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# Multi-objective optimization of pelletized coffee silver skin in flue gas torrefaction for producing premium solid fuel

Kanit Manatura [1,2,*], Supaporn Klinkesorn [1], Benjapon Chalermsinsuwan [3]

Namfon Samsalee [4], Sutthipoj Wongrerkdee [5] , Kitipong Jaojaruek [1], Adisak Pattiya [6] and Jun li [7]

1 Department of Mechanical Engineering, Faculty of Engineering at Kamphaeng Saen, Kasetsart University, Kamphaeng Saen campus, Nakhon Pathom 73140, Thailand
2 Energy and Environmental Engineering Center (EEEC) at Kamphaeng Saen, Faculty of Engineering at Kamphaeng Saen, Kasetsart University, Kamphaeng Saen campus, Nakhon Pathom 73140, Thailand
3 Department of Chemical Technology, Faculty of Science, Chulalongkorn University, Bangkok 10330 Thailand
4 Department of Applied Biology, Faculty of Sciences and Liberal Arts, Rajamangala University of Technology Isan, Nakhon Ratchasima 30000, Thailand
5 Department of Physical and Material Sciences, Faculty of Liberal Arts and Science, Kasetsart University Kamphaeng Saen Campus, Nakhon Pathom 73140 Thailand
6 Biomass Pyrolysis Frontier Research Group, Faculty of Engineering, Mahasarakham University, Kamriang, Kantharawichai, Maha Sarakham, 44150, Thailand
7 Department of Chemical & Process Engineering, University of Strathclyde, Glasgow, United Kingdom

*****Corresponding author

E-mail: kanit.ma@rmuti.ac.th

**Abstract**

Coffee silver skin, an organic residue from coffee production, demonstrates low solid fuel characteristics such as low bulk density and heating value, necessitating enhancements for solid fuel applications. Torrefaction in a flue gas environment (5% $O_2$, 15% $CO_2$, and a balance of $N_2$, v/v) is more energy-efficient than inert torrefaction, using recovered flue gas to improve fuel quality and process efficiency. Three input factors were assessed: temperature (200, 250, and 300 °C), residence time (30, 45, and 60 min), and gas media ($N_2$ and flue gas). Four performance metrics were evaluated: energy yield, upgrading energy index, specific energy consumption, and energy-mass co-benefit. Temperature significantly influenced most outcomes, except for energy-mass co-benefit, which was medium-dependent. Optimal torrefaction conditions achieving maximum energy yield (71.48%) and energy-mass co-benefit (5.30%) were identified at 200 °C for 30 min with flue gas. The torrefied material's properties include moisture content, volatile matter, fixed carbon, and ash content of 3.03%, 69.24%, 27.04%, and 1.01 %, respectively. Furthermore, the hydrophobicity of pelletized coffee silver skin notably increased under flue gas conditions, evident by a contact angle greater than 100°, indicating that flue gas torrefaction is a feasible approach for producing high-grade solid fuel.

**Keywords:** Flue gas torrefaction; Oxidative torrefaction; Optimization; Biochar

# 1. Introduction

The global "net zero" concept has recently gained significant attention worldwide due to the pressing issue of $CO_2$ emissions from fossil fuels, which are a major contributor to global warming. In 2022, global $CO_2$ emissions hit a record 36.1 gigatons (Gt), marking a substantial 63% increase from the 22.6 Gt reported in 1990 [1]. This escalating situation requires proactive interventions from governments and policymakers to achieve "carbon neutrality" and restrict the projected global temperature increase to 1.5°C by the mid-century [2]. Among the array of renewable energy sources, biomass stands out as a promising option for attaining carbon neutrality and promoting waste valorization [3,4]. This is attributed to its abundant availability, high yield, and carbon-neutral characteristics [5]. The estimated total biomass potential ranges from 200 to 700 exajoules (EJ) annually [6]. With the advent of carbon capture and storage (CCS) techniques, it is anticipated that the removal of $CO_2$ emissions could range from 151 to 1191 Gt globally by 2020, initiating in the early 21$^{st}$ century [2]. Nevertheless, the efficacy of utilizing biomass in industrial applications faces challenges, including high moisture content, low heating value, low density, and hydrophilicity [7]. Torrefaction, a thermal pretreatment technique, has emerged as a focal point of interest in both academic research and industrial sectors [8]. This method involves heating biomass at temperatures ranging from 200 to 300°C through a mild pyrolysis process [9], thereby enhancing solid fuel properties such as reduced moisture content, increased hydrophobicity, and elevated heating value [10].

Typically, this process occurs in a non-oxidative atmosphere, using $N_2$ and $CO_2$ as carrier gases [11]. Oxidative torrefaction is emerging as a cost-effective technique since it makes use of air, flue gas (from waste heat), or other gases with varying $O_2$ concentrations, eliminating the need for $N_2$ extraction [7]. A typical composition of flue gases includes $O_2$ (4–6% v/v), $CO_2$ (10–14% v/v), $H_2O$ (5–20% v/v), and the rest is $N_2$ [12]. The main factors influencing the torrefaction process are temperature, residence time, carrier gas, and heating rate, which impact mass yield, energy yield, and specific energy consumption [13,14]. This method could enhance the combustion-torrefaction system by reusing flue gas [15]. In oxidative torrefaction, both thermal degradation and exothermic reactions occur due to the presence of $O_2$, unlike inert torrefaction, which solely involves thermal degradation [16]. This leads to a faster reaction rate, reducing residence time, and improving energy efficiency [17]. The supplemental material summarizes literature reviews on oxidative torrefaction (Table 1S). Agricultural wastes, woody biomass, and microalgae are commonly used in oxidative and flue gas torrefaction studies. The impact of operational conditions, such as temperature, duration time, and carrier gas, on the properties of the torrefied product and production yield (solid yield, mass loss), as well as on torrefaction performance parameters (energy density, energy yield, energy-mass co-beneficial index), have been investigated [13]. This is followed by optimization analysis using response surface methodology (RSM) with a central composite design (CCD). Understanding the energetic performance characteristics, operational conditions, and optimization is crucial for practical applications of torrefaction. These factors offer insights into controlling process efficiency and producing high-quality torrefied products.

This study aims to analyze four key performance indicators for energetic torrefaction: energy yield (EY), upgrading energy index (UEI), specific energy consumption (SEC), and energy-mass co-benefit (EMCI). The objective is to identify the optimal conditions by considering the impact of operating temperature, residence time, and gas medium. Coffee is a globally popular beverage, with annual consumption growth averaging over 2% in the past decade. In Thailand, the period from 2016 to 2020 witnessed a similar trend in the coffee industry, with an average annual demand for coffee beans reaching around 79,000 tonnes [18]. The process of converting coffee cherries into coffee beans results in biowastes constituting 35-50 wt% [19], including by-products like husks, pulp, mucilage, parchment, silver skin, and spent coffee grounds [20,21]. Coffee silver skin, or coffee chaff, comprising approximately 4.3

wt% of the coffee cherry, is a by-product of the roasting process [22]. Despite its various uses, such as being a source of bioactive compounds, soil enhancer, fuel, fermentation substrate, and component in cement-based materials, coffee silver skin remains underutilized [23]. Utilizing biomass as biofuels for heating, cooling, and power applications can lead to reduced $CO_2$ emissions compared to fossil fuels, given their carbon-neutral properties. While $CO_2$ emissions from utilizing coffee silver skin as a biofuel in Thailand have not been documented, a study by Rahmah et al. pointed out that coffee pulp biomass could potentially cut $CO_2$ emissions by 49.69% to 72% [24]. Limited research exists concerning the statistical analysis of inert and flue gas torrefaction on torrefaction performance. This study hypothesizes that flue gas could improve coffee silver skin torrefaction performance, resulting in enhanced coffee silver skin properties. The study delves into performance indices such as EY, UEI, SEC, and EMCI to support the utilization of pelletized coffee silver skin as a solid biofuel for practical thermochemical applications. This approach aligns with the principles of a bio-circular-green (BCG) economy, offering promise for sustainable bioenergy advancement and supporting the Sustainable Development Goals (SDGs).

# 2. Materials and Methods

*2.1 Feedstocks and preparations*

Coffee silver skin (CS) was extracted from coffee beans at a community enterprise in Chiang Rai, Thailand. Measuring 5.37 wt% moisture content, CS consists of the inner shell of the coffee bean, depicted in supplementary Fig. 1S, illustrating its delicate nature. About 500 g of samples were ground into an 800 µm powder for 5 minutes with an SC-750T Ang grinder from China and sieved. The resulting powder was then shaped into pellets using a pellet machine, producing pellets with a diameter and height of 7 ± 0.1 mm and 25 ± 1 mm, respectively. Water was utilized as a binder during pelletization, mixed with the samples in a biomass-to-water ratio of 1:1.2 wt% to maximize pellet efficiency and prevent clogging in the pellet mill.

To maintain consistent moisture content and avoid any impact on physical and chemical properties, the pellets underwent a pre-torrefaction process. After drying in a hot air oven at 105 °C for 24 hours to eliminate surface moisture, the dried pellets were cooled in a controlled chamber to room temperature and stored in airtight plastic bags for further analysis. Proximate analysis was carried out using a NETZSCH TG209 thermal gravimetric analyzer from Germany to ascertain the moisture content (MC), volatile matter (VM), fixed carbon (FC), and ash (A) content of the raw and torrefied coffee silver skin pellets. The ultimate analysis involved determining the elemental compositions of carbon (C), hydrogen (H), and nitrogen (N) using a CHN elemental analyzer (Leco Truespec CHN-628, Netherlands) in compliance with ASTM D 5373–16 (2016) standards [25]. The oxygen (O) content was calculated based on the difference (O = 100 - C - H - N). Additionally, the heating value analysis was performed with an IKA C5000 bomb calorimeter following ASTM D 5865–13 (2013) guidelines [26].

### *2.2 Torrefaction System*

Approximately 100 g of pellets were introduced into the fixed-bed reactor equipped with a 2 kWe heater (Fig. 1). For flue gas torrefaction (FGT), a simulated flue gas containing 5% oxygen ($O_2$), 15% carbon dioxide ($CO_2$), and a balance of nitrogen ($N_2$) by volume is used as the medium. In contrast, $N_2$ alone was used in the inert torrefaction as a benchmark for performance comparison. Both gases were fed at a rate of 5 ml/min. The torrefaction temperature was maintained at 200, 250, and 300 °C for durations of 10, 30, and 50 min, respectively, with control managed by a PID controller. The heating rate was set at 20 °C/min by the electric heater located at the top of the reactor. A type K thermocouple placed at the reactor's center was utilized to measure the biomass temperature. It should be noted that heat loss between the reactor wall and the environment, as well as non-uniform temperature distribution, may impact biomass properties and torrefaction efficiency. The reactor was insulated with fiberglass, and positioning the sample vessel closer to the heater was aimed at mitigating non-uniformity effects. Post-process completion, the torrefied biomass was allowed to cool to ambient conditions before being collected. The torrefied coffee was stored in a sealed container and subsequently utilized for fuel and ANOVA analysis. The experiments were performed in duplicate, with mean values reported and further analyzed [26].

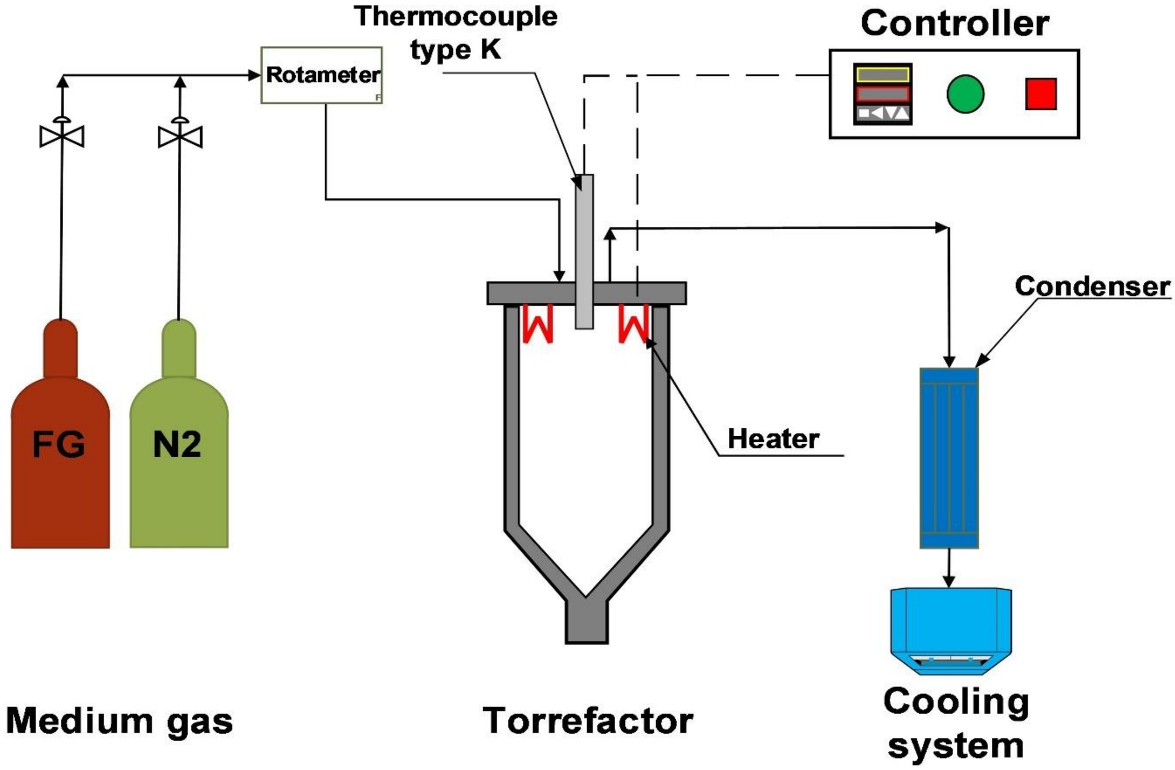



**Fig. 1 Biorender of the torrefaction System**

*2.3 Torrefaction Performances*

Four torrefaction indices were assessed: energy yield (EY), upgrading energy index (UEI), specific energy consumption (SEC), and energy-mass co-benefit (EMCI). EY measures the energy content in torrefied products in comparison to their original material and is computed by multiplying the solid yield by the energy density [27]. UEI indicates the ratio of

EY to the energy introduced into the system, providing a more thorough understanding of efficiency by considering energy input, in contrast to solely EY [9]. A higher UEI suggests enhanced energy conversion efficiency in torrefaction with reduced energy input. EMCI assesses torrefaction utilization from both energy and material standpoints, determined by the variance between EY and solid yield (SY) [13]. A higher EMCI is preferable, indicating that a lesser quantity of material produces more energy [9]. For instance, the EMCI values were 18.9 and 14.0 for the inert and oxidative torrefaction of eucalyptus, respectively [28]. These torrefaction indices were utilized to ascertain torrefaction efficiency [9], as depicted in Equations (1) – (6).

$$\text{SY (\%)} = \frac{m_{tor}}{m_{raw}} \quad (1)$$

$$\text{ED (-)} = \frac{HHV_{torr}}{HHV_{raw}} \quad (2)$$

$$\text{EY (\%)} = \text{SY} \times \text{ED} \quad (3)$$

$$\text{UEI (kW}^{-1}\text{h}^{-1}) = \frac{EY}{E_{elec}} \quad (4)$$

$$\text{SEC (kWh}\cdot\text{kg}^{-1}) = \frac{E_{elec}}{m_{tor}} \quad (5)$$

$$\text{EMCI (\%)} = \text{EY} - \text{SY} \quad (6)$$

The subscripts' raw' and 'tor' represent raw and torrefied coffee silver skin (CS), respectively. SY and ED denote solid yield (wt%, dry basis) and energy density, respectively. HHV refers to the higher heating value. $E_{elec}$ represents electric power consumption measured by a power meter.

*2.4 Statistical analysis*

An ANOVA was systematically employed to assess the influence of temperature, residence time, and medium types on coffee silver skin (CS) torrefaction performances using a cubic model. The analysis was conducted with Design-Expert software (Version 12; Stat-Ease, Inc.; USA). Temperature (200, 250, and 300 °C) and residence time levels (15, 30, and 60 min) along with two medium types (flue gas (FG) and $N_2$) were utilized. Temperature and time were coded as -1, 0, and 1, whereas the mediums were classified into FG and $N_2$. After torrefaction, responses like energy yield (EY), upgrading energy index (UEI), specific energy consumption (SEC), and energy-mass co-benefit index (EMCI) were examined in the ANOVA results. The goal for EY, UEI, and EMCI was to maximize their values, while SEC intended to minimize its value. These responses reflect the energetic indices in torrefaction, aiming for energy optimization [9,13]. The ANOVA outcomes and regression models were obtained with a confidence level of 95% ($p$ = 0.05) to evaluate the model's significance [27]. The resultant models indicated a significant effect on the system's responses, detailing the impact of various factors on the selected responses. The optimization of parameters was illustrated using contour plots, which serve as valuable tools in guiding the torrefaction process.

### *2.5 Contact angle analysis*

The contact angle measurement system, aimed at examining hydrophobic characteristics, consisted of a high-resolution camera and a light source. The torrefied sample underwent preheating at 105°C for 24 hours in an oven to remove surface moisture. For each trial, a 100 mg pellet sample was placed on the system's base. Initially, 15 µL of DI water was added to the sample's top surface. The high-resolution camera recorded the immediate contact angle, with subsequent variations documented at 1-second intervals to analyze the dynamic behavior [29,30].

## 3. Results and discussion

### *3.1 Physicochemical analysis*

Fig. 2 illustrates the proximate analysis on an as-received basis (AR) for coffee silver skin (CS) under inert and flue gas atmospheres. In general, fixed carbon (FC) and volatile matter (VM) values exhibit similarities in both atmospheres. Nevertheless, at severe torrefaction temperatures (250 and 300 °C), a higher FC was noted in the flue gas atmosphere, linked to the elevated oxygen content triggering increased depolymerization at these temperatures [13]. Table 1 displays a comparative examination of two distinct flue gas torrefaction conditions. The investigation suggests that a more rigorous condition and an augmented oxygen content in the flue gas, as per Lasek et al. [12], induce higher FC and lower VM compared to the current study. This disparity might be ascribed to more vigorous reactions during the dehydrogenation and deoxygenation processes [17].

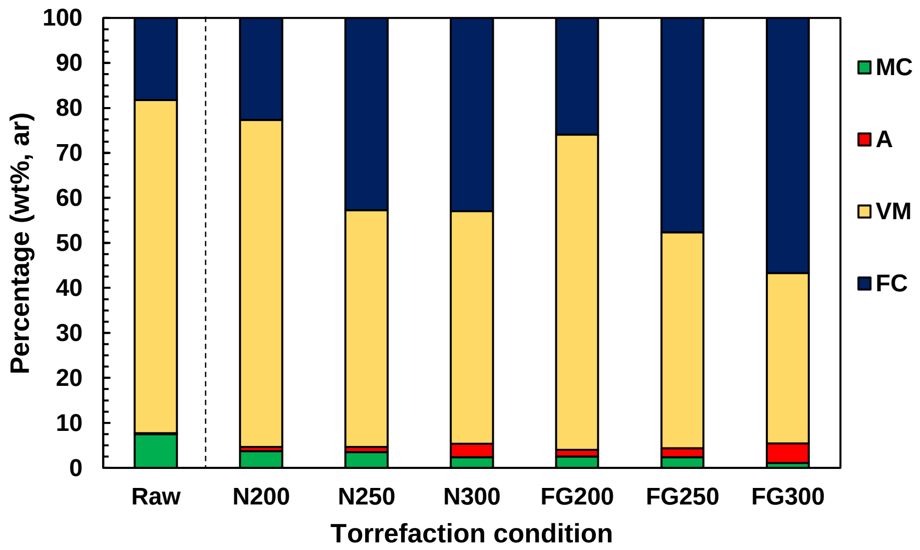



**Fig. 2 Proximate analysis of coffee silver skin under inert and flue gas atmosphere at 30 min**

**Table 1 Comparative the proximate analysis results with previous study**

| | **Lasek et al. [12]** | **This study** |
|---|---|---|
| Feedstock | Willow | Coffee silver skin |
| Medium (vol.%) | $O_2$: 8 | $O_2$: 5, $CO_2$: 15, $N_2$: 80 |
| Condition | T: 350 °C, t: 20 min | T: 200 °C, t: 30 min |
| MC (wt%) | 3.22 | 2.50 |
| VM (wt%) | 46.36 | 70.03 |
| FC (wt%) | 46.49 | 25.95 |
| A (wt%) | 3.93 | 1.51 |

The properties of the CS were improved by both atmospheres, as evidenced by the ratio of H/C and O/C as shown in the van Krevelen diagram in Fig. 3. These ratios approach the origin point of the plot when torrefaction occurs at higher temperatures (250 and 300 °C), indicating that the hydroxy group (-OH) was expelled (resulting in decrease hydrogen and oxygen atoms) and polymerization occurred (leading to an increase in carbon atoms). Consequently, the H/C and O/C ratios are lower. The flue gas torrefaction (FGT) is closer to the origin compared to inert torrefaction, with FGT exhibiting a steeper slope and a ratio of 1.22. This suggests that the oxygen content in FGT enhances oxidation reactions and devolatilization during torrefaction [7].

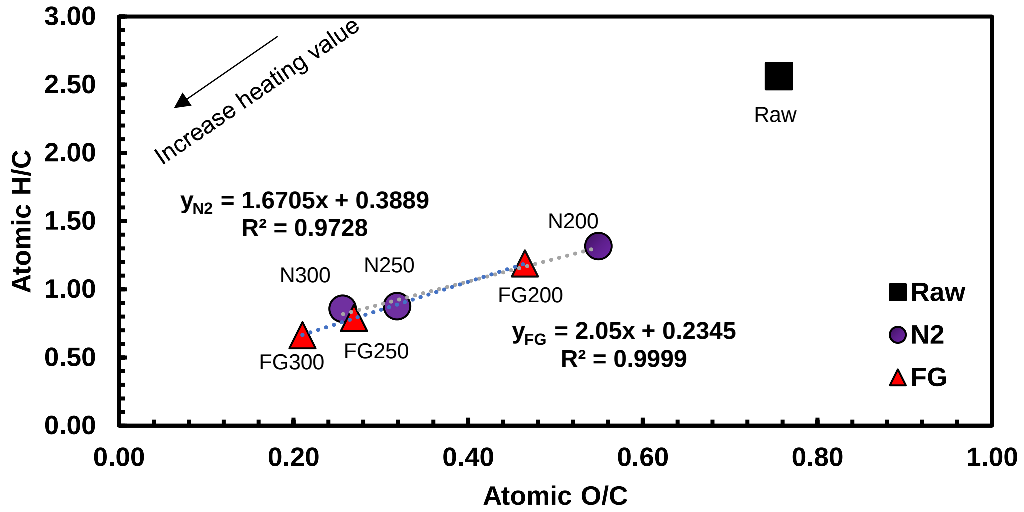
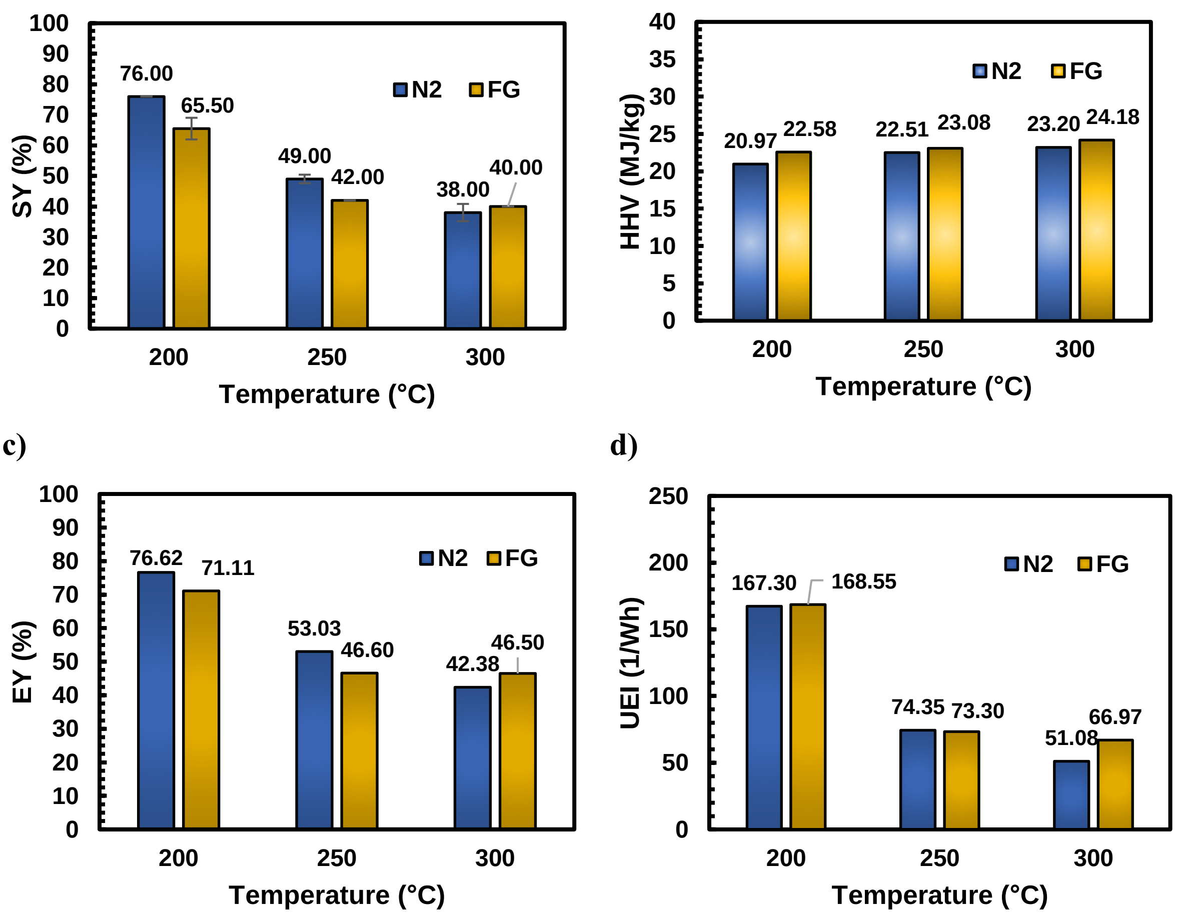



**Fig. 3 Van krevelen diagram for coffee silver skin in inert and flue gas torrefaction**

*3.2 Torrefaction performances*

Fig. 4 illustrates the impact of different medium types ($N_2$ and flue gas) and temperatures on torrefaction performance, specifically solid yield (SY), higher heating value (HHV) of torrefied samples, energy yield (EY), upgrading energy index (UEI), specific energy consumption (SEC), and energy-mass co-benefit (EMCI). With increasing torrefaction temperature, both solid and energy yield decrease, while the heating value and energy density increase. These changes stem from the combined torrefaction reactions, such as hemicellulose decomposition in conventional torrefaction during devolatilization (inert atmosphere) and partial oxidation in an oxidative atmosphere with a low oxygen level (5% vol.) [16]. As temperatures rise, energy consumption increases, resulting in higher SEC and lower UEI. Notably, the EMCI of flue gas at 200 °C is nearly nine times greater than that of inert torrefaction, indicating that at this lower temperature, there is a significant difference between EY and SY for flue gas compared to inert torrefaction [31]. This distinction implies that the oxygen present in the flue gas enhances the energy density of the sample, leading to a higher EY.

a) b)




e) f)

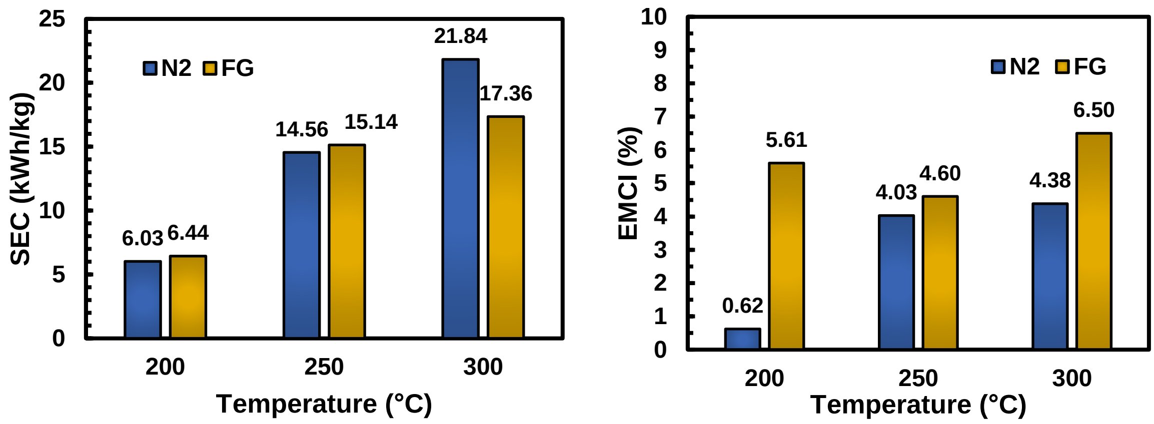



**Fig. 4 Torrefaction performances for coffee silver skin in inert and flue gas torrefaction at 30 min: a) SY, b) HHV, c) EY, d) UEI, e) SEC and f) EMCI**

*3.3 Statistical results*

The ANOVA results for the four response models (energy yield (EY), upgrading energy index (UEI), specific energy consumption (SEC), and energy-mass co-benefit (EMCI)) based on the effects of temperature, residence time, and media coded as A, B, and C, are presented in Table 2. The significance of the models was determined using *p*-value., F-value, and percentage contribution [27]. The models exhibited significantly low *p*-values (< 0.0001), indicating high significance, with $R^2$ values of 0.9664, 0.9869, 0.9849 and 0.8741, respectively. EY, UEI, and SEC were modeled by nonlinear (quadratic) equations (Eqs. (7) – (9)), whereas EMCI was represented by a linear equation (Eq. 10).

EY is primarily influenced by temperature (A), showing both linear (A) and quadratic ($A^2$) dependencies, contributing 85.05% and 13.36% to the model, respectively. EY demonstrates a negative correlation with temperature, indicated by the negative coefficient of A in Eq. (7). Additionally, EY is significantly affected by solid yield (SY), which is highly temperature-dependent due to the decomposition of light volatiles during torrefaction [32,27].

UEI is affected by temperature (A) and time (B), with A having a greater influence than B, followed by $A^2$ and AB with percentages of 57.49%, 25.69%, 10.53%, and 5.07%, respectively. The model includes linear terms for A and B, as well as interaction for AB, and a quadratic term for $A^2$, as shown in Equation (8). Both temperature (A) and time (B) exhibit a negative correlation with UEI, indicating a decrease over time. UEI, which represents the ratio of EY to power input, is notably affected by temperature according to Equation (7) and the power input measured in kW·h, which depends on the duration of power consumption during torrefaction. SEC is influenced by temperature (A), residence time (B), and medium (C), with A being the most significant factor, followed by B, AB, $A^2$, AC, and C. The main drivers, A and B, contribute to 90% of the variability in the model. The model, explained in Equation (9), includes linear terms for A, B, and C, interaction terms for AB and AC, and a quadratic term for $A^2$. Both A and B have positive coefficients, indicating a direct relationship with SEC, denoting power input per unit of torrefied biomass as defined in Equation (5) [14]. This association implies that power input is dependent on residence time, whereas the quantity of product is influenced by the SY, which is affected by temperature.

EMCI is significantly influenced by medium type (C) and temperature (A), contributing 58.85% and 19.6% to the model, respectively. Linear (A and C) and interaction (AC) relationships are identified in Eq. (10), with the priority order of C > A > AC, emphasizing the medium (C) as the most critical factor in EMCI. However, medium (C) is represented as a dummy variable with values of 1 and 2, indicating the effect on EMCI without providing the physical meaning. The positive coefficient for temperature suggests a proportional relationship with EMCI. This finding aligns with other studies on flue gas torrefaction [33,31]. Notably, factors contributing less than 1% are excluded from the models due to their minimal impact.

**Table 2 ANOVA results for EY, UEI, SEC and EMCI of coffee silver skin**

| Source | Sum of Squares | % Con.* | df | Mean Square | F-value | *p*-value |
|---|---|---|---|---|---|---|

| | | | | | | |
|---|---|---|---|---|---|---|
| ***EY*** | | | | | | |
| **Model** | 2868.10 | 100 | 9 | 318.68 | 25.59 | < 0.0001 |
| A-A | 2439.32 | 85.05 | 1 | 2439.32 | 195.87 | < 0.0001 |
| B-B | 4.25 | 0.15 | 1 | 4.25 | 0.3411 | 0.5753 |
| C-C | 0.9941 | 0.03 | 1 | 0.9941 | 0.0798 | 0.7847 |
| AB | 2.05 | 0.07 | 1 | 2.05 | 0.1646 | 0.6956 |
| AC | 22.94 | 0.80 | 1 | 22.94 | 1.84 | 0.2118 |
| BC | 0.0192 | 0.00 | 1 | 0.0192 | 0.0015 | 0.9696 |
| $A^2$ | 383.31 | 13.36 | 1 | 383.31 | 30.78 | 0.0005 |
| $B^2$ | 7.00 | 0.24 | 1 | 7.00 | 0.5625 | 0.4747 |
| ABC | 8.22 | 0.29 | 1 | 8.22 | 0.6602 | 0.4400 |
| **Residual** | 99.63 | | 8 | 12.45 | | |
| **Cor Total** | 2967.73 | | 17 | | | |
| **$R^2$** | 0.9664 | | | | | |
| ***UEI*** | | | | | | |
| **Model** | 31443.99 | 100 | 9 | 3493.78 | 66.75 | < 0.0001 |
| A-A | 18218.48 | 57.94 | 1 | 18218.48 | 348.05 | < 0.0001 |
| B-B | 7887.89 | 25.09 | 1 | 7887.89 | 150.69 | < 0.0001 |
| C-C | 187.08 | 0.59 | 1 | 187.08 | 3.57 | 0.0954 |
| AB | 1594.71 | 5.07 | 1 | 1594.71 | 30.47 | 0.0006 |
| AC | 28.92 | 0.09 | 1 | 28.92 | 0.5525 | 0.4785 |
| BC | 3.79 | 0.01 | 1 | 3.79 | 0.0723 | 0.7948 |
| $A^2$ | 3310.66 | 10.53 | 1 | 3310.66 | 63.25 | < 0.0001 |
| $B^2$ | 186.14 | 0.59 | 1 | 186.14 | 3.56 | 0.0961 |
| ABC | 26.32 | 0.08 | 1 | 26.32 | 0.5028 | 0.4984 |
| **Residual** | 418.76 | | 8 | 52.34 | | |
| **Cor Total** | 31862.75 | | 17 | | | |
| **$R^2$** | 0.9869 | | | | | |
| ***SEC*** | | | | | | |
| **Model** | 2008.69 | 100 | 9 | 223.19 | 58.11 | < 0.0001 |
| A-A | 1181.27 | 58.81 | 1 | 1181.27 | 307.58 | < 0.0001 |
| B-B | 639.19 | 31.82 | 1 | 639.19 | 166.43 | < 0.0001 |
| C-C | 20.87 | 1.04 | 1 | 20.87 | 5.43 | 0.0481 |
| AB | 89.58 | 4.46 | 1 | 89.58 | 23.32 | 0.0013 |
| AC | 24.42 | 1.22 | 1 | 24.42 | 6.36 | 0.0357 |
| BC | 0.0027 | 0.00 | 1 | 0.0027 | 0.0007 | 0.9795 |
| $A^2$ | 52.13 | 2.60 | 1 | 52.13 | 13.57 | 0.0062 |
| $B^2$ | 1.12 | 0.06 | 1 | 1.12 | 0.2926 | 0.6033 |
| ABC | 0.1035 | 0.01 | 1 | 0.1035 | 0.0270 | 0.8737 |
| **Residual** | 30.72 | | 8 | 3.84 | | |
| **Cor Total** | 2039.41 | | 17 | | | |

| | | | | | | |
|---|---|---|---|---|---|---|
| **$R^2$** | 0.9849 | | | | | |
| ***EMCI*** | | | | | | |
| **Model** | 60.29 | 100 | 9 | 6.70 | 6.17 | 0.0087 |
| A-A | 11.82 | 19.60 | 1 | 11.82 | 10.89 | 0.0109 |
| B-B | 3.37 | 5.59 | 1 | 3.37 | 3.11 | 0.1160 |
| C-C | 35.48 | 58.85 | 1 | 35.48 | 32.69 | 0.0004 |
| AB | 0.1128 | 0.19 | 1 | 0.1128 | 0.1039 | 0.7554 |
| AC | 9.03 | 14.98 | 1 | 9.03 | 8.32 | 0.0204 |
| BC | 0.0000 | 0.00 | 1 | 0.0000 | 0.0000 | 0.9957 |
| $A^2$ | 0.0078 | 0.01 | 1 | 0.0078 | 0.0072 | 0.9345 |
| $B^2$ | 0.3173 | 0.53 | 1 | 0.3173 | 0.2924 | 0.6034 |
| ABC | 0.1540 | 0.26 | 1 | 0.1540 | 0.1419 | 0.7162 |
| **Residual** | 8.68 | 19.60 | 8 | 1.09 | | |
| **Cor Total** | 68.97 | 5.59 | 17 | | | |
| **$R^2$** | 0.8741 | | | | | |

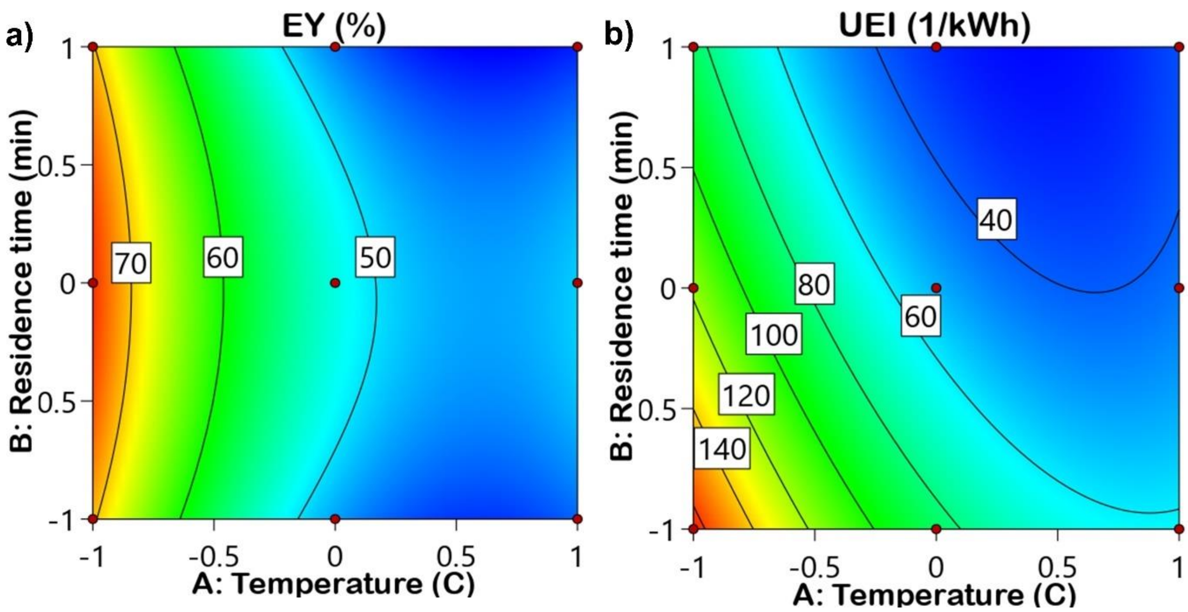
*% Cont = % contribution

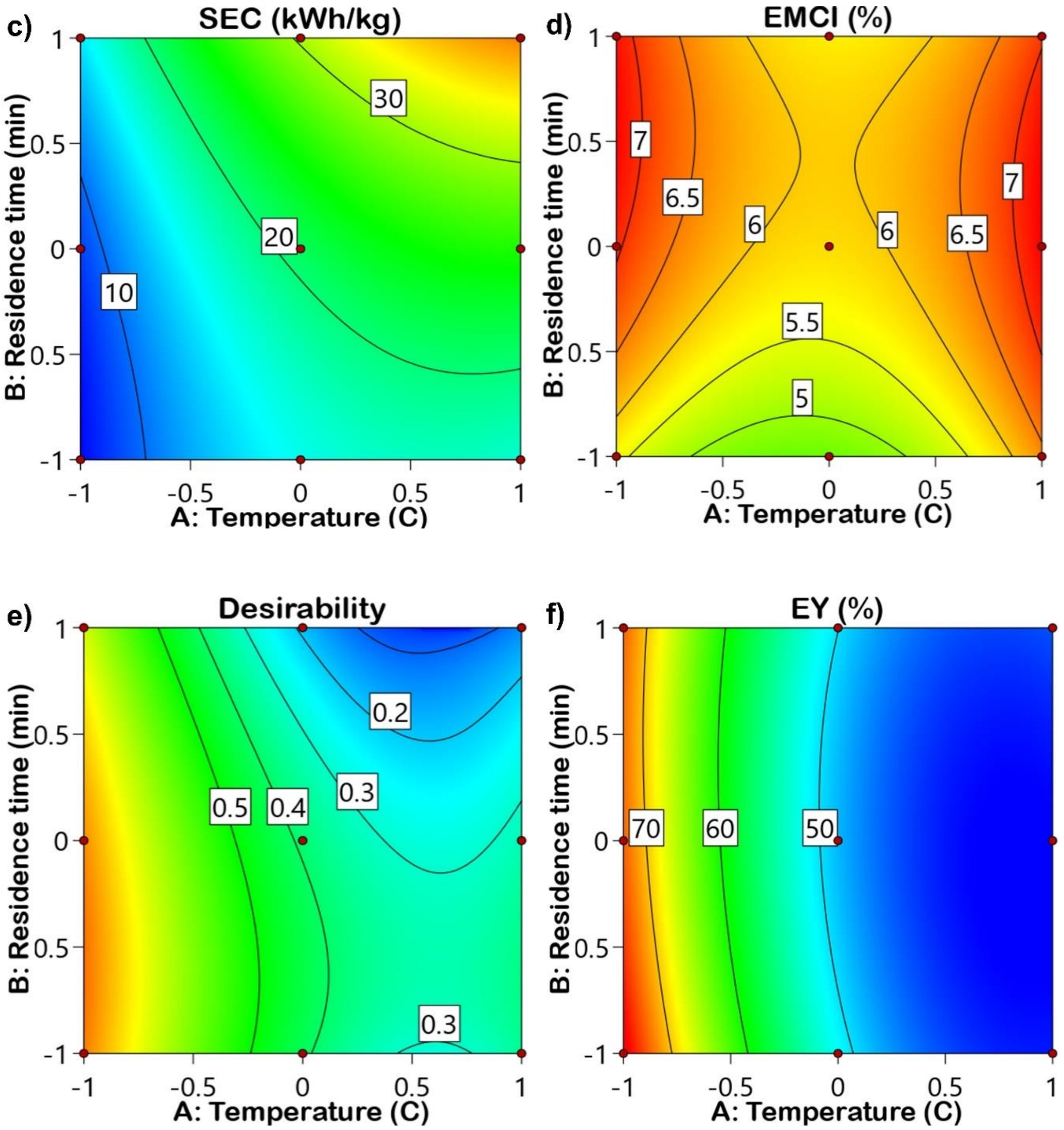
$$EY = 50.24 - 14.26A + 9.79A^2 \quad (7)$$

$$UEI = 48.62 - 38.96A - 25.64B + 14.12AB + 28.77A^2 \quad (8)$$

$$SEC = 22.74 + 9.92A + 7.30B - 1.08C + 3.35AB - 1.43AC - 3.61A^2 \quad (9)$$

$$EMCI = 5.13 + 0.9925A + 1.40C - 0.8675AC \quad (10)$$




c)
SEC (kWh/kg)
30
20
10
B: Residence time (min)
A: Temperature (C)
d)
EMCI (%)
7
6.5
6
6
6.5
7
5.5
5
B: Residence time (min)
A: Temperature (C)
e)
Desirability
0.2
0.3
0.5
0.4
0.3
B: Residence time (min)
A: Temperature (C)
f)
EY (%)
70
60
50
B: Residence time (min)
A: Temperature (C)

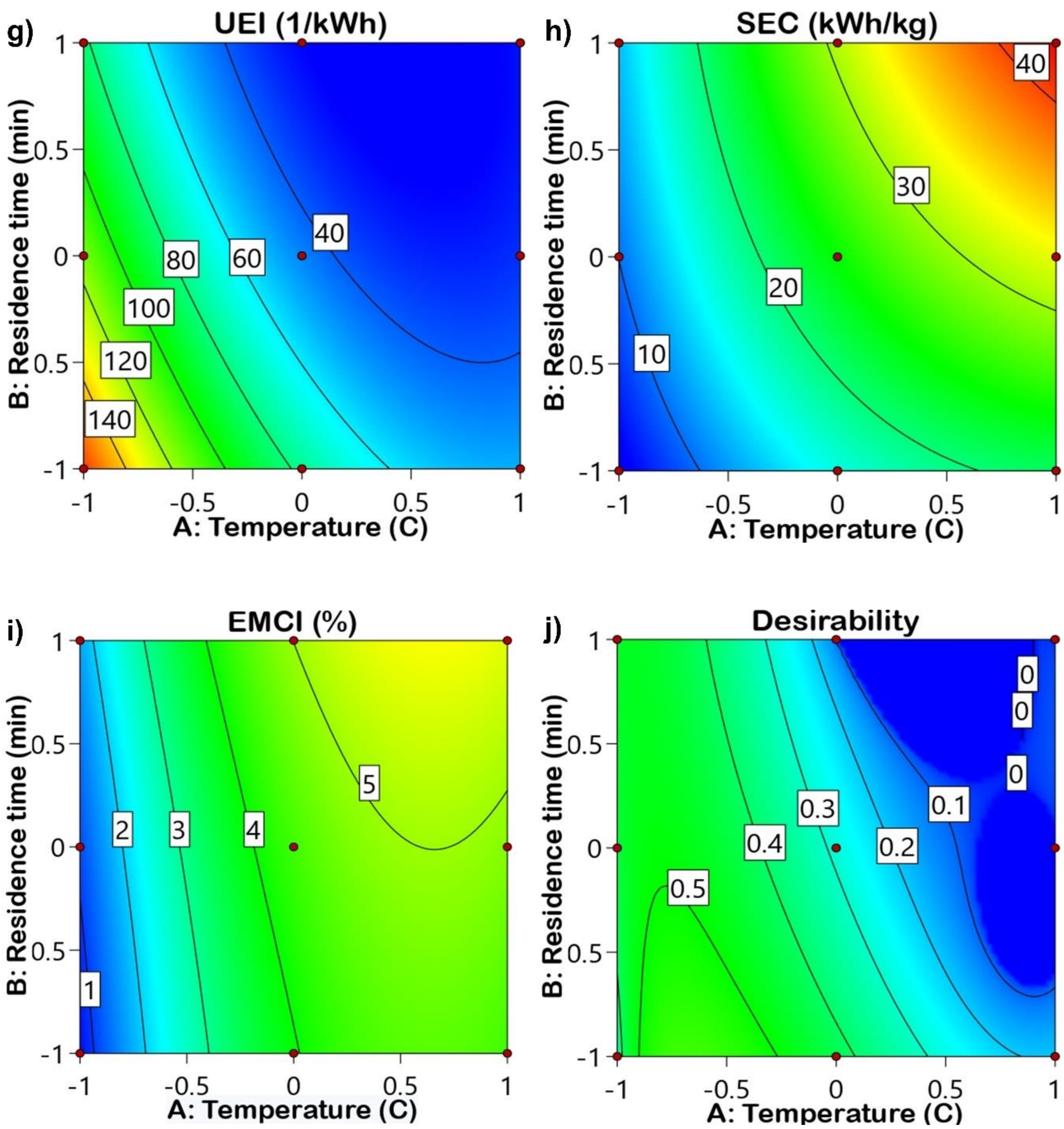



**Fig. 5 Contour plot for flue gas (a-e) and inert (f-j) torrefaction**

Fig. 5 presents contour plots for flue gas (a-e) and inert (f-j) torrefaction, illustrating the influence of two main variables—temperature (A) and residence time (B)— on the responses of energy yield (EY), upgrading energy index (UEI), specific energy consumption (SEC), energy-mass co-benefit (EMCI), and desirability, respectively. For flue gas torrefaction, the EY is sensitive to temperature, consistent with previous results (Table 2 and Eq. (7)). It is observed that higher EY is achieved at lower temperatures and shorter residence times, whereas increased temperature and time reduce EY due to greater material degradation. This phenomenon is attributed to hemicellulose decomposition and partial oxidation [16]. The UEI, which reflects energy efficiency by assessing energy input, demonstrates optimal values at lower temperatures and shorter durations, signifying enhanced energy conversion efficiency under these conditions. SEC, which quantifies the energy consumption during the process, is minimized at lower temperatures and shorter times, indicating that energy input can be substantially reduced with careful control of these parameters. The EMCI, reflecting both energy retention and material loss, consistently exhibits higher EMCI values, ranging from 5% to 7%, with optimal performance observed at short and long residence times across low to high temperatures, indicating a superior balance between energy retention and minimized mass loss. Lower temperatures and longer durations are recommended in the industrial context, while higher temperatures and longer durations are advised for solid fuel applications with a high energy content.

Inert torrefaction exhibits similar trends, although it generally yields lower UEI and higher SEC values compared to flue gas. It shows significantly lower EMCI values, ranging from 1% to 5%, with the lowest efficiencies occurring at shorter residence times and lower temperatures, indicating that flue gas torrefaction is comparatively more energy efficient. Both methods benefit from operating under milder conditions, but flue gas torrefaction offers a more favorable balance between energy efficiency and material conservation, as indicated by the superior UEI and SEC performance. The desirability analysis of flue gas and inert torrefaction shows that flue gas torrefaction is more desirable in a wider range of conditions, especially when temperatures are low and residence times are short, with values ranging from 0.2 to 0.5. The highest desirability for flue gas torrefaction (0.5) occurs at lower residence times and moderate temperatures, indicating that these conditions optimize the balance between energy efficiency and material conservation. In contrast, inert torrefaction shows significantly lower desirability, with values ranging from 0 to 0.5 and a sharp decline in performance as temperature increases. The inert process reaches its maximum desirability at lower temperatures and shorter residence times but remains less desirable overall compared to flue gas torrefaction. These results confirm that flue gas torrefaction provides a more flexible and efficient process, offering a wider range of optimal operating conditions, while inert torrefaction requires stricter control and offers less favorable outcomes in terms of process efficiency and desirability.

**Table 3 Optimum results from the simulation for the pelletized coffee silver skin**

| Parameters | Goal | Prediction | Experiment | Error (%) |
|---|---|---|---|---|
| A-Temperature (°C) | - | 200 | | |
| B-Residence time (min) | - | 30 | | |
| C-Medium | - | Flue gas | | |
| Desirability | - | 0.869 | | |
| EY (%) | Maximum | 71.48 | 71.11 | 0.52 |
| UEI ($kW^{-1}h^{-1}$) | Maximum | 163.35 | 168.55 | 3.18 |
| SEC ($kWh{\cdot}kg^{-1}$) | Minimum | 6.00 | 6.44 | 7.33 |
| EMCI (%) | Maximum | 5.30 | 5.61 | 5.85 |

Table 3 summarizes the optimal conditions derived from the analysis. The model suggests a temperature of 200 °C and a duration of 30 min, which corresponds to EY, UEI, SEC and EMCI values of 71.48%, 163.35 $kW^{-1}h^{-1}$, 6.00 $kWh{\cdot}kg^{-1}$ and 5.30%, respectively, under flue gas atmosphere. The experiment was repeated under these conditions to verify the model's accuracy of. EY, UEI, SEC and EMCI errors were found to be 0.52%, 3.18%, 7.33% and 5.85%, respectively. These errors may be attributed to inconsistent current during the furnace's startup stage. This validation affirms that the models are acceptable and can be effectively applied in related torrefaction systems.

*3.4 Hydrophobicity*

In this study, contact angle (CA) measurements were utilized to assess the hydrophilicity of CS (Fig. 6). The CA for raw CS initiates at 80° and decreases rapidly to 0°

within 24 seconds due to water adsorption by larger hemicellulose molecules during the test [34]. For both pretreated samples (FGT and NT), the CAs exceed 100°, indicating increased hydrophobicity and reduced water adsorption capability [35]. The CA for FGT remains stable between 120° and 140° for over 25 seconds. During torrefaction at 200 °C, the CAs for FGT and NT are similar at 120° and 127°, respectively. However, at higher temperatures (250-300 °C), FGT's CA increases to between 127 and 139°, indicating that surface oxidation removes more hemicellulose, thereby enhancing hydrophobicity and water resistance [36].

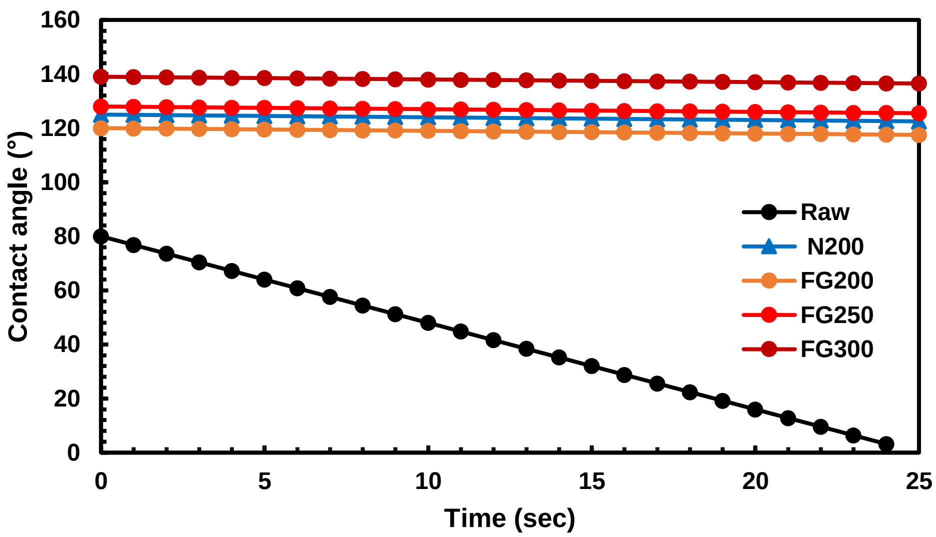



**Fig. 6 Contact angle for the pelletized coffee silver skin in flue gas torrefaction (FGT) and inert torrefaction (NT)**

# 4. Conclusions

The torrefaction of coffee silver skin in the flue gas was conducted to enhance coffee properties and the energetic efficiency of the process. Temperature was found to significantly impact EY, while both temperature and residence time primarily affected UEI and SEC. Flue gas as the medium type is mandatory for EMCI. Flue gas torrefaction at 200 °C for 30 minutes was identified as the optimal condition for EY, UEI, EMCI, and SEC, achieving the torrefied coffee properties, including MC, VM, FC, and A of 3.03%, 69.24%, 27.04%, and 1.01%, respectively. These findings support the use of waste and flue gas to improve energy efficiency for power and heat applications.

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## Author Contribution

Kanit Manatura: Conceptualization, Formal analysis, Funding acquisition, Validation, Project administration, Resources, Software, Supervision, Writing - original draft, Writing - review & editing.

Supaporn Klinkesorn: Data collection, Methodology.

Benjapon Chalermsinsuwan: Formal analysis, Investigation, Validation, Writing - review & editing.

Namfon Samsalee: Investigation, Writing - review & editing.

Sutthipoj Wongrerkdee: Writing - review & editing.

Kitipong Jaojaruek: Writing – review.

Adisak Pattiya: Writing – review.

Jun Li: Writing - review & editing.

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## Data Availability

The datasets analyzed during this study are available from the corresponding author on reasonable request.

## Declarations

Competing Interests The authors declare no competing interests.

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