

# Sustainable energy recovery through hydrothermal co-processing of municipal solid waste and laundry wastewater

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## ABSTRACT

The escalating challenges of municipal solid waste (MSW) and wastewater management in developing nations necessitate innovative solutions. This study introduces a novel approach by utilizing hydrothermal carbonization (HTC) to co-process traditional market solid waste and laundry wastewater, addressing both solid and liquid waste streams while simultaneously recovering energy. The research optimized the HTC process using response surface methodology (RSM) with a Box-Behnken design, examining the effects of temperature (180–220 °C), process time (30–90 minutes), and biomass waste-to-water ratio (0.5–1) on hydrochar properties. The results demonstrate that increasing temperature and process time enhance hydrochar's calorific value (up to 25.479 MJ/kg), energy densification ratio (up to 1.417), and fixed carbon content, indicating improved fuel characteristics. Optimal conditions (199.394 °C, 30 minutes, 1:1 ratio) were identified using Minitab 19 software. This study's novelty lies in its integrated approach to waste treatment and resource recovery. The findings suggest that HTC can effectively transform market waste and laundry wastewater into valuable hydrochar, offering a sustainable alternative to land-filling, mitigating environmental pollution, and providing a renewable energy source.

**Keywords:** hydrothermal carbonization, waste management, sustainable energy, coprocessing, msw, laundry wastewater, waste-to-energy conversion.

## INTRODUCTION

The increasing generation of municipal solid waste (MSW) and wastewater, exacerbated by insufficient management practices, presents a significant challenge for developing nations. This unsustainable scenario results in severe environmental degradation, public health hazards, and esthetic issues (Abubakar et al., 2022; Bouyakhssass et al., 2023; Djulianti and Ainun, 2018; Siddiqua et al., 2022; Sururi et al., 2017). In Indonesia, research has highlighted the urgency of this predicament, with organic waste – primarily food waste – and plastics constituting a substantial portion of MSW (Djaenudin et al., 2021; KLHK, 2023). This reality underscores the untapped potential of MSW as a resource for energy recovery. Alarming, a

significant proportion of MSW in Indonesia, approximately 69%, is still directed to landfills, where it generates toxic leachate and greenhouse gases and poses risks of landfill fires (Lokahita et al., 2019; Manurung and Santoso, 2019; Yudha Ramdhani et al., 2018). Traditional markets are notable contributors to this issue, ranking second only to households in terms of waste generation (KLHK, 2023). The inadequate waste management practices at these markets, combined with the high moisture content of organic waste, intensify leachate production, odor problems, and the risk of soil and water contamination (Abubakar et al., 2022; Kharola et al., 2022; Qi et al., 2022; Siddiqua et al., 2022; Widyarsana and Daniel, 2020). In addition to solid waste challenges, laundries contribute significantly to environmental concerns

through the generation of substantial volumes of wastewater. Daily greywater production, which includes laundry wastewater, is estimated to be between 137 and 153 L per person in urban Indonesian areas (Widyarani et al., 2022). Alarming, this wastewater accounts for approximately 10% of Indonesia's total urban wastewater generation (Gemala and Oktarizal, 2019; Zoroufchi Benis et al., 2021). Furthermore, an estimated 30% of wastewater from households and laundries is discharged untreated into waterways (Widyarani et al., 2022). The seemingly benign act of washing clothes can have significant repercussions on human health as untreated laundry wastewater introduces various contaminants, including surfactants, detergents, dyes, and persistent organic pollutants (Khalid et al., 2018; Mohamed et al., 2018; Oteng-Pepurah et al., 2018; Patil et al., 2020; Watiniasih et al., 2019). These pollutants pose considerable risks to human health, with long-term exposure potentially leading to skin irritations, respiratory issues, and more severe health complications (Melián et al., 2023).

The issue of environmental degradation is further compounded by energy crises and the depletion of natural resources, driven by increasing population pressures and high levels of anthropogenic activity. The International Energy Agency provides a sobering perspective, reporting that the global coal demand reached 8.03 billion tons in 2022, reflecting a concerning upward trend (IEA, 2022). Indonesia's coal consumption has surged dramatically, increasing from 20,910 tons per year in 2021 to approximately 86,587 tons per year in 2022 (IEA, 2022). This rising demand for a finite resource inevitably raises concerns regarding the sustainability of coal reserves. Global coal production peaked at 5.826 million tons of coal equivalent (Mtce) in 2021, with projections indicating a significant decline to 3.826 Mtce by 2030 (IEA, 2022). This trend calls for a decisive transition toward renewable energy sources and a circular economy approach. In this respect, technology for energy recovery can be a promising solution. This approach not only addresses energy crises but also promotes resource recovery and a more sustainable future (Moya et al., 2017).

Hydrothermal carbonization (HTC) has emerged as a promising thermochemical conversion process for biomass characterized by high moisture contents (Aragón-Briceño et al., 2021; Hitzl et al., 2015; Shafie et al., 2018; Shah et al., 2024). In this regard, the substantial quantities of

market solid waste and laundry wastewater present a viable biomass resource. This biomass can be harnessed as an alternative energy source while simultaneously mitigating the disposal of solid waste in landfills. However, the high moisture content of market solid waste typically necessitates a predrying process before conventional thermal treatments (Djaenudin et al., 2021). HTC circumvents this requirement by utilizing heat, pressure, and water, leading to reduced energy consumption and lower emissions compared with pyrolysis. HTC effectively processes raw materials with high moisture content, operating under wet conditions at elevated temperatures (180–250 °C) and pressures (2–10 MPa) (Hitzl et al., 2015; Shafie et al., 2018; Sulaiman et al., 2023., Funke and Ziegler, 2010; Wilk et al., 2021a, 2021b; Yan et al., 2023). Water maintained in a subcritical state dissolves the waste and acts as a catalyst during the process (Putra et al., 2020, 2021). HTC yields solid biofuel (hydrochar), liquid products, and gases.

A notable advantage of HTC is its capacity to simultaneously treat both solid and liquid waste within a single operation. This multifaceted approach enhances the efficiency of waste management systems. In addition, HTC allows for the potential substitution of clean water with wastewater during the process, representing a significant advancement in waste treatment sustainability. This innovative utilization of wastewater aligns with eco-friendly practices and substantially contributes to the optimization of waste processing strategies. Such advancements underscore the versatility and environmental benefits of HTC, positioning it as a promising technology for integrated solid and liquid waste treatment (Benavente et al., 2024; Picone et al., 2024; Putra et al., 2024a). However, despite various research initiatives aimed at integrating the processing of solid and liquid waste, the specific application of HTC tailored for the dual treatment of household waste – namely, garbage and laundry waste – has not yet achieved widespread implementation.

This study aims to identify the optimal conditions for producing hydrochar from traditional market solid waste (TMSW) while utilizing laundry wastewater (LWW) as a processing medium. A response surface methodology (RSM) with a Box–Behnken Design (BBD) was employed to investigate the effects of varying the hydrothermal process conditions. This systematic approach seeks to optimize the HTC process, potentially establishing an efficient and sustainable method

for transforming market waste into a valuable energy resource while simultaneously addressing the challenges of wastewater management.

## MATERIALS AND METHODS

### Materials

The mixed solid waste for this study was sourced from the Ciroyom traditional market in Bandung City, West Java, Indonesia. The waste composition profile of this market aligns with typical trends, featuring primarily organic waste (approximately 83%). This organic waste includes vegetable and fruit residues, cassava peels, and coconut fiber. The remaining waste composition comprises plastic (8%), paper (5%), and other materials (4%). These inorganic fractions include various plastic items, paper products, and styrofoam. Additionally, the laundry wastewater used for HTC was sourced from a laundromat in Bandung City. This wastewater contains organic matter, phosphates, and detergent residues, exhibiting a chemical oxygen demand of 400 mg/L, a phosphate level of 14.1 mg/L, and a detergent concentration of 33.1 mg/L.

### Experimental design and procedure

To prepare the waste for HTC, several pre-processing steps were undertaken. Initially, the organic and inorganic fractions of the market waste were separated. Subsequently, the organic fraction underwent size reduction to enhance the process efficiency, which may have involved shredding or grinding, depending on the specific equipment available. Inorganic waste components were sorted to isolate suitable materials for coprocessing, such as specific plastic types.

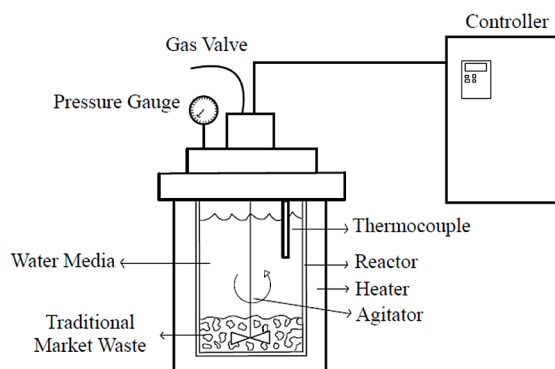
This study employed a batch-type HTC reactor fabricated from stainless steel (10-mm thickness, 1-L capacity) for the conversion of mixed traditional market waste into hydrochar. A schematic representation of the experimental apparatus is provided in Figure 1. To analyze the impact of key process parameters on hydrochar quality, a series of HTC experiments was conducted, systematically varying the following: temperature (180–220 °C), process time (30–90 min), and solid waste-to-wastewater ratio (TMSW-LSWW ratio) (0.5–1). These parameters significantly influence the properties of the resulting hydrochar

(Kantakanit et al., 2018a, 2018b; H. J. Kim et al., 2021). We utilized an RSM approach with a BBD to evaluate the effects of these operating conditions, optimize the HTC process, and determine the impact on the hydrochar properties, including the solid mass yield, energy densification ratio, and energy yield (Munir et al., 2018; Putra et al., 2022). The experimental matrix generated by the Minitab 19 software is shown in Table 1.

A homogeneous 250-g sample of blended traditional market waste was prepared for each experimental run. This feedstock was mixed with laundry wastewater in varying volumes (250, 375, and 500 mL) to achieve the desired TMSW-LWW ratios (1, 0.75, and 0.5, respectively). The reactor was then sealed, heated to the designated temperature, and held for the specified process time. Upon completion, the reactor was cooled, the excess gas was vented, and the contents were filtered to separate the solid (hydrochar) and liquid phases. The hydrochar was air-dried under sunlight for 24 h, followed by grinding and sieving to obtain a 60-mesh powder for subsequent characterization.

### Physical and chemical analyses

To assess the hydrochar quality, we determined its calorific values using ASTM D5865-12 (Standard Test Method for Gross Caloric Value of Coal and Coke) and proximate composition using ASTM D7582-15 (Standard Test Methods for Proximate Analysis of Coal and Coke by Macro Thermogravimetric Analysis). The hydrochar weight was measured gravimetrically. These values were used to calculate the process optimization parameters, including the mass yield (*MY*) (Equation 1), energy densification ratio (*EDR*) (Equation 2), and energy yield (*EY*) (Equation 3), in accordance with previous research (Azaare et



**Figure 1.** Schematic of the HTC apparatus

**Table 1.** Experimental design of HTC

Temperature (°C)	Process time (minute)	TMSW-LWW ratio
180	30	0.75
180	90	0.75
180	60	0.5
180	60	1
200	30	0.5
200	90	0.5
200	30	1
200	90	1
200	60	0.75
200	60	0.75
200	60	0.75
220	30	0.75
220	90	0.75
220	60	0.5
220	60	1

al., 2021a; Liu et al., 2024; Putra et al., 2024b; Shen et al., 2024; Wang et al., 2023; Wang et al., 2022):

$$MY = \frac{\text{mass of dry hydrochar}}{\text{mass of dry feedstock}} \times 100\% \quad (1)$$

$$EDR = \frac{\text{higher heating value [HHV] of hydrochar}}{\text{HHV of feedstock}} \times 100\% \quad (2)$$

$$EY = \text{mass yield} \times \text{energy densification ratio} \quad (3)$$

We supplemented the chemical analysis with qualitative scanning electron microscopy (SEM; Hitachi SU3500) to characterize the particle size, shape, distribution, and surface morphology of the obtained hydrochar. SEM analysis offered insights into the hydrochar porosity, potential contaminants, stability, and overall suitability for environmental applications.

### Statistical analysis

We employed analysis of variance (ANOVA) to investigate the impact of the HTC operating conditions on the hydrochar weight and heating value. RSM was implemented using the Minitab 19 software. A P-value of < 0.05 signified a statistically significant effect of the independent variables on the dependent variables. To evaluate the suitability of the response model, we conducted a lack-of-fit test. A P-value less than or equal to the significance level (0.05) indicated an unacceptable model; conversely, higher P-values suggested model adequacy. Optimal operating conditions

were determined by integrating the RSM and ANOVA results, with emphasis on the desirability value. This optimization objective function reflected the model's capacity to achieve targets based on predefined product quality criteria. A desirability value approaching 1 demonstrated optimization accuracy. Our optimization aimed to identify the HTC conditions yielding the maximum hydrochar weight and calorific value. Both optimization and data analysis were performed using the Minitab 19 software.

## RESULTS AND DISCUSSION

### Influence of HTC parameters on hydrochar properties: weight, calorific value, mass yield, energy densification ratio, and energy yield.

This section investigates the influence of the HTC operating parameters on the key characteristics of the resulting hydrochar: weight, calorific value (energy content), MY, energy densification ratio, and EY. Understanding these relationships is crucial for optimizing the HTC process and tailoring hydrochar properties for specific applications. Before the HTC treatment, the initial weight and calorific value of the feedstock were determined. This information served as a baseline for evaluating the changes induced by the HTC process. The initial wet weight of the waste was 250 g, corresponding to a dry weight of 108 g after natural drying under open-air sunlight for 24 h. This drying process resulted in a significant weight reduction of approximately 56.8%, primarily due to water evaporation. The calorific value of the dried raw material was measured to be 17.99 MJ/kg. These findings are consistent with previous research by (Putra et al., 2018), who reported a range of 11–25 MJ/kg for the calorific value of mixed urban waste. The observed weight loss during drying could be attributed to the evaporation of water content within the waste material. Drying processes induce evaporation decrease the water content and consequently reduce the weight of the dried material.

Hydrochar, a product derived from the thermal conversion of biomass in high-pressure water, is commonly known as a product of HTC. The properties of hydrochar, such as weight, calorific value, MY, EDR, and EY, are influenced by several factors, including temperature, process time, and waste-to-water ratio. Table 2 comprehensively presents the detailed effects of these three factors



**Table 2.** Dry weight, caloric value, solid mass yield, energy densification ratio, and energy yield of hydrochar

HTC process condition*	Dry weight of hydrochar (g)	Caloric value (MJ/Kg)	Solid mass yield (%)	Energy densification ratio	Energy yield (%)
180 °C_30'_0.75	65	20.394	60.185	1.134	68.240
180 °C_60'_1	59	20.402	54.630	1.134	61.965
180 °C_60'_0.5	57	20.751	52.778	1.154	60.889
180 °C_90'_0.75	56	20.809	51.852	1.157	59.988
200 °C_30'_1	55	21.878	50.926	1.216	61.944
200 °C_30'_0.5	52	21.729	48.148	1.208	58.165
200 °C_60'_0.75	50	22.084	46.296	1.228	56.842
200 °C_60'_0.75	49	22.679	45.370	1.261	57.208
200 °C_60'_0.75	50	22.795	46.296	1.267	58.674
200 °C_90'_1	50	23.093	46.296	1.284	59.440
200 °C_90'_0.5	48	23.246	44.444	1.292	57.441
220 °C_30'_0.75	47	23.252	43.519	1.293	56.258
220 °C_60'_1	47	23.501	43.519	1.307	56.860
220 °C_60'_0.5	45	23.601	41.667	1.312	54.673
220 °C_90'_0.75	40	25.479	37.037	1.417	52.465

**Note:** \* temperature\_time (minute)\_TMSW-LWW ratio.

on the hydrochar characteristics. The observed decrease in dry weight with increasing temperature and time in Table 3 paints a clear picture – this signifies a progressive intensification of biomass decomposition under more severe HTC conditions. Because the decomposition of biomass through HTC reactions is endothermic, higher temperatures provide the necessary energy for the breakdown of complex biomass structures into smaller molecules (Mendoza et al., 2021). This deconstruction process leads to the evolution of volatile compounds, such as light gases, organic vapors, and condensable liquids. These volatiles are not captured in the final hydrochar product, consequently contributing to the reduction in the dry weight (Chen et al., 2018; Aurnob et al., 2022; Román et al., 2018; Samaksaman et al., 2023).

The influence of the processing time on the dry weight further corroborates this phenomenon. Extending the HTC duration at a specific temperature allows for more extensive contact between the biomass and the hot water medium. This extended interaction time translates to a greater

degree of breakdown of the biomass structure, resulting in a further decrease in the dry weight. The declining trend observed in the solid MY directly reflects the efficiency of biomass conversion into hydrochar. Higher yields indicate greater proportions of the initial biomass being retained in the final hydrochar product. Conversely, lower yields signify a more significant conversion of biomass into volatile components, as corroborated by the decreasing dry weight (Román et al., 2018).

For instance, Table 2 reveals a substantial drop in dry weight from 65 g at 180 °C for 30 min (at a TMSW-LWW ratio of 0.75) to 40 g at 220 °C for 90 min (at the same ratio). This translates to a reduction of approximately 38%. Similarly, the solid MY plummets from 60.185% to 37.037% across these conditions. These observations highlight the pronounced effect of elevated temperature and extended processing time on the promotion of the decomposition of biomass and the reduction of the final hydrochar yield. The interplay between temperature and time orchestrates a symphony of deconstruction within

**Table 3.** ANOVA result of solid mass yield, energy densification ratio, and energy yield

Value	P-Value		
	Temperature (°C)	Process time (minute)	TMSW-LWW ratio
Solid mass yield (%)	0.000	0.006	0.167
Energy densification ratio	0.000	0.003	0.67
Energy yield (%)	0.003	0.045	0.177

the HTC reactor. Higher temperatures and longer durations act as catalysts, accelerating the breakdown of biomass and the release of volatile components. This translates to a decrease in both the dry weight and the solid MY of the final hydrochar product. Thus, understanding these trends is crucial for optimizing HTC processes to achieve the desired balance between the hydrochar yield and its properties.

Table 2 also reveals a clear trend of increasing calorific value with increasing temperature and extended processing time. For instance, the calorific value jumps from 20.39 MJ/kg at 180 °C for 30 min to 25.48 MJ/kg at 220 °C for 90 min (with a TMSS-LWW ratio of 0.75). This significant enhancement signifies the formation of more energy-dense carbonaceous structures within the hydrochar under harsher HTC conditions. The underlying mechanism for this trend lies in the devolatilization process. Higher temperatures and longer durations promote the decomposition of biomass, leading to the release of volatile compounds such as water, light gases, and organic vapors. This selective removal of oxygenated and hydrogen-rich components enriches the hydrochar with fixed carbon, a key contributor to its energy content (Irsan et al., 2019; Pauline and Joseph, 2020; Samaksaman et al., 2023).

The EDR, calculated as the calorific value per unit dry weight, serves as an indicator of energy concentration within the hydrochar. As shown in Table 3, the EDR exhibits an increasing trend with increasing temperature and time, ranging from 1.13 at 180 °C for 30 min to 1.42 at 220 °C for 90 min. This observation aligns with the rising calorific value and suggests that the hydrochar becomes more energy-dense relative to its original biomass weight under more severe HTC conditions. The EY, which incorporates both the MY and the EDR, presents a more comprehensive picture of the overall energy recovery from the biomass feedstock. Although the data in Table 3 demonstrate a decreasing trend in the EY with increasing temperature and time (highest at 68.24% at 180 °C for 30 min and lowest at 52.47% at 220 °C for 90 min), it is crucial to understand the underlying factors.

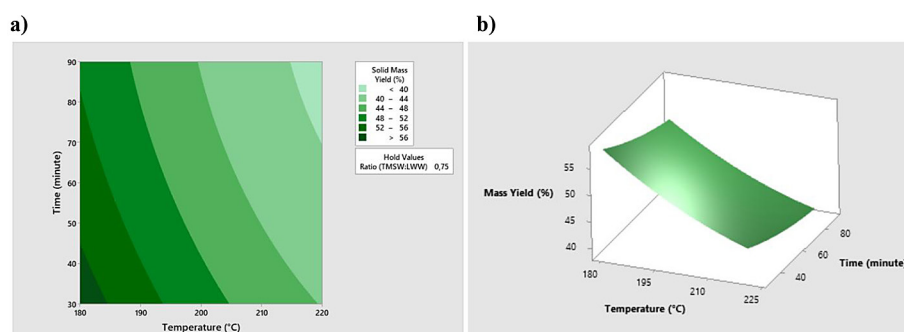
Despite the decrease in the MY with higher HTC severity, the significant increase in the calorific value helps maintain a relatively high EY. However, the combined effect of reduced MY and increased energy content ultimately leads to a slight decline in the EY under harsher conditions

(Putra et al., 2024c). This highlights the importance of striking a balance between maximizing the energy content (calorific value) and maintaining a reasonable MY for optimal energy recovery. The observed trends can further be explained by the alteration of the elemental composition within the hydrochar. Elevated temperatures contribute to the reduction of the O/C and H/C atomic ratios (Azaare et al., 2021b). This translates to a higher energy density in the hydrochar because of the enrichment of carbon, the primary contributor to its energy content.

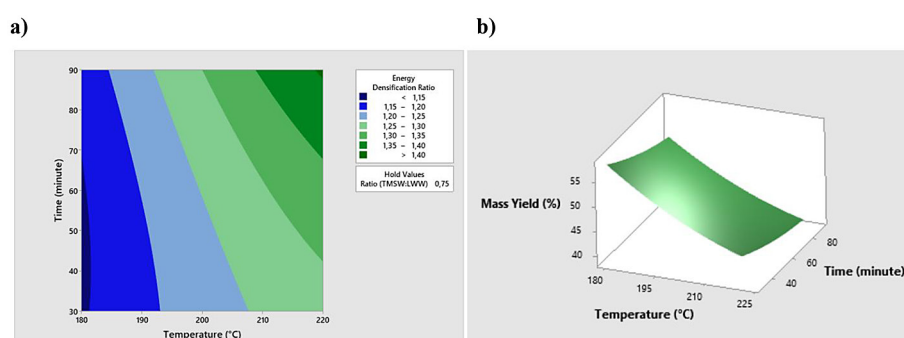
The calorific values obtained in this study (20.39–25.47 MJ/kg) fall within the range reported for subbituminous coals (Kim et al., 2017; Sattasathuchana et al., 2023; Yang et al., 2024). This observation underscores the potential of hydrochar as a comparable alternative to lower-grade fossil fuels. Although variations exist between studies due to diverse feedstock and HTC conditions, the results here demonstrate the promising potential of hydrochar as a renewable energy source.

### Interplay between HTC conditions and hydrochar properties: Insights from RSM and ANOVA

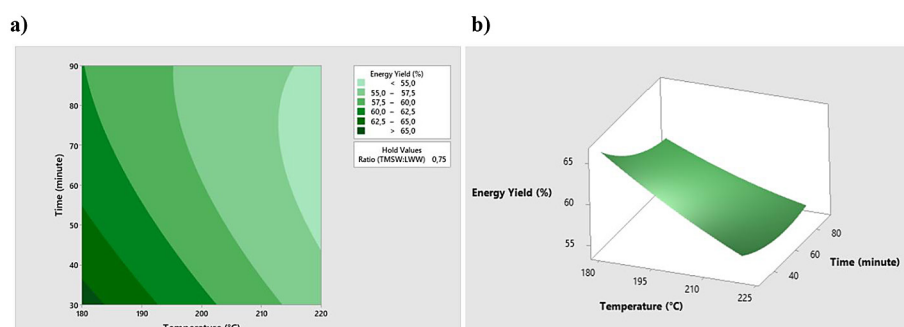
This section delves into the critical relationship between the HTC process parameters and the key hydrochar properties using RSM. The analysis utilizes 3D contour graphs and surface plots (Figures 2–4) to visualize these complex interactions. Figure 3 depicts the decreasing trend of the solid MY with increasing temperature, process time, and TMSW-LWW ratio. This can be attributed to the enhanced decomposition of the biomass feedstock under more rigorous HTC conditions. Elevated temperatures within the subcritical regime promote the breakdown of water-soluble components within the biomass, leading to the release of volatile compounds and a subsequent reduction in the final hydrochar weight (Irsan et al., 2019). The EDR, presented in Figure 3, reflects the energy content per unit dry weight of the hydrochar. As observed, the ratio exhibits an upward trend with increasing HTC severity. This aligns with the observed decline in the solid MY and enrichment of fixed carbon, the primary energy carrier within the hydrochar. Higher EDRs signify more concentrated energy sources, indicating the potentially greater value of hydrochar for various applications. Figure 4 depicts the EY of hydrochar across the investigated HTC



**Figure 2.** (a) Contour and (b) surface plot of the hydrochar solid mass yield



**Figure 3.** (a) Contour plot and (b) surface plot of the hydrochar energy densification ratio



**Figure 4.** (a) Contour plot and (b) surface plot of the hydrochar energy yield

conditions. The highest EY (68.24%) is achieved at 180 °C for 30 min with a TMSW-LWW ratios of 0.75. Conversely, the lowest yield (52.47%) occurs at 220 °C for 90 min with the same ratio. These seemingly contradictory observations underscore the importance of considering both mass and energy content when evaluating energy recovery efficiency. While temperatures and longer processing times increase the EDR, they also contribute to the decrease in the solid MY due to enhanced decomposition. This interplay leads to a slight decline in the EY under harsher HTC conditions. Here, optimizing the process parameters becomes crucial to achieve a balance between maximizing the energy content and maintaining a reasonable MY for optimal energy recovery.

The RSM analysis presented in Figures 2–4 sheds light on the intricate relationship between the HTC process parameters and the key hydrochar properties. Understanding these interactions enables researchers and engineers to optimize HTC processes for tailored hydrochar characteristics and can pave the way for the development of efficient and sustainable energy conversion strategies using HTC technology.

The statistical significance of the HTC process parameters on the hydrochar properties is explored using ANOVA, as can be seen in Table 4. The focus lies on the impact of temperature, process time, and waste-to-water ratio on the solid MY, EDR, and EY. The ANOVA test results reveal a crucial finding: the *P*-values for the

**Table 4.** Regression model of the hydrochar quality

Response	Regression model	Lack of fit
Mass yield (%)	$Y = 276 - 1.809 \text{ Temperature } (^{\circ}\text{C}) - 0.325 \text{ Process Time (Minute)} - 11.6 \text{ M/W Ratio} + 0.00357 \text{ Temperature } (^{\circ}\text{C}) * \text{ Temperature } (^{\circ}\text{C}) + 0.00081 \text{ Process Time (Minute)} * \text{ Process Time (Minute)} + 11.7 \text{ M/W Ratio} * \text{ M/W Ratio} + 0.00077 \text{ Temperature } (^{\circ}\text{C}) * \text{ Process Time (Minute)} - 0.000 \text{ Temperature } (^{\circ}\text{C}) * \text{ Ratio M/W Ratio} - 0.031 \text{ Process Time (Minute)} * \text{ M/W Ratio}$	0.051
Energy Densification ratio	$Y = -0.51 + 0.0144 \text{ Temperature } (^{\circ}\text{C}) - 0.00816 \text{ Process Time (Minute)} + 0.185 \text{ M/W Ratio} - 0.000032 \text{ Temperature } (^{\circ}\text{C}) * \text{ Temperature } (^{\circ}\text{C}) + 0.000012 \text{ Process Time (Minute)} * \text{ Process Time (Minute)} - 0.201 \text{ M/W Ratio} * \text{ M/W Ratio} + 0.000042 \text{ Temperature } (^{\circ}\text{C}) * \text{ Process Time (Minute)} + 0.00069 \text{ Temperature } (^{\circ}\text{C}) * \text{ M/W Ratio} - 0.00056 \text{ Process Time (Minute)} * \text{ M/W Ratio}$	0.607
Energy yield (%)	$Y = 184 - 0.85 \text{ Temperature } (^{\circ}\text{C}) - 0.545 \text{ Process Time (Minute)} - 15.4 \text{ M/W Ratio} + 0.00127 \text{ Temperature } (^{\circ}\text{C}) * \text{ Temperature } (^{\circ}\text{C}) + 0.00129 \text{ Process Time (Minute)} * \text{ Process Time (Minute)} + 8.3 \text{ M/W Ratio} * \text{ M/W Ratio} + 0.00186 \text{ Temperature } (^{\circ}\text{C}) * \text{ Process Time (Minute)} + 0.056 \text{ Temperature } (^{\circ}\text{C}) * \text{ M/W Ratio} - 0.059 \text{ Process Time (Minute)} * \text{ M/W Ratio}$	0.133

temperature and process time variables are statistically significant ( $P$ -value < 0.05) for all three response variables—solid MY, EDR, and EY. These statistically significant  $P$ -values indicate that the temperature and process time exert a substantial influence on the aforementioned hydrochar properties. This research further presents models for estimating the solid MY, EDR, and EY of the hydrochar products in Table 3. The lack-of-fit values for these models are critically evaluated. The obtained lack-of-fit values, 0.051, 0.607, and 0.133 for the solid MY, EDR, and EY, respectively, are all greater than the significance level.

This observation is highly encouraging. Because the lack-of-fit values exceed the significance level, we can confidently conclude that there is no significant lack-of-fit in any of the three models. In simpler terms, the models effectively capture the relationship between the investigated factors and the corresponding hydrochar properties without any systematic bias. This validates the suitability of these models for accurately predicting response variables under specified HTC process conditions. ANOVA successfully demonstrates the statistically significant influence of temperature and process time on key hydrochar properties. Furthermore, the lack-of-fit analysis confirms the suitability of the developed models for estimating solid MY, energy densification ratio, and EY. These findings equip researchers and engineers with reliable tools for optimizing HTC processes to achieve desired hydrochar characteristics for diverse applications.

### Optimization of the HTC process

The primary objective of this study is to identify HTC conditions that maximize the desired hydrochar properties: solid MY, EDR, and EY. The optimization process leverages the Minitab 19 software. This software facilitates the exploration of the HTC parameter space to identify conditions that yield hydrochar with optimal characteristics. By maximizing the desired response values (solid MY, EDR, and EY), the optimization process aims to reduce the operational time or costs while achieving the most favorable hydrochar properties. Table 5 presents optimized HTC process conditions for achieving the best hydrochar quality, identified using the Minitab 19 software.

Given the desirability value of 0.487 obtained by Minitab optimization, it is important to acknowledge that achieving a perfect value of 1 is often unrealistic because of inherent trade-offs between different response variables. Here, the value of 0.487 indicates a reasonable balance between maximizing the desired properties within the constraints of the optimization process. The concept of the desirability function plays a crucial role in the optimization process. This function quantifies the level to which the optimized results fulfill the specified optimization criteria (Akbari et al., 2023; Amenyeku et al., 2024; Periyavaram et al., 2023). A desirability value closer to 1 signifies a higher degree of success in achieving the desired hydrochar characteristics. In simpler terms, a value near 1 indicates the program's

**Table 5.** Optimization result of hydrothermal carbonization process

Temperature ( $^{\circ}\text{C}$ )	Process time (minute)	Ratio (waste media)	Energy yield (%) fit	Energy densification ratio fit	Mass yield (%) fit	Composite desirability
199.394	30	1	62.8743	1.21169	51.8391	0.487966



ability to produce hydrochar with properties that closely resemble the ideal target.

### Proximate and SEM analysis of the hydrochar

This section explores the impact of the HTC process parameters on the quality of the hydrochar as determined through proximate analysis (Figure 5). Proximate analysis provides valuable insights into the key components – moisture content, volatile matter, ash content, and fixed carbon – that influence the fuel properties of hydrochar (Amenyeku et al., 2024; Balmuk et al., 2023; Wang et al., 2023). The data in Figure 6 reveal a compelling trend. As the temperature, process time, and the amount of water used in the HTC process increase, so do the caloric value and fixed carbon content of the hydrochar. This observation aligns with the previously discussed deconstruction process. Higher temperatures promote the breakdown of cellulose and hemicellulose, leading to the

enrichment of fixed carbon—the primary contributor to the energy content. The observed increase in the caloric value, ranging from 20.39 to 25.47 MJ/kg, further substantiates this phenomenon. Conversely, the water and ash content of the hydrochar exhibits a decreasing trend with increasing HTC severity. The decrease in the water content can be attributed to the decomposition of water-soluble components like cellulose and hemicellulose at elevated temperatures. Furthermore, these components are known to bind water readily, so their decomposition leads to a reduction in the overall water content within the hydrochar. Meanwhile, the declining ash content can be explained by the release and dissolution of inorganic materials, typically present as ash, into the water medium during HTC (Deng et al., 2021). This highlights the ability of HTC to purify the biomass feedstock by removing undesirable ash components.

Figures 6 and 7 present SEM images of the hydrochar samples obtained at the lowest and

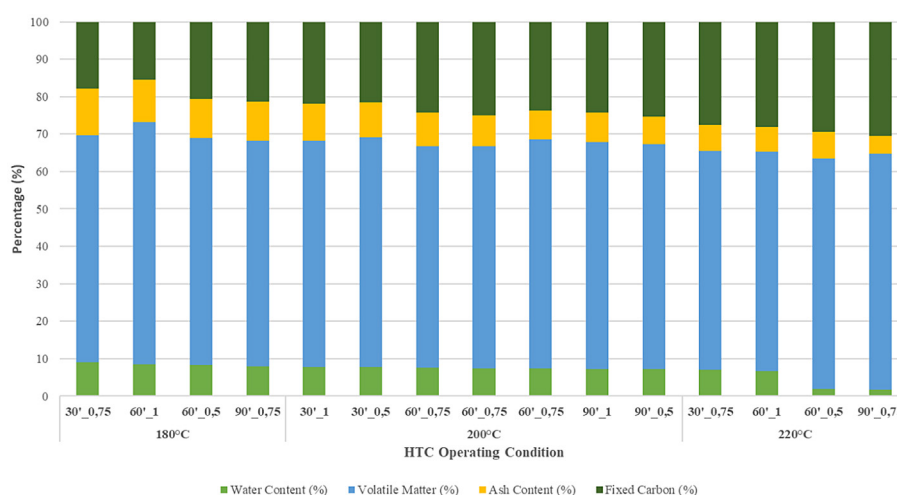


Figure 5. Proximate analysis of the hydrochar

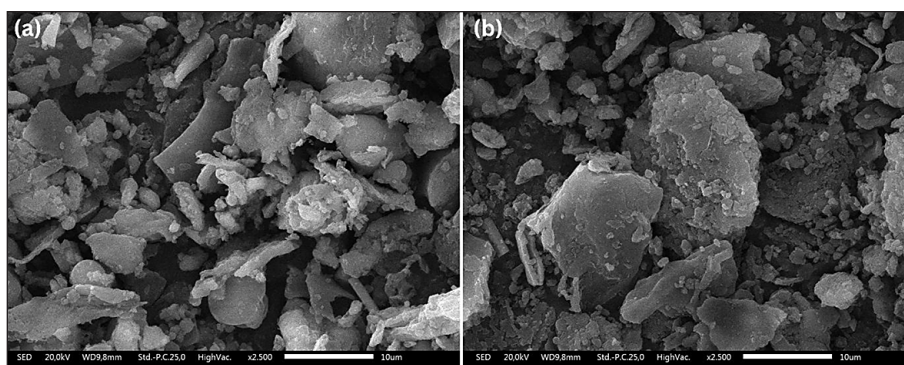
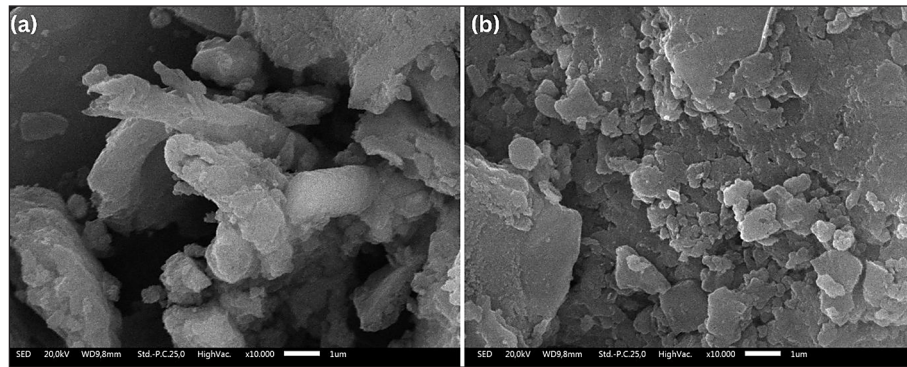


Figure 6. SEM analysis results: (a) temperature 180 °C, process time 30 min, ratio, 0.75; (b) temperature 220 °C, process time 90 min, ratio 0.75. Zoom: 2,500×

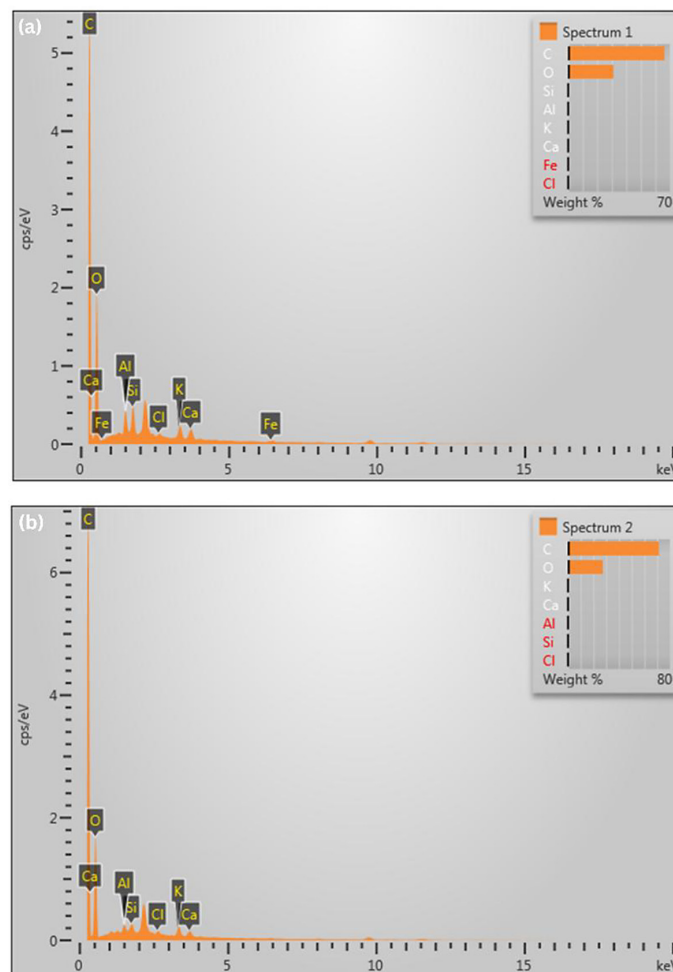


**Figure 7.** SEM analysis results: (a) temperature 180 °C, process time 30 min, ratio 0.75; (b) temperature 220 °C, process time 90 min, ratio 0.75. Zoom: 10,000×

highest operating conditions, magnified at 2.500× and 10.000×. These images provide valuable insights into the surface morphology and topography of the hydrochar. Interestingly, a clear trend emerges: as the operating conditions become more rigorous (higher temperatures and longer durations), the SEM images reveal the formation of

cracks on the hydrochar surface (Figure 7b). This observation suggests that HTC can induce significant structural changes within the hydrochar.

Figure 8 depicts the results of the energy-dispersive X-ray spectroscopy (EDS) analysis performed on the hydrochar samples. This technique provides information about the elemental



**Figure 8.** Spectrum EDS results: (a) temperature 180 °C, process time 30 minute, ratio 0.75; (b) temperature 220 °C, process time 90 minute, ratio 0.75

compositions of a material. As expected, carbon emerges as the dominant element, with its content ranging from 66.33wt% to 71.37wt%. This high carbon content aligns with the classification of hydrochar as a coal-like material with a high heating value (Chen et al., 2021; Eriska et al., 2017; Fakkaew et al., 2018). The observed decrease in the oxygen content alongside the increase in the carbon content further supports this notion. This trend can be attributed to the series of reactions occurring during HTC, such as hydrolysis, dehydration, and decarboxylation, which ultimately lead to the enrichment of carbon within the hydrochar. The findings from the proximate and microstructural analyses paint a comprehensive picture of how the HTC process parameters influence the quality of the hydrochar. The observed trends in the moisture, volatile matter, ash, and fixed carbon content highlight the ability of HTC to enhance the energy content of the hydrochar. Additionally, SEM analysis reveals the structural changes induced by HTC, whereas EDS confirms the enrichment of carbon, a key contributor to the high heating value of the hydrochar.

The comparative EDS spectra presented in Figure 8 provide compelling evidence of a thermally induced compositional alteration. Spectrum 1, obtained after processing at 180 °C for 30 minutes, reveals the presence of a multi-elemental composition including oxygen, calcium, aluminum, silicon, chlorine, and iron, suggesting a complex initial material. However, upon increasing the processing temperature to 220 °C and extending the duration to 90 minutes in Spectrum 2, a significant transformation occurs, marked by the complete disappearance of calcium and iron signals, alongside a relative increase in the weight percentages of the remaining elements (oxygen, aluminum, silicon, and chlorine). This observation strongly indicates that the elevated thermal conditions facilitate either the decomposition and volatilization of calcium and iron-containing compounds or their diffusion into the bulk material beyond the detection limit of surface-sensitive EDS. The higher total weight percentage in Spectrum 2 (approximately 80%) compared to Spectrum 1 (approximately 70%) further implies a relative enrichment of the remaining elements on the surface after the thermal treatment. The overall increase in the detected weight percentage in Spectrum 2 further supports a surface enrichment of the more thermally stable oxide or silicate components. These findings highlight the significant influence of processing temperature

and time on the surface elemental composition of the material, which could have implications for its resulting microstructure, phase distribution, and ultimately, its functional properties.

This study focused on the optimization of the HTC process itself. Further investigation into the effects of pre-treatment (such as sorting inert materials) and post-treatment (such as washing to reduce ash, pelletization for fuel standardization, or activation to produce activated carbon) was not performed; however, it constitutes an important next step for optimizing the resulting hydrochar for specific value-added applications.

### Proposed mechanistic pathway

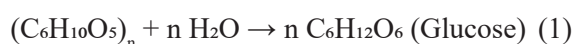
In general, HTC is a thermochemical process that converts wet lignocellulosic biomass into hydrochar, a solid carbon-rich material, through treatment with subcritical water at moderate temperatures (180–250 °C) and elevated pressures. This transformation occurs in an aqueous environment, where water acts both as a solvent and a reactant, facilitating significant changes in the chemical structure of the biomass constituents: cellulose, hemicellulose, and lignin. In our study, we investigate the interactions of lignocellulosic biomass with wastewater containing surfactants, using Linear Alkylbenzene Sulfonate (LAS) as a model for analyzing the complexity of reaction pathways in HTC systems.

Lignocellulosic biomass comprises three key polymers: cellulose, hemicellulose, and lignin. Cellulose is a linear polymer made of  $\beta$ -1,4-linked glucose units, which provides structural integrity to plant cell walls (Gu et al., 2015). Hemicellulose is a heterogeneous polymer composed of various sugars such as xylose and mannose, exhibiting a branched structure that makes it less crystalline and more susceptible to hydrolysis (Liu et al., 2019). Lignin is a complex, three-dimensional phenolic polymer that plays a critical role in providing structural stiffness and resistance to degradation in plants (Petrović et al., 2021). The structural diversity and varying bond strengths in these polymers influence their behavior during the HTC process and significantly contribute to the characteristics of the resulting hydrochar.

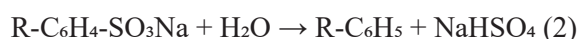
In our work, the integration of LAS into the HTC reaction introduces additional complexities. LAS, an anionic surfactant characterized by a hydrophilic sulfonate group and a hydrophobic alkyl chain, can interact with the lignocellulosic

components through both hydrophobic and ionic interactions (Zhu et al., 2015). The presence of these surfactants can alter the morphological and chemical properties of the biomass during HTC, affecting biomass conversion efficiency and the properties of the produced hydrochar.

Our proposed mechanism for the HTC of lignocellulosic biomass in the presence of LAS involves several key reaction stages. The initial phase consists of hydrolysis and degradation of lignocellulose, where subcritical water breaks glycosidic bonds in cellulose and hemicellulose, leading to the release of monosaccharides (W. Yan et al., 2014). The hydrolysis of cellulose can be represented by the following reaction:



In the case of hemicellulose, degradation produces a diverse range of monosaccharides such as xylose and manose, enriching the resultant reaction mixture with various sugar types, further complicating the reaction dynamics (Rosly et al., 2023). Lignin undergoes partial depolymerization, generating fragments with lower molecular weight that contribute to a higher content of oxygenated functional groups, such as  $-OH$  and  $-COOH$ , which can influence subsequent reactions and interactions with surfactants. The second phase concerns the degradation of LAS itself, which can undergo hydrolysis, generating alkylbenzenes and sulfate ions. The hydrolysis of LAS is described by the simplified reaction:



The degradation of LAS facilitates interactions with the increasingly complex lignocellulosic matrix, potentially affecting oxidation and functional group modification on the biomass (Liu et al., 2020). The interactions between the alkyl chains of LAS and the hydrophobic regions of lignin and hemicellulose may promote unique synergies during HTC, warranting further investigation to identify the intricate pathways at play.

During HTC of lignocellulosic waste co-processed with laundry wastewater, the interaction between degraded biomass fragments and LAS surfactants plays a crucial role in product formation. HTC-induced depolymerization of lignocellulose yields fragments with phenolic and carboxylate functional groups, while LAS, characterized by hydrophilic and hydrophobic domains, engages in both hydrophobic and electrostatic interactions with these fragments. Hydrophobic interactions

between LAS alkyl tails and non-polar regions of lignocellulose drive aggregation, while electrostatic interactions, potentially modulated by pH and bridging ions, occur between LAS sulfonate heads and charged functional groups on lignocellulose. These interactions significantly alter the morphology and surface properties of the biomass, influencing HTC reaction kinetics and hydrochar characteristics such as porosity and surface area. The subsequent stage involves the dehydration of monosaccharides into primary furan derivatives, such as hydroxymethylfurfural (HMF) and furfural. This dehydration can be expressed as follows:

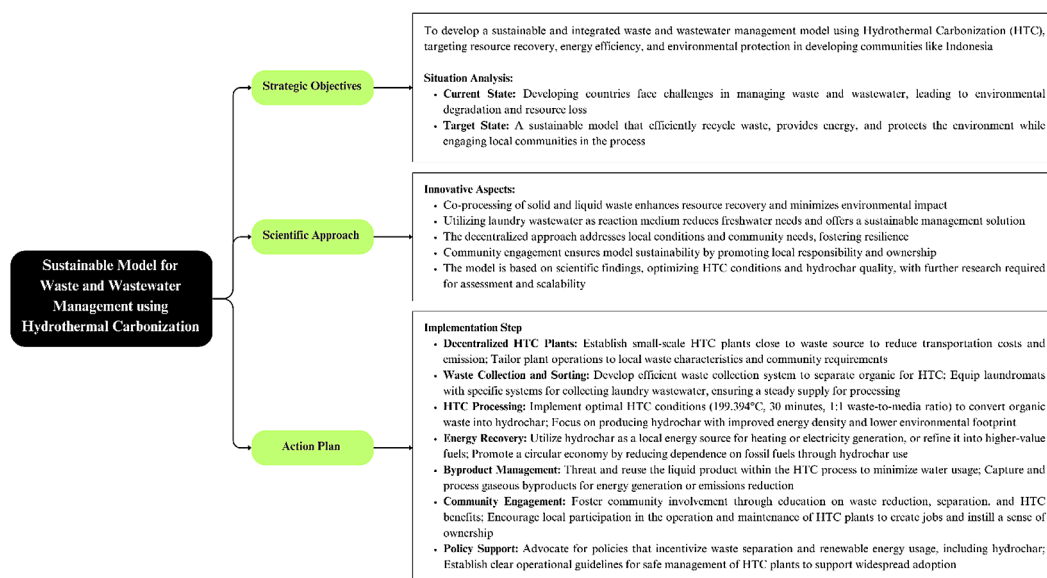


These furan derivatives may undergo further fragmentation, resulting in low-molecular-weight volatile compounds like acetic acid and gases, primarily  $CO_2$ , that contribute to the overall product distribution during HTC (Zhao et al., 2021). The presence of surfactants, like LAS, influences the solubility and stability of these intermediates in the aqueous phase, altering their reactivity and the final hydrochar composition (Ennis et al., 2017). As the HTC reaction progresses, the interactions of dehydrated sugars, lignin fragments, and surfactant degradation products lead to a range of condensation and polymerization reactions. These reactions are driven by collision frequency, temperature, and potential stabilization offered by surfactant molecules, which may preferentially aggregate certain reaction species and enhance hydrochar formation (Sun et al., 2023). Rates and mechanisms of these reactions can vary significantly based on reaction conditions, particularly temperature, which has been shown to significantly affect the structural properties of the resulting hydrochars (Yang et al., 2022). Thus, this study offers critical insights into the mechanistic pathways involved in HTC using complex feedstocks like lignocellulosic waste combined with wastewater surfactants. Future research will focus on deeper kinetic analyses and exploration of by-products to optimize the benefits of HTC in energy recovery and waste valorization, aiming towards ecosystem resilience and sustainability.

### Sustainable model for waste and wastewater management using HTC

A sustainable model (Figure 9) for integrated waste and wastewater management using HTC is proposed for communities in developing countries





**Figure 9.** Sustainable model for integrated waste and wastewater management using HTC

like Indonesia, with a focus on resource recovery, energy efficiency, and environmental protection. This model addresses the pressing need for efficient waste management and renewable energy generation in regions with inadequate waste management systems. This sustainable model for waste and wastewater management using HTC is proposed as an integrated solution for developing countries, specifically targeting traditional market waste and laundry wastewater. The model champions a decentralized approach by establishing small-scale HTC facilities near waste sources, such as markets and communal laundromats. This decentralized strategy aims to reduce transportation costs and emissions, enhance adaptation to local conditions, and foster active community involvement in waste management.

A key advantage of this model lies in the co-processing of solid organic waste from markets (like food scraps and packaging) and laundry wastewater. Laundry wastewater not only serves as a reaction medium in the HTC process, thereby reducing freshwater needs, but also has the potential to enhance process efficiency and hydrochar product quality due to its alkalinity and nutrient content. The model emphasizes optimizing HTC operating conditions to produce high-quality hydrochar as the primary output, which can be utilized as a renewable energy source, including for electricity generation. Implementation of this model involves several key steps, starting with the establishment of decentralized HTC facilities, developing efficient organic waste collection and

sorting systems, and forming “Ecolo-laundromats” for laundry wastewater collection. Furthermore, the model incorporates strategies for managing by-products, such as water and gases generated during the HTC process, focusing on recycling and reuse. The success of this model also heavily relies on supportive policies that incentivize waste separation and renewable energy utilization, as well as clear operational guidelines to ensure the safety and sustainability of HTC facility operations. This decentralized HTC model offers an innovative and practical solution to address the pressing waste and wastewater management challenges in developing countries. By focusing on a communal scale, co-processing, community engagement, and local resource utilization, this model has the potential to realize a sustainable waste management system, enhance resource recovery, reduce environmental impacts, and empower communities economically and environmentally.

However, scaling this hydrothermal co-processing of market waste and laundry wastewater to an industrial level presents significant challenges. Key hurdles include designing robust, continuous high-pressure/temperature reactors, managing heterogeneous and variable feedstock mixtures (including pre-treatment and slurry transport), and achieving energy efficiency, likely through effective heat recovery. Additionally, treating the complex byproduct water stream and ensuring overall economic viability against potentially high capital and operational costs are critical for successful large-scale implementation.

## CONCLUSIONS

The HTC of market waste using laundry wastewater as a medium offers a promising approach for waste management and renewable energy generation. This study demonstrates that HTC effectively converts market waste into a carbon-rich hydrochar product with potential as an alternative fuel to coal. The energy value and carbon content of the hydrochar increased with increasing temperature, process time, and water volume, whereas the solid MY, water content, ash content, and oxygen content decreased. The optimal HTC conditions for this study were a temperature of 199.394 °C, a 30-min processing time, and a 1:1 waste-to-media ratio. Further research is needed to investigate the influence of laundry wastewater on the hydrochar properties beyond carbon content, analyze liquid and gas byproducts, and optimize HTC processes for maximum sustainability and efficiency. This research highlights the potential of HTC with laundry wastewater to transform waste into a valuable resource and contribute to a more sustainable future.

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