Xanthones from Mangosteen (*Garcinia mangostana*): Multi-targeting Pharmacological Properties

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Objective: *This review focuses on mangosteen pericarp extracts, xanthones and derivatives for the future laboratory experiment and development in pharmacological aspects.*

Material and Method: *All relevant literature databases were searched up to 2 March 2014. The search terms included mangosteen, xanthone, mangostin, and gatanin in all of the human, animal, in vitro and in vivo studies. Anti-inflammation, antioxidant, antibacterial, anticancer and antiulcer properties of each substance were the key parameters.*

Results: *Xanthones are a group of oxygen-containing heterocyclic compounds including alpha-mangostin, gamma-mangostin, mangosteen extract, xanthone derivatives and synthetic xanthones, which provide remarkable and diverse pharmacological effects such as anticancer, antioxidant, anti-inflammatory and antimicrobial activities.*

Conclusion: *These xanthone compounds may play a major role in therapeutic treatment of the diseases but precise mechanisms of action are still unclear and need further investigation.*

Keywords: *Mangosteen, Xanthones, Mangostin, Anti-inflammation, Antioxidant, Antibacterial, Anti-cancer*

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Because of its delicious taste, mangosteen is a widely popular fruit within both Thailand and the international market. Traditionally, mangosteen has been long known as the "Queen of Fruits". It contains high nutritional values. In essence, it is a rich source of vitamins, minerals and fibers such as vitamin C, E, folate, calcium, potassium and magnesium[1]. In Thailand, most mangosteen trees are commonly grown in the Southern (70%) and Eastern (30%) regions. While mangosteen trees in the East will bear fruit and are ready for harvest from April to June, they do so from June to August in the South. As a result, this offers a total of fruit production available to the market for at least five consecutive months, with a total yield of about 298,000 tons[2].

Mangosteen (common name) or *Garcinia mangostana* Linn. (Botanical name) is in the family of Guttifarae. Its botanical characteristics are: a medium-sized fruit tree with 10-25 meters height and 25-35 cm in diameter with a single large oval leaf about 4.5-10 cm wide and 12-23 cm long. The fruit is approximately 3.5-7 cm in diameter with about 1 cm thick dark purplish brown shell and 4-8 white petals inside. The mangosteen plant is propagated by seeds[3].

Mangosteen is a source of mangostin, tannin, chrysanthemin, garcinone, gartanin, vitamin B1, B2 and vitamin C. It also contains bioactive substances called xanthones such as alpha and beta mangostin[4] for the use of indigenous medicinal plants of Southeast Asia. All parts of the mangosteen including fruit, root and bark can be used in the treatment of common cold, cystitis, diarrhea, dysentery, rash and gastrointestinal tract diseases. Additionally, it has been reported that mangosteen contains phytochemical compounds such as oxygenate and a variety of prenylated xanthones more than other plants[5].

Not only are mangosteens generally consumed as fruit, they contain a substance called xanthone, which is a flavonoid and polyphenon compound. It was found that xanthone extracted from the mangosteen fruit's pulp and seeds, called alpha-mangostin, beta-mangostin and garcinone B, can inhibit Mycobacterium tuberculosis[5]. According to the report of Thanaiwat (2009)[6], Mongkol (2009)[7] and Sakagami (2005)[8], xanthone has antioxidant, anticancer and antibacterial properties, respectively. A recent study by Sukma et al (2011)[9] also claimed that anti-inflammatory effects of mangosteen extract, called gamma-mangostin, can reduce inflammation in NG 108-15 cells. In addition, Bumrungpert A et al (2010)[10] found

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that alpha and gamma mangostin decrease the expression of genes controlling inflammation, TNF-alpha, IL-6 and CXCL10, caused by lipopolysaccharide in macrophage and adiposite cells with unclear mechanisms of action.

Xanthone extraction

Xanthone can be extracted from many plants, especially mangosteen. Briefly, the mangosteen peel is ground and extracted with solvent, based on the polar and non-polar properties of the substances. Therefore, different xanthones are extracted using different solvents. Nilar and Harrison (2002)[12] have extracted xanthone-garciniafuran from *Garcinia mangostana* using hot hexane as a solvent. Sakagami et al (2005)[8] extracted alpha and beta mangostin from *Garcinia mangostana* using methylene chloride and methanol, respectively. Suksamrarn et al (2002)[5] studied the isolation and identification of many types of xanthones using methanol extraction at the first step. The solvent was then extracted using liquid-to-liquid extraction technique. The water containing substances was then run through silica gel column chromatography. Three new xanthones (mangostenone A, mangostenone B, trapezifolixanthone) were found together with tovophyllin-B, α-mangostin, β-mangostin, garcinone B, mangostinone, mangostanol and flavonoid epicatechin. Xanthone extraction can be performed using silica gel chromatography and identified using Thin Layer Chromatography (TLC) or High Performance Liquid Chromatography (HPLC) and analyzed by UV, IR, MS and NMR[13,14].

Pilapa (2007)[15] investigated the stability of alpha mangostin, dried mangosteen extract (a = 0.41-0.46) and dried mangosteen extract solution (0.1%w/v) in ethanol. The samples then were stored in closed containers at 30, 50, 60 and 80 degrees Celsius for a period of 0-43 days and then analyzed for the amount of alpha mangostin by HPLA-PDA and analyzed for anti-oxidation capacity by DPPH and ORAC. Kinetic properties, decay and decrease in the antioxidant ability were calculated using the Arrhenius equation and the shelf-life was then predicted.

Degradation of alpha mangostin in dried powder extract from Southern region mangosteen demonstrated Arrhenius activated energy 42.29 kJ/M, which is higher than dried powder extract from Eastern region mangosteen (22.91 kJ/M). For the extracts solution, Southern region mangosteen has Arrhenius activated energy 78.42 kJ/M, which is lower than the extracts of plants from the East (97.05 kJ/M). Estimated half-life of alpha mangostin in dried powder extract is 660 days and in solution is 1,386 (South) and 6,930 days (East) using Arrhenium relation at 30°C.

Biological properties of xanthones

Anti-inflammation

Sukma et al (2011) recently discovered an anti-inflammatory effects of extracts from the mangosteen peel, gamma mangostin, in NG108-15 cells. The present study reported that increasing in the expression of serotonin 2, histamine H1 and the bradykinin 2 receptor gene may play a critical role in the treatment of inflammation, pain and neuropsychiatric symptoms[9]. Moreover, Bumrungpert et al (2010) reported that alpha and gamma mangostin extracted from mangosteen inhibited lipopolysaccharide induced inflammation of the human macrophage and adipocyte[10]. To an extent, the two xanthones also decreased TNF-alpha, IL-6 gene expression, but the precise mechanism remains unclear. Librowski et al (2006) reported the anti-inflammatory and analgesic ability of xanthone derivatives obtained in mice. The side effects did not differ compared with non-steroidal anti-inflammatory drugs (NSAIDs)[16].

Chomnawang et al (2007) studied the effect of the *Garcinia mangostana* in suppressing inflammation caused by acne. The results revealed that extracts of mangosteen inhibited free radicals and pro-inflammatory cytokines and decreased TNF-alpha production determined by ELISA technique[17].

Liu et al (2012) studied the anti-inflammatory action of alpha-mangostin on gene expression in a human myeloid leukemic cell line, U937. They found that alpha-mangostin inhibits lipopolysaccharide induced tumor necrosis factor (TNF)-alpha and interleukin (IL)-4 protein release and gene expression via MAPK, STAT1, c-Fos, c-Jun and EIK-1[18].

Bumrungpert et al (2010) reported that alpha and gamma mangostin extracted from mangosteen reduces lipopolysaccharide-mediated inflammation in MPhi (differentiated U937 cells) and insulin resistance in adipocytes, possibly by preventing the activation of MAPK, NF-kappa B, and AP-1, which are central to inflammatory cytokine production in white adipose tissue[10].

Antioxidant capacity

Hay et al (2004)[19] studied xanthones isolated from the bark of *Garcinia vieillardii* by grinding into a fine powder. Solvent is then extracted with cyclo-hexane, chloroform and ethyl acetate. Then the extract is concentrated by evaporation under reduced pressure

Table 1. Biological properties of xanthones

Xanthones	Biological properties	Reported by
Alpha-mangostin	Anti-inflammation	Bumrungpert et al (2010)
	Antibacterial	Sakagami et al (2005)
	Anticancer	Matsumoto et al (2003) Kim et al (2012) Johnson et al (2012) Aisha et al (2012)
	Antiulcer	Sidahmed et al (2013)
Gamma-mangostin	Anti-inflammation	Sukma et al (2011) Bumrungpert et al (2010)
	Anticancer	Balunas, M.J. et al (2008)
Mangosteen extract	Anti-inflammation	Chomnawang MT et al (2007)
	Anticancer	Aisha et al (2012)
Xanthone derivatives	Anti-inflammation	Librowski T et al (2005)
	Anticancer	Cheng G et al (2010) Teh et al (2013) Wang et al (2014)
	Alzheimer	Qin et al (2013)
Vieillardixanthone	Antioxidant	Hay et al (2004)
Issocudraniaxanthone A	Antioxidant	Hay et al (2004)
Mangiferin	Antioxidant	Pal et al (2013)

and then xanthones are isolated in the extract by HPLC using normal-phase column to isolate xanthones. The isolated xanthones, forbexanthone, isocudraniaxanthone A and 5,7-dihydroxychromone, 6-o-methyl-2- deprenylrheediaxanthone B and vieillardixanthone were tested for antioxidant activity by DPPH method. The two xanthones, vieillardixanthone and issocudraniaxanthone A were found to have the antioxidant capabilities equal to alpha-tocopherols and 2, 6-di-tert-butyl-4-hydroxyanisol (BHA).

Pal et al (2013) investigated the molecular mechanisms of the protective action of mangiferin, a natural xanthone, against lead-induced murine hepatic injury. It has been revealed that mangiferin exhibits both anti-oxidative and anti-apoptotic properties, hence, protects the organ in lead induced hepatic dysfunction[20].

Tangpong et al (2011) reported that xanthone derivative from *Garcinia mangostana* can prevent doxorubicin, a pro-oxidant, from causing mononuclear cells to increase the level of tumor necrosis factor-alpha (TNF-alpha). Pretreatment with xanthone suppressed doxorubicin-induced apoptotic indicators including protein carbonyl, nitrotyrosine, 4-hydroxy-2'-nonenal, p53, caspase-3 activity suggesting a novel anticancer therapy of xanthone[21].

Antibacterial properties

Sakagami et al (2005) reported that alpha-mangostin, isolated from the stem bark of *Garcinia mangostana* L., alone or in combination with gentamicin or vancomycin were active against vancomycin resistant Enterococci (VRE) and methicillin resistant Staphylococcus aureus (MRSA) respectively[8].

Anticancer properties

Matsumoto et al (2003) reported that xanthones from pericarps of *Garcinia mangostana*, alpha-mangostin, completely inhibits induction of apoptosis, and thus inhibits human leukemia HL60 cell growth[22].

Kurose et al (2012) studied the effect of alpha-mangostin extracted from mangosteen pericarp on apoptosis and cell cycle using the human breast cancer cell line MDA-MB231. The apoptosis is regulated by the PI3K/Akt signaling pathway and may be useful as a therapeutic agent for breast cancer with a p53 mutation and HER2/hormone-negative subtypes[23].

Johnson et al (2012) found that alpha-mangostin considerably decreases prostatic cancer cell viability in a dose-dependent manner with cyclins/cyclin-dependent kinases inhibition. In an in vivo mice model implanted with 22Rv1 cells, they demonstrated the ability of alpha-mangostin in tumor growth suppression compared with those with vehicle control[24].

Kim et al (2012) reported that alpha-mangostin induced autophagy activation in mouse intestinal epithelial cells. Conversely, co-administration of thapsigargin, which induced ER stress with alpha-mangostin, completely blocked the antitumor activity of alpha-mangostin itself. Therefore, alpha-mangostin

can be better used as an antitumor agent by autophagy activation rather than ER stress induction[25].

Aisha et al (2012) studied xanthones extracts (81% alpha-mangostin and 16% gamma-mangostin) prepared by crystallization of a toluene extract of *Garcinia mangostana* and analyzed by LC-MS. Anticolon cancer effect, cytotoxicity, apoptosis and antitumor properties were investigated on HCT 116 human colorectal carcinoma cells. The in vivo anticolon cancer activity was also investigated on subcutaneous tumors established in nude mice. They found that the extract has potent cytotoxicity due to induction of the mitochondrial pathway of apoptosis. In addition, tumor metastasis including cell migration, cell invasion and clonogenicity were also inhibited by the extract. It also significantly inhibits subcutaneous tumor HCT 116 colorectal carcinoma cell growth in nude mice suggesting potential anticolon cancer properties[26].

Balunaset et al (2008) studied the inhibitory effect of 12 pure xanthones isolated from *G. mangostana* on microsomal aromatase in breast cancer cells and reported that garcinone D, garcinone E, alpha-mangostin and gamma-mangostin exhibited dose-dependent inhibition. The most potent aromatase inhibitor was gamma-mangostin, but further investigation in vitro and in vivo studies is required[27].

Moreover, cluvenone, a synthetic xanthone analog was demonstrated to induce cell death via apoptosis and has similar cytotoxicity in multidrug-resistant and sensitive leukemia cells[28]. In addition, Guizzuntiet et al (2012) established that the direct target of cluvenone were in mitochondria and caused disruption of mitochondrial morphology in HeLa cell treatment[29].

Anti-ulcer properties

Sidahmedet et al (2013) studied the gastroprotective mechanism of alpha-mangostin in a rat model and found that it inhibited ethanol-induced gastric lesion and anti-Helicobacter pylori with antioxidant properties[30].

Xanthone derivatives

Librowski et al (2005) tested some synthesized xanthone derivatives for anti-inflammatory and analgesic properties in rats and found that compound MH-44, MH-41, MH-43 and MH-48 were observed to reduce edema and pain[16].

Qin et al (2013) studied a series of novel synthesized 1, 3-dihydroxyxanthone Mannich base derivatives on anticholinesterase activity. The result showed that most of the target compounds exhibited moderate to good inhibitory activities with the IC50 values at micromole level concentration against both acetylcholinesterase (AChE) and butyrylcholinesterase (BuChE), which are novel candidates for the treatment of Alzheimer's disease[31].

Cheng et al (2010) reported that 5, 6-dimethylxanthenone-4-acetic acid (DMXAA), a cell permeable xanthone derivative, has antitumor activity by activating cytosolic nucleotide-binding oligomerization domains (NOD) 1 and 2. In addition, DMXAA-induced chemokine CXCL10 mRNA and protein production in AB12 mesothelioma cell lines was observed, which can be blocked by RICK kinase inhibitor. These may play an essential role in biological properties[32].

Wang et al (2014) recently reported an anticancer activity of xanthone derivative, isoeuxanthone, in human cervical cancer cell (HeLa) and hepatocellular carcinoma cell line (HepG2). They found that oxiranylmethoxy substituted isoeuxanthone demonstrated the most effective cytotoxic activity against the two cancer cells than the other substituted xanthones and conceivably as a result of DNA damage[33].

Teh et al (2013) studied the cytotoxicity of xanthone dervivativeson nine cancer cell lines, Raji, SNU-1, K562, LD-174T, SK-MEL-28, IMR-2, HeLa, Hep G2 and NCl-H23. The result demonstrated that the diprenyl, dipyrano and prenylatedpyrano substituent groups of xanthone derivatives especially Mesuaferrin A, macluraxanthone and alpha-mangostin contributed in cytotoxicity of these cell lines[34].

Conclusion

Recently, mangosteens have become very popular as a dietary supplement. They have been under research study because they are a rich source of diverse and extraordinary natural compounds of xanthones with multi-targeting pharmacological properties such as anticancer, antioxidant, anti-inflammatory, antiulcer and antimicrobial activities. Some xanthones were synthesized and modified by several techniques to create novel compounds that may improve pharmacological effects and efficacy. However, synergism and interaction between xanthones and derivatives with other substances has not been extensively studied. Further investigations should be conducted to determine the precise mechanisms of action in all aspects, which can be developed into new medications.

Potential conflicts of interest

None.

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แซนโทนจากมังคุด: ฤทธิ์ทางเภสัชวิทยาที่หลากหลาย

ศราวุธ จินดารัตน์

วัตถุประสงค์: *การทบทวนวรรณกรรมนี้มุ่งเน้นที่สารสกัดจากเปลือกมังคุดแซนโทนและอนุพันธ์ สำหรับการทดลองในห้องปฏิบัติการในอนาคตและการพัฒนาในด้านเภสัชวิทยา*

วัสดุและวิธีการ: *ฐานข้อมูลวรรณกรรมทั้งหมดที่เกี่ยวข้องถูกค้นถึงวันที่ 11 กรกฎาคม พ.ศ. 2556 โดยคำที่ใช้ค้นหา ได้แก่ mangosteen, xanthone, mangostin และgatanin ทั้งในมนุษย์ สัตว์ และในหลอดทดลอง ต่อฤทธิ์การยับยั้ง และการอักเสบ สารต้านอนุมูลอิสระ ต้านเชื้อแบคทีเรีย ต้านมะเร็งและการรักษาแผล*

ผลการศึกษา: *แซนโทนเป็นกลุ่มสารประกอบ heterocyclic ได้แก่ แอลฟาแมงโกสทินแกมมาแมงโกสทิน สารสกัดเปลือมังคุด อนุพันธ์ของแซนโทนและสารสังเคราะห์ที่ให้ผลทางเภสัชวิทยาที่โดดเด่นและมีความหลากหลาย เช่น การป้องกันมะเร็งสารต้านอนุมูลอิสระ ต้านการอักเสบ และยาปฏิชีวนะ*

สรุป: *สารแซนโทนที่ได้มีการศึกษานี้น่าจะมีบทบาทสำคัญในการบำบัดรักษาโรคได้ แต่กลไกที่แน่นอนก็ยังไม่ชัดเจนและต้องทำการศึกษาต่อไป*