

بِسْمِ اللّٰهِ الرَّحْمٰنِ الرَّحِیْمِ

*IN THE NAME OF ALLAH, THE BENEFICENT,
THE MERCIFUL*

*Read! And thy Lord is Most Honorable and Most Benevolent,
Who taught (to write) by pen,
He taught man that which he knew not*

*(Surah Al-Alaq 30: 3-5)
Al-Quran*

**PHYTOCHEMICAL STUDIES ON *DURANTA*
REPENS LINN AND *EHRETIA*
OBTUSIFOLIA HOCHST**

**Thesis Submitted
for
The Fulfillment of the Degree of**

DOCTOR OF PHILOSOPHY

BY

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2004

Dedicated to

my most loving parents

Hafiza Iqbal

and

Raja Mohammad Iqbal Advocate

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2004

Summary

The drugs of plant origin have served through the ages as the mainstay for the treatment of human ailments. The knowledge of drugs derived from medicinal plants, therefore, goes back to pre-historic times. The man, as a savage, most has known by experience how to relieve his sufferings by the use of herbs growing around him. The historical analyses of traditional medicine reveals that in his search for food the primitive men began to distinguish those plants suitable for nutritional purposes from others possessing definite pharmacological actions.

Sizeable contributions towards the knowledge of plant based drugs have been made by various civilizations including the Muslim and Greek civilizations. In the Indo-Pak sub-continent there has been a fairly organized effort for pharmaco-chemical studies in the physiological active plant constituents. The H. E. J. Research Institute of Chemistry, which is among the finest institute of the world in natural product chemistry, founded in 1967 by the world renowned scientist late Prof. Salimmuzzaman Siddiqui. Currently it is led by the famous scientist of Pakistan Prof. Atta-ur-Rahman. Hundreds of medicinal plants are studied annually in the Institute for the discovery of plant based medicinal compounds.

The present Ph.D dissertation compiles the phytochemical investigation on two medicinal plants of Pakistan namely *Duranta repens* Linn. and *Ehretia obtusifolia* Hochst. The thesis, therefore, is presented in following two parts:

- ◆ PART A “Phytochemical studies on *Duranta repens* Linn.”
- ◆ PART B “Phytochemical studies on *Ehretia obtusifolia* Hochst.”

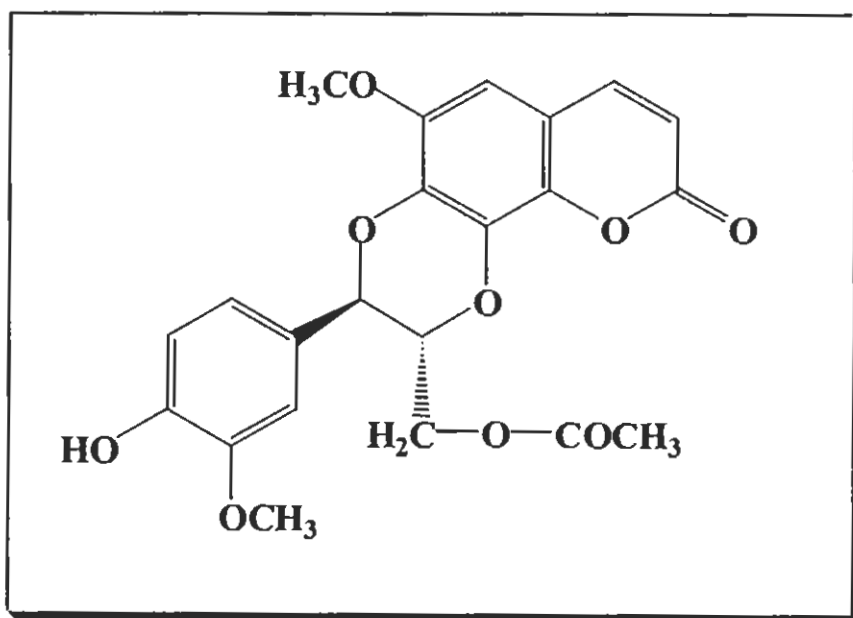
An Overview of Part A

“PHYTOCHEMICAL STUDIES ON *DURANTA REPENS* LINN.”

This part describes the isolation and structural elucidation of seven new compounds. In addition, three known compounds are reported for the first time from this species.

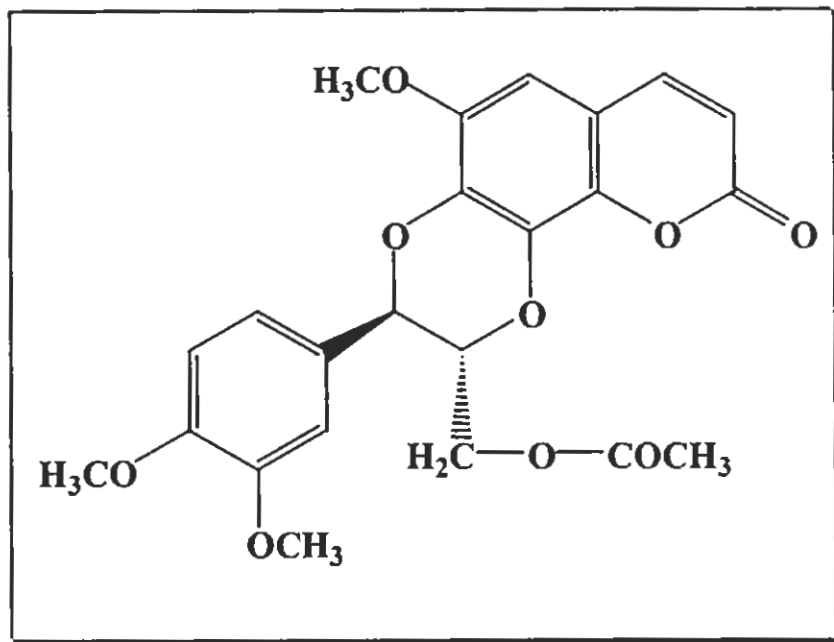
◆ New compounds isolated from *Duranta repens*

1) Durantin A (144)



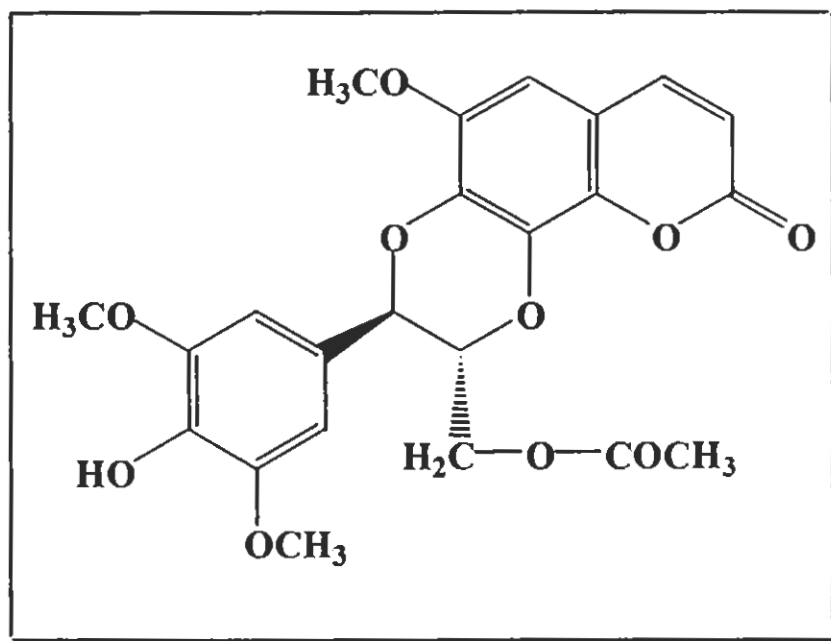
Heterocycles, **60**, 151-157, 2003

2) Durantin B (145)



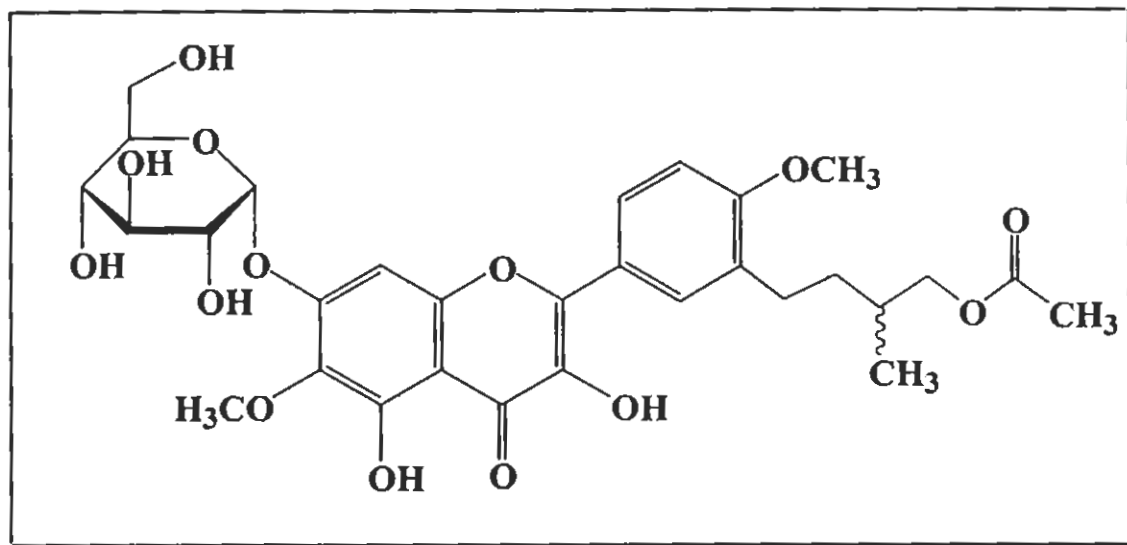
Heterocycles, **60**, 151-157, 2003

3) Durantin C (146)



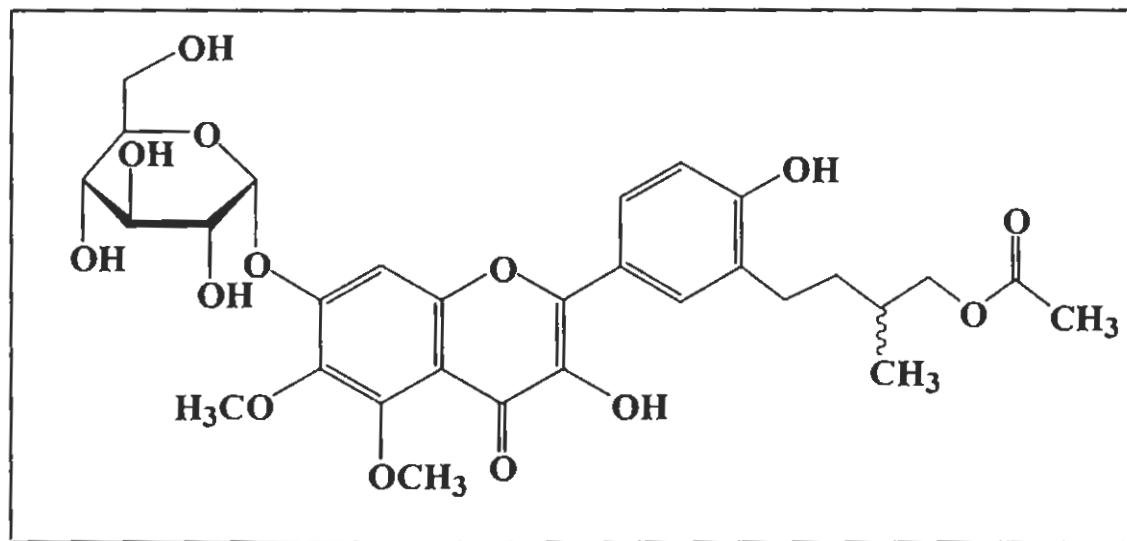
Heterocycles, **60**, 151-157, 2003

- 4) 7-O- α -D-Glucopyranosyl-3,5-dihydroxy-3'-(4''-acetoxy-3''-methylbutyl)-6,4'-dimethoxyflavone (147)



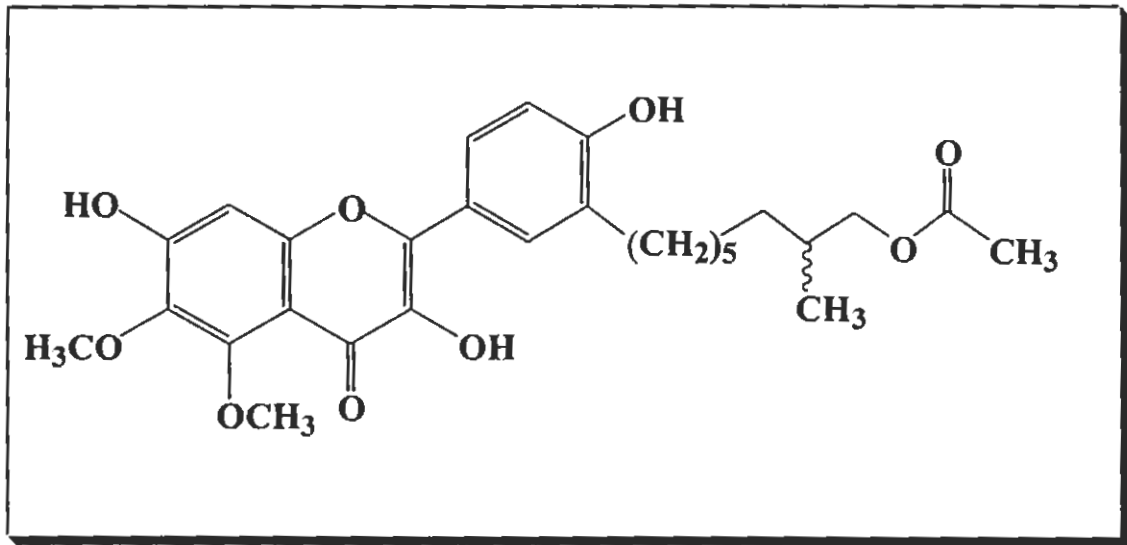
Chem. Pharm. Bull., 52, 785-789, 2004

- 5) 7-O- α -D-Glucopyranosyl-3,4'-dihydroxy-3'-(4''-acetoxy-3''-methylbutyl)-5,6-dimethoxyflavone (148)



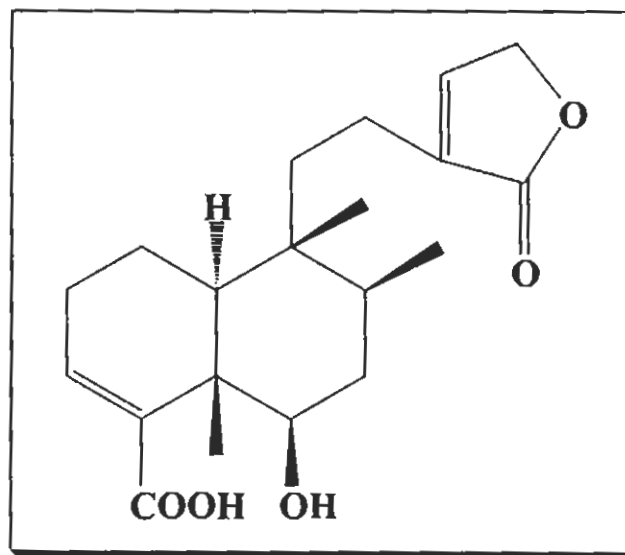
Chem. Pharm. Bull., 52, 785-789, 2004

- 6) 3,7,4'-Trihydroxy-3'-(8''-acetoxy-7''-methyloctyl)-5,6-dimethoxyflavone
(149)



Chem. Pharm. Bull., 52, 785-789, 2004

- 7) (-)-6 β -Hydroxy-5 β ,8 β ,9 β ,10 α -cleroda-3,13-dien-16,15-olid-18-oic
acid (150)



Chem. Pharm. Bull., 52, 785-789, 2004

◆ **Compounds isolated for the first time from *Duranta repens***

- 1) Cleomiscosin A (151)
- 2) (+)-Hardwickiic acid (152)
- 3) (+)-3,13-Clerodadien-16,15-olid-18-oic acid (153)

◆ **Compounds already reported from *Duranta repens***

- 1) (-)-6 β -Hydroxy-15,16-epoxy-5 β ,8 β ,9 β ,10 α -cleroda-3,13(16),14-trien-18-oic acid (154)
- 2) 6,7,8-Trimethoxycoumarin (155)
- 3) 5,4'-Dihydroxy-3,6,7-trimethoxyflavone (156)
- 4) 3,7,4'-Trihydroxy-3'-(4-hydroxy-3-methylbutyl)-5,6-dimethoxyflavone (157)
- 5) 3,7-Dihydroxy-3'-(4-hydroxy-3-methylbutyl)-5,6,4'-trimethoxyflavone (158)
- 6) Betulin (159)
- 7) Stigmasterol (160)
- 8) Stigmasterol 3-*O*- β -D-glucopyranoside (161)
- 9) β -Sitosterol 3-*O*- β -D-glucopyranoside (162)

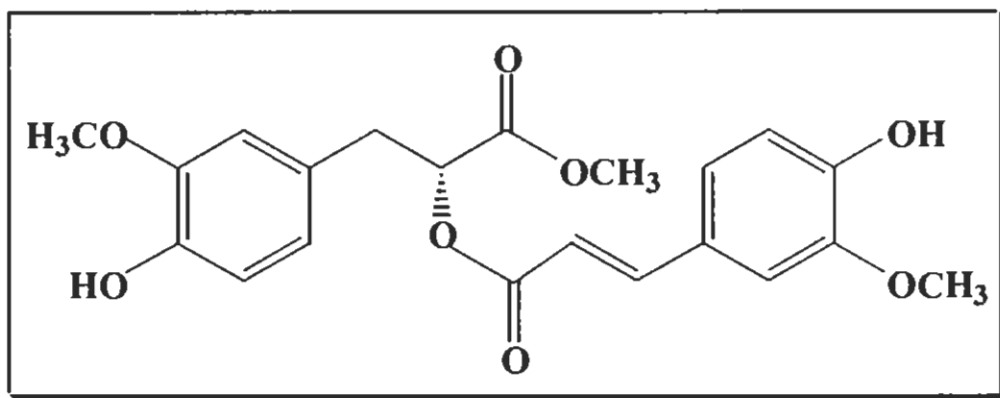
An Overview of Part B

“PHYTOCHEMICAL STUDIES ON *EHRETIA OBTUSIFOLIA* HOCHST.”

This part describes the isolation and structural elucidation of three new compounds along with seven known compounds reported for the first time from this species

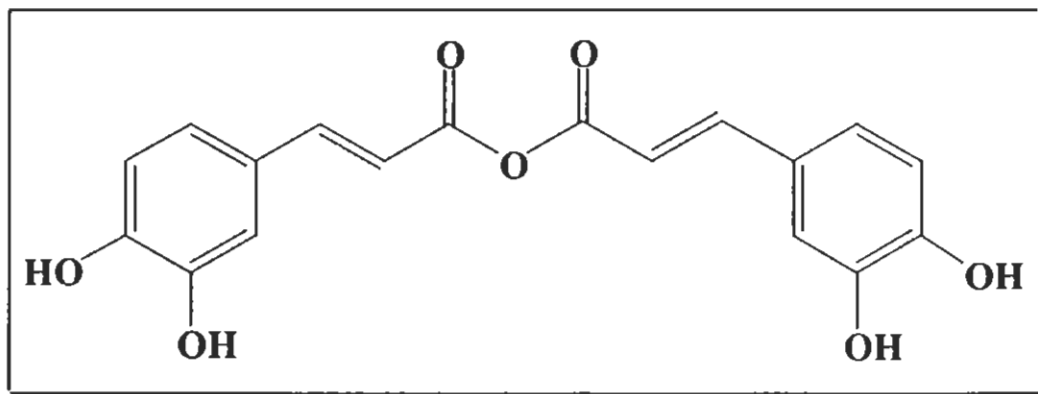
◆ New compounds isolated from *Ehretia obtusifolia*

1) Methyl 2-*O*-feruloyl-1 α -*O*-vanillactate (178)



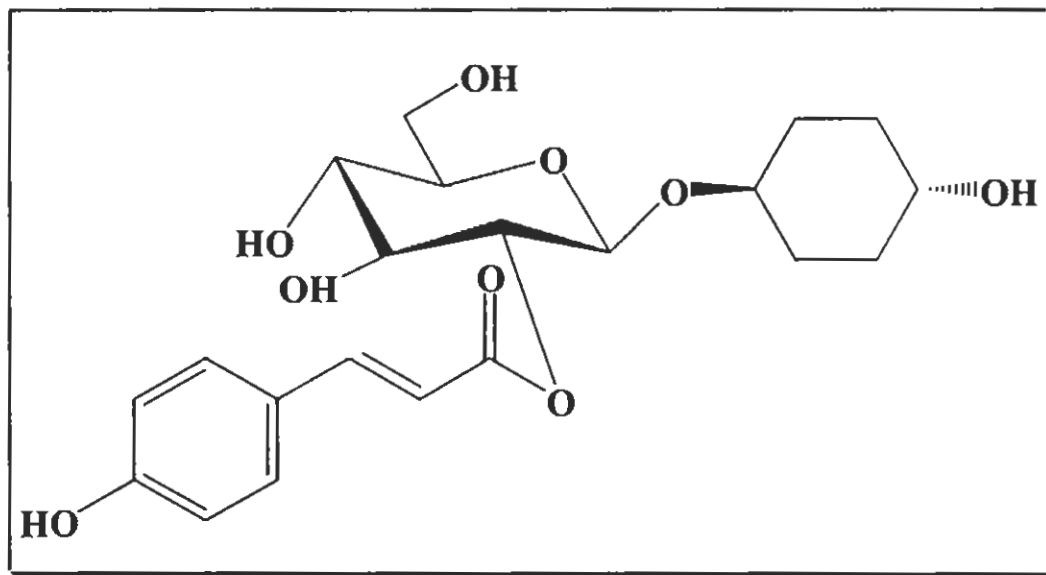
Chem. and Biodiver. (in press)

2) Caffeic anhydride (179)



Chem. and Biodiver. (in press)

3) *trans* 4-Hydroxycyclohexyl-2-*O-p*-coumaroyl β -D-glucopyranoside (180)



Chem. and Biodiver. (in press)

◆ **Compounds isolated for the first time from *Ehretia obtusifolia***

- 1) Methyl rosmarinate (181)
- 2) Rosmarinic acid (182)
- 3) Quercetin 3-*O*- β -D-glucopyranoside (183)
- 4) Quercetin 3- β -D-glucopyranosyl-4-*O*- β -D-glucopyranoside (184)
- 5) Ursolic acid (185)
- 6) β -Sitosterol (186)
- 7) β -Sitosterol 3-*O*- β -D-glucopyranoside (187)

Apart from the chemical work, pharmacological studies on the isolated compounds have also been undertaken to ascertain their therapeutic potentials.

خلاصہ

نباتی الاصل دوائیں زمانوں سے انسانی بیماریوں کے علاج کا اہم ترین ذریعہ رہی ہیں۔ چنانچہ ادویاتی پودوں سے دواؤں کے حصوں کا علم بھی باقبل از تاریخ ادوار سے تعلق رکھتا ہے۔ جب انسان غیر مہذب تھا تو اس وقت اس نے اپنے تجربے سے یہ سیکھا کہ اپنے ارد گرد اگنے والی جڑی بوٹیوں کے استعمال سے تکلیفوں کا علاج کیسے کیا جائے۔ روایتی دواؤں کے تاریخی تجزیوں سے ظاہر ہوتا ہے کہ خوراک کی تلاش کے دوران قدیم انسان نے عام غذائی پودوں اور معالجاتی اثرات کے حامل پودوں کے درمیان امتیاز کرنا سیکھ لیا تھا۔ پودوں سے حاصل کردہ دواؤں کے علوم میں اسلامی اور یونانی تہذیبوں سمیت مختلف تہذیبی ادوار میں کافی کام ہوا ہے۔ برصغیر ہندوستان میں پودوں کے لیے نمایاں طور پر منظم کوششیں کی گئی ہیں۔ ایچ ای جے ریسرچ انسٹی ٹیوٹ آف کیمسٹری ۱۹۶۷ء میں پاکستان کے عالمی شہرت یافتہ سائنس دان مرحوم پروفیسر سلیم الزماں صدیقی نے قائم کیا جو دنیا کے ”اصلی پیداواری کیمیا“ بہترین اداروں میں سے ایک ہے اور اب پاکستان کے مشہور سائنس دان پروفیسر ڈاکٹر عطاء الرحمن اس کے سربراہ ہیں۔ اس انسٹی ٹیوٹ میں ادویات مرکبات پر مشتمل نباتات کی دریافت کے میدان میں ہر سال سینکڑوں ادویاتی پودوں کا مطالعہ کیا جاتا ہے۔

زیر نظر بی ایچ ڈی مقالہ پاکستان کے دوا دویاتی پودوں کی نباتی کیمیائی تحقیقات پر مشتمل ہے جن کے نام یہ ہیں:

Ehretia obtusifolia Hochst. اور *Duranta repens* Linn.

یہ مقالہ دو حصوں پر مشتمل ہے جو درج ذیل ہیں:

◆ حصہ اول *Duranta repens* Linn. کا نباتی کیمیائی مطالعہ "

◆ حصہ دوم *Ehretia obtusifolia* Hochst. کا نباتی کیمیائی مطالعہ "

حصہ اول کا جائزہ

" *Duranta repens* Linn. کا نباتی کیمیائی مطالعہ "

اس حصے میں ۷ نئے مرکبات کی علیحدگی اور ساختی توضیحات بیان کی گئی ہیں۔ اس کے ہمراہ اس نوع سے تین معلوم مرکبات پہلی مرتبہ حاصل

کیے گئے ہیں۔

◆ *Duranta repens* سے علیحدہ کئے گئے نئے مرکبات

- ۱- ڈیورنٹین اے (۱۳۳)
- ۲- ڈیورنٹین بی (۱۳۵)
- ۳- ڈیورنٹین سی (۱۳۶)
- ۴- D- α -O- ϵ -گلوکوپائریزوسائل-۵،۳-ڈائی ہائیڈروکسی-۳- (۳-ایسیٹوکسی-۳- میتھائل بیوٹائل)-۶،۶-ڈائی میتھاکسی فلیون (۱۳۷)
- ۵- D- α -O- ϵ -گلوکوپائریزوسائل-۳،۳-ڈائی ہائیڈروکسی-۳- (۳-ایسیٹوکسی-۳- میتھائل بیوٹائل)-۶،۵-ڈائی میتھاکسی فلیون (۱۳۸)
- ۶- ۳،۴،۷-ٹرائی ہائیڈروکسی-۳- (۸-ایسیٹوکسی-۷- میتھائل اڈکٹائل)-۶،۵-ڈائی میتھاکسی فلیون (۱۳۹)
- ۷- (-)- β -۶-ہائیڈروکسی-۵- β ۸، β ۹، β ۱۰، α -کلیروڈا-۱۳،۳-ڈیہین-۱۵،۱۶-آئیملڈ-۱۸-اونیک ایسڈ (۱۵۰)

◆ *Duranta repens* سے پہلی مرتبہ علیحدہ کئے گئے مرکبات

- ۱- کلیومیسکو بن اے (۱۵۱)
- ۲- (+)-ہارڈی ویکل ایسڈ (۱۵۲)
- ۳- (+)-۱۳،۳-کلیروڈیڈائن-۱۵،۱۶-آئیملڈ-۱۸-اونیک ایسڈ (۱۵۳)

◆ *Duranta repens* سے پہلے سے منقول مرکبات

- ۱- (-)- β -۶-ہائیڈروکسی-۱۶،۱۵-ایپاکسی-۵، β ۸، β ۹، β ۱۰، α -کلیروڈا-۱۳،۳ (۱۶)-ٹرائین-۱۸-اونیک ایسڈ (۱۵۴)
- ۲- ۸،۷،۶-ٹرائی میتھاکسی کیومیرائین (۱۵۵)
- ۳- ۴،۵-ڈائی ہائیڈروکسی-۳،۶،۳-ٹرائی میتھاکسی فلیون ٹل (۱۵۶)
- ۴- ۳،۴،۷-ٹرائی ہائیڈروکسی-۳- (۳-ہائیڈروکسی-۳- میتھائل بیوٹائل)-۶،۵-ڈائی میتھاکسی فلیون (۱۵۷)
- ۵- ۷،۳-ڈائی ہائیڈروکسی-۳- (۳-ہائیڈروکسی-۳- میتھائل بیوٹائل)-۴،۶،۵-ٹرائی میتھاکسی فلیون (۱۵۸)
- ۶- بیٹیولائین (۱۵۹)
- ۷- سکلاشیرول (۱۶۰)
- ۸- سکلاشیرول ۳-D- β -O- ϵ -گلوکوپائریزوسائیڈ (۱۶۱)
- ۹- β -سیٹوشیرول ۳-D- β -O- ϵ -گلوکوپائریزوسائیڈ (۱۶۲)

حصہ دوم کا جائزہ

"*Ehretia obtusifolia* Hochst. کا نباتی کیمیائی مطالعہ"

اس حصے میں ۳ نئے مرکبات کی علیحدگی اور ساختی توضیحات بیان کی گئی ہیں۔ اس کے ہمراہ اس نوع سے ۷ مرکبات پہلی مرتبہ حاصل کئے گئے ہیں۔

◆ *Ehretia obtusifolia* سے علیحدہ کئے گئے نئے مرکبات

- ۱۔ میٹھائل O-۲-فیرولولینیل-O-1a-وینی لکٹینٹ (۱۷۸)
- ۲۔ کیفیک این ہائیڈرائیزڈ (۱۷۹)
- ۳۔ ٹرانس ۳-ہائیڈروکسی سائی کلوہیگروائل-p-O-۲-کومورائیل-D-β-گلوکو پائیرینوسائیڈ (۱۸۰)

◆ *Ehretia obtusifolia* سے پہلی مرتبہ علیحدہ کئے گئے مرکبات

- ۱۔ میٹھائل روز میرینٹ (۱۸۱)
- ۲۔ روز میرینک ایسڈ (۱۸۲)
- ۳۔ کیورسٹن D-β-O-۳-گلوکو پائیرینوسائیڈ (۱۸۳)
- ۴۔ کیورسٹن D-β-O-۳-گلوکو پائیرینوسائل-D-β-O-۳-گلوکو پائیرینوسائیڈ (۱۸۴)
- ۵۔ ارسولک ایسڈ (۱۸۵)
- ۶۔ β-سیٹوشیرول (۱۸۶)
- ۷۔ β-سیٹوشیرول D-β-O-۳-گلوکو پائیرینوسائیڈ (۱۸۷)

اس کے کیمیائی افعال سے ہٹ کر، علیحدہ مرکبات کا ادویاتی نقطہ نگاہ سے بھی مطالعہ کیا گیا تاکہ ان کی ادویاتی افادیت کا تعین کیا جاسکے۔