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Research Article

Comparative Study of Maceration and Ultrasonic Techniques in Coffee Oil Extraction Based on Energy Evaluation and Mass Transfer Value

Studi Perbandingan Teknik Maserasi dan Ultrasonik pada Ekstraksi Minyak Kopi Berdasarkan Evaluasi Energi dan Nilai Transfer Massa

Mega Mustikaningrum[1*)], Mohamad Endy Yulianto[2)], Laras Prasakti[3,4)]
[1)]*Universitas Muhammadiyah Gresik, Chemical Engineering Department, Gresik, East Java, 61121, Indonesia*
[2)]*Universitas Diponegoro, Industrial Chemical Engineering Department, Semarang, Central Java, 50275, Indonesia*
[3)]*Universitas Gadjah Mada, Chemical Engineering Department, Sleman, Yogyakarta, 55281, Indonesia*
[4)]*RWTH Aachen University, IME Process Metallurgy & Metal Recycling, 52062, Aachen, Germany*

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*Corresponding Author.
Email: megamustikaningrum@umg.ac.id

Abstract

Coffee is a significant commodity trend, with numerous coffee shops rapidly established in Indonesia. The development of this industry has led to an increased discharge of coffee grounds into the environment. The discarded grounds contain oil which can be optimized using various methods and serve as a raw material for biodiesel. Therefore, this study aimed to compare the maceration and ultrasonic methods of extracting coffee grounds. The comparison focused on yield, mass transfer value (diffusivity), and extraction speed constant based on the proposed mathematical model. The results showed a yield of 12.1% and 16%, for the maceration and ultrasonic methods, respectively. Diffusivity value was registered at 9.99 x 10^{-11} m^2/min and 9.8 x 10^{-10} m^2/min, while extraction speed constant values were discovered to be 0.2 m/min and 1.798 m/min, respectively. Additionally, the energy evaluation of ultrasonic extraction produced a Gibbs energy value of -3765.72 Joules.

Keywords: coffee grounds; extraction; Gibbs energy; maceration; mass transfer; ultrasonics

Abstrak

Kopi menjadi trend komoditi terbaru saat ini dengan maraknya coffee shop yang didirikan di Indonesia. Dengan berkembangnya industri ini, buangan ampas kopi ke lingkungan semakin bertambah. Buangan ampas kopi memiliki kandungan minyak kopi yang dapat dimanfaatkan salah satunya menjadi bahan baku biodiesel. Berbagai metode telah dilakukan untuk melakukan optimalisasi terhadap minyak kopi yang dihasilkan. Pada penelitian ini bertujuan untuk membandingkan dua metode ekstraksi ampas kopi yaitu metode maserasi dan ultrasonik. Perbandingan dilihat dari yield yang dihasilkan, nilai transfer massa dan konstanta kecepatan ekstraksi berdasarkan pengajuan model matematika. Yield minyak kopi hasil maserasi sebesar 12,1 % dan 16 % untuk metode ultrasonik. Nilai difusivitas untuk ekstraksi maserasi sebesar 9,99 x 10^{-10} m^2/min dan untuk ultrasonik sebesar 9.8 x 10^{-09} m^2/min. Untuk nilai konstanta kecepatan esktraksi untuk metode maserasi dan ultrasonik didapatkan sebesar 17,98 dan 200 m/min. Berdasarkan evaluasi energi pada ekstraksi ultrasonik didapatkan nilai energi Gibbs sebesar -3765,72 Joules.

***Kata kunci**: ampas kopi; ekstraksi; energi Gibbs; maserasi; ultrasonik; transfer massa*

1. Introduction

Coffee is a popular beverage that has become a trend and a staple lifestyle of urban communities. According to the Indonesian Central Statistics Agency, Indonesia is the third largest producer of this commodity worldwide [1]. Data from the Association of Indonesian Coffee Exporters (AICE) recorded a significant increase in coffee consumption across the country [2], reflecting the abundant availability of grounds. The increase in consumption has reached an average of 7% per year and 5 million 60-kilogram bags in 2020 - 2021. This led to the generation of substantial waste disposed into the environment.

On average, a ton of fresh coffee beans produces approximately 650 kg of grounds. The instant coffee industry accounts for approximately 50% of global ground production, leading to an annual quantity of coffee grounds of estimated 6 million tons [3]. This suggested that the availability of coffee grounds is quite abundant. Depending on the variety, the organic residue that comes from coffee grounds after the brewing process, commonly referred to as spent coffee grounds (SCG) has an oil content ranging from 10-15% by weight (%wt) [4]. Grounds discarded into the environment produce methane gas, which significantly contributes to climate change [5],[6].

Coffee oil presents a viable alternative as a raw material for biodiesel production due to the decreasing availability of fuel [7]. Every year, approximately 8 million tons of coffee are produced globally, and an estimated 1.3 billion L of biodiesel is derived from the oil, leading to increased fuel supply [8]. Despite this potential, Indonesia has yet to fully develop the benefits of coffee oil [9].

Various methods have been used to isolate coffee oil from grounds, including maceration [10][11], soxhletation method [12][13], supercritical liquid extraction [14], microwave-assisted extraction [15][16], and more recently ultrasonic extraction [17][18]. The diffusivity value of coffee oil in the maceration and ultrasonic methods, influenced by temperature and activation energy, was compared. The comparison included calculating the diffusivity of oil extraction under varying temperatures for both methods. This diffusivity value represents the rate of mass transfer of solute from high concentration to low concentration [19].

Studies on the development of a mathematical model for coffee oil extraction are limited. Therefore, data on kinetic and thermodynamic parameters are limited. This study presents kinetic and thermodynamic modeling to determine the constant value of extraction speed (kd), diffusivity (D), extraction equilibrium constant (K), and Gibbs energy. It is important to consider mathematical values such as diffusivity and rate constant which influence the transition rate of coffee oil from solid to liquid. The generated values can be utilized to conduct simulations, eliminating the need for additional data.

This study evaluates the energy feasibility of the extraction processes by analyzing the amount of entropy produced. The analysis showed whether the system occurs spontaneously or certain energy is required to facilitate the extraction process.

2. Research Methods

2.1 Tools and Materials

A complete series of maceration extractor equipment, comprised of three-neck flasks, a return cooler, thermometer, magnetic stirrer, hose, and pump, was

assembled as shown in Figure 1a. The sonicator series (110V/220V Digital Ultrasonic Cleaner 40Khz 50W Sonicator Washing Bath) includes a sonicator instrument and an Erlenmeyer for holding the sample, as detailed in Figure 1b. The raw materials used in this study were coffee grounds sourced from the Gresik area and 70% n-hexane technical solvent from Nirwana Surabaya.

2.2 Research Procedure

2.2.1 Maceration Method

A total of 100 grams of coffee grounds were dissolved in 600 mL of n-hexane solvent. Extraction was performed for 3 hours at a temperature setting of 70-80 ^{o}C using an extractor. An optimization stage included soaking the mixture for 24 hours in a closed glass cup for maceration. After extraction, the solvent was separated using distillation at 80-90 ^{o}C [20] until dripping stopped.

2.2.2 Ultrasonic Method

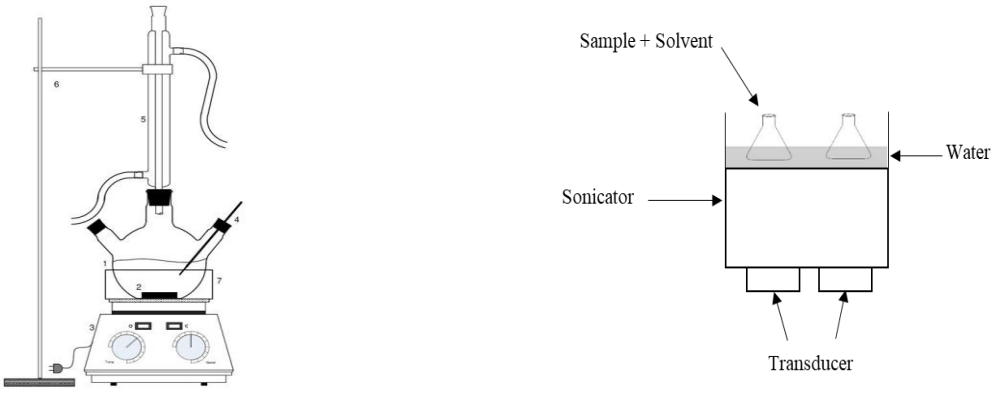
Approximately 10 grams of coffee grounds that had been prepared were dissolved in 200 mL of n-hexane solvent. Extraction was performed at 50 ^{o}C for 180 minutes in the sonicator. Following oil isolation, the distillation process was conducted to separate the n-hexane solvent from the coffee oil at 80-90 ^{0}C [21]. The distilled were weighed using balance analysis to determine the yield, calculated using equation (1).

$$\text{yield (\%)} = \frac{\text{weight coffee oil}}{\text{sample mass x coffee oil content in coffee grounds}} \text{x } 100\ \% \quad \textbf{..(1)}$$

2.2.3 Kinetic Data Retrieval

After extraction, samples were taken at a certain time range, and the data were analyzed and fitted to find the diffusivity parameter value. The equations and parameter values will be discussed further in a later section. The fitting will be done by minimizing the difference between the collected data and the calculation result.

In section 2.3, mathematical modelling produces partial second-order differential equations to represent the phenomena during the process. The differential equations will be discretized using numerical methods to produce ordinary differential equations (ODE). These equations are then able to be solved using the ode15s function in MatLab ver 2013b software. The results from the solved equations will then be fitted with experimental data using the lanolin function in MatLab to obtain unknown kinetic parameters.



(a) (b)

Figure 1. a) Series of maceration extractor tools, b) Series of ultrasonic extractor tools

2.3 Submission of Kinetic Models

Before developing the modeling, several assumptions were put forward, including that coffee ground particles had a spherical geometric shape with uniform porosity values. The calculations were only conducted on radial gradients without considering the axial direction. The modeling process also neglected temperature changes during extraction and filtration, assuming no variation in particle size and porosity [22]. Additionally, the diffusion equation was solved separately for each component of the bimodal particle size distribution, and the average particle size was used in the results. There were no particle swelling and changes in particle porosity over time.

The mass balance of coffee oil in the solvent is described in equation (2), while the mass balance in the coffee grounds solids is shown in equation (3). In both equations, the notation Ca* represents the concentration of coffee oil in the film layer, a value that cannot be precisely calculated. To address this, a partition equation was adopted as presented in equation (4). Where kd is the extraction speed constant (m/minute), m is the mass of coffee grounds (grams), V is the volume of n-hexane (L), Da is the diffusivity of coffee oil into the solvent (m^2/minute), Ca is the concentration of coffee oil in the solvent (mol/liter), Xa is the concentration of coffee oil in the solids (mol/gram), $\frac{\partial C_a}{\partial t}$ is the concentration of coffee oil in the solvent at any time (mol/liter.minute), $\frac{\partial X_a}{\partial t}$ is the concentration of coffee oil in solids at any time (mol/gram.minute), a is the surface area of coffee grounds calculated quantitatively based on the radius value (m^2), and K is the extraction equilibrium constant [23].

$$\frac{\partial C_a}{\partial t} = D_a \frac{\partial^2 C_a}{\partial r^2} - D_a \frac{2}{r}\frac{\partial C_a}{\partial r} - \frac{kd.a.m}{V}(C_a - C_a^*) \quad \text{.....(2)}$$

$$\frac{\partial X_a}{\partial t} = D_a \frac{\partial^2 X_a}{\partial r^2} - D_a \frac{2}{r}\frac{\partial X_a}{\partial r} + kd.a(C_a - C_a^*) \quad \text{.....(3)}$$

$$X_a = K\,C_a^* \quad \text{.....(4)}$$

To calculate the equation (2), (3), and (4), initial and boundary conditions needed to be established. The initial condition (IC) is expressed mathematically in equation (5). The proposed boundary condition (BC) is represented by equations (6) and (7), with Table 1 showing details of Xao.

At t = 0 → Xa = Xao and Ca=0 .....**(5)**

At r=0 → Xa=Xao; $\frac{\partial C_a}{dt} = 0$.....**(6)**

At r=r → $\frac{\partial X_a}{\partial t} = 0$; Ca=Cao .....**(7)**

Table 1. Calculated parameter values

No.	Variable	Value
1.	Mass (m)	
	Maceration	100 gram
	Ultrasonic	10 gram
2.	Volume (liter)	
	Maceration	100 mL
	Ultrasonic	200 mL
3.	Radius	0.000125 m
4.	Nr (number of iterations)	300
5.	Cao	0 mol/liter
6.	Xao	1889 mol/gram
7.	Surface area per coffee ground mass	0.01 m^2

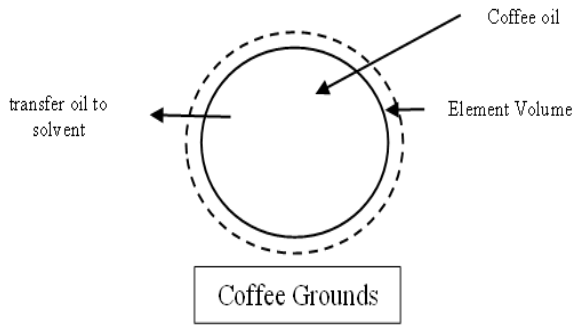


Figure 2. Illustration of the process of transferring oil to solvent

2.4 Energy Evaluation Model

To determine whether a process occurred spontaneously, the entropy factor and Gibbs free energy were considered. The enthalpy parameters (ΔH^o), Gibbs energy (ΔG^o) and entropy (ΔS^o) were calculated using the extraction kinetics equation, and tested at different temperatures.

Furthermore, thermodynamic studies were conducted at temperatures of 30, 40, and 50 °C. The equations for ΔG^o are presented in equation (8) and (9). The values of enthalpy and entropy was determined using linearity from equation (10), with ln K plotted on the y-axis and 1/T on the x-axis. The slope and intercept of the line provided the magnitude of the entropy and enthalpy values.

$$\Delta G^0 = -RT \ln K \quad \text{.....(8)}$$

$$\Delta G^0 = \Delta H^0 - T\,\Delta S^0 \quad \text{.....(9)}$$

$$\ln K = \frac{\Delta S}{R} - \frac{\Delta H}{RT} \quad \text{.....(10)}$$

3. Results and Discussion

3.1 Comparison of Coffee Oil Yields

The extraction process was used to isolate or separate the desired active compound from the raw material. It included stages such as (1) the solvent penetration into the solid matrix, (2) the solute dissolution in the solvent, and (3) the solute diffusion out of the solid matrix. In this case, the effectiveness of extraction depended on several factors such as temperature, the ratio of ingredients, the type of solvent, and the method applied [24].

This study was conducted using two different methods, namely maceration and ultrasonic. Figure 3 presents the differences in coffee oil yield after the extraction process. These differences arise from the distinct principles in the maceration and ultrasonics methods.

The maceration method was conducted at room temperature and it represented the simplest conventional method, where a liquid penetrated the cell walls of a plant and entered a cavity containing the active substance. This substance, present as a concentrated solution, diffused out due to the difference in concentration between the solution and the solute [25]. Based on this principle, maceration extraction usually requires an extended duration.

The yield of coffee oil from coffee grounds ranged from 5.7-16.0% [25]. Based on the results shown in Figure 3, the final yield obtained using the maceration method was 12. 42% wt, while the ultrasonic extraction produced 16% wt. The advantages of the ultrasonic method include faster processing time, and lower extraction power requirements, and the ability to perform extraction at lower temperatures [26]. The results are in line with the study conducted by Hibbert et al., where a coffee oil yield of 11.54% was obtained using the maceration method [15]. Meanwhile, the ultrasonic method produced 14.51% after 30 minutes [18].

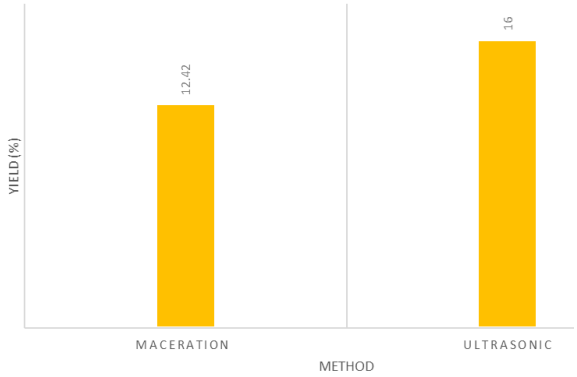


Figure 3. Differences in extraction results from maceration and ultrasonic methods

The ultrasonic method utilizes high-intensity sound waves to disrupt plant tissue through physical forces generated during the cavity mechanism, facilitating the release of coffee oil components into the solvent within a short time and enhancing diffusivity [27]. This phenomenon, known as erosion, causes the surface of the solid matrix to degrade, allowing the solvent to penetrate the plant tissue more effectively, leading to higher yields [28].

Based on mathematical calculations performed using MatLab, the decrease in coffee oil concentration at each time and position was detailed in Figures 4 and 5. These images were generated using the *imagesc* command in MatLab software, which visualized the relationship between the coffee grounds radius (x-axis), time (y-axis), and the concentration of coffee oil in the solid. The projection captured a radius range of 0 to 12 x 10^{-5} m. This visualization was intended to observe the distribution of oil reduction in solid coffee grounds.

Figure 4 explains the distribution of coffee oil at each position and time. In this case, the color gradation described the difference in concentration. Based on observation, towards the surface, the dense matrix of coffee oil was less than on the inside where the radius approached zero. This image was similar to the ultrasonic extraction phenomenon.

Based on the image, the distribution in the coffee matrix area using the ultrasonic method at a radius of 6 x10^{-5} m began to decrease significantly. In the maceration stage, subsidence was initiated at a radius of 8 x10^{-5} m. However, over the same distance range, the magnitude of the reduction in coffee oil concentration using the maceration method was not significant.

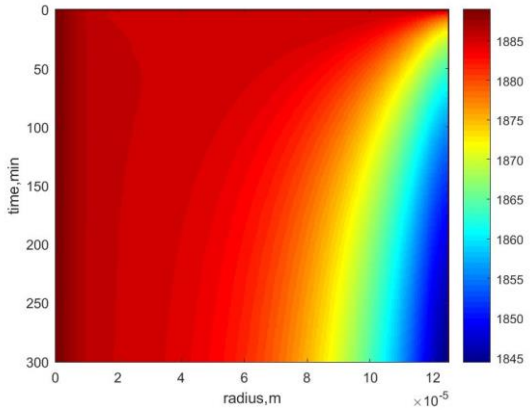


Figure 4. Distribution of coffee oil in a solid matrix using the maceration method

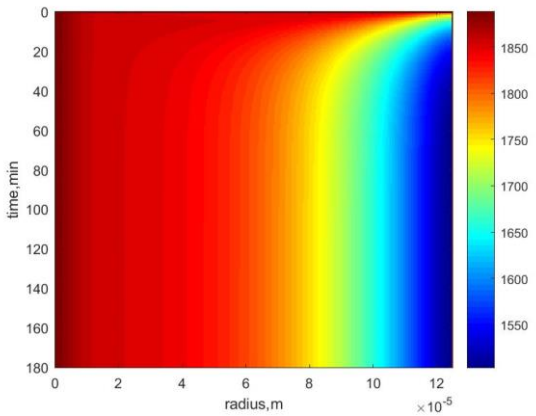


Figure 5. Distribution of coffee oil in a solid matrix using the ultrasonic method

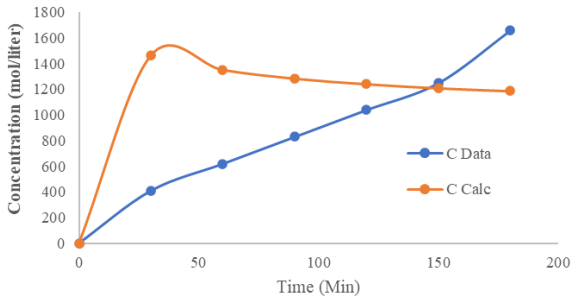


Figure 6. Comparison of research data vs calculations on maceration method

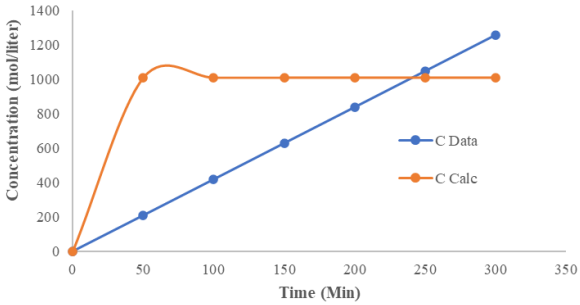


Figure 7. Comparison of research data vs calculations on ultrasonic method

The ultrasonic method can penetrate deeper pores. The result was obtained through data comparison and calculations. Figure 6 shows the comparison results of the maceration extraction method. Based on Figure 7, the final extraction result from the study data was 1659 mol/liter, while the calculated value was 1185.62 mol/liter. For the maceration method, the study data yielded 1259 mol/liter, compared to a calculated value of 1011.14 mol/liter.

Figures 6 and 7 show discrepancies between calculated and study data, which were relatively minor. In this case, the study had not identified an equilibrium point, resulting in a linear representation. This discrepancy explained the intersection of values in the image display, where the extraction process should ideally show a peak followed by a balanced line, as signified by the proposed mathematical model.

3.2 Differences in Diffusivity Results and Extraction Speed

In this study, the diffusivity of coffee oil into the solvent using hexane was quantitatively tested. It was important to acknowledge that the extraction time using the ultrasonic method was faster. The diffusivity value and extraction speed for each method were quantitatively evaluated. The results of the calculations were presented in Table 2, utilizing the parameter values listed in Table 1.

Table 2. Results of mathematical model calculations

Parameter	Extraction Method	
	Maceration	**Ultrasonic**
Diffusivity (m^2/min)	9.99×10^{-12}	9.8×10^{-11}
Extraction speed constant (m/min)	0.2	17.98

Diffusivity is the number of moles transferred per unit area, per unit time, and per unit concentration gradient. A molar flux in diffusion is comprised of two components attributed to mass movement and the relative speed of particles or molecules [29]. The increase in coffee oil diffusion observed using the ultrasonic method arises from the appearance of microacoustics during the extraction process. In this context, microacoustics refers to the effects of radiation pressure, gravity, cavitation, and acoustic pressure [30].

The acceleration of diffusivity and extraction speed was influenced by the presence of cavitation events produced by water bubbles. These bubbles are caused by alternating cycles of compression and rarefaction, which under certain conditions lead to cavitation, a phenomenon where bubbles burst. The phenomenon caused an increase in heat conditions of up to 5000 K and a high pressure of 1000 atm. This hot spot contributes to the acceleration of biochemical reactions, diffusion, and other phenomena [31].

The presence of cavitation caused several combinations of phenomena such as fragmentation, local erosion, pore formation, shear forces, increased absorption, and swelling index in the plant cellular matrix. This cavitation produces shock waves and accelerated collisions between particles that cause fragmentation of cellular structures. This rapid fragmentation dissolved bioactive components by decreasing particle size and increasing surface fluidity, thereby accelerating diffusivity within the solid matrix layer [32].

Ultrasonography, a phenomenon associated with ultrasonic extraction, enhances solvent absorption, thereby

increasing accessibility to bioactive compounds and subsequently raising diffusivity. This situation did not occur when using maceration extraction.

3.3 Ultrasonic Method Energy Evaluation

The purpose of energy evaluation is to determine the energy value produced by the ultrasonic method. This includes calculating the entropy and enthalpy values of the system. Based on Figure 8, the calculated enthalpy value was 28241.83 Joules, and the entropy was 99.09457 Joules/K. Using these values, the Gibbs energy was discovered to be -3765.72 Joules. The negative value signified that the extraction process was spontaneous and feasible [33].

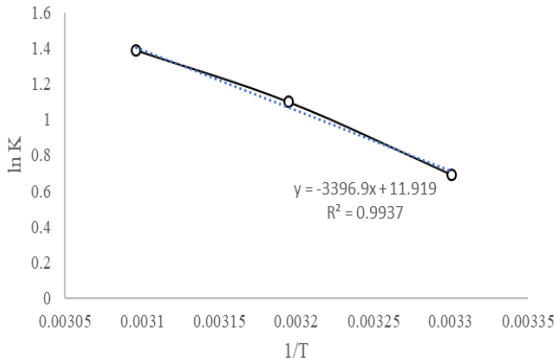


Figure 8. Plotting 1/T vs ln K to find energy parameter

4. Conclusion

In conclusion, the ultrasonic method outperformed maceration, yielding 16%, compared to 12%. The ultrasonic process also had a faster transfer value due to cavitation induced by sound waves, enhancing the diffusivity of coffee oil to the solvent. The Gibbs energy value showed that the process was spontaneous and efficient without requiring additional energy, making it feasible and energy-efficient.

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