การเปรียบเทียบการสกัดสารหอมจากดอกลั่นทม

Comparative study of scented compound extraction from *Plumeria obtusa* L.

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บทคัดย่อ

การศึกษาเปรียบเทียบวิธีการสกัดสารที่ให้กลิ่นหอมจากดอกลั่นทมด้วยวิธีการต่างๆ คือ วิธีการกลั่นสามวิธีได้แก่ การกลั่นด้วยน้ำ การกลั่นด้วยไอน้ำ และการกลั่นด้วยน้ำและไอน้ำ วิธีการสกัดด้วยตัวทำละลายอินทรีย์สองชนิดคือ เฮกเซนและปิโตรเลียมอีเทอร์ และวิธีการสกัดด้วยไขมันสองวิธีคือ การสกัดด้วยไขมันเย็น และไขมันร้อน และวิเคราะห์องค์ประกอบทางเคมีของสารหอมที่ได้จากแต่ละวิธีด้วยเครื่อง GC-MS จากการทดลองพบว่าเปอร์เซ็นต์ผลผลิตของสารหอมที่ได้จากการสกัดแต่ละวิธีคือ 0.0167, 0.0045, 0.0342, 0.4170, 0.3510, 0.3969 และ 12.2400 เปอร์เซ็นต์ ตามลำดับ เมื่อวิเคราะห์องค์ประกอบของสารหอมต่างๆ ที่สกัดได้จากดอกลั่นทม พบว่าสารประกอบหลักของน้ำมันหอมระเหยจากการกลั่นทั้งสามวิธี และแอบโซลูตจากการสกัดด้วยตัวทำละลายทั้งสองชนิด คือ benzyl salicylate ส่วนสารประกอบหลักของแอบโซลูตที่ได้จากการสกัดด้วยไขมันเย็นคือ linalool ในขณะที่การสกัดด้วยไขมันร้อนนั้นมีสารประกอบหลักคือ n-undecanoic acid

คำสำคัญ; ลั่นทม, การกลั่น, การสกัดด้วยตัวทำละลาย, การสกัดด้วยไขมัน, น้ำมันหอมระเหย, สารหอม, GC-MS, สารสำคัญ

ABSTRACT

Various of scented compound extraction methods from *Plumeria obtuse* L. flower; water distillation, steam distillation, water-steam distillation, hexane extraction, petroleum ether extraction, cold and hot enfleurage were studied. Chemical compounds of the extracts from each method were analyzed by GC-MS. The result showed percentage yields of the extracts were 0.0167, 0.0045, 0.0342, 0.4170, 0.3510, 0.3969 and 12.2400%, respectively. Main chemical component of essential oil from three distillation methods and absolutes of both solvent was benzyl salicylate, from cold enfleurage absolute was linalool and from hot enfleurage absolute was n-undecanoic acid.

Key word; *Plumeria obtuse* L., distillation, solvent extraction, enfleurage, essential oil, absolute, GC-MS, major compound

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INTRODUCTION

Plumeria obtusa L., the member of Apocynaceae Family, is commonly known as Frangipani or Temple tree. In Thailand, it is most famously called Lanthom. That is an evergreen small tree four to five metres tall. Leaves are simple, oval or obovate in shape and dark green. Apex of the leaf is rounded. Flowers are large, four to five centimeter across, showy, pleasantly fragrant and creamy white in colour with a yellow centre. Inflorescence is a cyme. Fruit is a double follicle with winged seeds. A milky sap is exuded from the branches when they are bruised or punctured. (Eggenberger and Eggenberger, 1994: Selvam, 2007: O' Sullivan, 2008)

Frangipani essential oil is used for several purpose such as an ingredient in cosmetics, excellent for aromatherapy uses, for example, scent candles, freshen potpourri, massage oils, and of course as a perfume to smell just truly great.(Woodspirits Natures Essentials., 2003; Aromatic Ltd. 2001-2008)

The fragrance of *Plumeria* flowers were previously studied on yield and chemical compounds from water distillation (Kamariah *et al*. 1999: Norsita *et al.* 2006) but there were not many reported about other extraction methods. Thus, objective of this experiment was to compare extraction methods and analyze chemical compound from floral scent.

MATERIALS AND METHODS

Extraction procedures

Distillation

Essential oil was extracted from fresh Frangipani flowers by three distillation methods: water, steam and water-steam (Ernest *et al* 1947: Oyen and Dung 1999). Three hundred grams of the flowers were used in each method and they were distilled for 4 hrs. After that, light yellow oil was collected on the water surface, and dehydrated with anhydrous Na_2SO_4. The oil was collected in an opaque bottle.

Solvent extraction

Fresh flowers, weight of 70 g, were soaked in 1 L of hexane or petroleum ether for 1 hr, then, the solvent was evaporated under reduced pressure at low temperature, and finally left the concrete yield. Afterward, the concretes were extracted by absolute ethanol, to yield the absolutes.(Ernest *et al* 1947)

Enfleurage

The cold enfleurage procedure was as follow; 200 ml of palm stearin was coated on a glass frame. After it was cooled and hardened, plant materials were placed and the scent components allowed to diffuse into the fatty layer for over a day. For hot enfleurage, fresh flowers were soaked in 400 ml of hot palm oil, heated at 60 - 70 ^{0}C for 30 min and placed to cool down, after that took in refrigerator for overnight. Both of processes were repeated with fresh plant materials. About 2 kg of fresh Frangipani flowers were used for cold enfleurage extraction and 100 g for hot enfleurage extraction. The flower and scent in the pomade from each method were extracted with cold absolute alcohol: The absolutes was obtained after the alcohol was evaporated. (Ernest *et al* 1947)

Chemical analysis

Identification of the chemical compounds in the extracts were performed by using GC-MS (QP 5050A Shimadzu) with a capillary columns DB-5 (5%-Phenyl)-methylpolysiloxane 0.25 μm, 60 m, 0.25 mm (i.d.), Type of detector was FID, Temperature programs were 80^0C for oventemperature, 200^0C for injector and 230^0C for detector. Injector volume was 1 μl, Carrier gas was Helium 99.999%,Total time was 40 min Solvent cutimes 5 min and Mass range 40 to 400 m/z.

RESULTS AND DISCUSSION

Distillation

Essential oil yields from water, steam and water-steam distillation were 0.0167, 0.0045 and 0.0342% respectively (Fig 1). Essential oils were light yellow with concentrated floral scent. The yield of water-steam distillation was much higher than other methods.

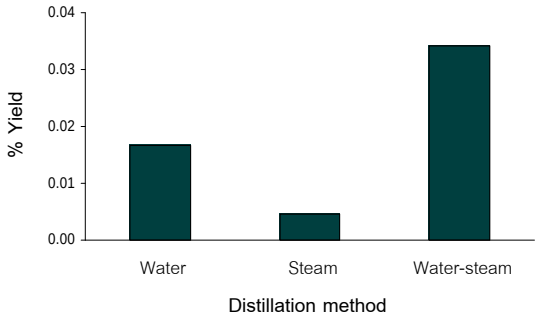

Fig 1 Comparison of yield of Frangipani oil from distillation

Chemical analysis of Frangipani oil by GC-MS revealed 19, 13 and 21 compounds in the oil getting from water, steam and water-steam distillation (Fig 2, 3, 4). Their major compounds were benzyl salicylate (31.32, 27.58 and 31.90%, respectively) (Table 1, 2, 3). Kamariah *et al.* (1999) also reported detection of benzyl salicylate (39%) as a main component in Franigipani oil getting from water distillation. Benzyl salicylate had a mild sweet odor whereas benzyl benzoate was almost odorless in *P. obtusa* (Norsita *et al.* 2006).

Solvent extraction

The yields of concrete and absolute from hexane extraction were 0.5377 and 0.4170%, and from petroleum ether extraction were 0.4351 and 0.3510%, respectively. Characteristics of concretes from both solvents were light yellow wax and had strong odor. Hexane absolute was light yellow liquid and mild odor of frangipani. Petroleum ether absolute was deep yellow, had strong odor and was more similar to fresh flower than that from hexane.

The chemical identification of hexane absolutes and petroleum ether absolutes by GC-MS showed that major compound of both absolutes were benzyl salicylate (44.69 and 42.63%, respectively). In hexane absolute, 17 compounds were identified while 13compounds were deteced in petroleum ether absolute. (Fig 5,6 and Table 4,5).

Enfleurage

Percentage yields of cold enfleurage absolute and hot enfleurage absolute were 0.3842 and 12.24, respectively. Both absolutes were soft fat and light yellow.

On GC-MS analysis, thirteen compounds were detected in cold enfleurage absolute and linalool was found to be main component (table 6, Fig 7). In hot enfleurage absolute, twelve compounds were detected but n-undecanoic acid was its main component.(Table7, Fig 8) Only there compounds; linalool, benzyl benzoate, and benzyl salicylate were found in both absolutes.

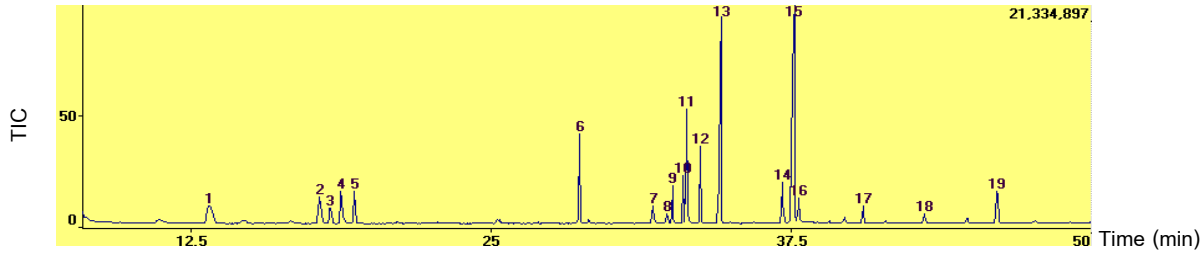

Fig 2 Chromatogram of essential oil from water distillation

Table 1 Chemical compounds of essential oil from water distillation

Peak No.	Retention time (min)	%Relative peak area	Possible compounds
1	13.280	4.72	linalool
2	17.877	4.36	(Z)-geraniol
3	18.314	2.02	(Z)-citral
4	18.762	3.79	(E)-geraniol
5	19.318	3.66	(E)-citral
6	28.702	4.85	(Z)-beta-farnesene
7	31.773	1.22	1-hexadecene
8	32.346	0.33	2-methylpentadecane
9	32.577	1.97	alpha-farnesene
10	33.022	2.61	(Z)-farnesol
11	33.192	6.70	(E)-farnesol
12	33.746	4.37	(E)-farnesal
13	34.601	18.90	benzyl benzoate
14	37.155	2.71	1-octadecanol
15	37.659	31.32	benzyl salicylate
16	37.865	1.65	eicosane
17	40.531	1.33	(E)-farnesyl acetate
18	43.072	0.61	(E)-farnesyl acetate
19	46.100	2.89	heneicosane

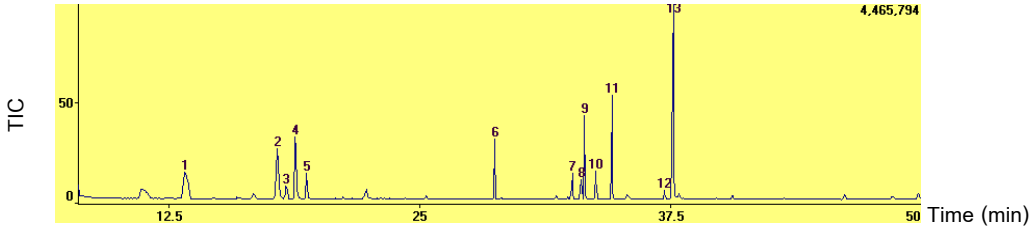

Fig 3 Chromatogram of essential oil from steam distillation

Table 2 Chemical compounds of essential oil from steam distillation

Peak No.	Retention time (min)	%Relative peak area	Possible compounds
1	13.327	11.18	linalool
2	17.923	13.70	(Z)-geraniol
3	18.363	2.54	(Z)-citral
4	18.813	12.15	(E)-geraniol
5	19.363	4.60	(E)-citral
6	28.753	4.68	(Z)-beta-farnesene
7	32.627	2.27	alpha-farnesene
8	33.072	1.41	(Z)-farnesol
9	33.235	6.95	(E)-farnesol
10	33.789	2.36	(E)-farnesal
11	34.609	9.89	benzyl benzoate
12	37.206	0.69	1-octadecanol
13	37.669	27.58	benzyl salicylate

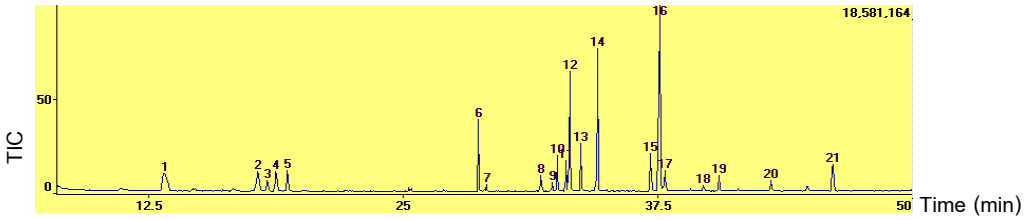

Fig 4 Chromatogram of essential oil from water-steam distillation

Table 3 Chemical compounds of essential oil from water-steam distillation

Peak No.	Retention time (min)	%Relative peak area	Possible compounds
1	13.316	6.49	linalool
2	17.886	4.33	(Z)-geraniol
3	18.327	1.66	(Z)-citral
4	18.777	3.12	(E)-geraniol
5	19.324	3.09	(E)-citral
6	28.718	5.03	(Z)-beta-farnesene
7	29.104	0.39	(E)-beta-farnesene
8	31.790	1.40	1-hexadecene
9	32.360	0.45	2-methylpentadecane
10	32.592	2.62	alpha-farnesene
11	33.035	2.14	(Z)-farnesol
12	33.210	9.29	(E)-farnesol
13	33.754	3.42	(E)-farnesal
14	34.592	13.28	benzyl benzoate
15	37.166	3.04	1-octadecanol
16	37.658	31.90	benzyl salicylate
17	37.880	1.95	eicosane
18	39.763	0.46	unknown[1]
19	40.549	1.69	unknown[2]
20	43.096	1.05	(E)-farnesyl acetate
21	46.121	3.20	heneicosane

Unknown[1] m/z 41(100) 51(36.9) 68 (33.2) 77(40.6) 93(71.8) 105(35.1) ,Unknown[2] m/z 41(100) 51(28.7) 69(24.6) 77(29.9) 93(51.3) 105(34.3)

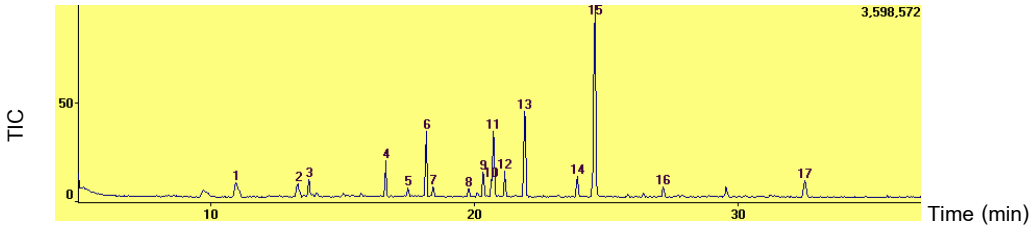

Fig 5 Chromatogram of hexane absolute

Table 4 Chemical compounds of hexane absolute

Peak No.	Retention time (min)	%Relative peak area	Possible compounds
1	10.981	3.66	linalool
2	13.345	3.93	(Z)-geraniol
3	13.751	3.10	(E)-geraniol
4	16.661	2.72	(E)-geranylacetnoe
5	17.510	0.69	2,4-di-t-butylphenol
6	18.199	5.53	(Z)-nerolidol
7	18.462	0.84	unknown[1]
8	19.807	0.88	1-hexadecene
9	20.363	2.56	alpha-farnesene
10	20.682	1.74	(Z)-farnesol
11	20.754	7.22	(E)-farnesol
12	21.177	2.84	(E)-farnesal
13	21.936	11.67	benzyl benzoate
14	23.927	2.80	1-octadecanol
15	24.610	44.69	benzyl salicylate
16	27.181	1.43	unknown[2]
17	32.554	3.69	isoeicosane

Unknown[1] m/z 41(100) 43(87) 55 (48) 56(25) 57(35) 68(24) 83(14) 97(15)

Unknown[2] m/z 41(100) 50(15.8) 51(23) 53(17) 68(21) 69(31) 77(33) 93(35) 105(32)

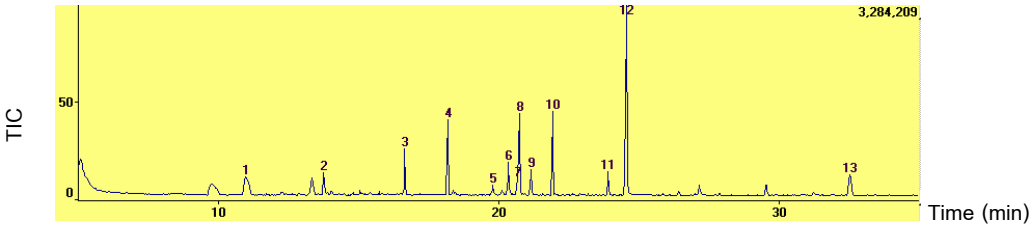

Fig 6 Chromatogram of petroleum absolute

Table 5 Chemical compounds of petroleum absolute

Peak No.	Retention time (min)	%Relative peak area	Possible compounds
1	10.993	6.04	linalool
2	13.753	4.44	(E)-geraniol
3	16.658	3.96	(E)-geranylacetnoe
4	18.196	6.53	(Z)-nerolidol
5	19.801	1.16	1-hexadecene
6	20.361	3.39	alpha-farnesene
7	20.678	1.54	(Z)-farnesol
8	20.751	9.49	(E)-farnesol
9	21.171	2.72	(E)-farnesal
10	21.929	10.64	benzyl benzoate
11	23.920	3.33	1-octadecanol
12	24.587	42.63	benzyl salicylate
13	32.534	4.12	Isoeicosane

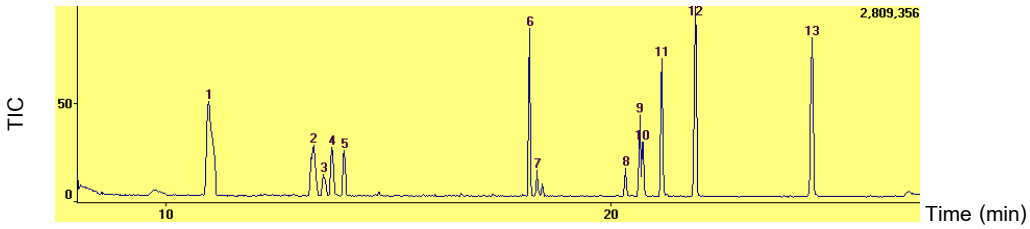

Fig 7 Chromatogram of cold enfleurage absolute

Table 6 Chemical compounds of cold enfleurage absolute

Peak No.	Retention time (min)	%Relative peak area	Possible compounds
1	10.962	23.13	linalool
2	13.323	8.31	Z)-geraniol
3	13.542	1.87	(Z)-citral
4	13.732	4.69	(E)-geraniol
5	14.006	4.14	(E)-citral
6	18.182	8.16	(Z)-beta-farnesene
7	18.354	0.81	(E)-beta-farnesene
8	20.344	1.42	(E)-nerolidol
9	20.665	4.67	(Z)-farnesol
10	20.733	3.40	(E)-farnesol
11	21.165	9.04	(E)-farnesal
12	21.925	14.73	benzyl benzoate
13	24.551	15.62	benzyl-salicylate

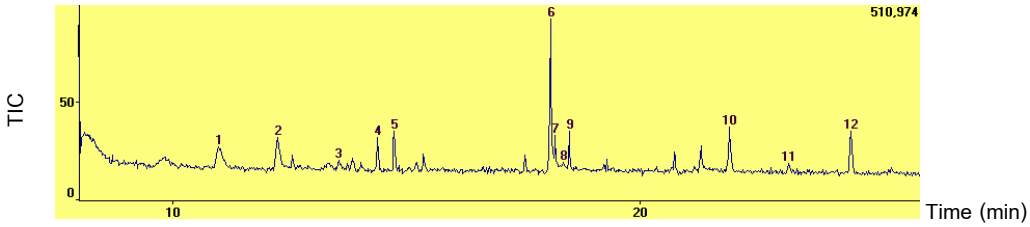

Fig 8 Chromatogram of hot enfleurage absolute

Table 7 Chemical compounds of hot enfleurage absolute

Peak No.	Retention time (min)	%Relative peak area	Possible compounds
1	10.989	3.80	linnalool
2	12.254	3.94	octanoic acid
3	13.550	4.01	unknown[1]
4	14.381	6.69	(E,E)-2,4-decadienal
5	14.746	8.49	(E,E)-2,4-dodecadienal
6	18.094	31.75	n-undecanoic acid
7	18.195	7.91	farnesol
8	18.358	4.84	geranyl valerate
9	18.492	5.94	ethyl pentadecanoate
10	21.919	9.72	benzyl benzoate
11	23.193	3.22	unknown[2]
12	24.514	9.70	benzyl salicylate

Unknown[1] m/z 40(33) 41(37) 43.00 (70)44(56) 51(36) 55(100) 70(29) 71 (23) 79(23) 83 (37) 98 (42) 26(23)

Unknown[2] m/z 41(33) 43(100) 45 (20) 57 (26) 58 (53) 59(11) 60(15)69(17) 71(13) 149 (11.20)

CONCLUSIONS

The quantity, characteristics and chemical compounds of Frangipani oil and absolutes were different because of extraction methods. Percentage yields of Frangipani oil from water-steam, water and steam distillation were 0.0342, 0.0167, and 0.0045%, of hexane and petroleum ether absolutes were 0.4170 and 0.3510%, of cold and hot enfleurage were 0.3842 and 12.24%, respectively. The main compound of essential oil and absolute from solvent extraction was benzyl salicylate, from cold enfleurage was linalool and from hot enfleurage was n-undecanoic acid. Frangipani absolutes by using hexane extraction is appropriate method to produce Frangipani absolutes in pilot scale for used as perfume or cosmetic materials because this method is cheaper and more convenient than enfleurage method and it gave higher percentage of yield than distillation method. The pure essential oil from water-steam distillation is fitting for aromatherapy such as massage oil.

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