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### The Indonesian *Zanthoxylum acanthopodium* DC. : Chemical And Biological Values

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**Abstract:** The Indonesian *Zanthoxylum acanthopodium* DC. is one of main species of this genus that grows in Sumatera. It is known as andaliman in Batak society. In Indonesia, andaliman is widely grown in the plateau of North Sumatera. This spice is used for seasoning Batak's traditional cuisine. The fruit of *Z. acanthopodium* DC. is round and small, green when young, red to dark color, and black when it is dried. Andaliman fruits demonstrate unique sensory properties which are aromatic compounds with a bitter taste and distinctive spicy. *Zanthoxylum acanthopodium* DC. has not been used as a medicinal plant. Traditionally, *Zanthoxylum* fruits are used for treating digestive, asthma and bronchitis, relieve pain, heart disease, mouth diseases, teeth and throat, as well for treating diarrhea. Andaliman fruits extracts had been reported to have antimicrobial activity, antioxidant, antiinflammatory, xanthine oxidase inhibitor, and possessed cytotoxic activity. Metabolites isolated from Indonesian *Zanthoxylum acanthopodium* DC. so far were substituted amide, volatile components, and polar terpenoids. Some of the metabolites had shown antioxidant and xanthine oxidase inhibitor properties.

**Keywords :** andaliman fruits, biological activity, chemical compounds, *Zanthoxylum acanthopodium* DC.

#### Introduction

The history records Indonesia as one of the country's largest producer of spices in the world which has led European nations to come to the archipelago. Spices had been considered very valuable because the properties are not only as a flavoring or spice in cooking, but also as traditional medicine and had effect for keeping the body warm. Indonesia are wealthy in spices, the popular ones are ginger, cloves, nutmeg, cinnamon, and pepper. The exoticism spices of this country have attracted the attention of food industries and scientific researches to be further explored because of their activity as natural antioxidants.

One type of Mother Earth's spices that has not been widely utilized (limited only to primary commodities) is andaliman. This one is a popular spice used in various types of Batak's traditional cuisines such as Sangsang, Na Niura, Na tinombur, and Arsik. The spicy taste is bitter than chili. There is a specific taste when pounded together with chili, causing a distinctive aroma of essential oils, producing a thrilling sensation that stimulates the production of saliva and produces a right sense in the tongue of the Batak society, and also raising the appetite. These things has led the foods to be bland without the presence of andaliman fruits.

Andaliman was found growing wild in Tapanuli, North Sumatera at 1500 meters above sea level. The fruits shape are like pepper, small round, green, but becomes black when they are dry. Andaliman's names is

varieted by region. It is called andaliman in Toba, tuba in the area of Simalungun and Dairi, and Siyarnyar in South Tapanuli.

As a spice, andaliman fruits have preservative power that makes Batak's cuisines last longer. This property was thought to be related to the presence of antimicrobial and antioxidant compounds. The antioxidant activity in food can be used to protect the fats / oils against oxidative damage. Andaliman fruits also contains vitamin C and E, which are useful to stimulate body endurance.



Figure 1. Andaliman at the age 2 years



Figure 2. Andaliman at the age 4 years

#### Overview of Andaliman (*Zanthoxylum acanthopodium* DC.)

The plant of Andaliman was described by<sup>1</sup> as follows: shrub or small tree low-branched, upright to 5 m, yearly plant. Trunk, branches, and twigs are thorny. Scattered leaves, stemmed, pinnate compound leaf litter odd, 5-20 cm long and 3-15 cm wide, there are oil glands. Compound interest is limited, children umbrella menggarpu compound, small; flat rate basis or conical shape; 5-7 petals free, 1-2 cm long, pale yellow colour; androgynous, stamens 5-6 sitting on the basis of flowers, reddish anthers, stigmas 3-4, the embryo of fruit is apocarpous and symbiotic to others. The fruit is round, 2-3 mm diameter, green light, dark red; one seed per fruit, hard skin, shiny black color.

Andaliman's stem has a lot of branches and many small pieces leaves. The whole trunk and branches, from the bottom to the tip, were fulfilled with sharp thorns like a rose thorn. However, thorns of andaliman are large and sturdy. Andaliman fruits appear / grow among thorns.

Andaliman plant systematics are as follows<sup>2</sup>:

World	: <i>Plantae</i>
Division	: <i>Spermatophyta</i>
SubDivision	: <i>Angiospermae</i>
Class	: <i>Dicotyledoneae</i>
Sub-class	: <i>Rosidae</i>
Ordo	: <i>Sapindales</i>
Family	: <i>Rutaceae</i>
Genus	: <i>Zanthoxylum</i>
Species	: <i>Zanthoxylum acanthopodium</i> DC.



Figure 3. Andaliman Branches



Figure 4. Andaliman Flower



Figure 5. Andaliman Fruits

*Zanthoxylum acanthopodium* DC is known in Indonesia as andaliman. This plant has the native name in the region of North Sumatera which is intir-intir, and foreign names which are Sichuan pepper or Indonesian lemon pepper<sup>3</sup>. In Japan there are spices sansho (*Zanthoxylum piperitum* DC) that have similar shape and fruit aromas with andaliman fruits. Moreover, in some countries, there are spices known as *Z. simulans*, and *Z. bungeanum* Maxim that have scents and volatile compounds which are also similar to andaliman<sup>4</sup>.

In Indonesia, the typical andaliman plants were found in the region of North Sumatra. In the world, these plants are scattered among others in northern India, Nepal, East Pakistan, Myanmar, Thailand, and China<sup>5,6</sup>. Andaliman grows wild in the area with an altitude of 1500 meters above sea level<sup>1</sup> at a temperature of 15-18° C<sup>5,6</sup>. Andaliman fruits are commonly used as a spice in Batak's traditional cuisine. The fruits contain aromatic compounds with a bitter taste and distinctive spiciness. The fruits produce the effect of thrilling sensation on taste buds and cause the tongue felt numb<sup>7</sup>.

Andaliman is wild plants and difficult to cultivate, grown on the farm or land of a new openings in the wilderness. Andaliman is not planted, such as chili, pepper, and other vegetables. Usually andaliman grow simply<sup>4</sup> but the germination is low. Plants that grow naturally derived from the seeds that are dispersed by birds (after eating andaliman fruits). Farmers also obtain seeds inadvertently from the burning weeds location in the old plants area<sup>1</sup>.

Species of *Zanthoxylum* generally have a spicy flavor and bitterness which are more stinging when the fruit was perfectly ripe. The specificity of the andaliman plant is its ability to give a unique sensory properties that is trigeminal biting sensation on the tongue, in addition to a pleasant aroma<sup>4</sup>.

### The genus of *Zanthoxylum*

Family of Rutaceae has 150 genera and 1500 species which are distributed in temperate and tropical regions. This plant group is widely common in South Africa and Australia. Some of the main genus are *Citrus*, *Zanthoxylum*, *Ruta*, *Murraya*, and *Fortunella*<sup>2</sup>.

*Zanthoxylum* genus comprises about 250 species, in the form of trees and shrubs, including the family Rutaceae that grows in warm temperate and subtropical regions around the world. The fruits of several species are used to make the spice Sichuan pepper. Historically, the bark is used widely for dental pain, colic, and rheumatism<sup>8</sup>.

*Zanthoxylum* is a genus that is rarely studied. Some of the main species of this genus are *Zanthoxylum piperitum* DC (*Z. sansho*) in China, Japan, Korea, *Z. simulans* (*Z. bungei*) in China and Taiwan, *Z. bungeanum* Max. in China, *Z. schinifolium* Sieb. et Zucc. in China and Korea, *Z. nitidum* Roxb (DC) in China and Southeast Asia, *Z. limonella* in India and Southeast Asia, *Z. armatum* DC (*Z. alatum* Roxb.) in the Himalayas, East Asia, and Southeast Asia, *Z. avicennae* (Lamk) DC (*Z. tidorensis*) in China, Southeast Asia, Indonesia and *Z. acanthopodium* DC. in the eastern Himalayas, China, Southeast Asia, and Sumatera<sup>3</sup>.

Parts of the plant that is often used is the fruits. In Korea, aromatic seeds of *Z. schinifolium* species are preferred for use, although the pericarp can also be used. Fresh young leaves of Sichuan pepper tree are usually used in Japan as a spice and for decoration<sup>3</sup>.

Dried fruits of genus *Zanthoxylum* have aromatic scents like lemon. Some species have diverged taste, for example, *Z. alatum* (spicy), *Z. schinifolium* and *Z. avicennae* which both have the scents of *anise*. Almost all the species have a very pungent and biting that ultimately generate numbness in the tongue, except *Z. schinifolium* because it has a little sting quality<sup>3</sup>.

### The Chemical Values of *Zanthoxylum acanthopodium* DC.

Most of the volatile components of the genus *Zanthoxylum* is terpenoids. One of the most widely component in the essential oil of Indonesian *Z. acanthopodium* is geranyl acetate (35%), while the other components are limonene, citronellal, and linalol.

Some of the chemical constituents of the andaliman fruits had published, among others, by <sup>4</sup> who isolated the substituted amide with the solvent ethyl acetate-ethanol. The results of spectrophotometric analysis showed that the nitrogenous component have a molecular weight of 247 with molecular formula C<sub>16</sub>H<sub>25</sub>ON. The component was an aliphatic carbon chain which are either in the form of methyl, methylene, and metin, branched and had some double bonds. Oxygen bound in the form of carbonate. Interpretation of the data indicated that the component was 2E, 6Z, 8E, 10E-N-(2'-metilpropil)-dodekatetraenamida (Sanshool) with the configuration as shown in figure 6. The compounds from *n*-butanol extract of andaliman fruits which collected from North Sumatera, Indonesia had been isolated and elucidated<sup>11</sup>. Based on spectrum data, they were polar terpenoids, 17-((23E,26E)-23- ethylidene - 29 - hydroxy-29-(2-hydroxypropoxy)-25-methyldec-26-en-20-yl)-13,15-dimethyl-gona-1,3,5(10)-trien-3-ol and cholest-5-en-3β-yl β-D-glucopyranoside (Figure 8). New flavone glycoside compounds were isolated from the methanol extract of *Z. acanthopodium* DC. fruits that grows in India<sup>10</sup>.

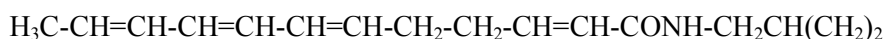


Figure 6. (Source : <sup>9</sup>)

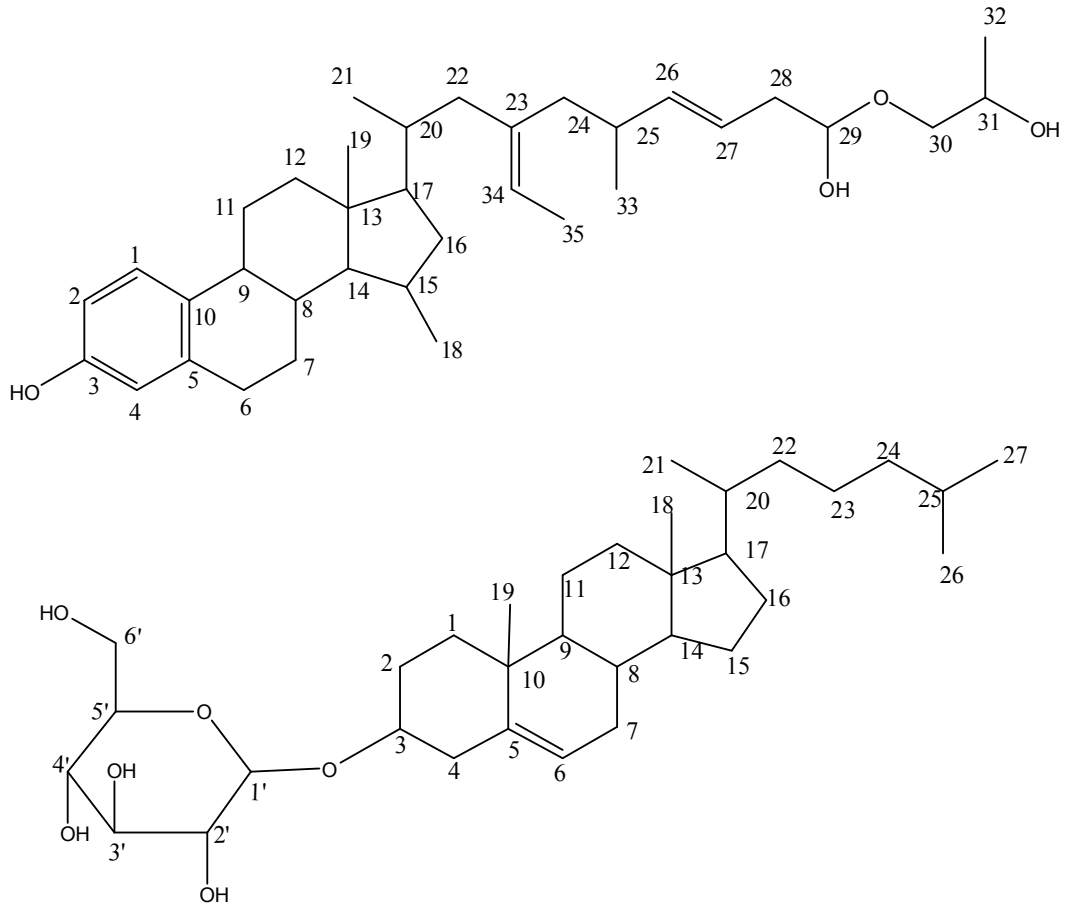


Figure 7. (source: 11)



Figure 8. Andaliman Plant Ready to Harvest

## The Biological Values of Genus *Zanthoxylum* and Andaliman Plant

Traditionally, *Zanthoxylum* fruits are used for treating digestive, asthma and bronchitis, relieve pain, heart disease, mouth diseases, teeth and throat, as well for treating diarrhea. Root bark and leaves are used to cure stomachache, toothache, cough, and venereal diseases, rheumatism and lumbago. *Zanthoxylum* have some biological activity such as larvicides, anti-inflammatory, analgesic, antioxidant, antibiotic, hepatoprotective, antiparasitidal, cytotoxic, antiproliferative, anthelmintic, antiviral, and antifungal anticonvulsants<sup>12</sup>.

Monofilidin compounds that can be isolated from *Z. monophyllum* showed antimicrobial activity<sup>13</sup>. Isolation of compounds from *Zanthoxylum usambarensis* dichloromethane extract of roots and bark produce two alkaloids compounds that physiologically active, namely canthin-6-one as a fungicide and pelitorin as insecticide<sup>14</sup>. Alkaloid compounds derived from dichloromethane extract of the roots and bark of *Zanthoxylum chiloperone* var. *angustifolium* showed antifungal activity against *Candida albicans*, *Aspergillus fumigatus* and *Trichophyton mentagrophytes*<sup>15</sup>. Seven benzofenantridin alkaloids derived from the bark extract of *Zanthoxylum rhoifolium* showed antimalarial activity against *Plasmodium falciparum*<sup>16</sup>. Essential oils of *Z. armatum* exhibited a strong inhibition of the growth of fungal mycelium. Antitumor activity of leaf essential oil of *Z. rhoifolium* been studied in vitro and in vivo. Toothpaste containing *Z. nitidum* extracts decrease the incidence of dental plaque and improve dental health. Benzofenantridin alkaloid isolated from the stem bark of *Z. tetraspermum* showed antibacterial activity. Ethanol extract of *Z. armatum* leaves had hepatoprotective activity and had been evaluated against rat<sup>12</sup>.

Terpenilcoumarin compounds which had been isolated from the stem of *Z. shinifolium*, aurapten and kolinin of *Z. schinifolium* skin had inhibitory activity against the monoamine oxidase enzyme (MAO) associated with Parkinson's disease. Furanocoumarin as psoralen compounds isolated from the fruits of *Z. americanum* had antiplatelet activity and inhibitory activity of DNA replication in hepatitis B virus<sup>17</sup>.

3,5-diasetylumbulin flavonoids that has been isolated from *Z. integrifolium* fruits have antiplatelet activity. Aliphatic amide  $\alpha$ -sanshool, isolated from *Z. liebmannianum*, was known as an anthelmintic. A typical example of aromatic amides isolated from *Z. syncarpum*, sincarpamide, active as antiparasitidal<sup>17</sup>.

Analysis of genetic diversity of *Zanthoxylum acanthopodium* DC. , which had been collected from India, by using markers Amplified Fragment Length Polymorphism (AFLP) had been carried out<sup>18</sup>. Genomic DNA study of eight *Z. acanthopodium* collections, after amplified, obtained six primer pairs selectively used to detect polymorphism and found 23 markers of specific species of *Z. acanthopodium*. The markers can be used to resolve problems faced by the pharmaceutical industry to obtain raw materials for drugs, phytofarmaka, or foodstuffs, beverages and others. The relatively low level of similarity of *Z. acanthopodium* showed *Z. acanthopodium* had a high genetic diversity.

Andaliman had been reported to have antimicrobial activity. Based on the experiments results, the extract of andaliman fruits can inhibit the growth of microbes that are pathogenic and destroy foodstuffs such as *Escherichia coli*, *Salmonella typhimurium*, *Bacillus cereus*, *Staphylococcus aureus*, and *Pseudomonas fluorescens*, and *Aspergillus flavus*<sup>19</sup>. Essential oil from Andaliman leaves inhibited the growth of *Colletotrichum gloeosporioides* and *Botryodiplodia theobromae*<sup>20</sup>. Other research is to test the effect of andaliman fruits extract on fertility and the development of the mouse embryo showed andaliman fruits extracts were antifertil<sup>21</sup>. The activity of ethanol extract of andaliman fruits as antiinflammatory has been studied in vitro and proved the ability of ethanol extract of the andaliman fruits to inhibit tumor necrosis factor, interleukin-6, and cyclooxygenase which plays an important role in the inflammatory process<sup>22</sup>.

Several studies had proven that andaliman fruits extract had antioxidant activity. Antiradical activity of ethanol extract of andaliman fruits with a concentration of 200 ppm indicates inhibition of 61.81%<sup>23</sup>. The antioxidant activity of andaliman fruits extracts was also proved stable against various conditions of temperature and pH in some food systems<sup>24</sup>. N-butanol extract of andaliman fruits was tested to have antioxidant activity with IC<sub>50</sub> value of 53.51 mg / mL and xanthin oxidase enzyme inhibitory activity with IC<sub>50</sub> value of 3.69 mg / mL<sup>12</sup>. The compounds that have been characterized from andaliman fruits along with biological activity were the terpenoids polar compounds that have been studied to have antioxidant activity and xanthin oxidase inhibition<sup>12</sup>. Petroleum extract of andaliman fruits also possessed cytotoxic activity on T47D cell line with IC<sub>50</sub> 149,4  $\mu$ g/mL<sup>25</sup>.

## **Cultivation of Andaliman<sup>7</sup>**

As noted in the previous chapter that it is difficult to cultivate andaliman because these plants live naturally (wild) at the newly opened forest by burning woods. This plant propagation are not known economically and practically, whether vegetative or generative. Vegetative propagation is less efficient due to the lack of parent trees, while the generative is difficult to develop because the seeds germinate and difficult andaliman kecambahnya low power.

As time goes by, many farmers in Tapanuli, North Sumatra are working on it since the sale of andaliman very helpful to the family income even if only as a side income in addition to coffee and rice. Andaliman farming is still relatively simple and traditional, ranging from land management process, plants treatment up to harvesting is not done by using modern technology.

### **Plant seeding**

The farmers generally start to crop andaliman with a nurseries before planting in the field. Seeds were then dried and then soaked in water to determine wether the seeds are good or not. Andaliman good seeds are the seeds that were sinking, the ones that floated to the top will be discarded because they are not suitable for breeding.

The size of seedbed shady land is prepared according to the number of andaliman seeds. Andaliman seeds are sown, backfilled with soil but not too thick, dry weeds or grass land are sown in the nursery, then burned so that the skin more quickly broken. Nursery are allowed to grow andaliman for one month ago then moved into a polybag which already contains soil or compost for 2 months and moved into the prepared soil.

### **Preparation of Land**

Before planting, it is necessary to have the land management, which is first to do is clear the land for planting and the weeds so as not to interfere with the process of planting and land preparation. Prepared planting hole with a size of 1.5 x 1.5 meters and the distance of the hole is about 5-6 meters from other plants because the plants will grow taller each year and extending to the side.

### **Investment**

Planting process is the process of transplanting andaliman plants from polybags to land prepared. It needs cover crops such as coffee and incense as a place for andaliman intercropping. Fertilizer was added after the soil is processed.

### **Fertilization**

The process of fertilization is very important to speed up the process of plant growth. The applied fertilizers consist of compost, usually given two times a year in March and in September when the rainy season comes to speed up the process of growth and produce good fruits. Compost is given about 2 kg per tree, while chemical fertilizer is given if the farmer has enough capital that is as much as 1-2 ounces per tree. The dose of fertilizer will increase each year along with the development of the plants.

### **Maintenance**

Maintenance of the plant is done to prevent pests such as weeding. According to farmers, this plant is rarely attacked, although there is usually moldy (white patches) on plants and the maintenance is done manually.

### **Harvest**

Farmers do harvesting after the age of 3 years from the time of planting. Productive age is less than 7 years. Andaliman fruits that can be harvested is the green ones as small as pepper. Harvesting is done manually by picking by hand. It is required concentration and high caution in the process of picking because andaliman

plants have many thorns in every branches. Andaliman freshly picked fruits should be wrapped in banana leaves, because it will be broken if left open.

## Conclusion

A common feature of most Indonesian *Z. acanthopodium* species which had been studied is the wonderful capacity to produce volatile oils and terpenoids. These vary in composition and show different properties. The authors had discussed the chemical and biological values of the genus *Zanthoxylum* and the Andaliman species in medicine and their general potential in drug development.

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