These are mainly the tetrahydrobenzyl-isoquinoline type (Table 12) and the indole type (eg. hypaphorine). Hypaphorine (LX) has been reported to be present in almost every *Erythrina* species studied and is probably ubiquitous.[78]

TABLE 10. Some *Erythrina* alkaloids of the dienoid type.



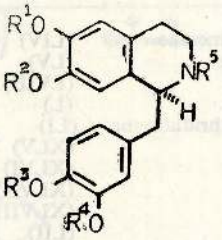
		R^1	R^2	R^3
Erysopine	(XLV)	H	H	CH_3
Erythraline	(XLVI)	—CH_2—		CH_3
Erysodine	(XLVII)	H	CH	CH_3
Erysotrine	(XLVIII)	CH_3	CH_3	CH_3
Erysonine	(XLIX)	H	CH_3	H

TABLE 11. Some *Erythrina* alkaloids of the alkenoid type.

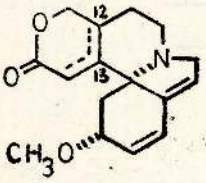


		R^1	R^2	R^3	R^4
Erythratinone	(L)	CH_3	CH_3	=O	OCH_3
3-Demethoxyerythratidinone	(LI)	CH_3	CH_3	=O	H
Erythratine	(LII)	—CH_2—		OH	OCH_3
Erythramine	(LIII)	—CH_2—		H	OCH_3

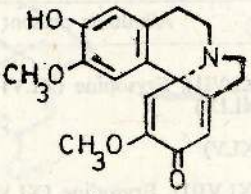
TABLE 12. Some tetrahydrobenzylisoquinoline alkaloids of *Erythrina* species.



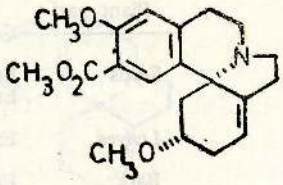
		R^1	R^2	R^3	R^4	R^5
N-Norprotosinomenine	(LIV)	H	CH_3	CH_3	H	H
Protosinomenine	(LV)	H	CH_3	CH_3	H	CH_3
N-Nororientaline		CH_3	H	H	CH_3	H
Orientaline		CH_3	H	H	CH_3	CH_3



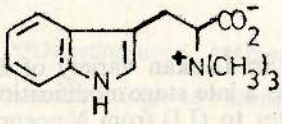
(LVI) Δ^{12}
(LVII) Δ^{13}



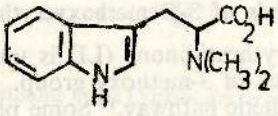
(LVIII)



(LIX)



(LX)



(LXI)

The pods of the Indian variety of *E. lithosperma* Blume has yielded 12 alkaloids[56] belonging to 3 classes, benzylisoquinoline, *Erythrina* and indole (see Table 13) including 2 biogenetically significant alkaloids (+) *N*-norprotosinomenine (LIV) and erysodienone (LVIII). Alkaloids present in other parts of the Indian plant are summarised in Table 14.[53,60] Investigation of the leaves of *E. lithosperma* Blume (smooth variety, a thorny variety is also available) collected in Sri Lanka has revealed the absence of the latter 2 alkaloids.[15,16] However, from the local species 2 new alkaloids structurally related to erysodienone, *viz.* erythratidinone (L) and 3-demethoxyerythratidinone (LI) have been isolated. Among the other alkaloids present were erythratine (XLVI) and erysotrine (XLVIII) (see Table 13).

Chemical analysis of the leaves of *E. fusca* Lour. from Sri Lanka has revealed the presence of only erysotrine (XLVIII) in isolable quantities.[15]

TABLE 13. Alkaloids of Indian and Sri Lankan varieties of *E. lithosperma* Blume.

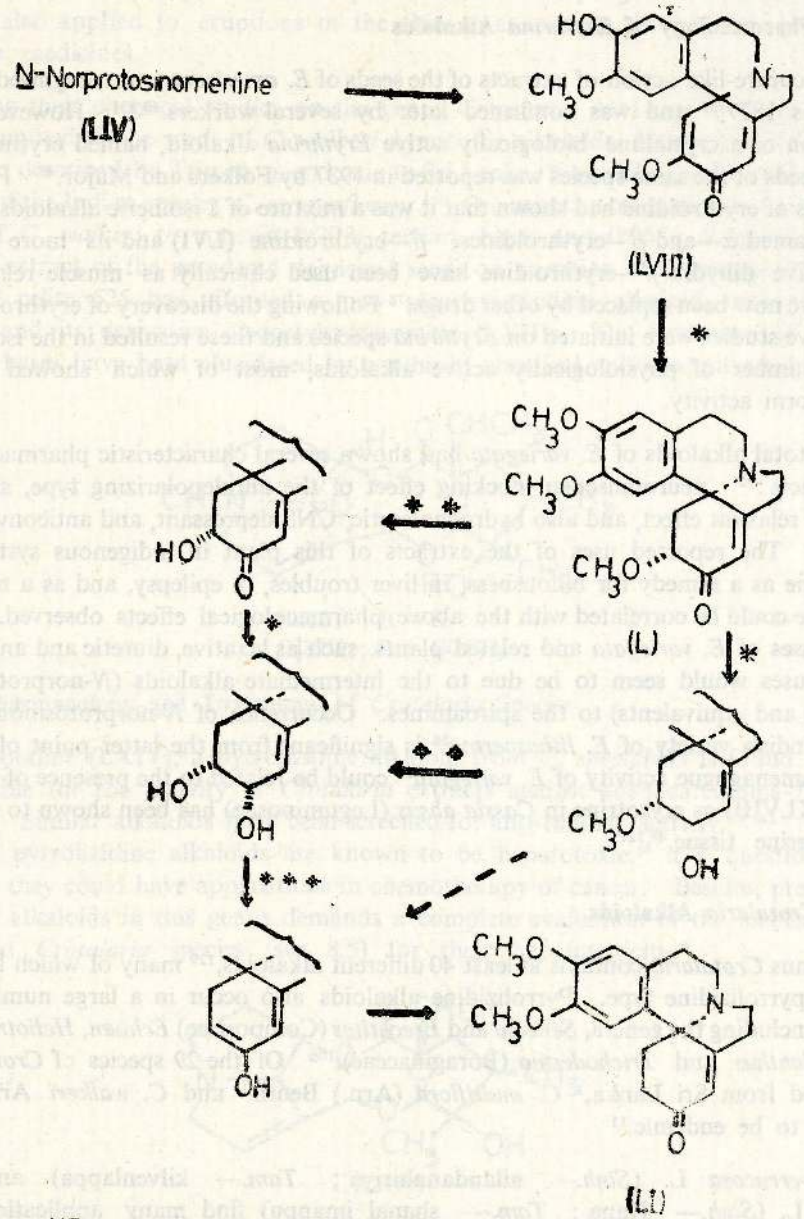
Alkaloid			Pods of Indian variety ($\times 10^{-5}$%)	leaves of Sri Lankan variety ($\times 10^{-2}$%)
Class	Identity			
Benzylisoquinoline	*N*-Norprotosinomenine	(LIV)	6.7	—
	Protosinomenine	(LV)	2.8	—
Erythrina	Erysodienone	(LVIII)	1.8	—
[Rearranged benzyl	Erythratidinone	(L)	—	12.0
soquinoline or	3-Demethoxyerythratidinone	(LI)	—	1.6
spiroamine]	Erysopine	(XLV)	4.8	—
	Erythraline	(XLVI)	5.8	Trace
	Erysodine	(XLVII)	7.6	—
	Erysotrine	(XLVIII)	5.2	22.0
	Erythratine	(LII)	1.6	—
	Erythramine	(LIII)	1.4	—
	β-Erythroidine	(LVI)	2.7	—
Indole	*N*, *N*-Dimethyltryptophan	(LXI)	3.7	—
	Hypaphorine	(LX)	1.9	—

TABLE 14. Alkaloids of various parts of the Indian *E. lithosperma*.

Plant part	Alkaloids present
Seeds	Erysodine (XLVII), Erysopine (XLV) Erysonine (XLIX)
Leaves	Erysopine (XLV)
Bark	Erysotrine (XLVIII), Erysodine (XLVII), Erythramine (LIII), Erythratine (LII)

8.3. Biosynthesis of 3-Demethoxyerythratidinone

3-Demethoxyerythratidinone (LI) is unique in the Sri Lankan variety of *E. lithosperma* in its lack of 3-methoxy group. This implies a late stage modification of the normal biosynthetic pathway.[15] Some plausible routes to (LI) from *N*-norprotosinomenine (LIV) *via* erysodienone (LVIII) is depicted in Scheme 5.



*Reduction ; **Demethylation ; ***Dehydration.

Scheme 5. Biosynthetic routes to 3-Demethoxyerythratidinone in Sri Lankan *Erythrina lithosperma*

8.4. Pharmacology of *Erythrina* Alkaloids

The curare-like action of extracts of the seeds of *E. americana* was recognised as far back as 1877,[39] and was confirmed later by several workers.[49,131] However, first isolation of a crystalline biologically active *Erythrina* alkaloid, named erythroidine from seeds of the same species was reported in 1937 by Folkers and Major.[49] Further analysis of erythroidine had shown that it was a mixture of 2 isomeric alkaloids which were named α—and β—erythroidines. β—erythroidine (LVI) and its more potent derivative dihydro-β—erythroidine have been used clinically as muscle relaxants, but have now been replaced by other drugs.[78] Following the discovery of erythroidines, intensive studies were initiated on *Erythrina* species and these resulted in the isolation of a number of physiologically active alkaloids, most of which showed some curariform activity.

The total alkaloids of *E. variegata* had shown several characteristic pharmacological effects :[55] neuromuscular blocking effect of the antidepolarizing type, smooth muscle relaxant effect, and also hydrocholeretic, CNS depressant, and anticonvulsant effects. The reported uses of the extracts of this plant in indigenous system of medicine as a remedy for biliousness, in liver troubles, in epilepsy, and as a nervine sedative could be correlated with the above pharmacological effects observed. The other uses of *E. variegata* and related plants, such as laxative, diuretic and antiasthmatic uses would seem to be due to the intermediate alkaloids (*N*-norprotosinomenine and equivalents) to the spiroamines. Occurrence of *N*-norprotosinomenine in the Indian variety of *E. lithosperma*[56] is significant from the latter point of view. The emmenagogue activity of *E. variegata*[10] could be related to the presence of erysotrine (XLVIII) as erysotrine in *Cassia absus* (Leguminosae) has been shown to stimulate uterine tissue.[48,127]

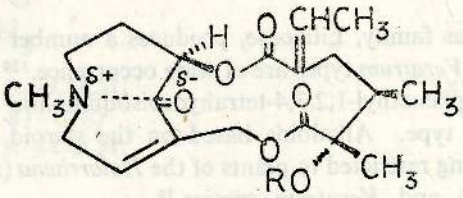
8.5. *Crotalaria* Alkaloids

The genus *Crotalaria* contains at least 40 different alkaloids,[129] many of which belong to the pyrrolizidine type. Pyrrolizidine alkaloids also occur in a large number of plants including the genera, *Senecio* and *Erechtites* (Compositae) *Echium*, *Heliotropium* *Trachelanthus* and *Trichodesma* (Boraginaceae).[106] Of the 29 species of *Crotalaria* recorded from Sri Lanka,[1] *C. multiflora* (Arn.) Benth. and *C. walkeri* Arn. are known to be endemic.[11]

C. verrucosa L. (*Sinh.*— nilandanahiriya ; *Tam.*— kilvenlappa) and *C. juncea* L. (*Sinh.*— Hana ; *Tam.*— shanal imappu) find many applications in the indigenous system of medicine.[10] The leaves and seeds of these plants are said to have a cooling effect and blood-purifying action and hence used in cases of fever and skin diseases. *C. verrucosa* which is used more often in medicine than *C. juncea* is employed also as a bitter and as an expellent of bile. The juice of the tender leaves

is applied both internally and externally for scabies and impetigo. The leaf preparation is also applied to eruptions of the skin so as to clean them before applying curative medicines.

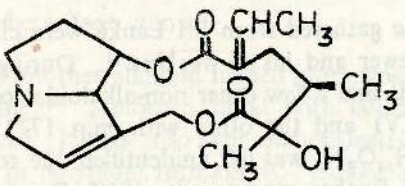
During their chemical studies on the genus *Crotalaria*, Atal and his co-workers have examined,[160] the seeds of *C. walkeri* Arnott for alkaloids. Although this species has been described by Trimen as endemic to Sri Lanka,[164] according to Hooker it is a variety of the Indian species *C. semperflorens*.[160] Quantitative estimation had revealed seeds of *C. walkeri* to contain 0·20% tertiary bases and 0·06% *N*-oxides. The ethanol extract of the powdered dehusked seeds on partition chromatography over buffered celite 525 has afforded a new secopyrrolizidine alkaloid, crotaverrine (LXII) and its derivative, O-acetylcrotaverrine (LXIII). The structures of these tertiary bases have been elucidated by combined chemical and spectral analyses.



(LXII) ; R = H
(LXIII) ; R = $COCH_3$

8.6. Pharmacology and Toxicology of *Crotalaria* Species

Monocrotaline (LXIV), a pyrrolizidine alkaloid from *C. spectabilis* is found to be responsible for the activity of *Crotalaria* extracts against adenocarcinoma-755 in mice.[100] Similar alkaloids have been screened for anti-tumour activity.[34,145] However, as pyrrolizidine alkaloids are known to be hepatotoxic,[84] it is questionable whether they could have applications in chemotherapy of cancer. Besides, presence of these alkaloids in this genus demands a complete evaluation of the medicinally employed *Crotalaria* species (see 8.5) for their hepatotoxicity.*



(LXIV)

*Investigation of these and other plant species used in native medical practice for hepatotoxicity and the occurrence of pyrrolizidine alkaloids has been undertaken by Prof. S. N. Arseculeratne of the Dept. of Microbiology, Peradeniya Campus in collaboration with the author.

Poisoning by the pyrrolizidine alkaloids has recently been reviewed by Bull, who suggests that the condition should be called pyrrolizidine alkaloidosis.[25] The pyrrolizidine alkaloids are not all poisonous. For an alkaloid to be toxic it must have a double bond between C-1 and C-2.[142] The cyclic diesters (e.g. (LXIV)) are twice as toxic as the open diesters, and 4 times as toxic as the open monoesters. Esters of branched-chain acids are toxic while esters of straight chain acids are not.[143] It has been suggested that the alkaloids themselves are not hepatotoxic, but are converted in the liver to toxic pyrrole-like derivatives which react with tissue constituents to form bound pyrroles which either remain in the tissues or are excreted in the urine.[109]

9. Alkaloids of Liliaceae

9.1 Introduction

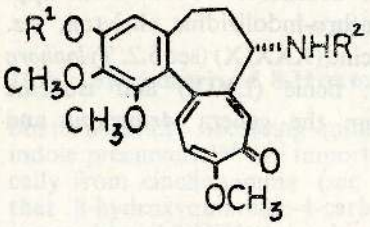
The monocotyledonous family, Liliaceae, produces a number of alkaloidal types of which colchicine and *Veratrum* types are of wide occurrence.[129] Colchicine group of alkaloids have a 1-phenethyl-1,2,3,4-tetrahydroisoquinoline origin and *Veratrum* group is of steroidal type. Alkaloids based on the steroid nucleus are not very widely distributed, being restricted to plants of the *Holarrhena* (Apocynaceae, see 4.2), *Solanum* (Solanaceae) and *Veratrum* species.[18]

Abeywickrema has listed 18 species of Liliaceae[1] belonging to 14 genera containing 2 endemic species, *Urginea rupicola* (Trim.) Trim. ex Hook. f. and *Asparagus zeylanicus* (Baker) Hook. f.[11] The non-endemic *Gloriosa superba* L. (*Sinh.*— Niyagala) grows wild in Sri Lanka. This plant is known to ancient medical writers in Sanskrit as "Gharbha ihatin"— that which causes abortion. Decoctions containing *G. superba* have found considerable use in native medicine particularly as abortifacients.[26,48] The roots are used for snake-bites and scorpion stings and the starch from roots is given internally for gonorrhoea.[29]

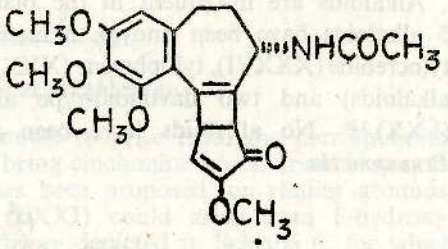
9.2 Alkaloids of *Gloriosa superba*

The tubers of *G. superba* gathered from Sri Lanka were chemically investigated as far back as 1915 by Clewer and his co-workers.[31] During their studies they were able to isolate 2 alkaloids and a few other non-alkaloidal constituents. One of the bases was colchicine (LXV) and the other with m.p. 177° to 178°C and probable molecular formula, $C_{33}H_{38}O_9N_2$ was left unidentified due to the non-availability of sufficient quantities for further studies. In 1968, Dunuwille, Balasubramanium and Bibile reinvestigated the mature and tender tubers, the seeds and the flowers of *G. superba*.[43,44] Their examination of the mature tubers which commonly cause poisoning in the rural areas of Sri Lanka, had revealed colchicine (LXV) to be the major alkaloid (0.025%). However, the colchicine content was remarkably low

compared to that found in the tubers of European (*ca.* 0.20%) and African (0.12%) origin. Colchicine was also found to occur in the tender tubers, seeds and flowers. Three other alkaloids, namely *N*-formyl-*N*-desacetylcolchicine (LXVI) (0.003%) 2-demethylcolchicine (LXVII) and lumicolchicine (LXVIII) were also isolated from the mature tubers. They also found several other unidentified alkaloids to be present in trace amounts.



(LXV) ; $R^1 = CH_3$, $R_2 = COCH_3$
(LXVI) ; $R^3 = CH_3$, $R^2 = CHO$
(LXVII) ; $R^1 = H$, $R^2 = COCH_3$



(LXVIII)

Seeds of *G. superba* obtained from Sri Lanka were also analysed by Santavy *et al.* [110] by paper chromatographic techniques and was shown to contain colchicine (LXV), *N*-formyl-*N*-deacetylcolchicine (LXVI) and 2-demethylcolchicine (LXVII).

9.3 Pharmacology and Toxicology of *G. superba*

Numerous fatalities due to ingestion of *G. superba* in decoctions used as abortifacients are on record.[168] Poisoning has also occurred in Sri Lanka through the tubers being mistaken for those of the yams of sweet potato (*Ipomeoa batatas*).[44,59] Jayatilaka and Balasubramanium have investigated[90] the digestive tract histopathology in rats poisoned by the extracts of *G. superba* tubers and have found that the lesions are similar to those poisoned by colchicine (LXV) which was shown to be an alkaloidal constituent of these tubers (see above).

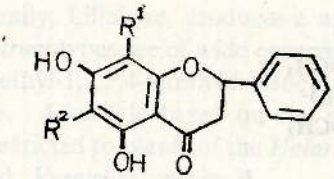
Subbaratnam obtained another alkaloid named gloriosine from *G. superba* during the isolation of colchicine.[154] Amoroso found both colchicine and gloriosine to have an anti-mitotic effect.[6] It has also been demonstrated that gloriosine is more effective than colchicine in polyploid formations. Colchicine has been used in the treatment of gout for many years. It has also been used as a cytotoxic drug in the treatment of inoperable carcinoma.[23] Therefore, in view of its wide use in therapy, experiments have been conducted on mice, rats and sheep with the aim of discovering its action and toxicity. Alopecia (loss of hair) caused by treatment with colchicine[135,136] and due to the ingestion of tubers of *G. superba* is known.[59]

10. Alkaloids of Moraceae

10.1 Introduction

Moraceae in Sri Lanka has 34 species distributed in 11 genera.[164] Of these, 5 species in the genera *Allaeanthus* (more correctly *Broussonetia*[32]), *Artocarpus* and *Ficus* are reported to be endemic. The former two genera contain one each of endemic species and *Ficus* has 3 species which are endemic.[11]

Alkaloids are infrequent in the plant species of Moraceae. From *Ficus* spp. 5 alkaloids have been known, 3 having a phenanthro-indolizidine skeleton, *viz.* tylocrebine (XXXVI), tylophorine (XXXIII) and septicine (XXXIX) (see 5.2. *Tylophora* alkaloids) and two flavonoid-type alkaloids, *viz.* ficine (LXIX) and isoficine (LXX).[129] No alkaloids have been reported from the genera *Artocarpus* and *Broussonetia.*



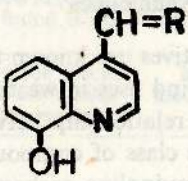
(LXIX) R^1=  , R^2=H

(LXX) R^1=H , R^2= 

10.2 Alkaloids of *Broussonetia zeylanica* (*Allaeanthus zeylanicus*)

The genus *Broussonetia* in Sri Lanka is represented by a single species *B. zeylanica* Thwaites (*Sinh.*—Alandu) which is endemic to the country.[11] This graceful tree, the very tough inner bark-fibres of which were used for string, seems to have become rare as its lowland forest habitat has disappeared.[32]

Although *B. zeylanica* had no claims of medicinal applications, when screened some of its extracts exhibited significant antimicrobial activity against three common pathogenic organisms ; *Candida albicans*, *E. coli* and *Staphylococcus aureus*.[71] Prompted by this, an investigation directed towards the isolation and identification of the active constituent had led to the recognition of a new alkaloid, 8-hydroxy-quinoline-4-carboxaldehyde (LXXI) in 0.25% yield from the dried timber. This alkaloid was found to be active against *C. albicans* and *S. aureus*. This was the first report of a naturally occurring 8-hydroxyquinoline (oxine). However, 8-hydroxy 4-quinolone has been recently reported as a constituent of the ink of giant octopus, *Defleini martimi*.[153]

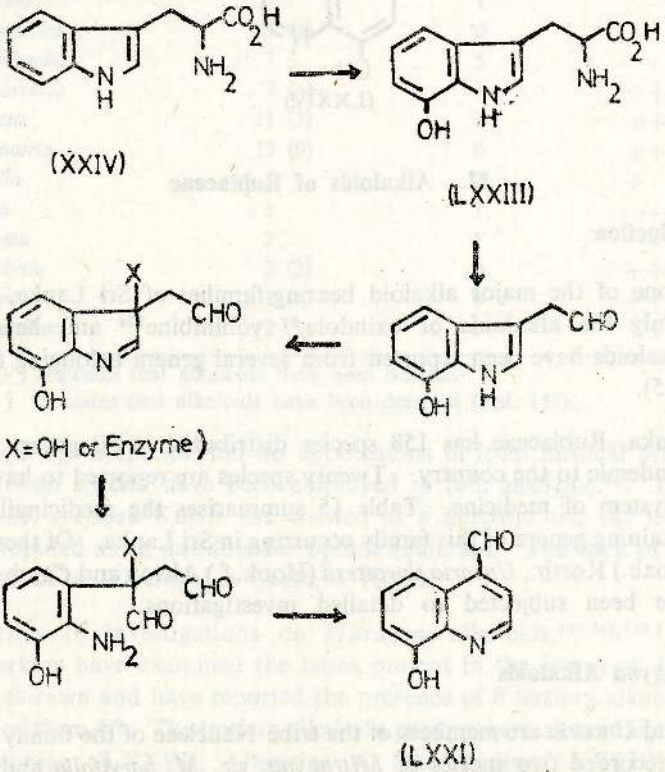


(LXXI) ; R = O

(LXXII) ; R = N—NH—C(=S)—NH_2

10.3 Biosynthesis of 8-Hydroxyquinoline-4-carboxaldehyde

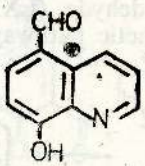
Some naturally occurring quinolines are known to arise from the corresponding indole precursors,[104] an important example being cinchonine which arises biogenetically from cinchonamine (see 11.6). It has been proposed, on similar grounds, that 8-hydroxyquinoline-4-carboxaldehyde (LXXI) could arise from 8-hydroxytryptophan (LXXIII) by a biosynthetic pathway depicted in Scheme 6, for which chemical analogues are known.[165]



Scheme 6. Proposed biosynthetic route to 8-Hydroxyquinoline-4-carboxaldehyde from *Broussonetia zeylanica*.

10.4 Pharmacology of 8-Hydroxyquinolines

8-Hydroxyquinoline and its derivatives are known to have an array of antimicrobial properties and a number of them find uses in western medicine as topical antiseptics and disinfectants.[33] The possible relationship between complex formation property and tuberculostatic activity of this class of compounds had prompted synthesis and testing of a number of 8-hydroxyquinoline carboxaldehydes and their thiosemicarbazones.[24] 8-Hydroxyquinoline-5-carboxaldehyde (LXXIV) was found to be the most effective against human type H-37 Rv strain of *Myobacterium tuberculosi*.[94] However, the effective doses were found to be toxic to mice. Pharmacological evaluation of 8-hydroxyquinoline-4-carboxaldehyde (LXXI) and the thiosemicarbazone (LXXII) has suggested that the introduction of —CHO and —CH=N—NH —$CSNH_2$ groups causes a decrease in tuberculostatic activity.[24]



(LXXIV)

11. Alkaloids of Rubiaceae

11.1 Introduction

Rubiaceae, one of the major alkaloid bearing families of Sri Lanka, is known to contain mainly the alkaloids of oxindole,[57] yohimbine[124] and heteroyohimbine types.[57] Alkaloids have been reported from several genera belonging to this family (see Table 15).

In Sri Lanka, Rubiaceae has 158 species distributed in 50 genera of which 74 species are endemic to the country. Twenty species are reported to have uses in the indigenous system of medicine. Table 15 summarises the medicinally useful and alkaloid containing genera of this family occurring in Sri Lanka. Of these, *Mitragyna parvifolia* (Roxb.) Korth., *Uncaria thwaitesii* (Hook. f.) Alston and *Cinchona ledgeriana* Moens. have been subjected to detailed investigations.

11.2 *Mitragyna* Alkaloids

Mitragyna and *Uncaria* are members of the tribe Naucleae of the family Rubiaceae.[20] Trimen has recorded two species of *Mitragyna*, *viz. M. parvifolia* and *M. tubulosa* (Arn. *ex* Bedd.) Kuntze from Sri Lanka.[164]

TABLE 15. Medicinally used and alkaloid containing genera of Rubiaceae found in Sri Lanka.

Genus	No. of species in Sri Lanka (No. endemic)	No. of species used in indigenous medicine	Presence of Alkaloids*
Anthocarpus	1	1	++
Borreria	3	1	++
Canthium	7 (5)	2	++
Cinchona	0	—	++
Gardenia	3	1	—
Hedyotis	29 (22)	1	++
Ixora	5 (2)	1	++
Knoxia	4 (1)	0	++
Lasianthus	9 (9)	0	++
Morinda	3	3	+
Mitragyna	2	0	++
Mussenda	2	2	—
Nauclea	1	1	—
Neonauclea	1 (1)	0	+
Oldenlandia	7	5	—
Ophiorrhiza	7 (4)	1	++
Pavetta	11 (3)	1	++
Psychotria	13 (9)	0	++
Randia	5	2	+
Rubia	1	1	—
Tarenna	2	1	—
Tricalysia	2 (2)	0	++
Uncaria	1 (1)	0	++
Urophyllum	2 (1)	0	++

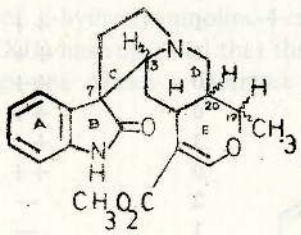
*(++) indicates that alkaloids have been isolated
(+) indicates that alkaloids have been detected (Ref. 157)

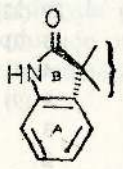
Although *Mitragyna* species find no applications in local medical practice, some Asian and African species have been employed in folk medicine.[137] In Thailand, the leaves of *M. speciosa* Korth. are chewed as a narcotic and the leaves of *M. parvifolia* are reputed as an antidote for opium addiction. The bark of *M. africana* Korth. is used as a febrifuge in Western Africa.

In their series of investigations on *Mitragyna* alkaloids,[147,148,149,150] Shellard and his co-workers have examined the bases present in the leaves of *M. parvifolia* collected in Kekirawa and have reported the presence of 8 tertiary alkaloids and the *N*-oxides of 4 of them.[149] The tertiary alkaloids present were akuammigine (LXXV), tetrahydro-alstonine (LXXVI), Uncarine C [pteropodine (LXXVII)], Uncarine D [speciophylline (LXXVIII)], Uncarine E [isopteropodine (LXXIX)], Uncarine

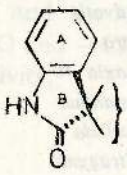
F(LXXX), coryantheidol (LXXXI) and dihydrocorynantheol (LXXXII). The *N*-oxides were characterised from their mass spectra and by sulphurous acid reduction to the corresponding parent tertiary alkaloids which were identified by *TLC* with authentic samples in a number of developer solvent systems. Characterised in this manner were the *N*-oxides of akuammigine, speciophylline, Uncarine F and dihydrocorynantheol (see Table 18).

TABLE 16. Some pentacyclic ring E heterooxindole alkaloids of Sri Lankan Rubiaceae.



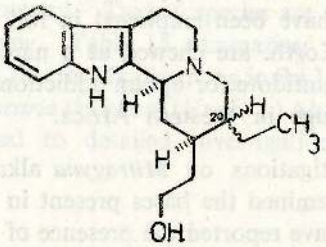


C(7)-R



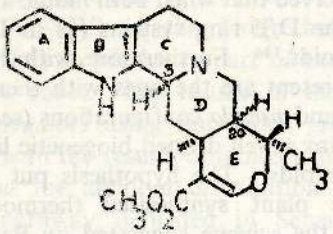
C(7)-S

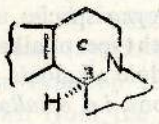
		C(7)	C(3)-H	C(19)-H	C(20)-H
Uncarine C	(LXXVII)	R	α	β	α
Uncarine D	(LXXVIII)	S	β	β	α
Uncarine E	(LXXIX)	S	α	β	α
Uncarine F	(LXXX)	R	β	β	α
Mitraphylline	(LXXXIV)	R	α	β	β
Isomitraphylline	(LXXXV)	S	α	β	β
Formosanine	(LXXXVIII)	R	α	α	β



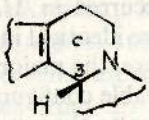
		C(20)—H
Corynantheidol	(LXXXI)	α
Dihydrocorynantheol	(LXXXII)	β

TABLE 17. Some terpenoid indole alkaloids of Sri Lankan Rubiaceae.



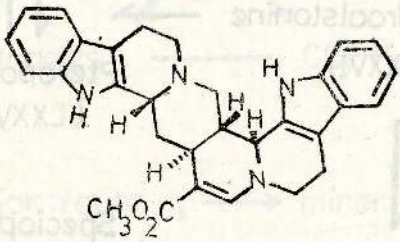


allo, normal



epiallo pseudo

		C(3)-H	C(20)-H
Akuammigine	(LXXV)	β	α
Tetrahydroalstonine	(LXXVI)	α	α
Ajmalicine	(XXI)	α	β
Isoajmalicine	(LXXXIII)	β	β



Roxburghine D (LXXXIX)

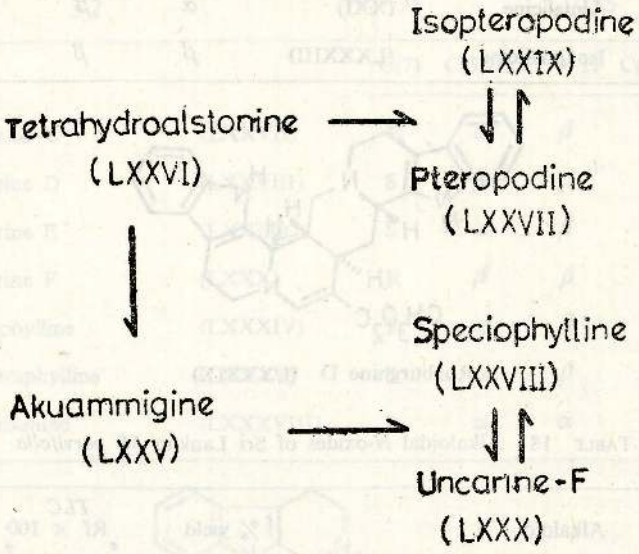
TABLE 18. Alkaloidal *N*-oxides of Sri Lankan *M. parvifolia*

Alkaloid		% yield	TLC Rf × 100 *	†
Speciophylline	*N*-oxide	1.33	14	4
Uncarine—F	*N*-oxide	1.33	36	28
Akuammigine	*N*-oxide	1.66	38	46
Dihydrocorynantheol	*N*-oxide	0.67	50	56

*Using Silica gel G/methanol
†Using Silica gel G/chloroform– methanol, 6 : 1

11.3. Biosynthesis of *Mitragyna* Alkaloids

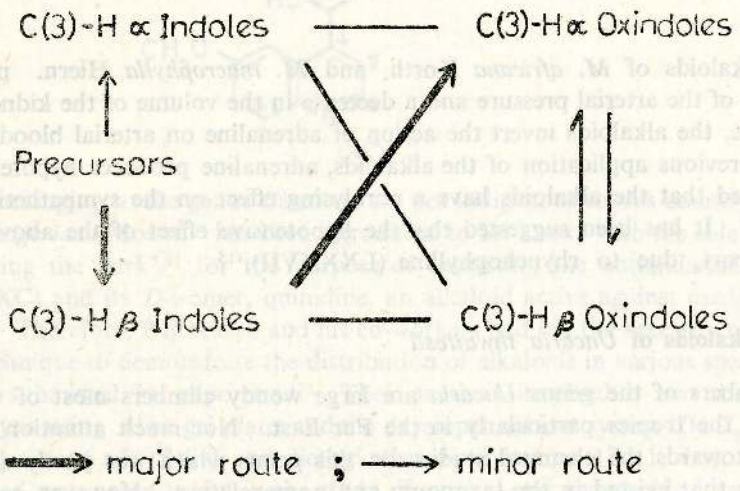
Mitragyna species are known to produce both indole and oxindole groups of alkaloids. In 1969, Shellard, Phillipson and Gupta observed that when both indole and oxindole alkaloids occurred in *Mitragyna* species, the D/E ring systems (as in LXXIV and LXXIII) were identical in both types of alkaloids.[146] Furthermore, with the exception of *M. speciosa*, the major indole alkaloids present are the ones with thermodynamically least stable configurations, *i.e. epiallo* and *pseudo* configurations (see Table 17). This gave rise to the possibility that there was a well defined biogenetic link between the formation of indole and oxindole alkaloids. The hypothesis put forward by Shellard *et. al.*[146] states that : (*a*) the plant synthesised thermodynamically more stable indole alkaloids according to the scheme suggested by Battersby and Hall[17] for the biogenesis of indole alkaloids, (*b*) these alkaloids then isomerised into the corresponding thermodynamically less stable configuration, and (*c*) all the indole alkaloids were then converted to the corresponding oxindole alkaloids. The pattern of alkaloids in *M. parvifolia* obtained from Sri Lanka is shown in Scheme 7, which is different to the alkaloid pattern found in *M. parvifolia* samples from Burma and Uttar Pradesh state of India.



Scheme 7. Alkaloidal pattern in *Mitragyna parvifolia* from Sri Lanka.

During these biosynthetic investigations, Shellard and Houghton fed ajmalicine (XXI) and 3-isoajmalicine (LXXXII) into young plants of Sri Lankan *M. parvifolia* and in both cases obtained mitraphylline (LXXXIV) and isomitraphylline (LXXXV). This and a similar series of biosynthetis experiments led to the modification of the originally postulated hypothesis (see above). Further experiments with labelled precursors supported this modification of the original hypotheses for the biosynthesis of oxindole alkaloids in *Mitragyna* species as indicated in Scheme 8.

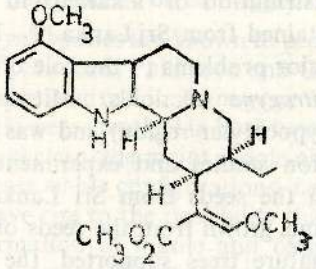
The Chelsea group has also examined the distribution of alkaloids in young plants of *M. parvifolia* grown from the seeds obtained from Sri Lanka.[148] During this study they were able to resolve one of the major problems ; the role of mitraphylline (LXXXIV) in the biosynthesis of *Mitragyna* alkaloids. Mitraphylline was found only in the lower part of the stem (hypocotylar region) and was absent in leaves or roots. The results of these isolation studies and experiments with labelled precursors using the plants grown from the seeds from Sri Lanka were considered with the results obtained using the plants grown from the seeds of Uttar Pradesh and the alkaloid distribution in the mature trees supported the above metabolic pathway indicated in Scheme 8.



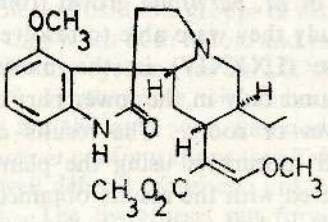
Scheme 8. Biosynthesis of Oxindole alkaloids in *Mitragyna* species.

11.4. Pharmacology of *Mitragyna* Alkaloids

Some *Mitragyna* alkaloids are known to have pharmacological activity. Mitragynine (LXXXVI) is a general protozoal poison but is ineffective against bacteria or pathogenic protozoa. It has a depressant effect in isolated tissues, facilitates the passage of impulses in the autonomic nervous system and increases the excitability in the medulla and probably of motor centres.[69] Mitraphylline (LXXXIV), an alkaloid reported from a number of *Mitragyna* species, resembles mitragynine in its pharmacological activity but is less effective.[108] Mitraphylline is also hypotensive.[121] It is interesting to note that, in connection with the employment of *Mitragyna* extracts as febrifute (see above), rhynchophylline [mitrinermine (LXXXVII)] exhibits a significant antipyretic action.[123,137] Rhynchophylline was also found to be toxic to paramecium.[121]



(LXXXVI) Mitragynine



(LXXXVII) Rhynchophylline

The alkaloids of *M. africana* Korth. and *M. macrophylla* Hiern. produce a reduction of the arterial pressure and a decrease in the volume of the kidney. Like yohimbine, the alkaloids invert the action of adrenaline on arterial blood pressure. After a previous application of the alkaloids, adrenaline produces hypotension. It is supposed that the alkaloids have a paralysing effect on the sympathetic nervous system.[122] It has been suggested that the hypotensive effect of the above extracts are, in part, due to rhynchophylline (LXXXVII).[115]

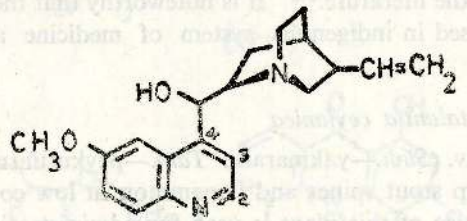
11.5. Alkaloids of *Uncaria thwaitesii*

The members of the genus *Uncaria* are large woody climbers most of which are found in the tropics particularly in the Far East. Not much attention has been directed towards the chemical studies on this genus which was partly due to the confusion that existed in the taxonomy and nomenclature. However, recently the genus *Uncaria* has been revised and 39 species out of a total of 120 have been recognised. A number of alkaloids were isolated and identified from these species by Phillipson and Hemingway by means of sensitive chromatographic and spectroscopic techniques.[125] The distribution of more than 60 alkaloids reported from *Mitragyna* and *Uncaria* species have been recently summarised by Herath.[83]

The only Sri Lankan species of the genus *Uncaria*, *U. thwaitesii* (Hook. f.) Alston which is endemic to the country has been investigated recently for alkaloids.[81,82,83] From the bark basic extract, four indole alkaloids *viz.*, formosamine (LXXXVIII), mitraphylline (LXXXIV), roxburghine D (LXXXIX) and another roxburghine isomer with a m.p. 215°C have been isolated in the yields of 0.009, 0.19, 0.002 and 0.003% respectively. The occurrence of these less polar bases formosamine, mitraphylline and a roxburghine isomer with a m.p. 215°C all in trace amounts has also been demonstrated in the timber extracts of *U. thwaitesii*. Identification of these alkaloids was based on spectral analysis and chemical reactions.

11.6. Studies on *Cinchona* Alkaloids

The four principal *Cinchona* alkaloids, quinine (XC), quinoline, cinchonine and cinchonidine, all contain a quinoline ring linked by a secondary alcohol at its fourth position to a quinuclidine ring. Quinidine is the dextrorotatory optical isomer of quinine (XC) ; cinchonidine is quinine minus the methoxy group and cinchonine is the *D* isomer of cinchonidine. All 4 *Cinchona* alkaloids have schizonticidal activity, but quinine is the only one used in the treatment of malaria because of its high absorption property.



(XC)

Plants belonging to the genus *Cinchona* are not indigenous to this country. However, *C. ledgeriana* Moens. has been introduced to Sri Lanka with the sole intention of exporting the bark[5,91] for the purpose of extracting the antimalarial alkaloid, quinine (XC) and its *D*-isomer, quinidine, an alkaloid active against cardiac arrhythmias.[114] Therefore, Wijesekera and his co-workers had felt the necessity to devise a simple technique to demonstrate the distribution of alkaloids in various specimens of *Cinchona* submitted by exporters.[171] Their method involved a two dimensional *TLC* analysis using silica gel plates and the developer solvent system $CHCl_3$–MeOH–17% NH_3 (24 : 6 : 0.05 v/v) and diethyl ether-diethylamine (17 : 1, v/v). Further, this method may prove useful in the study of varietal and geographical variation of alkaloid content in *Cinchona* samples.

Quinine (XC) is one of the least toxic alkaloids known. Non-fatal cases of poisoning have been caused from its use as an antimalarial,[111] but fatalities were usually due to its use as an abortifacient.[58]

Extracts of *C. ledgeriana* have been screened for the occurrence of biogenetic precursors of indole alkaloids and 5-carboxystrictosidine (XXIX) has been isolated (see 4.5).[36]

12. Alkaloids of Rutaceae

12.1. Introduction

The Rutaceae family of plants produces some 13 basic alkaloid types[129] and the acridan-9-one (acridone) group of alkaloids represents a class specific to this family. Recently, the number of acridone alkaloids known has doubled,[92] but in all cases so far reported, the structures have been represented by modifications on a single acridone ring system.

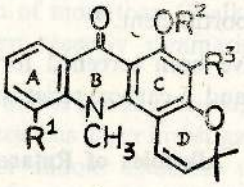
Rutaceae in Sri Lanka has 40 species distributed in 18 genera,[1] of which 4 species are endemic.[11] Fourteen non-endemic species of Rutaceae find applications in the indigenous system of medicine (see Table 2).[3] Further, this family contains such useful plants as lime (*Citrus aurantifolia* L.), lemon (*C. limonium* L.), orange (*C. sinensis*) and curry leaf (*Murraya koenigii*, *Sinh.*—karapincha). *Acronychia* represents an important genus in the family Rutaceae as acronycine (XCI) isolated from different species of this genus has shown the broadest experimental anti-tumour activity of any alkaloid studied.[93] However, no evidence for clinical trials with this alkaloid has yet been found in the literature.[140] It is noteworthy that the bark of *A. laurifolia* (*Sinh.*—ankenda) is used in indigenous system of medicine as an application to sores and ulcers.[10]

12.2. Alkaloids of *Atalantia ceylanica*

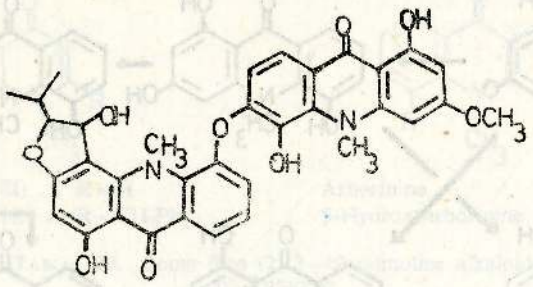
A. ceylanica (Arn.) Oliv. (*Sinh.*—yakinaran : *Tam.*—peykuruntu) is a much branched bush armed with sharp stout spines and is common in low country and in eastern province. The leaf juice of this plant is used for administration of pills and is an emperical remedy to prevent attacks of ague.[10]

Fraser and Lewis have recently investigated *A. ceylanica* obtained from Sri Lanka.[51,52] From the wood extractives of this plant they isolated 2 major acridone alkaloids, 3,12-dihydro-6,11-dihydroxy-3,3,12-trimethylpyrano (2,3-C) acridine-7one (XCII) and its 5-(3-methylbut-2-enyl) derivative (XCIII) in the yields of 0.04 and 0.003% respectively.[52] The structures of these two alkaloids were established from the analysis of their spectral data. These 2 alkaloids are close relatives of the acridone alkaloid acronycine (XCI) (see above). Occurrence of (XCII) as a plant product is particularly interesting since it has been tentatively reported as one of the metabolites isolated from rat bile after dosage with acronycine.[155]

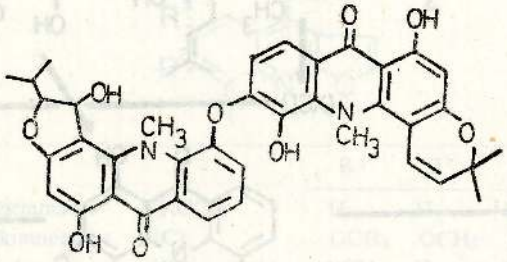
TABLE 19. Some acridone alkaloids from *A. ceylanica*.



	R¹	R²	R³
(XCI)	H	CH_3	H
(XCII)	OH	H	H
(XCIII)	OH	H	$-CH_2-CH=<$



(XCIV)



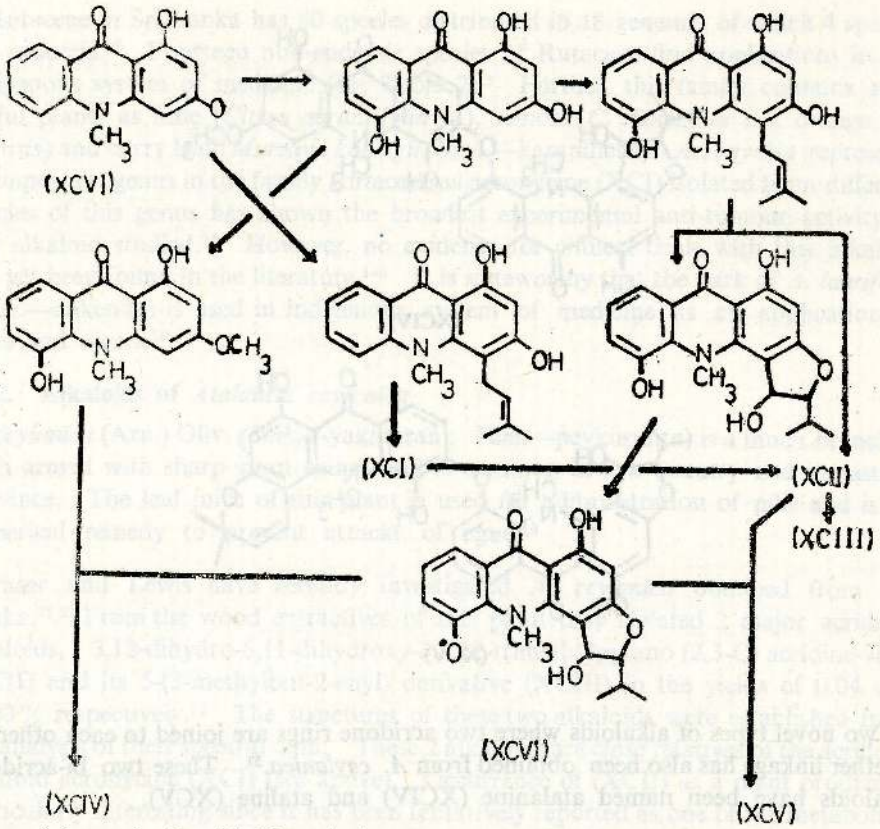
(XCV)

Two novel types of alkaloids where two acridone rings are joined to each other by an ether linkage has also been obtained from *A. ceylanica*.[51] These two bi-acridone alkaloids have been named atalanine (XCIV) and ataline (XCV).

12.3. Biosynthesis of *Atalantia* Alkaloids

1, 3-Dihydroxyacridan-9-one (XCVI) could be regarded as the biogenetic precursor for all the known acridone alkaloids.[76] Possible biosynthetic routes from this precursor to the 4 alkaloids obtained from *A. ceylanica* are given in Scheme 9. Biogenetically, the introduction of the 5-hydroxy function into acronycine (XCI) to obtain (XCII) probably occurs at a later stage, perhaps *via* an arene oxide intermediate.[74]

Biogenetic considerations have aided Fraser and Lewis to locate the ether linkages in the two novel bi-acridone alkaloids, ataline (XCV) and atalanine (XCIV) isolated from *A. ceylanica*.[51] In both alkaloids, the ether linkage was assigned to C-5[1]—C-6 since all the acridone alkaloids isolated with hydroxy groups in ring A (see structures XCII and XCIII) have it located at 5 position. In this position, it would enable the radical intermediate (XCVI, Scheme 9) to couple with another acridan-9-one at the C-6 position, *i.e. ortho* to its hydroxy group. However, the authors have stated that alternative sites for the ether linkage cannot be excluded.

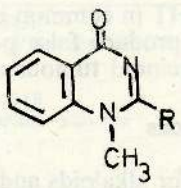


Scheme 9. Possible biosynthetic routes to the alkaloids of *Atalantia ceylanica*.

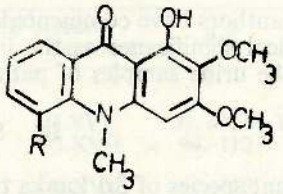
12.4. Alkaloids of *Glycosmis bilocularis*

The genus *Glycosmis* has 2 species in Sri Lanka, *G. bilocularis* Thwaites and *G. pentaphylla* (Retz.) Corr. The former, a very rare shrub found in low country and Mihintale in the North Central province, is endemic to the country and has been recently analysed for its constituent alkaloids.[22] Although this plant has no reported medicinal applications, the non-endemic *G. pentaphylla* (*Sinh.*—dodanpana ; *Tam.*—kulapannai) is used in snake-bites and given in cases of low fever.[28]

From the basic extract of the leaves and stems of *G. bilocularis*, Bowen and Perera,[22] had isolated 7 alkaloids out of which one is a new acridone alkaloid. The known alkaloids isolated were of acridone, quinazolone and furo (2,3-b) quinoline types. The quinazolone alkaloids identified were glycorine (XCVII) and arborine (XCVIII) whereas furo (2,3-b) quinolines included dictamnine (XCIX), skimmianine (C) and kokusaginine (CI). The acridone type which is characteristic of the family Rutaceae included the known alkaloid arborinine (CII) and a new base 5-hydroxyarborinine (CIII). It is important to note that the presence of acridones hydroxylated at C-5 appears to be a characteristic of the Sri Lankan Rutaceae (refer XCII, XCIII XCIV, and XCV).

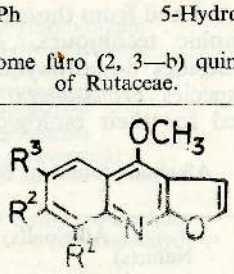


Glycorine (XCVII) ; R = H
Arborine (XCVIII) ; R = CH_2Ph



Arborinine (CII) ; R = H
5-Hydroxyarborinine (CIII) ; R = OH

TABLE 20. Some furo (2, 3—b) quinoline alkaloids of Rutaceae.



		R^1	R^2	R^3
Dictamnine	(XCIX)	H	H	H
Skimmianine	(C)	OCH_3	OCH_3	H
Kokusaginine	(CI)	OCH_3	H	OCH_3

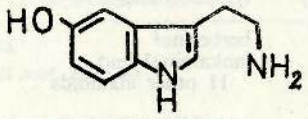
12.5. Alkaloids of *Micromelum ceylanicum*

Micromelum ceylanicum Swingle, the only species of this genus found in Sri Lanka which has been wrongly reported by Trimen[164] as *M. pubescens* (*Sinh.*—wal-karapincha ; *Tam.*—kakaipalai) is a small tree common in low country especially in the dry region. This plant has also been examined by the same authors[22] and they have isolated an alkaloid whose structure is presently under investigation.

13. Miscellaneous Studies

13.1. 5-Hydroxytryptamine in Edible Fruits

5-Hydroxytryptamine (5HT) or serotonin (CIV) which is known to be derived from the amino acid tryptophan (XXIV) is considered to be an important biogenetic amine.[103] Prompted by a report[166] of its occurrence in banana, and clinical consequences, Dharmatilaka and Kottegoda have investigated a number of edible fruits from Sri Lanka[38] for its presence. Their method of analysis involved the preparation of extracts of these fruits by a known procedure[169] and an assay for 5HT by a method developed by Kottegoda[97] on the heart of brackish water molluscs, after identification by paper chromatography. It was found that anamalu plantain, papaw (*Carica papaya*) and Jambola (*Citrus decumana*) contained appreciable amounts of 5HT.



(CIV)

The authors have commented that the presence of 5HT in common edible fruits is of clinical significance as the ingestion of these may produce false positive results when the urine samples of patients with suspected carcinoid tumour are examined.

14. Summary and Conclusions

The plant species of Sri Lanka that have been studied for alkaloids and the alkaloids isolated from these are summarised in Table 21. A total of 22 plant species including 9 endemics have been investigated and from these 68 alkaloids have been isolated and 11 detected by chromatographic techniques. Amongst the alkaloids isolated, 10 were found to be new. Besides 8-hydroxyquinoline-4-carboxaldehyde (LXXI) obtained from the endemic species *Broussonetia zeylanica* (Moraceae), these new bases have not been evaluated for their biological activity.

TABLE 21. Alkaloids found in Sri Lanka Plants

FAMILY Plant species	Alkaloid(s) present Name(s)	Formula(e)	Ref.
ANCISTROCLADACEAE			
Ancistrocladus hamatus[1]	ancistrocladine	(I)	66
	hamatine[3]	(III)	66
APOCYNACEAE			
Holarrhena mitis[1]	conessine	(V)	19, 27, 101
	iso-conessimine	(VI)	27
	holadienine	(IX)	27
	conkurchine	(XIII)	27
	holarrhenine	(VII)	27
	holafebrine	(X)	27
	holarrhimine	(XI)	27
	N-3-methylholarrhimine	(XII)	27
	mitiphylline	(XV)	89, 102
	N-desmethylmitiphylline	(XVI)	89, 102
	triacanthine	(XIV)	89, 101, 102
	conamine	(VIII)	101
Catharanthus roseus and *C. pusillus*	vincaleukoblastine[2,4]	(XIX)	70, 152
	leurocristine[2,4]	(XX)	70, 152
	ajmalicine[2,4]	(XXI)	70, 152
	vincamine[2,4]	(XXII)	70, 152
(*Vinca* spp.)	5α-carboxystrictosidine	(XXIX)	36
	strictosidine	(XXVIII)	36
Rauwolfia serpentina	5α-carboxystrictosidine	(XXIX)	36
	strictodesine	(XXVIII)	36
Kopsia fruticosa	ajmalicine[2,4]	(XXI)	7
ASCLEPIADACEAE			
Tylophora asthmatica	tylophorine	(XXXIII)	126
	tylophorinine	(XXXIV)	126
	tylophorinidine	(XXXV)	126
T. cordifolia	tylophorinine[2]	(XXXIV)	126
T. flava[1]	tylophorine[2,4]	(XXXIII)	126
	tylophorinine[2]	(XXXIV)	126
BERBERIDACEAE			
Berberis tinctoria	berberine[4]	(XL)	144
	lankamine[3] and 11 other alkaloids		144
LAURACEAE			
Neolitsea fuscata[1]	isoboldine	(XLIII)	72

FAMILY Plant species	Alkaloid(s) present Name(s)	Formula(e)	Ref.
LILIACEAE			
Gloriosa superba	colchicine[4]	(LXV)	31, 43, 44, 110
	N-formyl-*N*-deacetyl -colchicine	(LXVI)	44, 110
	2-demethyl colchicine	(LXVII)	44, 110
	lumicolchicine	(LXVIII)	44
LEGUMINOSAE			
Erythrina lithosperma	erythratidinone	(L)	15, 16
	3-demethoxyery-thratidinone	(LI)	15, 16
	erythraline[4]	(XLVI)	15, 16
	erysotrine	(XLVIII)	15, 16
E. fusca	erysotrine	(XLVIII)	15
Crotolaria walkeri[1]	crotaverrine[3]	(LXII)	160
	O-acetylcrotaverrine	(LXIII)	160
MORACEAE			
Broussonetia zeylanica[1] (= *Allaeanthus zeylanicus*)	8-hydroxyquinoline -4-carboxaldehyde[3,4]	(LXXI)	71
RUBIACEAE			
Mitragyna parvifolia	akuammigine	(LXXV)	149
	tetrahydroalstonine	(LXXVI)	149
	Uncarine C	(LXXVII)	149
	Uncarine D	(LXXVIII)	149
	Uncarine E	(LXXIX)	149
	Uncarine F	(LXXX)	149
	corynantheidol	(LXXXI)	149
	dihydrocorynantheol	(LXXXII)	149
	akuammigine *N*-oxide		
	Uncarine D *N*-oxide		
	Uncarine F *N*-oxide		149
	dihydrocorynantheol *N*-oxide		149
Uncaria thwaitesii[1]	Uncarine A	(LXXXVIII)	81-83
	mitraphylline[4]	(LXXXIV)	81-83
	roxburghine D	(LXXXIX)	81-83
Cinchona ledgeriana	quinine[2,4]	(XC)	171
	quinidine[2,4]		171
	5α-carboxystrictosidine	(XXIX)	36
RUTACEAE			
Atalantia ceylanica[1]	acridone alkaloid A[3]	(XCII)	52
	acridone alkaloid B[3]	(XCIII)	52
	atalanine[3]	(XCIV)	51
	ataline[3]	(XCV)	51
Glycosmis bilocularis[1]	glycorine	(XCVII)	22
	arborine	(XCVIII)	22
	dictamnine	(XCIX)	22
	skimmianine	(C)	22
	kokusaginine	(CI)	22
	arborinine	(CII)	22
	5-hydroxyarborinine[3]	(CIII)	22
Citrus decumana	serotonin[2]	(CIV)	38

[1]endemic to Sri Lanka
[2]alkaloids detected but not isolated
[3]new alkaloids
[4]pharmacologically active alkaloids.

Although Sri Lanka has a rich flora and alkaloids are of medicinal significance, it seems surprising that only a little effort had been directed towards the isolation of this important group of metabolites from local plants. Recently, a comprehensive phytochemical study of Sri Lanka plants had been presented.[159] This report contained only 2 alkaloid bearing species out of the 101 plant species that have been investigated. On the other hand, an extensive screening programme had shown that out of the 464 plants tested at least 201 contained alkaloids which is above the average incidence anticipated.[157,158] However, this is not unexpected from the plants of tropical region,[172] especially as the collections have been made mainly from alkaloid rich families.

The importance of research in the field of plant alkaloids with potential medicinal applications need not be overemphasized. Since at least 830 flowering plants are unique to the island, the prospects of obtaining new drugs from this source would appear to be good. It may therefore, seem strange that pharmacologists, unlike botanists and chemists in Sri Lanka have given little attention to native plants.

There is no good reason to believe that if plants are examined at random, the chemicals extracted from them would provide a useful collection of medicinal agents. However, if folkloric claims are used in conjunction with preliminary screening programmes there is a greater chance of obtaining new drugs from local plants.

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J. Natn. Sci. Coun. Sri Lanka 1978 6 (1) : 89—90

ලිපිවල සාරාංශ - සිංහල පරිවර්තන

දේශීය රනිල වර්ග කීපයක් ගැන කරනු ලබන අධ්‍යයනයක්

II. සයනෝජනක ග්ලූකෝසයිඩ

ජැන්ස්, ඊ. ආර්. සහ පිරිස්, නිර්මලා

J. Natn. Sci. Coun. Sri Lanka 1978 **6**(1) : 1—9

ශ්‍රී ලංකාවේ බොහෝ විට දක්නට ලැබෙන 50 කට ආසන්න රනිල බෝග ඇටවල සයනයිඩ් සහ සයනෝජනක ග්ලූකෝසයිඩ් නිබෙදැයි රසායනික වශයෙන් පරීක්ෂණයකට භාජනය කරන ලදී. පාඩියෝලූස් ලූනාටුස් (ලිමා බෝංචි) ගණයට අයත් රනිල වර්ග හතරක පමණක් සයනෝජනක ග්ලූකෝසයිඩ් සැහෙන ප්‍රමාණයක් අඩංගුවී ඇති බව සොයාගන්නා ලදී. සයනෝජනක ග්ලූකෝසයිඩ් ඉවත්කළ හැකි පිරිසැකසුම් ක්‍රම කීපයක් සාර්ථක ලෙස අත්හද බලන ලදී. විග්නා සිනෙන්සිස් (පඳුරු මෑ වර්ග) පාඩියොලූස් වල්ගරිස් (වල් බෝංචි) පිසුම් සතයිවුම් (මෑ වර්ග) යන රනිලයන්ගේ දේශීය ප්‍රභේදවල අඩංගු සයනෝජනක ග්ලූකෝසයිඩ් ප්‍රමාණය වෙනස් ස්ථාන වල වැවෙන රනිල ප්‍රභේදවල ඇතැයි වාර්තාගත වී ඇති ප්‍රමාණයට වඩා අඩු බව සොයාගන්නා ලදී.

සරමේ සහ කලිසමේ මයිනහම් ප්‍රසාධය

බස්නායක, වී. සහ නිකොලස්, ඒ. ඩි.

J. Natn. Sci Coun. Sri Lanka 1978 **6**(1) : 11—14

පහළ ගාත්‍රවල හමේ යාංචනය කෙරෙහි සරම සහ කලිසම් යන ඇඳුම් වර්ග දෙක නිසා ඇතිවන මයිනහම් ප්‍රසාධය, පැරඩික්ලොරෝබෙන්සීන් නම් වාෂ්පශීල ද්‍රව්‍යයෙහි කොටස්වල බර අඩුවීම අනුව මැන බලන ලදී. කකුලේ කලවා හා ජංඝාවලට බඳින ලද වයර් කූඩුවක් තුළ ඉහතකී ද්‍රව්‍යය තබා ටිනාඩි 30 ක් ඇවිදීමට සලස්වන ලදී. බර අඩුවීමේ අනුපිළිවෙල මි. ග්‍රෑ. 100 ක් විය. කලිසම් හා සරම අතරද කලවා හා ජංඝා අතරද සැලකිය යුතු වෙනසක්, බර හානියට අදළව ඇති නොවිය. මයිනහම් ප්‍රසාධය ඇතිවන්නේ ඇඳුමට පහළින් ඇතුල්වන වාතය නිසා නොව, ඇඳුමට යටින් **සඞ්ඝාසීක** ලෙස හටගන්නා වාතයක් නිසා යයි කිව හැක.

සෙසරෝ මධ්‍යනය උදෙසා නිරපේක්ෂ අභිසාරිකා සාධක

යෝගචන්ද්‍රන්, සි.

J. Natn. Sci. Coun. Sri Lanka 1978 **6**(1) : 15—22

මේ ලිපියෙහි සාකච්ඡාවට භාජනය කොට ඇත්තේ (A) ගැටලුවේ ඒකතරා ව්‍යාප්තියකි. එනම්: $f[x]$ නිරපේක්ෂ ලෙස k අනුපිළිවෙළට අයත් සෙසරෝ සීමකය ලෙස පවතින අවස්ථාවලදී $f(x)\ g(x)$ ගුණිතය නිරපේක්ෂ ලෙස r අනුපිළිවෙළට අයත් සෙසරෝ සීමකය ලෙස පැවතීමට $g(x)$ අභිසාරිකා සාධකයට බලපාන අවශ්‍ය හා ප්‍රමාණවත් නියම යන් කවරේද ? $p > -1, p + q > -1, r, k \in N, r \geqq k$ සහ f, g යන මේවා සුදුසු ස්ථානීය නියමයන් සපුරා-ලන විට ඇතැම් l උදෙසා $f(x) \sim lx^p/c, k/$ වශයෙන්ද, ඇතැම් l උදෙසා $f(x)\ g(x) \sim lx^{p+q}/c, r/$ වශයෙන්ද පවතින විට අවශ්‍ය හා ප්‍රමාණවත් නියමයන් අපට සොයාගත හැකිය. $P = Q = 0$ වශයෙන් පවතින කල්හි, මේ ගැටළුවේදී අපට හමුවන නියමයෝ නිශ්චිත වශයෙන්ම (A) ගැටළුවට උචිතතා නියමයන්ම වෙති.

{1, 5, 10} යන අංකයන්ගේ කුලකය

බාලසුන්දරම්, එච්.

J. Natn. Sci. Coun. Sri Lanka 1978 **6** (1) : 23—26

{1, 5, 10} යන අංකයන්ගේ කුලකයට අයත් ගුණයක් නම් එම කුලකයේ කවර හෝ අංක දෙකක ගුණිතයෙන් එකක් අඩුකළ විට එම ගුණිතය වර්ගයක් බවට පත්වීමයි. එහෙත් {1, 5, 10, *c*} වැනි කුලකයකට අයත් එම ගුණයම ඇති *c* ධන නිඛිලයක් නොපවතින බව මේ ලිපියෙන් පෙන්වා දී ඇත.

තඹ සහ එහි මිශ්‍ර ලෝහ කැල්සියම් හයිපොක්ලෝරයිඩ්වල ගිල්වීමෙන් ඇතිවන විබාදනය ගැන අධ්‍යයනයක්

අමිරුද්දින්, ඒ. එම්. සහ වඩලා, එස්. එල්.

J. Natn. Sci. Coun. Sri Lanka 1978 **6**(1) : 27—38

සෙන්ටිග්‍රේඩ් 35 ක උෂ්ණත්වයකදී නඹ සහ එහි මිශ්‍ර ලෝහ එකොළහක් සියයට 60 ක ක්ලෝරීන් අඩංගු සියයට 20 ක කැල්සියම් හයිපොක්ලෝරයිට් ලයික්වෝරයක ගිල්වීමෙන් පසුව විබාදන ආචරණය අධ්‍යයනය කරන ලදී. විභව-කාල වක්‍ර, ගැල්වනෝස්ථිතික, ඇනෝඩික හා කැතෝඩික ධ්‍රැවණ ආචරණය සහ සම්මත බර හානි දත්තද මෙම අධ්‍යයනයට උපයෝගී කරගන්නා ලදී. මෙම පරීක්ෂණ තත්ත්වයන් යටතේ සියලුම මිශ්‍ර ලෝහ ස්වයං නිෂ්ක්‍රියතාවට පත්වූ බව සොයා ගන්නා ලදී. ආරක්ෂිත පටලය යැදීමට ගතවූ කාලය පැයකට අඩුවිය. මිශ්‍ර ලෝහවල විබාදන විභවය 0.650 සිට 0.680 V (Vs SCE) දක්වා විහිදී ගියේය. ඇනෝඩික ධ්‍රැවණයෙන් නිෂ්ක්‍රියතාව බිඳී ගොස්, සම්භර මිශ්‍ර ලෝහවල කැලැල් හටගැනීමද, තවත් මිශ්‍ර ලෝහවල පොදුවේ විබාදනය ඇතිවීමද දැනගත හැකිවිය. දෙවනු කී ආචරණය ඇතිවූයේ දියවන සුලු දෙයක් හටගැනීම නිසාය. පාර-නිෂ්ක්‍රිය ප්‍රදේශයේ ටැපල් බැවුම් අනුපාතය 0.120 සිට 0.254 mV දක්වා විහිදී තිබිණි. කැතෝඩික ධ්‍රැවණ වක්‍රවලින් ටැපල් ප්‍රදේශ දෙකක් දැකගත හැකිවිය. ඒවා අම්ලකර ඌනාන ප්‍රතික්‍රියාවට අනතුරුව හටගත් ජලකර විකසන ප්‍රතික්‍රියාව නිසා ඇතිවුණ බවක් පෙනුණි. ද්‍රාවණයෙහි සම්පූර්ණයෙන් ගිල්වා කරන ලද පරීක්ෂණ යටතේ ලැබුණ බර හානි දත්තවලින් පෙනී ගියේ නිකල් ටින් ලෝකඩ (උසස් වර්ගයේ නිකල්) ලෝහයට ප්‍රබල ප්‍රතිරෝධක ශක්තියක් ඇති බවය. මෙම මිශ්‍ර ලෝහයෙහි විබාදන අනුපාතය වරින්වර ද්‍රාවණයෙහි ගිල්වීමෙන් තුන් ගුණයකින්ද අර්ධ ගිල්වීම් තත්ත්වයන් යටතේ එකලොස් ගුණයකින්ද වැඩිවිය.

ශ්‍රී ලංකාවේ ඇතැම් ශාකයන් තුළ අන්තර්ගත ඇල්කලොයිඩ් වර්ග—රසායන විද්‍යාත්මක හා ඖෂධවේදිය අධ්‍යයනයක්

ගුණසිලක, ඒ. ඒ. එල්.

J. Natn. Sci. Coun. Sri Lanka 1978 **6**(1) : 39—86

ශ්‍රී ලංකාවේ ඇතැම් ශාකවල අන්තර්ගත ඇල්කලොයිඩ් සම්බන්ධයෙන් මෙතෙක් කර ඇති අධ්‍යයන එකතු කොට, රසායන විද්‍යාත්මක හා ඒවා සංශ්ලේෂණ කරුණු ගැන විශේෂ අවධානයක් ඇතිව, සමීක්ෂණයකට භාජනය කොට ඇත. විභාග කරන ලද ශාකවර්ග ප්‍රධාන කුලවලට බෙදී එම කුලයට අයත් ශාකවලින් ලැබෙන ඇල්කලොයිඩ් වර්ගවල කෙටි විස්තරයක්ද පරීක්ෂාවට බදුන් වූ ශාකයන්ගේ ලුහුබු උද්භිද විද්‍යා විස්තරයක්ද දී ඇත. ස්වදේශීය වෛද්‍ය විද්‍යාව සඳහා යහ/හෝ බටහිර වෛද්‍ය විද්‍යාව සඳහා භාවිතයට ගන්නා ශාක සම්බන්ධයෙන්, වර්තමාන ඖෂධවේදයට අදළ පරිදි **එම** ශාකවල ඇති ඇල්කලොයිඩ් ගැන සහ ඒවා භාවිත කළ යුතු ආකාරය ගැනද විස්තර කිරීමට **ප්‍රයත්නයක්** දරා ඇත.

J. Natn. Sci. Coun. Sri Lanka 1978 6(1): 91—93

இந்த இதழின் கட்டுரைகளின் சுருக்கங்கள்

உள்ளூர் அவரையினங்கள் சிலவற்றைப்பற்றிய ஆய்வுகள்

II. சயனசன் தோன்றும் குளுக்கோசைட்டுகள்

ஜான்ஸ், ஈ. ஆர். ; பீரிஸ், நிர்மலா

J. Natn. Sci. Coun. Sri Lanka 1978 6(1): 1—9

இலங்கையில் பெரும்பாலும் வளருகின்ற சுமார் 50 அவரையினங்களின் கொட்டைகளில் சயனேட்டும் சயனசன் தோன்றும் குளுக்கோசைட்டுகளும் இருக்கக்கூடிய நிலைகளைப்பற்றிய இரசாயன பகுப்பாய்வொன்று மேற்கொள்ளப்பட்டது. பாசியோலுஸ் லுனேட்டுஸ் (லீமா போஞ்சி) இனஞ் சார்ந்த அவரையங்கள் நான்கில் மட்டுமே குறிப்பிடத்தக்க அளவு சயனசன் தோன்றும் குளுக்கோசைட்டு உண்டென்பது புலனுயிற்று. சயனசன் தோன்றும் குளுக்கோசைட்டுக்களை அகற்றும் சீர்முறைகள் சிலவும் வெற்றிகரமாகப் பரிசோதிக்கப்பட்டன. விக்னசினென்சிஸ் (பயற்றஞ்செடிவகை), பாசியோலுஸ் வல்கறிஸ் (காட்டு மொச்சை) பிசும் சத்தைவும் (பயற்றங் கொடிவகை) ஆகிய அவரையங்களின் தேசிய இனங்களில் அடங்கும் சயனசன் உருவாகும் குளுக்கோசைட்டின் அளவானது ஏனைய இடங்களில் வளரும் அவரையினங்களில் உண்டெனக் கூறப்பட்டுள்ள அளவிலும் குறைவான தென்பதும் கண்டறியப்பட்டுள்ளது.

சாரம், காற்சட்டை என்பவற்றின் துருத்தி விளைவு

பஸ்நாயக்கா, வீ. ; நிகலாஸ், ஆர். ஏ. டீ.

J. Natn. Sci. Coun. Sri Lanka 1978 6(1): 11—14

கீழே காலுறுப்புத்தோல்சார் காற்றேட்டத்தின் மீது சாரம், காற்சட்டை ஆகிய உடுப்புகள் இரண்டின் காரணமாக ஏற்படும் துருத்திவிளைவு, பராதிக்லோரோ பென்சீன் என்ற ஆவியாகும் பொருளின் துண்டுகளின் எடை இழப்பினைத் துணையாகக் கொண்டு அளவிடப்பட்டது. தொடை, கணைக்கால் ஆகியவற்றோடு கட்டப்பட்ட கம்பிக் கூண்டினுள்ளோ மேற்கூறிய இரசாயனப் பொருளை இட்டு 30 நிமிடங்கள் நடக்க ஏற்பாடு செய்யப்பட்டது. எடை குறைந்து செல்லும் ஒழுங்குமுறை 100 மி.கி. ஆகவிருந்தது. காற்சட்டைக்கும் சாரத்துக்குமிடையிலும் தொடைக்கும் கணைக்காலுக்குமிடையிலும் எடை இழப்புசார் குறிப்பிடத்தக்க வித்தியாசம் இருந்ததாகத் தெரியவில்லை. துருத்தி விளைவானது கீழிலிருந்து உடுப்புக்குள் நுழையும் காற்று காரணமாக வன்றி, உடுப்புக்குள்ளேயே இருந்து தோன்றும் ஒரு வகையான காற்று காரணமாகவே ஏற்படுகிறதென்பது புலனுகும்.

சிசாரோ இடைகளுக்கான தனி ஒருங்கற் காரணிகள்

யோகசந்திரன், சி.

J. Natn. Sci. Coun. Sri Lanka 1978 **6**(1) : 15—22

இக் கட்டுரையில் ஆராயப்பட்டுள்ள பிரசினம் (*A*) பிரசினத்தின் ஒரு வகையான பிரிவாகும் : $f(x)$ தனிப் பெறுமானமுடையதாய் k வரிசைக்குரிய சிசாரோ எல்லைப்பாடாக இருக்கும் போதெல்லாம் $f(x)\ g(x)$ பெருக்கம் தனிப் பெறுமானமுடையதாய் r வரிசிசைக்குரிய சிசாரோ எல்லைப் பாடாக இருப்பதற்கு $g(x)$ ஒருங்கற் காரணிக்கு வேண்டிய மற்றும் போதுமான நிபந்தனைகள் யாவை ?

$p > -1$, $p + q > -1$, $r, k \in N$, $r \geqq k$, உம் f, g உம் ஏற்ற ஓரிட நிபந்தனைகளைப் பூர்த்திசெய்யுமிடத்து, சில l தொடர்பில் $f(x) \sim lx^p/c,\ k/$ ஆகவும் சில l தொடர்பில் $f(x)\ g(x) \sim 1'x^{p+q}/c,\ r/$ ஆகவும் இருக்கும்போது வேண்டிய மற்றும் போதுமான நிபந்தனைகளை நாம் கண்டு பிடிக்கலாம். $P = Q = 0$ ஆக இருக்கும்போது இப் பிரசினத்தில் நாம் எதிர்நோக்கும் நிபந்தனைகள் சரியாகவே (*A*) பிரசினத்திற்கு வேண்டிய நிபந்தனைகள் ஆகும்.

{1, 5, 10} ஆகிய எண்களின் தொடை

பாலசுந்தரம், எம்.

J. Natn. Sci. Coun. Sri Lanka 1978 **6**(1) : 23—26

{*1, 5, 10*} ஆகிய எண் தொடைக்குள்ள உடைமை என்னவென்றால் அத்தொடையின் எந்த எண் இரண்டின் பெருக்கத்திலிருந்தும் ஒன்று கழித்தவிடத்து, அப்பெருக்கம் ஒரு சதுரமாக இருப்பதாகும். ஆனால் {*1,5,10 c*} போன்ற ஒரு தொடைக்குரிய அதே உடைமையைக் கொண்ட c நேர்த்தொகையீடொன்று இல்லை என்பதை இக் கட்டுரை காட்டுகின்றது.

செம்பு, மற்றும் அதன் கலப்புலோகங்கள் கல்சியம் ஐப்போகுளோரைட்டுக் கரைசலில் அமிழ்த்தப்படுவதனால் ஏற்படும் அரிப்பு பற்றிய ஆய்வுகள்

அமிருதீன், ஏ. எம். ; சல்வா, எஸ். எம்.

J. Natn. Sci. Coun. Sri Lanka 1978 **6**(1) : 27—38

35 சென்றிகிறேட் வெப்பநிலையில் செம்பு, அதன்பதினைருவகையான கலப்புலோகங்களை 60 வீத குளோரீன் உள்ள 20 வீத கல்சியம் ஐப்போகுளோரைட்டுக்கரைசலில் அமிழ்த்தியபின் ஏற்பட்ட அரிப்பு நடத்தை ஆராயப்பட்டுள்ளது. அழுத்த-நேரவளையிகளும், கல்வனோநிலையியல், அனோட்டுகதோட்டுமுனைவாக்க நடத்தையும் வழக்கமான எடை இழப்புத்தரவுகளும்,

இவ்வாய்வின்போது பயன்படுத்தப்பட்டன. உபயோகிக்கப்பட்ட பரிசோதனைகளின் கீழ் எடுத்துக்கொண்ட கலப்புலோகங்கள் யாவும் சுயமாகவே தாக்கப்படா நிலை அடைந்தன. பாதுகாப்பான படலம் உருவாவதற்கு ஒரு மணித்தியாலத்திற்குச் சற்றுக் குறைவான காலம் தேவைப்பட்டது. கலப்பு லோகங்களின் அரிப்பு அழுத்தம் 0·650 முதல் 0·680 V (Vs SCE) வரையான பரப்பெல்லையைக் கொண்டிருந்தது. அனோட்டு முனைவாக்கம் காரணமாகத் தாக்கப்படாநிலை உருகுநிலைந்து, கலப்புலோகங்கள் சிலவற்றில் தழும்புக் குழிகள் ஏற்பட்டன. ஏனைய கலப்புலோகங்களில் பொதுவாக அரிப்பு ஏற்பட்டது. பின்னர் கூறப்பட்ட நடத்தையானது கரையுந்திறன் கொண்ட ஒரு பொருள் உருவானதால் ஏற்பட்டது. தாக்கப்படாநிலை கடந்த பகுதியின் தாபல் சாய்வுளிகிதங்கள் 0·120 முதல் 0·254 mV வரை பரந்திருந்தன. கதோட்டுமுனைவாக்கவளையிகளின் மூலம் இரண்டுதாபல் பகுதிகள் கண்டறியப்பட்டன. அவை ஒட்சிசன் தாழ்வுறுத் தாக்கத்தையடுத்துண்டான ஐதரசன் கூர்ப்புத்தாக்கம் காரணமாக ஏற்பட்டன என்பது புலனுகியது. கரைசலில் முழுமையாகவே அமிழ்த்துவிட்டு செய்த பரிசோதனைகளின் மூலம் கிடைத்த எடைஇழப்புத்தரவுகள் வாயிலாக நிக்கல்வெள்ளீய வெண்கல (உயர்ரக நிக்கல்) உலோகம் பெரிதும் தடைத்திறன் கொண்டுள்ளதென்பதும் தெளிவாகியது. இக்கலப்புலோகத்தின் அரிப்புவீதமானது இடைக்கிடையில் கரைசலில் இடுவதனுல் மூன்று மடங்கிலும் பகுதிஅமிழ்த்தல் நிலைகளின் கீழ் பதினெரு மடங்கிலும் அதிகரித்தது.

இலங்கையில் சில தாவரங்களில் காணப்படும் அற்கலோயிட்டுகள் — இரசாயனவியல் மற்றும் மருத்துப்பொருளியல் ஆய்வு

குணதிலக்க, ஏ. ஏ. எல்.

J. Natn. Sci. Coun. Sri Lanka 1978 **6**(1) : 39—86

இலங்கையில் இதுவரை செய்யப்பட்டுள்ள ஆய்வுகளின் அடிப்படையிலே சில தாவரங்களில் அடங்கும் அற்கலோயிட்டுக்களை பற்றிய இரசாயனவியல் மற்றும் உயிரியற்றெகுப்புசார் ஆய்வொன்று இங்கு மேற்கொள்ளப்பட்டுள்ளது. ஆய்வுக்குட்பட்ட தாவரங்களைப் பிரதான குடும்பங்களின் அடிப்படையில் காட்டு வதோடல்லாமல் அக்குடும்பத்துக்குரிய தாவரங்களிலிருந்து பெறக்கூடிய அற்கலோயிட்டுகள் பற்றிய ஒரு விளக்கமும் ஆய்வுக்குட்பட்ட தாவரங்களின் தாவரவியல் விளக்கமும் கொடுக்கப்பட்டுள்ளன. இந்நாட்டு மருததுவக் கலைக்காகவும் அல்லது மேனாட்டு மருத்துக் ககலைக்காகவும் பயன்படுத்தப்படும் தாவரங்களைப் பொறுத்தவரை, நவீன மருத்துப் பொருளியலுக்கமைவாக, அத்தாவரங்களிலுள்ள அற்கலோயிட்டுகளைப்பற்றியும் அவற்றைப் பயன்படுத்த வேண்டியமுறை பற்றியும் எடுத்துரைக்க முயன்றுள்ளது.

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Contents

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