Low-temperature process for green tea drying using zeolite adsorption integrated fluidized bed dryer

S U Handayani, V Paramita, M E Yulianto, A P Siswanto
Industrial Technology Department, Vocational School, Diponegoro University

Corresponding author: sriutamihandayani@gmail.com

Abstract. This work studied the optimization of low-temperature drying by using a laboratory scale and applied it on the pilot plant scale of fluidized bed dryer integrated with zeolite adsorption. The drying time was observed regarding the airflow rate, temperature and zeolite-tea leave ratio. The green tea leaves moisture content was specified gravimetrically until constant weight can be obtained. Applying 70°C as the drying temperature, the green tea leave is secure for a faster drying rate and can be avoided from the catechin epimerization and thermal degradation. Applying 0.40 m/s as airflow rate can remove the water surface faster than 0.50 m/s of airflow rate, although it needs a longer time to drag out the inner moisture content of the solid. The pilot-plant scale required longer time (40 minutes) on the first falling rate period than laboratory scale (30 minutes), although it gave faster second falling rate period at the same airflow rate (0.50 m/s), temperature (70°C) and zeolite-tea leave ratio (25:75).

1. Introduction
Drying is a process of removal of moisture from the materials into surrounding environment air by vaporizing the water content [1-5]. High temperature drying for the wet fermented dhool (macerated tea shoot) applied by endless chain pressure (ECP) dryer (100 °C) and fluid bed dryer (140 °C) were providing unfavorable quality. The drying of tea is purposed to decrease the moisture content of treated tea to reach about 3% wb to extend the shelf life during storage. In order to increase the fluid bed efficiency, modified the inlet air supply into three-stage was also applied. The multi-stage fluid bed dryer with re-circulation is found to have the best combination of characteristics 1. The drying kinetics of canola in fluidized bed dryer (FBD) were studied in the range of 30–100 °C and concluded that the drying rate in FBD was found to be high at increased temperatures [2].

Many researchers gave their pay attention to the dryer heating sources or dryer types in order to obtain the optimum and best drying conditions which are specific to the materials. The favorable low temperature for drying is around 40–70°C, especially for heat-sensitive materials [6-7]. In order to optimize energy in low-temperature drying, zeolite adsorption performed favorable quality products [5].

Different from fermented tea, green tea (unfermented tea) includes heat-sensitive materials. Tea catechin ((–)-epicatechin, (–)-epigallocatechin, (–)-epicatechin gallate, and (–)-epigallocatechin gallate) epimerized, from epi- to nonepi- structures, in the pH 8 buffer solution at 70 °C for 20 min [8]. Green tea catechin also easily epimerized during brewing, sterilization and long-term heating during preparation [9]. The epimerization possibly occurred when heating at 40 °C during storage in a few days [10]. In order to dry at low temperatures, humidity serves as a fundamental role in the drying of materials. When less humidity applied in the inlet air, the drying materials possibly finished faster comparing with high humidity applied for inlet air [11]. Airstream flow rate speeds up the drying process by bringing
the high humidity air forward into outlet air and taking dry air on the materials. Circulating fluidization offers a promising technology for drying a post-harvest product, rather than discard the used hot air [12]. Moreover, decreasing the high humidity air before it was circulated plays an important role. Therefore, modifying and scaling up the re-circulated low-temperature drying equipped by zeolite adsorption is an interesting area to explore because it provides shorter drying time due to low humidity hot air applied and increase the quality of dried materials [4-5]. Present work studies the optimization of low-temperature drying by using a laboratory scale of fluidized bed dryer and applies on the pilot plant scale of fluidized bed dryer, both dryers are equipped zeolite adsorption. The drying time was observed regarding the airflow rate, temperature, and zeolite-tea leave ratio.

2. Methods

2.1. Materials
Green tea was obtained from local tea plantation owned by Rumpun Sari Medini (Kendal, Indonesia). Zeolite and sodium hydroxide was bought from Bratachem (Semarang, Indonesia).

2.2. Drying of green tea leave
The tea leaves were dried in the fluidized bed drier equipped with the zeolite as adsorber. The pilot-plant scale of fluidized bed drier was composed of a cylindrical chamber with a 600 mm ID and 2000 mm height. The humidity of air was adsorbed by zeolite before blown into the drying chamber. The zeolite was placed on an adsorber box, sized 30 x 30 x 15 cm (Figure 1). In addition, the zeolite also placed on the drying chamber with the ratio of zeolite and green tea at 75:25 and 25:75. The airflow rates were 0.40; 0.45 and 0.50 m/s, inlet drying temperatures were 40, 55 and 70 °C and the outlet air temperatures were decreased with the decreasing of airflow rates, in the range of 31-39°C.

![Figure 1](image-url). Schematic of circulating fluidized bed dryer integrated with zeolite adsorption [4].

2.3. Zeolite activation
It is required to activate the natural zeolite before applying it for drying, in order to increase the water adsorption capacity and remove any impurities containing on the zeolite. Methode applied was referring to the previous work [4].

2.4. Response surface methodology (RSM)
In order to executed RSM, the design of experiments (DoE) applied was central composite designs (CCD) by using Software Statistica 8.0 (StatSoft, Dell Software, Texas, United States). A second-order
model can be constructed efficiently with this methodology [13]. CCD are first-order (2N) designs augmented by the additional center and axial points to allow estimation of the tuning parameters of a second-order model.

\[ Y = R_0 + \sum_{i=1}^{k} R_i X_i + \sum_{i=1}^{k} R_i^2 X_i^2 + \sum_{i=1, j<i}^{k} \sum_{j=2}^{k} R_i X_i X_j + \varepsilon \]  

(1)

where \( X_i, X_j \) is input variables that affect the response \( Y \); \( R_0, R_i, R_{ii} \) and \( R_{ij} \) \((i = 1-k, j = 1-k)\) is a known parameter, and \( \varepsilon \) is a random error. A second-order model is designed so that the variance of \( Y \) is constant for all points equidistant from the center of the design.

\[ X_i = (\frac{X_i - X_0}{\Delta X_i}) \]  

(2)

where \( X_i \) is the value of the code, \( X_0 \) is the actual value at the center point and \( \Delta X_i \) is the value of the pace of change [14]. The design involves 2N factorial points, 2N axial points and 1 central point (9 runs). The dependent variable was moisture content, while the independent variables were drying temperature \( (X_1) \) and time \( (X_2) \).

**Table 1.** Run parameter for laboratory-scale optimization at zeolite-tea leave ratio of 75:25

| Run | Coded values | Real values |
|-----|--------------|-------------|
|     | X1 (°C)     | X2 (min)    | X1 (°C) | X2 (min) |
| 1   | -1          | -1          | 40      | 30       |
| 2   | -1          | +1          | 40      | 90       |
| 3   | +1          | -1          | 70      | 30       |
| 4   | +1          | +1          | 70      | 90       |
| 5   | -α          | 0           | 33.8    | 60       |
| 6   | +α          | 0           | 76.2    | 60       |
| 7   | 0           | -α          | 55      | 18       |
| 8   | 0           | +α          | 55      | 102      |
| 9   | 0           | 0           | 55      | 60       |

2.5. Moisture content analysis

The moisture content of green tea leaves was determined by the gravimetrical method. Green tea leaves (120 g) was placed in a fluidized bed dryer and heated at determined temperature (Table 1). The samples were taken every 15 minutes and weighed in an analytical balance (OHAUS Pioneer TM PA2102C, Ohaus Instruments Co., Ltd., Shanghai, China) until constant weight can be obtained. The value of moisture content was determined using Equation (l), as follows:

\[ WC = \frac{w_t - w_n}{w_t} \]  

(3)

where \( WC \) is the weight of water in the tea leaves, \( w_t \) is the weight of tea leaves at t minutes and \( w_n \) is the weight of tea leaves at constant weight. Formatting the title

3. Result and discussion

3.1. Temperature optimization on the drying time regarding the moisture content

The drying temperature and time were optimized by applying the response surface methodology (RSM). The design of experiments (DoE) plays an important rule in this application. It selected the point in which the main response should be evaluated and identified the design variables which gave large effects for further investigation. Applying multiple regression analysis on the experimental data, the following second-order polynomial equations were found to represent the moisture content (db, \( Y \)) on Equation (2), as follows:
\begin{equation}
Y = 0.491 - 0.682X_1 - 0.541X_2 + 0.047X_1^2 + 0.138X_2^2 + 0.186X_1X_2
\end{equation}

Figure 2. Three-dimensional response surface for the moisture content of green tea leave regarding the temperature and time of drying at 0.50 m/s of air flow rate with zeolite-tea leave ratio of 75:25.

Figure 2 shows the effect of temperatures on the moisture content at 0.50 m/s of airflow rate and zeolite-tea leave ratio of 75:25 regarding the drying time. The longer the drying time and the higher the temperature decreased the moisture content of tea leaves, significantly. The higher temperatures at a constant airflow rate gave the possibility to the bulb temperature during drying could be reached faster. Although, the difference in temperature between the air stream and the wet surface of the solid (i.e. green tea leave) was higher on the higher temperature applied. However, the temperature of the wet surface increased faster when applying lower temperatures. Faster drying time (105 min) can be reached by applying a higher drying temperature (70 °C).

Figure 3. Pareto chart (a) and observed vs predicted values (b) for the moisture content of green tea leaves dependent variable regarding the temperature and time of drying at 0.50 m/s of airflow rate with zeolite-tea leave ratio of 75:25.
Pareto chart (Figure 3a) also mentioned that the drying temperature gave larger effects (21.1095) than the time required for drying (16.7339). This result is in agreement with the conclusion of the drying rate was found to increase significantly with increasing temperature. Moreover, the predicted data were fitted well with the observed data, as shown in Figure 3b. This result was also evidenced by the value of R-square of 99.47%. Since green tea leaves are containing sensitive temperature components (e.g., catechin), it is required to avoid the epimerization and thermal degradation of catechin and applied 70 °C as the drying temperature of green tea leaves is necessary. Even though green tea leaves already inactivated before drying, the enzyme activity of polyphenol oxidase or hydroperoxidases can possibly occur yet. Applying higher drying temperature than 70 °C during drying, can possibly increase the risk of catechin epimerization, thermal degradation and also reduce the effects of drying caused by the case hardening on the outer layer of the dried product in the early stage of drying. The optimum value of temperature was 70 °C for 105 min, in order to avoid the epimerization and thermal degradation of catechin. The moisture content possibly reached 0.005 (dB) from moisture content initial of 1.423 (dB) by applying 0.5 m/s of airflow rate and zeolite-tea leave ratio of 75:25.

3.2. Scaling-up the laboratory into a pilot-plant model

Since in the relatively dilute vertical gas-solid suspensions, the shear of the particle phase could be negligible to the overall pressure gradient, the global flow behavior of the recirculating FBD is largely independent of particle collisions. In the absence of interparticle forces or electrostatics, continuum equations for gas-solid suspensions derived by five dimensionless parameters. These parameters determined the operational characteristics of the bed (Froude number and solid loading), the combination of gas and particle properties (Reynold number and density ratio) and the ratio of solid loading to the bed diameter \( \frac{L}{D} \).

\[
Fr = \frac{u}{\sqrt{g \frac{D}{d}}} \\
L = \frac{G}{\rho g u} \\
Re = \frac{du \rho g}{\mu} \\
R = \frac{\rho_s - \rho_g}{\rho_g}
\]

where, \( u \) represents the superficial gas velocity; \( G \) is the average solid flux; \( \rho_s \) and \( \rho_g \) are the densities of the gas and the material of the particles, respectively; \( \mu \) is the gas viscosity; \( L \) and \( D \) is the length and diameter of bed, respectively, and \( g \) is the acceleration of gravity.

Thus, the global hydrodynamics found in the laboratory-scale model can be reproduced in a pilot-plant model of identical aspect ratios by matching values of Fr, L, Re, R, and \( \frac{L}{D} \). Algebraic manipulations of these numbers yield the following relations between the conditions of re-circulating FBD in the pilot-plant model (subscript 1) and those in the laboratory model (subscript 0):

superficial gas velocity \[
\frac{u_1}{\nu_1^{1/3}} = \frac{u_0}{\nu_0^{1/3}}
\]

particle size \[
\frac{d_1}{\nu_1^{2/3}} = \frac{d_0}{\nu_0^{2/3}}
\]

particle density \[
\frac{\rho_s1}{\rho_g1} = \frac{\rho_s0}{\rho_g0}
\]

solids flux \[
\frac{G_1}{\rho_g1^{2/3} \mu_1^{1/3}} = \frac{G_0}{\rho_g0^{2/3} \mu_0^{1/3}}
\]
characteristic bed dimension \( \frac{D_1}{v_1^{2/3}} = \frac{D_0}{v_0^{2/3}} \)  \( \text{(13)} \)

A wide range of kinematic viscosities, \( v_0 \), is obtained by fluidizing the tea leaves with adjustable air flow rate on laboratory scale model. By virtue of equation 11, new value of \( v_1 \) obtained analogous to that in the diameter of \( D_0 \) \( \text{[12]} \). In this study, we assume that the laboratory scale model operates at constant properties under the following conditions: particle mean diameter, \( d_0 = 0.84 \) mm and density, \( \rho_0 = 1 \) gr/cm\(^3\); inlet air temperature, \( T_0 = 70^\circ \text{C} \) (\( \rho_0 = 1,029 \) gr/cm\(^3\), \( \mu_0 = 0.021 \) mPa.s); superficial gas velocity \( u_0 = 3.5 \) fps, solid flux, \( G_0 = 2 \) to \( 36 \) kg/m\(^2\)/sec and solid sphericity, \( \phi_0 = 0.1 \).

To ensure hydrodynamic similitude, the particle-size-distribution (PSD) relative to the mean and the particle sphericity should also be identified in the pilot-plant model and its laboratory-scale model. Because the sphericity, \( \phi \), affects the global hydrodynamics of fluidized beds through the product, \( \phi d \), then adjusted this product in each test and modified equation (8) as follows \( \text{[12]} \):

\[
\phi \frac{d_1}{v_1^{2/3}} = \phi \frac{d_0}{v_0^{2/3}}
\]

(14)

The pilot-plant scale of fluidized bed drier was developed by scaling up from its laboratory scale and composed of a cylindrical chamber with a 600 mm ID and 2000 mm height. It provided the ratio of height to the bed dryer diameter at 3.33. The ratio of height to the bed dryer diameter in the range of 4.4–7.3. However, coarser material (\( d > 0.25 \) mm) was recommended for using a teeter bed with shallow beds (height up to 1 m). This bed uses low multiples of the minimum fluidization velocity \( \text{[18]} \).

3.3. Comparing the drying rate on the laboratory and pilot-plant scale of fluidized bed dryer equipped by zeolite adsorption

In order to fasten the drying time during the laboratory-scale running parameter, the zeolite-tea leaves ratio of 75:25 was applied. However, they were also mentioned that this condition incurred the extra costs and energy for zeolite regeneration. Applying a high ratio of zeolite on the pilot-plant scale will not match the economical reason for the high capacity of dried green tea production. Furthermore, this study modified the ratio of zeolite-tea leaves at 25:75 when trialed at the pilot-plant scale in order to decrease the long-time of payback period without ignoring the product quality demands.

The moisture content and the drying rate of tea leaves in the laboratory and pilot-plant scale of fluidized bed dryer are shown in Figures 4a and 4b, respectively. The initial moisture content at the pilot-plant scale (257%, dB) was higher than the initial moisture content of tea leaves on the laboratory scale (211%, dB). It could be possible since the pilot-plant scale has 21 times larger capacity than the laboratory scale. Figure 4a shows the faster-decreasing moisture content in the first 30 minutes on the laboratory scale than the pilot-plant scale, then following by a relatively stable decrease of moisture content on a longer time.

Figure 4b showed that A to B is the constant-rate period, B to C is the first falling rate period and C to D is the second falling rate period of drying. At the initial stages of drying, the tea leaves were containing high moisture content (211 and 257 % (dB) for laboratory and pilot plant scale, respectively) and covering the entire surface of the material as a continuous film. During the constant-rate period, the rate of drying on the drying operations was mostly constant due to increasing the temperature of the material up to reach the wet-bulb temperature of the air. Therefore, mainly superficial water can be removed and independent of the moisture content. The range of the constant drying rate highly depends on the area exposed on the drying process, the difference in temperature between the air stream and the wet surface of the materials, and also the air velocity. At 0.50 m/s of airflow rate, the pilot-plant scale required a longer time (40 minutes) on the first falling rate period, since they need a longer time to reach the bulb temperature. However, the second falling rate on the pilot-plant scale gave a faster drying rate period than the laboratory scale at the same airflow rate. The rate of moisture removal after 75 min was high up to a moisture content of 5.2 % (dB) and 2.4% (dB) for laboratory scale and pilot plant scale, respectively. It indicates that the higher capacity of the apparatus required a longer time of the air temperature to perfectly covered the surface of wet temperature. After this condition approached, the
transfer of the moisture content from wet solid to the drying chamber can occur easily. The prediction of the drying rate is important for designing a drying system.

Figure 4. Effect of airflow rate on the moisture content (a) and drying rate (b) at 0.50 m/s of airflow rate and 70 °C of temperature with zeolite-tea leave ratio of 25:75. ■, laboratory scale; ▲, pilot plant scale. A–B: constant rate period; B–C: first falling rate period; C–D: second falling rate period; †: laboratory-scale; ‡: pilot plant scale.

4. Conclusion
The pilot-plant scale required longer time (40 minutes) on the first falling rate period than laboratory scale (30 minutes), although it gave faster second falling rate period at the same airflow rate (0.50 m/s), temperature (70° C) and zeolite-tea leave ratio (25:75).

Acknowledgment
This work was supported by the Grant-in-Aid for Scientific Research (No. 141-68/UN7.5.1/PG/2015) form the Ministry of Education and Culture of Indonesia.

References
[1] Temple S J, Boxtel A J B V 2000 J Agr Eng Res. 77(4) 401–407
[2] Gazor H R, Mohsenimanesh A 2010 Czech J. Food Sci. 28(6) 531–537
[3] Dutta P P, Baruah D C 2014 Appl Therm Eng. 63(2) 495 – 502
[4] Handayani S U, Yulianto M E, Senen, Paramita V 2015 Res J Appl Sci Eng Technol. 9(12) 1128–1131
[5] Atuonwu J C, Stratem G V, Deventer H C V, Boxtel A J B V 2011 Chem Eng Trans. 25 111–116
[6] Kumar P S, Sagar V R 2014 J Food Sci Technol. 51(8) 1540–1546
[7] Manikantan M R, Barnwal P, Goyal R K 2014 J Food Sci Technol. 51(4) 813–819
[8] Ishino N, Yanase E, Nakatsuka S 2010 Biosci Biotechnol Biochem. 74(4) 875–877
[9] Yoshida Y, Kiso M, Goto T 1999 Food Chem. 67 429–433
[10] Wang H, Helliwell K 2000 Food Chem. 70(3) 337–344
[11] Pusapati R T, Rao T V 2014 Fluidized bed processing: A review IJRPB 2(4) 1360 –1365
[12] Chang H, Louge M 1991 Hydrodynamic scale-up of circulating fluidized beds, Eleventh International Conference on Fluidized Bed Combustion ASME Montréal Available at http://grainflowresearch.mae.cornell.edu/circulating_fluidization/papers/montreal.pdf (accessed March 23, 2017)
[13] Montgomery D C 1997 Design and Analysis of Experiments 4th edition John Wiley & Sons New York p. 98–173
[14] Box G E P, Hunter W G, Hunter J S 1978 Statistics for experimenters: an introduction to design, data analysis and model building, John Wiley & Sons New York p. 10-35
[15] Box G E P, Draper N R 1987 Empirical model-building and response surfaces John Wiley & Sons New York p. 253–280
[16] Suzuki M, Sano N, Yoshida R, Degawa M, Miyase T, Maeda-Yamamoto M 2003 J Agric Food Chem. 51(2) 510–514
[17] Song B J, Manganais C, Ferruzzi M G 2015 Food Chem. 173 305–312
[18] Levenspiel O 1969 Fluidization Engineering, John Wiley & Sons New York p. 14–81
[19] Akhtaruzzaman M, Ali M R, Rahman M M, Ahamed M S 2013 J Bangladesh Agril Univ. 11(1) 153–158