Optimization Of Trigona Honey And Coating Material Formulations For Antioxidants And Physical Properties Of Microencapsulation Of Coriander Seed Extract Using The D-Optimal Mixture Method

Irfan Muttaqin[1*] ,Yusman Taufik[2] ,Yusep Ikrawani[3]

[1] Student Magister Teknologi Pangan Universitas Pasundan, Bandung, Indonesia
[2,3] Lecturer Magister Teknologi Pangan Universitas Pasundan, Bandung, Indonesia
*Corresponding Author:
Email: irfanmuttaqin567@gmail.com

Abstract
The purpose of this study was to produce an optimal formulation of coriander seed extract microencapsulation products using the Mixture D-Optimal Design Expert program.The study consists of two stages. The first stage was the analysis of the coriander seed extract powder using the spray drying method without the use of coating materials and honey. The second stage was to determine the optimal formula using the D-Optimal Mixture method of Design Expert program through measurement of chemical responses (antioxidant activity, and water content), physical response. (particle size). The optimum formula obtained based on the Design Expert program with the D-Optimal Mixture method has a desirability value of 1 with the prediction value of each response being 395.124 ppm antioxidant activity, 2.329 % water content, and 2.967 µm particle size.

Keywords: *Seed extract, coriander, Design Expert, antioxidant and microencapsulation.*

I. INTRODUCTION

Free radicals are reactive compounds that are produced from the body's metabolic processes and are able to react with molecules in direct contact by attracting electrons. If the number of free radicals in the body increases, they can cause fat oxidation, damage to proteins and DNA. The impact of free radicals can cause various diseases in the body.Along with the development of science and technology, research on various food ingredients containing bioactive compounds is increasing. One of the benefits of bioactive compounds that have been studied is as an antioxidant which has a positive effect on healing various diseases. Based on research that has been done, various bioactive compounds that function as antioxidants can be used to cure various degenerative diseases such as hypercholesterolemia, diabetes, cancer, hypertension, and so on.Trigona coriander seeds and honey have a fairly high source of antioxidants such as phenolic compounds, flavonoids, vitamins A, C and E. Coriander (C. sativum) is a herbal, aromatic plant, and belongs to the Apiaceae family. Coriander has a long history of nutritional and herbal medicinal uses including anti-inflammatory, analgesic, anticonvulsant, blood pressure lowering, cholesterol lowering, digestive and sedative (Laribi et al., 2015).Honey is a bee product whose main components are fructose and glucose and contain protein, amino acids, vitamins, enzymes, minerals and other minor components (Burlando & Cornara, 2013).

Honey is rich in phenolic content such as quercetin, caffeic acid phenethyl ester (CAPE), acacetin, kaempferol, galangin which act as natural antioxidants (Khalil & Sulaiman, 2010). Antioxidants in trigona honey are beneficial for various types of diseases related to polyphenolic components, peptides, enzymes and organic acids. Bee products are also used in gastrointestinal, cardiovascular, inflammatory and neoplastic conditions (Eteraf & Najafi, 2013).It is widely known that the nutrients or active substances contained in foodstuffs have certain properties, including being easily damaged by the environment and the natural properties of the ingredients themselves. One of the efforts to protect and maintain these nutrients is to use microencapsulation. The microencapsulation method is one of the most widely used techniques for protecting bioactive compounds from various environmental factors such as evaporation, oxidation, temperature degradation, humidity, and light, therefore, can extend shelf life. product and avoid damage. (Wathoni et al., 2019).In the microencapsulation technique, the material used in the coating can be a polymer. Polymers have certain physicochemical properties so they have different structures and characteristics.

The polymer used must be capable of providing a cohesive film with the core material, must be chemically miscible, but must not react with the core (inert), and must have properties suitable for the coating purpose. (Baena-aristizábal et al., 2019). The coating materials used generally are maltodextrin and arabic gum.Maltodextrin functions in providing resistance to oxidation and increases the solubility of an encapsulate, while gum arabic functions in increasing the stability of the emulsion (Fasikhatun, 2010). However, microcapsules coated with protein isolate have the highest encapsulation efficiency with greater retention and higher antioxidant activity among all wall materials (Abrahao, et al, 2019). Maltodextrin used as a coating needs to be combined with soy protein isolate to increase the stability of the emulsification. Soy protein isolate has good solubility, emulsification properties, water binding capacity, foaming properties, good film forming ability, relatively easy to digest, and safe for consumption (Chen, et al., 2011).This research uses the Design Expert program, the Mixture D-Optimal method, which is used to help optimize products or processes. The Design Expert program with the D-Optimal Mixture method has advantages over other data processing programs, namely the accuracy of this program is quite high, it is more flexible, and it also provides statistical features that make it easier to operate (Tiaraswara, 2016).

II. METHODS

This research was conducted through two stages, namely stage 1 research, and stage 2 research.

- **Phase 1 Research**

Phase 1 of the research was the analysis of coriander seed extract products that had not been coated with coating material and honey using the spray dryer method. The analysis that will be carried out includes analysis of antioxidant levels, analysis of water content, analysis of particle size.

- **Phase 2 Research**

Phase 2 of the research is the design and response formulation which will be formulated using the Design Expert ver.13 Mixture D-Optimal method as a changing variable and a fixed variable first. The changed variables were maltodextrin, protein isolate, and trigona honey while the fixed variable was 60% coriander seed extract. The fixed variable is the raw material component which is assumed not to affect the response that will be obtained from each formula. The variable changed is the raw material component which is assumed to have an influence on the response produced in each formula.

Table 1. Variable Design Changes

Var. Changed	*Low*(%)	*high*(%)
Maltodextrin*	5	10
*Protein isolates**	10	15
Trigon honey	15	20

Table 2. Output formula to analyze

A	Maltodextrin	ISPs	HONEY
A1	5%	10%	25%
A2	7.12028%	12.1717%	20.708%
A3	8.1296%	10%	21.8704%
A4	10%	10%	20%
A5	5%	15%	20%
A6	5%	13.0365%	21.9635%
A7	7.17259%	14.0053%	18.8221%
A8	6.07803%	10.9937%	22.9282%
A9	8.4589%	15%	16.5411%

A10	9.18251%	12.2393%	18.5782%
A11	10%	13.6%	16.4%
A12	5%	10%	25%
A13	5%	15%	20%

The variable design changes can be observed in Table 1 and the output of the design results is in Table 2. The responses measured were chemical responses (antioxidant levels, moisture content) and physical responses (particle size).

III. RESULTS AND DISCUSSION

Phase 1 Research Results

Phase 1 of the research was the analysis of coriander seed extract products that had not been coated with coating material and honey using the spray dryer method. The results of stage 1 research can be observed in the following table.

Table 3. Stage Research Results 1

No	Response	Coriander extract
1	Antioxidant Level (ppm)	1184.03 ppm
2	Water content (%)	6. 4694 %
3	Particle Size (μm)	2745μm

Phase 2 Research Results

The stages of this research are carried out by makingExtract of coriander seeds The output of the research design is the design of changing variables along with the percentage of total changing variables used in the product and the range of limits on the Design Expert Mixture D-Optimal method. The output of the research design is in the form of 13 different formulations.The thirteen formulations will be analyzed chemically and physically, all data from the analysis will be inputted into the application so that the optimal formulation will be obtained from the Design Expert.

A. Results of Chemical Response Analysis

1. Antioxidant Levels

Based on the results of the analysis of variance (ANOVA), the p value "prob>F" in the Sp Quartic vs Quadratic model was obtained, namely <0.0002 (smaller than 0.05) indicating that 13 formulations were used as the initial model for significant analysis of the response to antioxidant levels. However, in the sample codes A7 and A9, the formulas generated from the Design Expert program cannot be used as products because of the high viscosity produced, the drying process cannot be carried out using spray drying.

Table 4. Results Of Response Analysis Of Antioxidant Levels

Sample Code	Antioxidant
A1	764.17 ppm
A2	643.69 ppm
A3	526.09 ppm
A4	695.21 ppm
A5	739.96 ppm
A6	918.36 ppm
A7	0
A8	748.44 ppm
A9	0
A10	616.60 ppm
A11	574.69 ppm
A12	764.65 ppm
A13	740.48 ppm

Determination of the order model for each response is based on value*sequential p-value*the lowest. However, under certain conditions, the model order is also determined based on the value of adjusted R2 , predicted R2 , and models with higher polynomials. The lowest sequential p value is found in the order model Sp

Quartic vs Quadratic (0.0014) and adjusted R2, predicted R2, and models with higher polynomials.The value of R2 is 0.9958 which indicates that the independent variables in this case A (Maltodextrin), B (Protein Isolate) and C (Trigona Honey) have an effect of 99.58% on the dependent variable, namely the response to antioxidant levels and the rest is influenced by factors -other factors outside the independent variables.

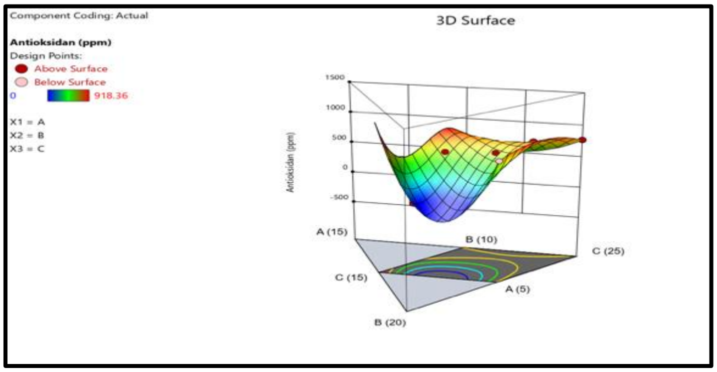

Fig 1. Graphic Contour Plot Response Up to Antioxidant

The distribution of the results of the response analysis of antioxidant levels in the 13 coriander seed extract microencapsulation formulas that were used as the initial model can be displayed in a three-dimensional graph in the form of a 3D Surface Plot (Figure 1). Scattered dots indicate the position of each of the 13 formulations based on the response of antioxidant levels to the Sp Quartic vs Quadratic order model. The formula with the lowest antioxidant content is placed in the red graph area followed by orange, yellow, green, light blue to dark blue indicating a higher antioxidant response. According to Jalil, et al., (2009), Honey from stingless bees contains compounds such as protacatechuic acid (PCA), 4-hydroxyphenylacetic acid, and cerumen which function as antioxidants. Trigona honey has hydrogen peroxide, flavonoids, phenolic compounds, and antibacterial peptides which act as antibacterials.According toWangensteen et al (2004) Coriander seeds contain flavonoids and vitamins which play a role in lowering cholesterol and as antioxidants. In phase 1 of the study, antioxidant levels were measured in coriander seed extract powder using the spray drying method without the use of coating materials, with antioxidant levels of 1184.03 PPM. The antioxidant value without using a coating material is very low, in contrast to using a coating material and the addition of trigona honey solution can increase the antioxidant content in microencapsulation of coriander seed extract with an average value of antioxidant content of 526.09 ppm -918.36 ppm.

2. Water content

The results of the analysis of variance (ANOVA) showed that the p value "prob>F" in the Sp Quartic vs Quadratic model was 0.0051 (smaller than 0.05). The p value "prob>F" which is smaller than 0.05 indicates that the 13 formulations used as the initial model for significant analysis of the response to water content, however, in sample codes A7 and A9, the formulas produced from the Design Expert program cannot be used as products due to their high the resulting viscosity, the drying process cannot be carried out using spray drying.

Table 5. Water Content Response Analysis Results

SAMPLE CODE	WATER CONTENT
A1	2.58%
A2	3.41%
A3	2.77%
A4	3.02%
A5	3.05%
A6	3.35%
A7	0
A8	4.77%
A9	0
A10	2.25%

A11	2.30%
A12	2.59%
A13	3.12%

The order model assigned to the response to water content is Sp Quartic vs Quadratic. The order model is determined based on the sequential p value of 0.0066. and adjusted R2, predicted R2, and models with higher polynomials.The R-square value in response to water content is 0.9768. This value indicates that the independent variables in this case A (Maltodextrin), B (Protein Isolate) and C (Trigona Honey) have an effect of 97.68% on the dependent variable, namely the response of water content and the rest is influenced by other factors outside the variable free.

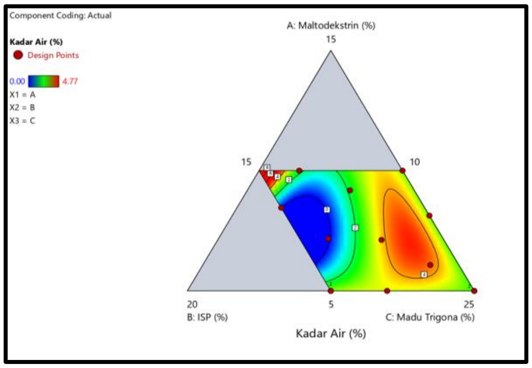

Fig 2. Graph Contour Plot Response Moisture Content

The results of the analysis of water content activity and seen based on the Graph Contour Plot and 3D Surface Plot with the Sp Quartic vs Quadratic order model show that the red graph area tends to lead to the trigona honey variable with an average water content in the 13 formulas ranging from 2.25 – 4.77 % of this value is better than without the addition of coating material which is equal to 6.4694%. The higher the water content value indicates the unwanted water content based on SNI 01-4320-1996 water content in powder products, which has a water content at the level of 3.0-5.0%.Based on the decrease in water content, it can be said that the higher the concentration of the coating material in the emulsion, the lower the water content of the resulting product. This is in accordance with the results of research by Saikia et al. (2015) who showed that microcapsules with a core material to coating material ratio of 1:10 had a higher water content than microcapsules with a ratio of 1:20.Moisture content is the main parameter in determining the quality of dry microcapsule products related to product durability and shelf life (Desmawarni, 2007). The low water content in microcapsules makes them more resistant to microbiological damage and damage due to hydrolysis of the oil contained in the microencapsulation (Simanjuntak, 2007).Trigona honey has characteristics such as being thinner and amber brown in color. The taste of trigona honey tends to be more acidic because it has a pH of 3.05-4.55. The water content of honey is more, which is around 30-35%.

B. Physical Response

1. Particle Size

Based on the results of the analysis of variance (ANOVA) the p value "prob>F" in the special quartic vs Quadratic model was 0.0014 (smaller than 0.05) which showed 13 formulations were significant in response to particle size, but in sample codes A7 and The A9 formula produced from the Design Expert program cannot be used as a product because of the high viscosity produced, the drying process cannot be carried out using spray drying.

Table 6. Particle Size Analysis Results

SAMPLE CODE	Particle SIZE
A1	6.048µm
A2	5,580µm
A3	6.462µm
A4	6.903µm

A5	17.470µm
A6	6.541µm
A7	0
A8	7.297µm
A9	0
A10	5.243µm
A11	2.984µm
A12	6.048µm
A13	17.470µm

The R-square value on the particle size response is 0.9882. This value indicates that the independent variables in this case A (Maltodextrin), B (Protein Isolate) and C (Trigona Honey) have an effect of 98.82% on the dependent variable, namely the particle size response and the rest is influenced by other factors outside the variable free.

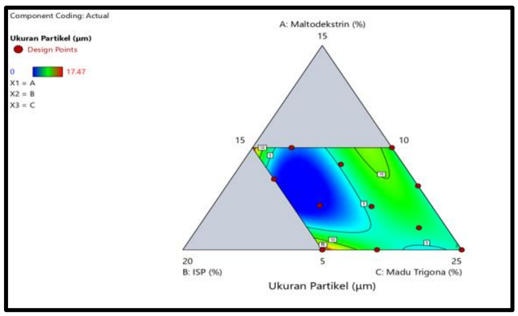

Fig 3. Particle Size Response Contour Plot Graph

The results of particle size analysis and views based on the Graph Contour Plot and 3D Surface Plot with the Sp Quartic vs Quadratic order model show that the red graph area tends to lead to protein isolate and maltodextrin variables with the average particle size in the 13 formulas ranging from 2.984 – 17.470 µm size value The particle size will be higher depending on the large amount of coating material concentration. Materials can be said to be microparticles if the material is 2-5000 in sizeµm (Jyothi., et al. 2010).Particle size has an important role in microencapsulation because it can affect particle stability, bioavailability of active ingredients, release characteristics. The process of forming microencapsulation sizes in this study used an ultra turrax homogenizer. The speed of the homogenizer used was 10,000 rpm for 30 minutes. One of the factors that affect the particle size of a material is the speed of agitation.

The higher the speed of agitation, the smaller and more uniform the size of the particles formed.In addition, the coating materials used in this study, namely protein isolate and maltodextrin are the two materials used in the microencapsulation process. Both of these materials have an important role in microencapsulating particle size, protein isolate can help increase particle stability and control particle size by forming a homogeneous film and is able to strengthen particle structure by preventing damage or cracks in particles during the production and storage process, while maltodextrin can help increase solution viscosity and preventing particles from agglomerating. In addition, maltodextrin can help protect the active ingredients from moisture and oxidation during the production and storage processes, thereby increasing the stability of the particles.

C. Selected Formulas

The selected formula is the optimal solution or formulation predicted by the Design Expert of the D-Optimal Mixture method based on an analysis of the chemical response (antioxidant content and moisture content), physical response (particle size) The formula optimization process is carried out to obtain a formula with the most optimal response. The most optimal response is obtained with a desirability value close to 1. Optimization stagei which was carried out gave nine formula solutions with desirability values of each 1 ; 1 ; 1 ; 0.960 ; 0.898 ; 0.642 ; 0.545 ; 0.283. The optimal formula solution is obtained from the results of running the Design Expert program ver. 13 of 13 formulas that are likely to give optimal results. Based on the formula solution, a formula that gives a high desirability value is selected which will then be recommended by the Design Expert ver. 13.Analysis resultss performed on the nine solutions recommended by the Design Expert application ver. 13, the most optimal formula was selected with a desirability value close to one (1), namely solution 1 which had a formula of 6.572% maltodextrin, 13.239% protein isolate, and 20.189% trigona honey as variable variables with a predicted response value of antioxidant levels of 395.124 ppm, water content 2.329 %, and a particle size of 2.967 µm.

IV. CONCLUSIONS

Conclusion

- Based on the results of research using the Design Expert ver. 13.0 The D-Optimal Mixture method can determine the optimal formulation for the microencapsulation characteristics of coriander seed extract. Responses that influenced the 13 formulations used as the initial model were antioxidant content, water content, and particle size.
- Based on the optimization results, an optimal formulation is obtained which is recommended by the Design Expert ver. 13.0 The D-Optimal Mixture method has a desirability value of 1 which consists of a combination of 6.572% maltodextrin, 13.239% protein isolate, 20.189% trigona honey as variable variablewith predicted response value for antioxidant content of 395.124 ppm, water content of 2.329%, and particle size of 2.967 µm.

Suggestion

- It is necessary to observe the microstructure using a Scenning Electron Microscopy (SEM) tool to see the external appearance of the microencapsulated powder which is related to the release rate of the active ingredients in the microencapsulated coriander seed extract.
- There needs to be a temperature comparison in the process of making coriander seed extract microencapsulation in order to know which temperature is the best in producing antioxidant activity.
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