# Optimization of Cinnamaldehyde Production from Cinnamon Leaf (Cinamomum burmanii Nees ex Bl)

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#### **ABSTRACT**

Cinnamaldehyde was the major constituent of the bark and leaf oil of Cinnamomum burmannii Blume. The first production of C. burmannii is the bark powder for flavoring industry and the second product is essential oil. The essential oils as well as cinnamon flavor is a potent antimicrobial and antifugal activities, ayuverdic medicine and the antipyretic. The production and functional properties of essential oil from bark or leaf of C. burmannii depend on cinnamaldehyde content. The objective of this experiment was to establish optimum processing condition of cinnamaldehyde production from cinnamon leaf. The essential oil of cinnamon leaf was produced by water and steam distillation method with three factor experiments: bulk density, power distillation and distillation time. The Optimal of three factor process were evaluated using Response Surface Methodology with mathematic, statistic and matlab programs. The result showed that the optimal production of cinamaldehyde compound from the leaf of C. burmannii were producing at bulk density 16.769 kg/m<sup>3</sup>, power to produce of steam 1252 watt, and distillation time 3.027 hour.

Keywords: C. burmannii leaf, water-steam distillation, essential oil, cinnamaldehyde

#### INTRODUCTION

Cinnamonum is a large genus with many species of which yield a volatile oil on distillation (Gunther, 1987 and Purseglove et.al, 1987). The composition and value of the essential oil of C. burmannii depend on the species and the part of the plant, which is utilized. The most important Cinnamonun oils in the world trade are those from C. verum (Cinnamon bark and leaf oils). C. cassia (cassia oil) and C. camphora (sassafras and Ho leaf oils). The latter spices provide oils which are utilized as source of chemical isolates (Anonym, 1995).

Cinnamomum burmannii is the main source of Indonesian cassia, in which there is a considerable export trade. The general composition of C. burmannii is similar to that of C. cassia, and also has a relatively high mucilage content (Purseglove et.al, 1987). Indonesian cassia is much more important as a spice than as a source of oil. It enters international trade along with Chinese cassia (Rismunandar and farry, 2001). The essential oils produced from leaf of C. burmannii is categorized to cassia oil and is not leaf oil because the constituent of cinnamaldehyde content (Purseglove et.al. 1987: Anonym, 1995).

Cinnamon bark oil possesses the delicate aroma of spicy, sweet and pungent taste. Its major constituent is cinnamaldehyde but other minor components impart the characteristic odor and flavor. It is employed mainly in the flavoring industry where it is used in meat and fast food seasonings, sauces and pickles, baked goods, confectionery, cola-type drinks, tobacco flavors and in denal and pharmaceutical preparations (Anonym, 1995; Hirasa and Takemasa, 1998). Perfumery application is far fewer than in flavoring because the oil has some skin-sensitizing properties, it has limited use in some perfumes.

Cassia oil is distilled from a mixture of leave, twigs and fragments of bark. Cinnamaldehyde is the major constituent and is used mainly for flavoring cola-type trinks, with smaller amounts used in bakery products, sauce, confectionery and liqueurs. Like cinnamon bark oil, its skin sensitizing properties and is limit used as a ragrance.

Cinnamon leaf oil has warm, spicy, but rather harsh odor, lacking the rich body of the bark oil. Its major constituent is eugenol rather than cinnamaldehyde. It is used as flavoring agent for seasonings and savory macks. As a cheap fragrance it is added to soaps and insecticides. The high eugenol content also makes it faluable as source of chemical for subsequent conversion into iso-eugenol, another flavoring agent (Anonym, 1995; Senanayake et.al, 1978).

Cinnamaldehyde is a volatile component give the characteristic aromas of the cinnamon spices. Cinnamaldehyde, also produces a slight pungent sentation via the trigeminal nerves (Fisher and Scoot, 997). Another experiment reported that Flavor of cinnamon have used in ayurvedic teraphy in India encent and traditional Chinese medicine (Hirasa et.al, 1998; eng et.al, 2001). Shang et.al (2002), reported that sinnamaldehyde from cinnamomum sp. Oils have exsibited the strong termiticidal property.

The use of the chemicals to enhance the safety of nany foods is of great interest to the food industry. The stability of some foods against attack by microorganisms is due to the fact that the contain naturally occurring substances with antimicrobial activity. Some pices are known to contain essential oils that possess intimicrobial activity, such as eugenol in clove and

cinnamaldehyde in cinnamon (Mau et. al, 2001). Friedman et.al (2000), Concluded from their research that those foods with a high cinnamaldehyde content might protect both the food and/or the consumer against infection by human pathogens. Friedman et.al (2000) was reported cinnamaldehyde in the cinnamon flavor to be effective against human pathogens such as Escherichia coli and Salmonella are stable to food-processing conditions including baking, cooking, frying and micro waving as well as to prolonged storage after incorporation into food such as apple juice, baked product and poultry.

Water and steam distillation is the technique employed to commercially process cinnamon for oil production (Guenther, 1987). In general, the separation component in distillation process depends on boiling point. During the distillation, boiling point was influenced by another variable processes such as bulk density, power distillation and distillation time. Until now, no systematic studies have been published for investigated the influence of distillation variables to the recovery cinnamaldehyde component. Response Surface methodology is a powerful experimental procedure for optimizing multiple, interrelated parameters. In this method, experiments are conducted to discover which values of the parameters optimize a response. The predicted optimal value for the independent variable can be found from the estimated surface (Liu et.al, 2000; Balanos et.al, 2003).

The bark of *C. burmannii* was used as spices and in the production of essential oils, but cinnamon leafs just of waste, are not being used for production of essential oils (Rismunandar and Farry, 2001). The objective of this study was to optimize the optimum processing condition of cinnamaldehyde production from cinnamon leaf.

# **MATERIALS AND METHODS**

Materials: Leaves of C. burmannii were purchased from Cangkringan, Kaliurang, Yogyakarta. Leaf cinnamon was getting occurs of the same old tree that's 10

years old. The raw material chosen of a green leaves.

Sample Preparation: the leaves left after trimming the cut stems, as well as those obtained from pruning operations, provide the raw material for production of cinnamon leaf oil. They were allowed to sorting material, separated with twigs and fragments of bark, and than allowed to dry for 7 day (approximately 16,75% dry matter) before distillation.

Experimental Design: The processing variables were applied in the experiment, specific volume samples material in the retort, the heat power to gaining steam and distillation time. Respond Surface Methodology was used to explore the effect of processing variables and their combinations on production cinnamaldehyde. Initial specific volume sample in the retort ranged from 8,5 to 25,0 kg/m³, the heating power ranged from 1182 to 1364 watt and distillation time ranged from 2,5 to 4,0 hour. RSM basically uses an experimental design to fit a model by least square analysis (Montgomery, 1991). A total of 15 different combinations of three processing variables (Table 1) established to evaluate optimization for production of cinnamaldehyde from cinnamon leaf of distillation experiments.

Table 1. Combination design process of optimization for production cinnamaldehyde

Treatment	Bulk density (kg/m³) X	Heating power (watt) Y	Distillation time (hour) Z
i	25 (1)	1365(1)	4(0)
2	25(1)	1182(-1)	4(0)
3	8,5(-1)	1365(1)	4(0)
4	8,5(-1)	1182(-1)	4(0)
5	25(1)	1273(0)	5.5(1)
6	25(1)	1273(0)	2.5(-1)
7	8,5(-1)	1273(0)	5.5(1)
8	8,5(-1)	1273(0)	2.5(-1)
9	17(0)	1364(1)	5.5(1)
10	17(0)	1364(1)	2.5(-1)
11	17(0)	1182(-1)	5.5(1)
12	17(0)	1182(-1)	2.5(-1)
13	17(0)	1273(0)	4(0)
14	17(0)	1273(0)	4(0)
15	17(0)	1273(0)	4(0)

Distillation Experiment: Dry matter before distillation were cut and than immediately carried out to processing. At the end of each distillation, essential oil from distillation kept in a cool room at 4°C until analysis. Analysis of oil was carried out after the end of distillation experiment and completed within 15 day. The end of this study, a distillation experiment with optimal condition was also performed. This sample (essential oil) was used to validate data obtained from software and to evaluate effect of processing variables during distillation. For the physico-chemical analysis, the essential oil was purification with added Na<sub>2</sub>SO<sub>4</sub> anhydrous.

Analysis: The relative percentage of cinnamaldehyde contained in leaf oil was analyzed by a GC Q gas-liquid chromatography ion trap mass spectrometer. Chromatography was performed using a (0,25 mm x 30 mm, 0,25 mm film, DB-5). Fused silica coloum with an average helium carrier gas flow set to a constant velocity of 40 cm/s. The split ratio of the column was 80:1. The injector temperature was set at 280°C. The column oven temperature was held at 80°C for 5 min, then programmed to 270°C at 10 °C for 5 min. The spectrometer was operated in the electron impak mode with a source temperature of 280°C. Total ion current profiles were used for quantitation.

#### RESULTS AND DISCUSSION

The leaves of *C. burmannii* have a hot taste and emit a spicy odor when cutting, therefore after cutting, the raw material immediately carry out to distillation retort cause its influence the yield oil production and competition chemical compound. The result for analyzed used GC-MS showed that cinnamaldehyde is a major constituent. It is similar with the result report study by the other experiments (Agusta, 2001; Ketaren, 1987; Hirasa and Takemasa, 1998; Purseglove, 1987).

On Table 2. showed the yield of cinna-maldehyde production from cinnamon leaf by water-steam distillation. The difference percentage of cinnamaldehyde compound from this experiments because the essential

oil is mixture with a much constituent compound and they have a deference boiling point. In distillation process, boiling point was influenced with extraction oil from materials, therefore with different condition process, range distillation time and steam production, will influence production of cinnamaldehyde. The boiling point of cinnamaldehyde compound ranged from 135°C to 246°C (Budaverari et.al, 1996; O'neil et.al, 2001). The result report from another experiment explained that cinnamaldehyde production from cinnamon bark is influenced with time distillation and the size crushed material (Lisawati et.al, 2002).

The essential from cinnamon leaf possesses a more desirable aroma and flavor. Cinnamaldehyde have influence of characteristic aroma of cinnamon oil and it's a major of constituent compound (Fisher and Scoot, 1997). The optimal of cinnamaldehyde production from this research have evaluated by RSM was 71,313%.

Lisawati et.al (2000), reported that the cinnamaldehyde production from bark cinnamon by distillation was 17,65% until 32,81%. The percentage cinnamaldehyde compound in leaf oil was higher than in bark oil, from this research can conclude that leaf oil is enough potential to produced cinnamaldehyde compound.

Figure (1) shows the result of optimization condition for production cinnamaldehyde from cinnamon leaf. The interaction of variables process distillation can performance by response surface and plot contour graph. The mathematical models obtained according to the maximum percentage cinnamaldehyde (Y) was:

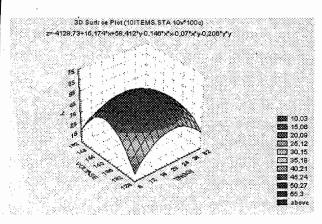
$$Y = 69.413 - 4.879x - 6.768y - 7.119z - 15.847x^{2} - 21.832y^{2} - 15.988z^{2} - 7.011xy - 8.122xz + 11.450yz$$

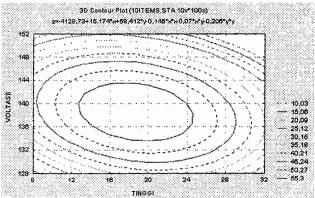
The negative slopes from the model show that directly affect the bulk density, heating power (voltage), time

Table 2. The result of production cinnamaldehyde compound from distillation of cinnamon leaf

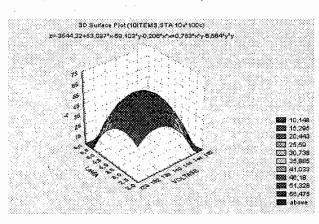
Treatment	Bulk density kg/m³ X	Heating power (watt) Y	Distillation time (hour) Z	Oil yield (%)	Percentage cinnamalde-hyde yield Y(%)*
1	25 (1)	1365(1)	4(0)	0.509	21.788
2	25(1)	1182(-1)	4(0)	0.416	46.628
3	8,5(-1)	1365(1)	4(0)	0.406	30.860
4	8,5(-1)	1182(-1)	4(0)	0.349	27.657
5	25(1)	1273(0)	5.5(1)	0.568	10.729
6	25(1)	1273(0)	2.5(-1)	0.416	39.958
7	8,5(-1)	1273(0)	5.5(1)	0.319	51.441
8	8,5(-1)	1273(0)	2.5(-1)	0.373	48.181
9	17(0)	1364(1)	5.5(1)	0.749	27.167
10	17(0)	1364(1)	2.5(-1)	0.311	19.760
11	17(0)	1182(-1)	5.5(1)	0.665	20.524
12	17(0)	1182(-1)	2.5(-1)	0.616	58.918
13	17(0)	1273(0)	4(0)	0.615	66.136
14	17(0)	1273(0)	4(0)	0.639	76.906
15	17(0)	1273(0)	4(0)	0.686	65.196

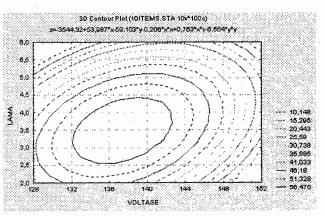
# The optimal result of distillation time = -0.02977 = 3.354 hour



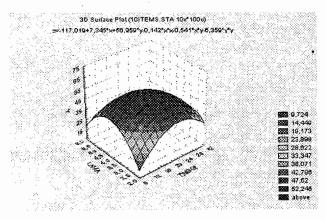


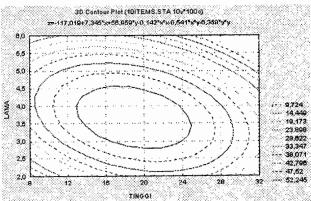
# The optimal result of bulk density = $-0.0271 = 16.769 \text{ kg/m}^3$





# The optimal result of heating power = -0.2287 = 1252 watt





tillation and the interaction of term, respectively, ien each variable levels increased, the response maxim percentage cinnamaldehyde (Y) decreased. The ponse decreased when the variable levels were modid from -1 to 1. The positive slope 11.450 indicated a ximum percentage cinnamaldehyde increase with reasing voltage-time distillation interaction term z).

The optimal result of cinnamaldehyde production aluation for bulk density,, heating power and distilion time were 16.769 kg/m³, 1252 watt and 3.027 ur, respectively. The yield optimal of cinnamaldehyde oduction was 71,313%. The validity of application 3M for optimization of cinnamaldehyde production is 95.144%.

The price and functional properties of cinnamon leaf s were depend on the contained cinnamaldehyde mpound in their essential oils. Cinnamaldehyde comunds in the flavor cinnamon could be used for the timicrobial agents, ayuverdic therapy, and medicine ramedical and antitermitic potential. At the present, and antitermitic potential in the present, and concentration of cinnamaldehyde comunds in cinnamon leaf oils are considered to be imratant to understand potential in health foods.

# **CONCLUSIONS**

The leaf oil of C. burmanii contained cinnamal-hyde is the major compound. This is deferent perntage of cinnamaldehyde in yield oils of difference riables was used at processing. With respect to oduce, the best results were obtained, in the range of ecific volume materials in the retort 16.769 kilogram/meter, at heating power 1252 watt and time distillant 3.554 hour, production cinnamaldehyde from the ocess was possessed 71,313%. That's to be enough tential to production. At the present, the nature and incentration of cinnamaldehyde compounds in cinnamal potential in health foods.

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# LITERATURE CITED

- Anonym,1995.Cassia. http://www.botanical.com/botanical/mgmh/c/cassia32.html
- Anonym, 2000. Cinnamomum Oils (Including Cinnamon and Cassia) http://www.fao.org/docrep/V5350e/V5350e04.htm
- Balanos, N.L.J., Islas, J.H., Alvarez, B.E. and R.R. Martinez. 2003. Mixed Culture Optimization for Margold Flower Ensilage via Experimental Design and Response Surface Methodology. J. Agric. Food Chem. 2003, 51, 2206-2211.
- Budaveri, S.; O'neil, M.J.; Smith, A.; Heckelman, P.E and J.F, Kinneary., 1996. *The Merck Index*. The Merck and CO. Inc. USA, 386.
- Friedman, M., Kozukue, N and L.A., Harden. Cinnamaldehyde Content in Foods Determinated by Gas Chromatograpgy-Mass Spectrometry. J.Agric. Food Chem. 2000, 48, 5702-5709.
- Friedman, M., Henika, P.R., Mandrell, R.E. Antimicrobial Activities of Plant Essential Oils and Other Plant Compounds Against Strains *Escherichia coli* and *Salmonella entrica*. J. Food Sci. 2000, submitted for publication
- Fisher, C. and R.T. Schott. 1997. Food Flavors, Biology and Chemistry. Departement of Animal and Food Sciences and Phychology. University of Delaware. New York, USA
- Guenther, E., 1987. Essential oils Vol.1., terjemahan oleh Ketaren, S., Penerbit Universitas Indonesia., 23-60

- Hirasa, K. and Takemasa, M.1998. "Spices and Technology". Lion Corporation, Tokyo, Japan, p. 182 186
- Jeng, L.M, Chiu, P.C and C.H, Pao. Antimicrobial effect of Extracts from Chinese Chieve, Cinnamon and Corni Fructus. J. Agric. Food Chem. 2001, 49, 183-188.
- Katzer, G. 1999. Ceylon Cinnamon (*Cinnamomum zeylanicum* Blume). WDG, Australia.
- Lisawati, Y., Sulianti, B.S. and Chairul. 2002. Pengaruh Waktu Destilasi dan Derajat Kehalusan (Mesh) Serbuk Kulit Kayu Manis (*Cinnamomum burmanii Nees ex* BL) terhadap Kadar Sinamilaldehida pada Minyak Atsirinya. Majalah Farmasi Indonesia, p: 123-132
- Liu, F.F., Ang, C.Y.W and D. Spinger. Optimization of Extractions for Active Components in *Hypericum perforatum* Using Response Surface Methodology. J. Agric. Food Chem. 2000, 48, 3364-3371.
- Montgomery, D.C., 1991. "Design and Analysis of Experiment". Third Edition, John Wiley & Sons, Singapore.

- Mau, J.L., Chen, P.C and P.C. Hseih. Anti microbial Effect of Extracts from Chinese Chive, Cinnamon and Croni Fructus. J. Agric. Food Chem. 2001, 49, 183-188.
  - O'neil, M.J., Smith, A. and P.E. Heckelman. 2001. "The Merck Index: an Encyclopedia of Chemicals, Drugs and Biologicals". Merck and Co.Inc. NJ.
  - Purseglove, J.W., Brown, E.G., Green, C.L. and S.R.J. Robbins. 1987. "Spices". Volume I. Longman Scientific & Technical Couplised in the United States with John Wiley & Sons. Inc. New York, p: 144-145.
  - Rismunandar dan B.P. Farry. 2001. "Kayu Manis, Budi Daya dan Pengolahan". Penebar Swadaya, Jakarta.
  - Senanayake, U.M., Lee.T.H., Wills, R.B.H. Volatile Constituents of Cinnamon (*Cinnamomum* zeylanicum) Oils, J. Agric. Food Chem. 1978, 26, 822-824
  - Shang, T.C and S.C. Sen. Antitermitic Activity of Leaf Essential Oils and Components from Cinnamomum osmophleum. J. Agric. Food Chem. 2002, 50, 1389-1392.