

2010–2020 年红厚壳属植物化学成分及生物活性研究进展

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[摘要] 红厚壳属植物资源丰富, 植物中富含香豆素类、双苯吡酮类、黄酮以及萜类等化合物, 具有抗肿瘤、抗 HIV-1、抗炎、抑菌、抗氧化等药理作用。通过查阅和分析相关文献, 对近 10 a 红厚壳属植物的化学成分和药理作用进行全面整理, 旨在为该属植物的进一步研究和开发利用提供参考。

[关键词] 红厚壳属; 化学成分; 药理活性

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A Review on Chemical Components and Bioactivities of Calophyllum Plants

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[Abstract] Plants in Calophyllum genus are rich in compounds including coumarins, xanthenes, flavonoids and terpenoids, with pharmacological activities on anti-tumor, anti-HIV-1, anti-inflammatory, antibacterial and antioxidative effect. In order to provide directions for the further research of the Calophyllum genus, we reviewed studies on the chemical constituents and pharmacology from year 2010–2020.

[Key words] Calophyllum; Chemical constituent; Pharmacological activity

红厚壳属 (Calophyllum) 植物为藤黄科 (Guttiferae) 常青乔木, 全世界有 180 ~ 200 种, 主要分布在马来西亚、菲律宾、印度、斯里兰卡等亚热带地区, 其次则分布在南美洲和大洋洲^[1]。我国主要有 4 种: 红厚壳 (Calophyllum inophyllum linn)、薄叶红厚壳 (Calophyllum membranaceum)、兰屿红厚壳 (Calophyllum blancoi) 和滇南红厚壳 (Calophyllum polyathum), 主要分布在海南、广西南部、云南南部及台湾^[2]。

自 1993 年 Patil 等^[3]从 Calophyllum inophyllum 中分离得到具有显著的抑制 HIV 逆转录酶活性的香豆素类化合物 inophyllum B 和 inophyllum P 以来, 国外学者对红厚壳属植物的化学成分进行了大量

的研究, 分离得到香豆素类 (Coumarins)、双苯吡酮类 (Xanthenes)、黄酮类 (Flavonoids)、萜类 (Terpenoids) 等多种类型化合物^[4-5]。经现代药理学研究表明, 红厚壳属植物药用活性成分具有抗肿瘤、抗 HIV-1、抗菌、抗炎、抗氧化等多种生物活性, 极具研究和开发价值。因此, 本文就红厚壳属植物的化学成分和生物活性研究作一综述。

1 红厚壳属的化学成分与分布

红厚壳属植物主要含有香豆素类、双苯吡酮类、黄酮类、萜类等多种类型化合物。

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1.1 香豆素类 (Coumarins)

香豆素类化合物具有苯骈 α -吡喃酮母核, 可视为邻羟基桂皮酸形成的内酯, 环上常含有羟基、烷氧基、苯基、异戊烯基等取代基。红厚壳属植物

中的香豆素类成分的结构按内酯环上取代基不同, 可分为三类: 丙基取代、苯基取代、甲基取代香豆素^[6], 见表 1。

表 1 红厚壳属植物中的香豆素类化合物

编号	化合物名称	来源	参考文献
1	Apetalolide	C. dioscurii	[7]
2	Methyl inophyllum P	C. dioscurii	[7]
3	Calopolynolide B	C. depressinervosum	[8]
4	Benjaminin	C. benjaminum	[9]
5	Gracilenins A	C. gracilentum	[10]
6	Gracilenins B	C. gracilentum	[10]
7	Gracilenins C	C. gracilentum	[10]
8	Inophyllum H	C. inophyllum	[11]
9	Inophyllum I	C. inophyllum	[11]
10	Inophyllum J	C. inophyllum	[12]
11	12-O-Butylinophyllum D	C. inophyllum	[11]
12	12-O-Ethylinophyllum D	C. inophyllum	[11]
13	(-)-12-Methoxyinophyllum A	C. inophyllum	[12]
14	(+)-12-Methoxyinophyllum H-1	C. inophyllum	[12]
15	(-)-12-Methoxyinophyllum H-2	C. inophyllum	[12]
16	Hoseimarin	C. hosei	[13]
17	Incrassamarin A	C. incrassatum	[1]
18	Incrassamarin B	C. incrassatum	[1]
19	Incrassamarin C	C. incrassatum	[1]
20	Incrassamarin D	C. incrassatum	[1]
21	Iso-soulattrolide	C. symingtonianum	[14]
22	Soulamarin	C. soulattri	[15]
23	Wallimarin T	C. wallichianum	[16]
24	7,4'-Dihydroxy-6,8-dimethoxy-4-phenylcoumarin	C. polyanthum	[17]
25	7-Hydroxy-6,8,4'-trimethoxy-4-phenylcoumarin	C. polyanthum	[17]
26	7-Hydroxy-4'-methoxy-4-phenylcoumarin	C. polyanthum	[17]
27	6,7-Dihydroxy-4'-methoxy-4-phenylcoumarin	C. polyanthum	[17]
28	α -Hydroxytomentolide A	C. apetalum	[18]
29	5,7-Dihydroxy-6-(3-methylbutyryl)-4-phenylcoumarin	C. sclerophyllum	[19]
30	7,8-Dihydro-5-hydroxy-7,8-dimethyl-4-propyl-2H,6H-benzo[1,2-b:5,4-b']dipyrans-2,6-dione	C. incrassatum	[1]

1.2 双苯吡酮类 (Xanthenes)

双苯吡酮类又称咕吨酮、氧杂蒽酮、苯骈色原酮等, 具有苯骈 γ -吡喃酮环的基本骨架。双苯吡酮类化合物具有多样的取代基结构及不同的取代模式, 且其母核容易发生扭曲变化, 也有一

些双苯吡酮类化合物^[20], 见表 2。

1.3 苯并二氢吡喃-4-酮类 (4-chromanones)

苯并二氢吡喃-4-酮又称 4-二氢色原酮, 红厚壳属中的苯并二氢吡喃-4-酮类化合物, 见表 3。

表 2 红厚壳属植物中的双苯吡酮类化合物 (1)

编号	化合物名称	来源	参考文献
1	1,6-Dihydroxy-5,7-dimethoxyxanthone	<i>C. ceriferum</i>	[22]
2	1,7-Dihydroxy-2,3,8-trimethoxyxanthone	<i>C. ceriferum</i>	[22]
3	1,3,7-Trihydroxy-8-methoxyxanthone	<i>C. inophyllum</i>	[23]
4	1, 3, 6-Trihydroxy-5-methoxy-xanthone	<i>C. membranaceum</i>	[24]
5	1, 3, 7-Trihydroxyxanthone	<i>C. membranaceum</i>	[24]
6	1-Hydroxy-6, 7-dimethoxyxanthone	<i>C. membranaceum</i>	[24]
7	1, 4, 5-Trihydroxyxanthone	<i>C. membranaceum</i>	[24]
8	2-Deprenyl-rheediaxanthone B	<i>C. inophyllum</i>	[23]
9	2-Hydroxy-1-methoxyxanthone	<i>C. inophyllum</i>	[23]
10	2-Hydroxy-1,8-dimethoxyxanthone	<i>C. inophyllum</i>	[23]
11	3, 5- Dihydroxyxanthone	<i>C. austroindicum</i>	[24]
12	4-Hydroxy-3-methoxyxanthone	<i>C. membranaceum</i>	[24]
13	6-Hydroxy-2,3-dimethoxyxanthone	<i>C. inophyllum</i>	[23]
14	7-Hydroxy-1,3-dimethoxyxanthone	<i>C. inophyllum</i>	[23]
15	1,6-Dihydroxy-5-methoxy-8-(2-methyl-2-oxo-3-butenyl)-40,40,50-trimethyl-dihydrofurano(20,30:3,4)xanthone	<i>C. inophyllum</i>	[25]
16	7,9,12-Trihydroxy-2,2-dimethyl-2H,6H-pyrano[3,2-b]xanthen-6-one	<i>C. elatum</i>	[26]
17	12b-Hydroxy-des-D-garcigerrin A	<i>C. elatum</i>	[26]
18	Dulciol E	<i>C. elatum</i>	[26]
19	Ananixanthone	<i>C. venulosum</i>	[27]
20	Buxixanthone	<i>C. buxifolium</i>	[28]
21	Caloxanthone I	<i>C. venulosum</i>	[27]
22	Caloxanthone O	<i>C. inophyllum</i>	[21]
23	Caloxanthone P	<i>C. inophyllum</i>	[21]
24	Caloxanthone Q	<i>C. inophyllum</i>	[29]
25	Caloxanthone T	<i>C. inophyllum</i>	[23]
26	Calophylxanthones A	<i>C. membranaceum</i>	[30]
27	Calophylxanthones B	<i>C. membranaceum</i>	[30]
28	Calotetrapterin A	<i>C. tetrapterum</i>	[31]
29	Calotetrapterin B	<i>C. tetrapterum</i>	[31]
30	Calotetrapterin C	<i>C. tetrapterum</i>	[31]
31	Calothorexanthone	<i>C. thorelii</i>	[32]
32	Calaxanthone C	<i>C. calaba</i>	[34]
33	Fuscaxanthone C	<i>C. benjaminum</i>	[9]
34	Garbogiol	<i>C. thorelii</i>	[32]
45	Globuxanthone	<i>C. thorelii</i>	[32]
46	Isogarciniaxanthone E	<i>C. elatum</i>	[26]

表 2 红厚壳属植物中的双苯吡酮类化合物 (2)

编号	化合物名称	来源	参考文献
37	Inophinnin	C. inophyllum	[35]
38	Inophinone	C. inophyllum	[36]
39	β -Mangostin	C. hosei	[13]
40	Nodusuxanthone	C. nodusum	[23]
41	Osajaxanthone	C. hosei	[13]
42	Phyllatrin	C. soulattri	[37]
43	Soulattrin	C. soulattri	[38]
44	Symphoxanthone	C. elatum	[26]
45	Rheediaxanthone A	C. inophyllum	[36]
46	Thorexanthone	C. thorelii	[32]
47	Tovopyrifolin C	C. venulosum	[27]
48	Venuloxanthone	C. venulosum	[27]
49	Xanthochymone B	C. depressinervosum	[8]

表 3 红厚壳属植物中的苯并二氢吡喃 -4- 酮类化合物

编号	来源	来源	参考文献
1	Apetalic acid	C. brasiliense	[39]
2	Brasiliensic acid	C. brasiliense	[40]
3	Isobrasiliensic acid	C. brasiliense	[40]
4	Blancoic acid	C. castaneum	[19]
5	Isoblancoic acid	C. castaneum	[19]
6	Calopolyanic acid	C. polyanthum	[41]
7	Isocalopolyanic acid	C. polyanthum	[41]
8	Isorecedensic acid	C. polyanthum	[41]
9	Calopolyanic acid methyl ester	C. membranaceum	[30]
10	Isopinetoric acid methyl ester	C. membranaceum	[30]
11	Isoapetalic acid	C. antillanum	[43]
12	Calodryobalanoic acid	C. dryobalanoides	[42]
13	Calolongic acid	C. Pinetorum& C. teysmannii	[43- 44]
14	Isocalolongic acid	C. Pinetorum&C. teysmannii	[43- 44]
15	Caloteysmannic acid	C. teysmannii	[44]
16	Caloinophyllin A	C. inophyllum	[45]
17	Caloinophyllin B	C. inophyllum	[46]
18	Isopinetoric acid III	C. pinetorum	[43]

2 药理作用

2.1 抗肿瘤活性

2010 年, Dai 等^[21]从 C. inophyllum 中分离得的双苯吡酮类化合物 caloxanthone O 对人胃癌细胞 (SGC-7901) 有细胞毒性, 其 IC₅₀ 为 22.4 μg/mL。2011 年, 魏代静等^[23]从 C. inophyllum 中分离得的双苯吡酮类化合物 1, 3, 5-trihydroxy-2 -

methoxy - xanthone 同样对人胃癌细胞 (SGC-7901) 的增殖显示出生长抑制活性, 其 IC₅₀ 值为 1.8 × 10⁻⁵ mol/L。

2017 年, Shanmugapriya 等^[47]研究发现 C. inophyllum 果实提取物不仅可以在 G₀/G₁ 期和 G₂/M 期诱导细胞周期停滞和凋亡, 而且可通过增强细胞内活性氧, 干扰正常的线粒体膜电位和激活 Caspase-3 来影响线粒体的凋亡途径, 进而对

MCF-7 细胞具有显著的抗肿瘤活性, IC_{50} 值为 $23.59 \mu\text{g/mL}$ 。

2018 年, Sichaeem 等^[46]使用 MTT 比色法评估从 *C. inophyllum* 分离的 18 个化合物对人口腔表皮样癌细胞 (KB) 和人宫颈腺癌细胞 (HeLa S-3) 的体外细胞毒性活性, 其中双苯吡酮类化合物 1, 6-dihydroxy-7-methoxyxanthone 对人口腔表皮样癌细胞 (KB) 和人宫颈腺癌细胞 (HeLa S-3) 显示中等活性, IC_{50} 值分别为 25.8 和 $18.1 \mu\text{M}$ 。Kar Wei 等^[48]对 *C. teysmannii* 中分离出双苯吡酮类化合物 ananixanthone 以及通过对其乙酰化, 甲基化和苄基化作用得到的四个衍生物进行人胃癌细胞 (SNU-1) 和人慢性髓性白细胞 (K562) 细胞毒性活性测试。测试显示, Ananixanthone 的细胞毒性优于其他衍生物, IC_{50} 值分别为 (8.97 ± 0.11) 和 $(2.96 \pm 0.06) \mu\text{g/mL}$, 侧链的修饰并没有增强其对这两种癌细胞系的细胞毒性。

2.2 抗炎活性

2011 年, Ee 等^[35]从 *C. inophyllum* 茎皮分离得的双苯吡酮类化合物 inophinnin, 其具有抗炎活性。2012 年, Tsai 等^[49]在脂多糖 (LPS) 诱导的 RAW 264.7 细胞上测试了 *C. inophyllum* 丙酮提取物的抗炎活性, 结果表明 *C. inophyllum* 提取物通过抑制 iNOS 和 COX-2 的表达以及 miR-146a 的表达而具有很强的抗炎作用。2014 年, Zakaria 等^[50]研究发现 *C. inophyllum* 果实提取物具有抗炎作用, $50 \mu\text{g/mL}$ 的 *C. inophyllum* 粗提物对环氧合酶和脂氧合酶活性的抑制率分别为 77% 和 88%。2017 年, Nguyen 等^[51]发现 *C. inophyllum* 种子的主要成分香豆素 Calophyllolide 具有抗炎活性, 其能诱导抗炎细胞因子 IL-10 的表达, 同时抑制促炎细胞因子 IL-1 β , IL-6, TNF- α 的表达。

2.3 抗菌活性

2010 年, Mishra 等^[52]发现与 *C. inophyllum* 茎皮氯仿提取物相比, 甲醇提取物对革兰氏阳性菌和革兰氏阴性菌有明显的抗菌作用。研究表明, *C. inophyllum* 茎皮的甲醇提取物在 $25.0 \mu\text{g/mL}$ 浓度对铜绿假单胞菌、金黄色葡萄球菌和表皮葡萄球菌具有显著的抗菌作用, 在 $50.0 \mu\text{g/mL}$ 浓度对地衣芽孢杆菌、枯草芽孢杆菌、大肠杆菌和肺炎克雷伯菌具有显著的抗菌作用。2012 年, Alkhamaiseh 等^[53]对 *C. canum* 三种不同极性组分提取物进行蜡状芽孢杆菌、金黄色葡萄球菌、大肠杆菌、铜绿假单胞菌、白色念珠菌和新生隐球菌抗菌试验, 发现其中正己烷组分对蜡样芽孢杆菌的活性最高, MIC 和 MBC 分别为 $12.5 \mu\text{g/mL}$ 和 $100.0 \mu\text{g/mL}$,

DCM 组分对金黄色葡萄球菌的活性更强, MIC 为 $50.0 \mu\text{g/mL}$ 。2018 年, Susanti 等^[54]采用纸片扩散法检测从 *C. symingtonianum* 的茎皮中分离得到的香豆素类化合物 12-O-ethylinophyllum D 和 iso-soulattrolide 对金黄色葡萄球菌、蜡状芽孢杆菌、大肠杆菌和铜绿假单胞菌的抑菌活性, 结果表明两个化合物对铜绿假单胞菌均有抑制作用, MIC 为 $111.0 \mu\text{g/mL}$, iso-soulattrolide 对金黄色葡萄球菌也具有抑制活性, MIC 为 $25.0 \mu\text{g/mL}$ 。

2.4 抗氧化活性

2012 年, Prasad 等^[55]发现, 从 *C. inophyllum* 叶中分离得的三萜类化合物 Canophyllic acid 和黄酮类化合物 Calophyllic acid 与 Isocalophyllic acid 的混合物具有很好的抗氧化活性。2015 年, Aminudin 等^[56]研究发现 *C. symingtonianum* 和 *C. depressinervosu* 甲醇提取物均具有清除 DPPH 自由基能力, *C. symingtonianum* 心材提取物的 IC_{50} 为 $(5.17 \pm 0.04) \mu\text{g/mL}$, *C. symingtonianum* 叶提取物的 IC_{50} 为 $(15.70 \pm 1.43) \mu\text{g/mL}$, *C. depressinervosu* 树皮提取物的 IC_{50} 为 $(5.17 \pm 0.04) \mu\text{g/mL}$ 。2019 年, Rachmani 等^[33]测试了 *C. soulattri* 叶乙醇提取物的总黄酮含量和清除 DPPH 自由基能力, 研究显示总黄酮含量为 $(25.677 \pm 0.046) \%$, IC_{50} 为 $(39.63 \pm 14.99) \text{ppm}$ 。

3 小结

综上所述, 随着分离、纯化和鉴定技术的发展与应用, 国外学者不仅从红厚壳属植物中不断分离出新的香豆素、双苯吡酮类、黄酮等化学成分, 而且对其药理活性进行了大量研究, 对开发天然安全有效的抗肿瘤、抗 HIV-1、抗炎、抗菌和抗氧化药物具有重要意义。在前人的基础上对该属植物的化学成分及其药理活性进行深入研究, 对于合理开发利用红厚壳属植物资源具有重要意义。

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