Mechanism of Microwave Assisted Hydrodistillation Studied Through Heat Analysis

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Abstract—In this study, the efficiency of heating mechanism of microwave assisted hydrodistillation (MAHD) in the extraction of cinnamon bark oil was investigated. The optimum conditions used to analyse the heating performance were 8:1 ratio of water to cinnamon bark powder and fixed 250 W of irradiation power. The increasing temperature in the cinnamon bark matrix was dependent on the solvent, physical, dielectric and heating properties of the cinnamon matrix. Due to the high dielectric properties of water it accelerated the process of extraction. However, after the cinnamon matrix reached the boiling point, the density of solvent decreased which led to decrease in the dielectric properties. Volume rate of heat generation and penetration depth of microwave was also evaluated. The rate of volume heat generation reduced when the exposure time increase which is related to the reducing dielectric properties of cinnamon matrix. The penetration depth was calculated to support the data of dielectric properties. This study therefore produced an in depth justification necessary to understand the heating mechanism of MAHD in extraction of cinnamon bark essential oil.

Keywords—MAHD; dielectric properties; volume rate of heat generation; penetration depth

1. INTRODUCTION

The novelty of microwave assisted hydrodistillation (MAHD) extraction technique has been explored in many fields, especially in researches involving extraction of medicinal plants and herbs. This is often implored either to obtain similar or most often better quality of essential oil yield compared to those obtained using conventional methods, at less energy consumption as well as rapid extraction time. In order to maximize these advantages of microwave extraction, study on the influence of dielectric properties of microwave extraction is very important. The capability of a specific material or solvent to absorb microwave energy and dissipate heat energy depends on the dielectric constant (ε') and dielectric loss (ε'') of the material. Dielectric constant is the ability of a plant sample matrix to absorb irradiation energy whereas dielectric loss is conversion of irradiation energy into thermal energy [1]. Dielectric properties possessed by solvents are very essential in microwave heating performance. Solvents which exhibit high dielectric properties are capable of absorbing more microwave energy and convert it into thermal energy which invariably could enhance the heating up of the plant matrix. On the other hand, solvents with low dielectric properties may not be able to support the effective heating of the plant matrix [2]. Based on this, in MAHD process, the solvent which has high dielectric properties would absorb more microwave energy, leading to the production of heat inside the plant material which is thereafter transferred outward. This process is known as volumetric heating. On the contrary, conventional method heat surfaces first before the inner part of the plant material gets heated. Therefore, the plant material release the essential oil effectively and rapidly through good heat distribution of microwave heating [3]. The major focus of microwave extraction research is to improve the techniques of extraction towards more feasible, efficient and high productivity. Therefore, temperature analysis study is required to be carried out in order to understand the principle of heating using microwave. In this study, the efficiency of microwave was investigated on the parameters of volume rate of heat generation and penetration depth of cinnamon bark matrix. Besides that, the efficiency of water as a solvent in the extraction of cinnamon bark oil through MAHD was investigated by analysing the dielectric properties.

2. MATERIALS AND METHODS

A. Modified Domestic Microwave Oven

Fig. 1 depicts the pictorial diagram of a modified domestic microwave oven used for the extraction of cinnamon bark oil. For this extraction purpose, microwave oven (model- Samsung MW71E), was used with the following specifications: power consumption capacity (1150 W), maximum microwave power output (800 W), power source (250v, 50Hz), frequency of microwave irradiation (2450 MHz) and dimension of microwave cavity (306 x 211 x 320 mm). For the temperature analysis of cinnamon matrix (water + cinnamon powder), the pre-soaked cinnamon bark powder was transferred into 1 litre round bottom flask which was set up inside the cavity of the modified domestic microwave oven prior to the experiment. Pico log TC-08-USB model data collector was used in this experiment to collect the temperature of the cinnamon bark matrix until it reached the boiling point. Three temperature sensors of the Pico Log device was immersed inside the cinnamon bark according to the following position which is bottom< middle< top as illustrated in the Fig. 1. Temperature per second was recorded for the minimum (200 W) and maximum (250 W) of microwave power level. The temperature selection as well as amount of solvent used for the study were based on previous study on extraction of cinnamon bark oil through MAHD [4].

B. Density Measurement

The density of cinnamon bark matrix at water to cinnamon bark powder ratio of 8:1 was measured by using a gas pycnometer (model Micromeritics Accupyc II 1340). Initially, the equipment was calibrated by inserting the sample cup with 1 cm^2 standard sphere. After calibration, test samples were filled to about 2/3 full of test sample (cinnamon bark matrix at ratio of 8:1 of water-to-cinnamon bark powder) and the sample mass was recorded. After recording the mass, micromeritics software was used to analyse the density of the sample. The mean values of the density of cinnamon bark matrix for five replicates was recorded.

C. Bomb Calorimeter

The specific heat capacity of cinnamon bark powder was measured using bomb calorimeter (model Parr 1341 plain jacket calorimeter). Prior to the experiment, 1g of cinnamon bark powder was weighed and placed in the combustion cup and 10 cm length of titanium ignition wire was connected through the hole of oxygen combustion bomb (model Parr 1108). After this the bomb head was placed into the bomb and then filled with oxygen from a standard cylinder less than 20 atm of pressure. The combustion bomb was placed inside the water-filled bucket of bomb calorimeter and the lid was closed. A belt was used to connect the stirrer and the motor. When the set up was completed, the ignition power was switched on as well as the motor. Temperature values were recorded from the initial temperature until 3 constant temperatures was attained. The specific heat capacity was calculated by using the following equation:

$$C_p = \frac{q}{m \, x \, \Delta T} \tag{1}$$

wwhere,
C_p : specific heat capacity at constant pressure of cinnamon powder in cal/ g. ^{0}C
q : energy used to burned the cinnamon powder in cal
m : mass of cinnamon powder in g
ΔT : temperature increase in ^{0}C

D. Volume Rate of Heat Generation

Volume rate of heat generation is a critical term for analysing the efficiency of microwave heating properties. This term interrelates temperature rise and dielectric properties of cinnamon matrix, which also will be applied in energy balance equation. Volume rate of heat generation of the cinnamon matrix was calculated by using the following formulae:

$$Q_{mw} = \rho_{mix} \, x \, C_{p\,mix} \, x \, \frac{dT}{dt} \tag{2}$$

where,
Q_{MW} : volume rate of heat generation, cal/ cm^3.sec
ρ_{mix} : density of cinnamon mixture, g/cm^3
$C_{p\,mix}$: heat capacity of cinnamon matrix, cal/ ^{0}C.g
$\frac{dT}{dt}$: rate of temperature increase, ^{0}C/ sec

In applying equation (2), the density (ρ) and specific heat capacity (C_p) of the cinnamon mixture was calculated by the following simple mixing rules:

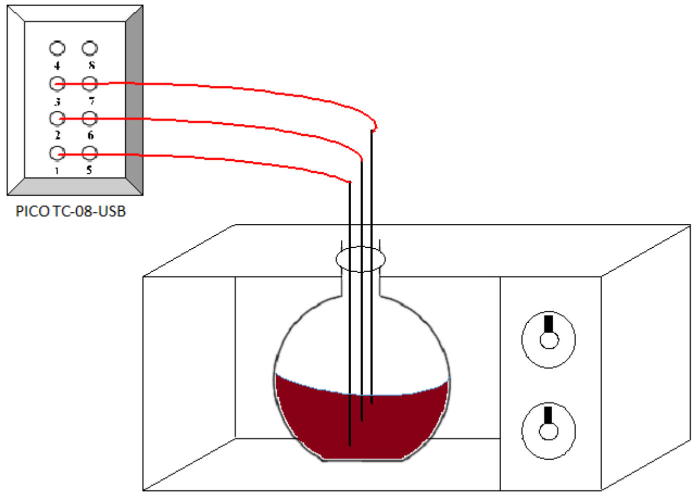

Figure 1: Modified domestic microwave oven with Pico TC-08-USB data collector

$$P_{mix} = \rho_W \emptyset + \rho_s\,(1 - \emptyset) \tag{3}$$

$$C_{p\,mix} = C_{p,W} \emptyset + C_{p,s}\,(1 - \emptyset) \tag{4}$$

where,
Φ : Volume fraction

E. Dielectric Properties

Generally, the rate of heat generation is influenced by the dielectric properties. Three dielectric properties that are predominantly used in microwave heating are tangent loss, dielectric constant and dielectric loss. Tangent loss can be measured by dividing dielectric loss by dielectric constant. The dielectric constant and dielectric loss of water was calculated by using the following model [5] illustrated in equation 5 and 6 respectively. The value of dielectric constant and dielectric loss of cinnamon bark powder was reported as 2.8 and 0.38 respectively by [6] at 2450 MHz. Simple mixing rule was applied to calculate the total dielectric properties.

$$\varepsilon'_w = 85.215 - 0.33583T \tag{5}$$

$$\varepsilon''_w = 320.685T^{-1.0268} \tag{6}$$

where,
ε'_w : dielectric constant of water
ε''_w : dielectric loss of water
T : temperature (^{0}C)

F. Penetration Depth

Penetration depth, D_p at a particular frequency is influenced by the dielectric properties of material matrix in an inverse manner. The depth of penetration is defined as the depth into the cinnamon matrix at which the power density has decrease to 1/e (about 37%) of its surface value. The following equation shows the penetration depth formulae used in this experiment:

$$D_p = \frac{\lambda_o\, x\, \sqrt{\varepsilon'}}{2\pi\varepsilon''} \tag{7}$$

3. RESULTS AND DISCUSSION

A. Temperature Analysis

Fig. 2 illustrates the temperature profile of a mixture of water and cinnamon powder at the ratio of 8:1 and microwave power level of 250 W in relation to the increasing exposure time. From this graph, an exponential trend can be seen from 0 sec to 120 sec. The sharp exponential trend obtained herein can be attributed to properties of the solvent (water) which was used in the extraction process. Water is a polar solvent with great capability to absorb large microwave energy. Reason for this is that water has high dielectric constant [2, 7] which means water has the high capability to store electrical energy in an electric field (microwave cavity). This also indicates water has high dipolar moment meaning the polarity of water bond (H-O-H) is strong

[8]. As a consequences of that, water absorb more microwave energy and dissipate the microwave energy into heat energy. This heat energy may increase the temperature of the cinnamon matrix inside the microwave cavity. In addition, the temperature of the cinnamon mixture further increase with respect to the increasing irradiation time in particular when the irradiation time increase from 180 sec to 660 sec. However, upon increasing the irradiation time of the microwave beyond 720 sec, the temperature seemed to reach steady state. This is because at this time, the cinnamon mixture had reached its boiling point (100^0C).

Fig. 3 illustrates the temperature increase with respect to increasing irradiation time at a fixed microwave power of 250 W. The rate of temperature increase was calculated by dividing the temperature increase with irradiation period. As can be seen from the graph, the temperature increase becomes gradually reducing as the irradiation power increases; the only exception being the initial increase after just about 60 secs. This initial increase can be attributed to the quick heating system of the microwave due to the high polarity of water which might have caused it absorb microwave energy at a fast rate. However, the rate of temperature increase start to decline after 60 sec, due to the decreasing of dielectric properties of water [7, 9], which will be discussed extensively in the next section under volumetric rate of heat generation. Besides that, there might be other possible factors which may influence the decrease in the rates of temperature increase including physical properties of water, such as the boiling point, density and heat capacity [2]. For instance, when the boiling point of the cinnamon matrix increase, the mass of the cinnamon matrix began to decrease as some portion of the water start to evaporate from the cinnamon matrix during extraction process. Hence, the density and heat capacity of the cinnamon matrix also may decrease as the mass decrease [10]. The level off values of these parameters might negatively influence the rate of temperature increase in relation with extended irradiation time.

In another vein, various microwave power levels could also influence the ability of the water to absorb microwave energy. The ability of the materials to absorb microwave energy is closely associated to the pulse cycle. For instance, at higher microwave power level (250 W), the system reveals longer pulse time which resulted into larger rate of temperature increase (0.2807 ^{0}C/sec) at initial irradiation time of 60 sec as depicted in Fig. 3. However, at lower microwave power level (200 W), the pulse time is short and low rate of temperature increase (0.1050 ^{0}C/sec) compared to 250 W power level was recorded at the same initial irradiation time of 60 sec as illustrated in Fig. 4. Hence it takes longer time to raise the temperature for the mixture to reach its boiling point due to low percentage of microwave absorbance by water at 200 W, as reported elsewhere [2]. Similar observation were also reported by [11] that longer pulse time at higher microwave power level had strongly influence the rise in rate of temperature and microwave power absorbance. Likewise, this current research also follows the same observation like [2, 7, 11] where at higher microwave power level longer pulse time and higher rate of temperature suitable to absorb more microwave energy was obtained. This findings demonstrated that the 250 W is the best microwave power in extraction of cinnamon bark oil at 8:1 of water to cinnamon powder ratio.

B. Volumetric Rate of Heat Generation

In this research, the volume rate of heat generation was used to establish energy balance equation for microwave irradiation system. The volume rate of heat generation is directly proportional to density, heat capacity and rate of temperature increase of cinnamon mixture. Fig. 5 illustrates the volume rate of heat generation of cinnamon matrix as a function of irradiation time at 250 W of microwave power level and 8:1 water-to-cinnamon powder ratio. This graph shows that the relation of volume rate of heat generation is inversely proportional to exposure time after 60 sec. At the initial 60 sec of irradiation period, the volume rate of heat generation reached highest point which is 0.0200 kcal/ cm^3.s rapidly. However, as the irradiation time was prolong, the volume rate of heat generation began to decline and reach plateau state at 0.0006 kcal/ cm^3.s. Similar observation has been reported by [2, 7] that at the higher microwave level, the volume rate of heat generation decrease in relation with increasing microwave radiation time after 60 sec. Reason for that could be because water contain non-symmetric molecules that could induce temporary dipole movement inside the electromagnetic field and increase the volume rate of heat generation rapidly. However, the friction between a dipole molecule and surrounding molecules produce heat with the aid of external electric field. Therefore, when the temperature increase as a result of prolonged irradiation time, the dipole situated within the electric field rotates to align itself in the changing microwave field and thus the dielectric properties of water decline. As a result, the capability of water to absorb microwave energy also reduces. According to [12], the dielectric properties of a polar solvent is fully dependent on the movement of dipole molecules within the structure. On the other hand, heating temperature determines the ability of a solvent to absorb sufficient microwave energy and dissipate the microwave energy into heat energy and influence along the dielectric properties of the solvent [13-15].

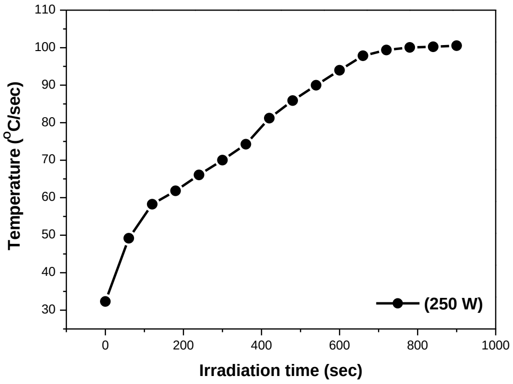

Figure 2: Temperature profile of cinnamon bark matrix at fixed irradiation power of 250 W and water to raw material ratio of 8:1

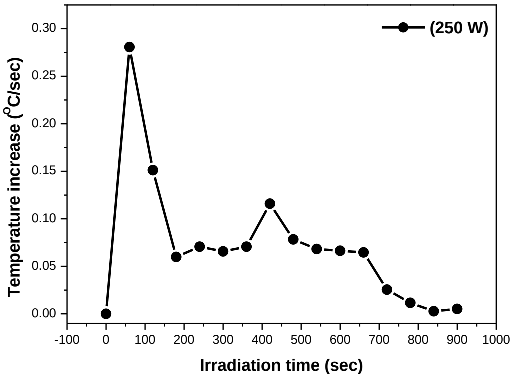

Figure 3: Temperature increase of cinnamon bark matrix at fixed irradiation power of 250 W and water to raw material ratio of 8:1 for various irradiation time

C. Dielectric Properties

Since water has high dipolar moment, it may absorb quickly and strongly the microwave energy and convert it into thermal energy, which may help to heat the test sample efficiently. Besides strongly absorbing the microwave irradiation energy, the particular solvent is also required to transform the absorb microwave energy into heat energy. The ability of the water to absorb the microwave energy and turn it into heat energy at a given temperature and frequency is known as loss tangent (tan δ). Loss tangent is expressed by dividing dielectric loss with dielectric constant (ε''/ ε'). Dielectric constant (ε') describes the capability of the cinnamon matrix molecules to be polarized by the applied electric field [16] while dielectric loss or also known as loss factor (ε'') is the magnitude value of the absorbed microwave irradiation energy converted into thermal energy [3]. Fig. 6 and 7 depicts the dielectric constant and dielectric loss of cinnamon matrix as a function of increasing temperature at 250 W of microwave power and 8:1 of water-to-raw material ratio respectively. As can be seen from both graphs, there is almost similar trends obtained where dielectric properties including dielectric constant and dielectric loss decrease with respect to temperature rise. Similar observation has been reported by [10] where, as the temperature increase the dielectric properties of water and rosemary essential oil were decline. The dielectric properties of cinnamon matrix are strongly influence by plant moisture content and density. According to the research of [10], usually the radiated microwave energy is strongly absorbed at the higher moisture content location due to the high percentage of water present which has high dielectric properties. This finding also revealed that high dielectric properties was exhibited by test samples that contain high moisture content at the constant density. Whereas, after the water loss during the evaporation process before extraction (low moisture content; mass of plant material reduce; density reduce as well) from the test samples at high temperature shows lower value of dielectric properties. As for current studies, the moisture content of the cinnamon bark is approximately 9.2%. Therefore, the dielectric properties which are dielectric constant and dielectric loss were high at the initial stage of extraction rate. However, as the temperature rise, the water contained inside the plant material start to evaporate and lead to water loss. Hence, the mass of the cinnamon matrix solution decrease by reducing the moisture content inside the plant material and therefore the density also decreases. As a result, the dielectric properties of cinnamon matrix decrease as the temperature increase.

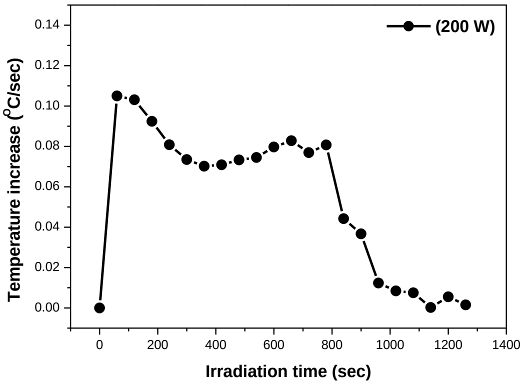

Figure 4: Temperature increase of cinnamon bark matrix at fixed irradiation power of 200 W and water to raw material ratio of 8:1 for various irradiation time

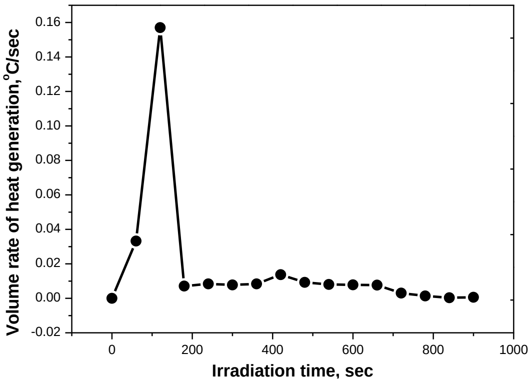

Figure 5: Volume rate of heat generation of cinnamon matrix extracted through MAHD at water: cinnamon powder ratio of 8:1 and microwave power level of 250 W for various microwave irradiation time

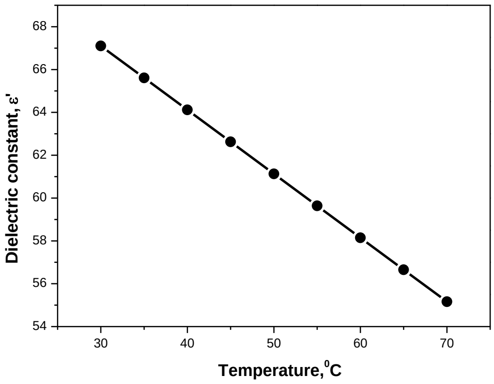

Figure 6: Dielectric constant (ε') at various temperature of cinnamon bark matrix extracted through MAHD at 250 W of irradiation power and 8:1 ratio of water to cinnamon bark powder

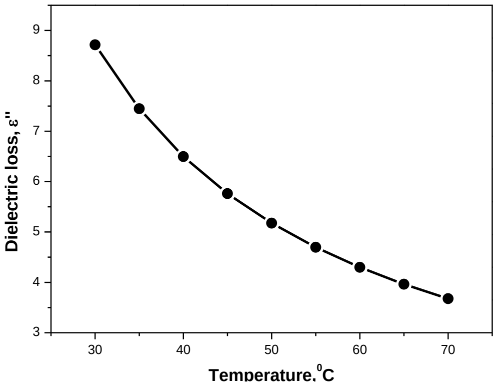

Figure 7: Dielectric loss (ε'') at different temperature of cinnamon bark matrix extracted through MAHD at 250 W of irradiation power and 8:1 ratio of water to cinnamon bark powder

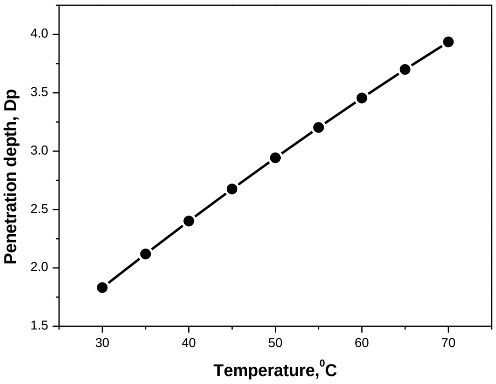

Figure 8: Penetration depth, Dp, at various temperature of cinnamon bark matrix extracted through MAHD at 250 W of irradiation power and 8:1 ratio of water to cinnamon bark powder

D. Penetration Depth

When microwave radiation energy pass through or penetrates the cinnamon matrix solution, the energy absorbed by the cinnamon matrix at a rate is associate with the loss tangent. Penetration is unlimited to the materials which are transparent to radiation energy while there is no penetration occurrence for the materials which are opaque or reflective like metals. The penetration factor of irradiation energy is inversely proportional to the loss tangent of cinnamon matrix at a provided frequency. Depth of penetration, (D_P) is the depth that has reduce about 37% of the initial value of power flux at its surface. Penetration depth of irradiation energy is dependent on the dielectric properties of plant matrix and the frequency. Fig. 8 shows the effects of penetration depth as a function of increasing temperature. This experiment was conducted at the optimum parameters of microwave extraction which are 250 W of microwave power and 8:1 ratio of water-to-cinnamon powder. The depth of penetration values were calculated to support the previous section findings of dielectric properties. Reason for that is because results of penetration depth gives the most convenient and efficient way to explain the dielectric constant and dielectric loss of a plant material and properties of the microwave heating. Besides that, the penetration depth data has also been used to compare the ability of the plant sample to absorb radiation energy. In this study, the graph depicted a linear relationship between penetration depth and irradiation time. Noteworthy, the results obtained for penetration depth is contrary to what was obtained from the previous section for dielectric properties. This is in agreement with the report stating that penetration depth is inversely proportional to the dielectric loss [2]. A large value of penetration depth indicates that the plant material absorb very low percentage of microwave energy while for the case of short penetration depth shows surface heating is predominant[15].

4. CONCLUSION

In this study, the performance of microwave heating at optimum conditions successfully provided new option in extracting cinnamon bark oil from *Cinnamomum Cassia* through microwave assisted hydrodistillation method. This method can be also be applicable to extraction of plants and herbs materials that consist essential oil. Here, the higher temperature gradients present at 250 W of microwave power level prove that more microwave energy was absorbed at high irradiation power which accelerated the extraction of cinnamon bark oil. However, the rate of temperature increase and rate of volume heat generation of cinnamon bark matrix declined as the irradiation period was prolonged. This was observed as the dielectric properties of cinnamon bark matrix reduced as a function of increasing exposure time leading to the decrease in the rate of temperature increase and volume rate of heat generation. Penetration depth values were calculated to analyses the ability of cinnamon matrix to absorb microwave energy. The highest penetration value measured in this experiment was 3.94 and this value was high enough to absorb the sufficient amount of microwave energy which effectively enhanced the extraction process of the cinnamon matrix. Therefore, from this study, it can be concluded that microwave assisted hydrodistillation method is a versatile technique which may be explored for extraction processed. Moreover, with MAHD, there is the possibility for property-enhanced products compared to other conventional methods which are mainly based on convection and conduction technique.

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