Effect of torrefaction on fuel properties of biopellets

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A B S T R A C T

The study aimed to determine the effects of torrefaction on the fuel properties of pellets. Therefore, firstly, torrefaction parameters of rose (*Rosa Damascena* Mill.) oil distillation solid waste and red pine sawdust were determined through the torrefaction optimization process in terms of temperature and holding time. Then, using the selected torrefaction parameters, 14 different raw and torrefied pellets containing RP, PS, and Turkish Elbistan Lignite were prepared in different weight ratios. Finally, the fuel properties of the prepared raw and torrefied pellets, namely dimensions, proximate analyses, higher heating values, tensile strength, durability, abrasive resistance, and water uptake resistances, were investigated. The findings demonstrated that the higher heating values and carbon content of raw biomass samples increased while their volatile matter content decreased. The use of lignite at high concentrations led to an increase in ash content and a decrease in the strength and durability of pellets, which should be emphasized. In addition, red pine sawdust was used in place of solid waste from rose oil distillation solid waste to produce pellets with greater strength. All pellet mixtures with torrefaction had higher heating values and energy densities despite the fact that their mass and energy efficiency had decreased. It was determined that torrefaction increased the pellets' resistance to absorbing water and gave them a more hydrophobic structure. Thus, it was determined that torrefaction could enhance the crucial fuel parameters of the biomass samples.

1. Introduction

The industrial revolution contributed to economic development, resulting in an increase in employment, production of manufactured goods, and consumption of the population. Although the economic growth promoted by the governments is an essential indicator of the level of welfare, the negative side of economic growth is the irreversible environmental damage due to rising energy-intensive production [1]. In recent decades with the economic expansion and booming population, the use of natural resources, energy consumption, and CO_2 emissions have been on the rise in parallel. CO_2 is one of the most abundant greenhouse gases and is mainly responsible for global warming. The IPCC reports drawing attention to the rapid increase of global CO_2 levels after the industrial revolution [2]. Currently, the global CO_2 emission is about 37 billion metric tons, and the energy sector contributing 35 %, is one of the

biggest responsible for these emissions [3]. The global energy sector mainly depends on the combustion of conventional fossil fuels. Fossil fuels meet 80 % of global energy demand in 2021 [4]. It has been frequently discussed the adverse effect of the energy sector on climatic change, and conclude that if we have to diminish CO_2 emissions and therefore combat global warming, we have to use alternative carbon-neutral energy sources [5–7]. Using carbon-neutral energy sources is crucial, contributing to achieving the long-time Paris Agreement goals, which maintain the global temperature increase to 2 °C compared to the pre-industrialization period.

Türkiye first accepted the net zero emissions goal for 2053 and then signed the Paris Agreement on October 7, 2021 [8]. To reach the 2053 target, Türkiye plans to mitigate its emissions by 41 % from its current state by 2030 [9]. Increasing carbon-neutral energy investments can be suggested as a viable solution to achieving emission targets, providing new employment opportunities, and sustaining economic growth [10,11]. Among them, biomass is defined as a continuously available energy source due to its huge quantity and ease to reach its sources in nature [12,13]. Therefore, it could be a good alternative. Although the usage of biomass as a new alternative energy source is not possible to meet the whole energy needs of our country, biomass fuels will help to solve the domestic energy needs of people living in rural areas where 40 % of the country's population lives [14]. Some biomass types, like woody origin biomass, are more suitable for combustion in rural areas since there is no need for initial investment using the same equipment as fossil fuels. However, with their moisture content and properties such as low bulk density and, poor grindability, low energy density, woody biomass may not be economically feasible for direct combustion [12,15,16] as well as improper storage conditions can cause environmental problems and lead to high transportation costs [17]. Pelletization is the firstly applied process to decline the moisture content and accordingly increase the density of biomass. The torrefaction is a thermal pretreatment process applied to pelletized biomass to enhance fuel and burning properties. The hemicellulosic or lignocellulosic content of the biomass slowly decomposes during torrefaction, usually at temperatures between 200 and 300 °C in an inert environment. Biofuel loses 20–25 % of its weight during the process while gaining roughly 25–30 % of its energy density [18]. Torrefied biomass has a higher energy content in terms of higher heating value (HHV), better hydrophobicity, rot resistance, and a long storage period [19,20]. In addition, among the thermochemical conversion processes of biofuels, torrefaction is accepted as the process with the lowest global warming potential [20]. As a result, torrefaction plays an essential role in lowering our energy deficit and boosting energy production from current biomass sources with reduced carbon emissions for Türkiye. In light of these figures, co-combustion of low-quality local coals together with torrefied biomass in Türkiye will provide environmentally friendly disposal of biomass and coal blends with improving their heating value [20]. The central rose (Rosa Damascena Mill.) growing countries are Türkiye and Bulgaria. Türkiye produces more than 65 % of the world's rose oil, rose concrete, and rose absolute. Each year about 2 tons of rose oil, 10 tons of rose concrete, and 2 tons of absolute from 4100 ha are produced in Türkiye [21]. Almost the entire production area of rose flowers is located in the province of Isparta in Türkiye. As a result of this activity, an important amount of rose oil distillation waste is produced. Annual produced rose oil distillation wastes have been calculated as approximately 26 000 tons in Isparta [22]. The solid wastes resulting from the production of the rose oil distillation are uncontrollably discharged into soil opened around the facility. This situation constitutes an important source of pollution for surface soil and groundwater in Isparta. On the other hand, the amount of sawn timber produced sawn wood from coniferous trees was 6 150 000 m^3 in 2021 in Türkiye [23]. Therefore, waste wood from coniferous trees and rose oil processing wastes constitute an essential part of the woody biomass potential of Türkiye. Consequently, it is vital to know the torrefaction parameters to increase woody biomass's heating properties, activities, and hygroscopic characteristics [24].

Optimization is essential in the torrefaction process, which increases the fuel and combustion process's efficiency and reduces the storage cost while producing energy from renewable energy sources. Temperature and holding time are the essential factors of the torrefaction optimization process because these two parameters are used for designing and constructing industrial torrefaction systems [25]. Although there are few studies on the pelletization and torrefaction of rose oil distillation wastes and their various mixtures [17, 26] to the best of our knowledge, there is no study in the literature that examines the fuel properties of pellets produced as a result of blends of torrefied rose pulp with lignite. Furthermore, there are no studies evaluating the torrefaction parameters of rose oil distillation wastes to improve the fuel properties of this waste. Therefore, firstly torrefaction parameters of the rose (Rosa Damascena Mill.) oil distillation solid waste (RP) and red pine sawdust (PS) were examined, and then by using the selected torrefaction parameters, 14 different raw and torrefied pellets containing rose oil distillation solid waste, red pine sawdust, and Turkish Elbistan Lignite were

Table 1
Fuel mixing ratios of the prepared pellets (by weight).

Pellets	PS (%)	RP (%)	L (%)
50PS-50RP-0L	50	50	0
49PS-49RP-2L	49	49	2
48PS-48RP-4L	48	48	4
45PS-45RP-10L	45	45	10
0 PS-25RP-75L	0	25	75
0 PS-75RP-25L	0	75	25
98PS-0RP-2L	98	0	2
96PS-0RP-4L	96	0	4
90PS-0RP-10L	90	0	10
0 PS-98RP-2L	0	98	2
0 PS-96RP-4L	0	96	4
0 PS-90RP-10L	0	90	10
100PS-0RP-0L	100	0	0
0 PS-100RP-0L	0	100	0

prepared in different weight ratios. Finally, the fuel properties of the prepared raw and torrefied pellets, namely dimensions, proximate analyses, higher heating values (HHVs), tensile strength, durability, abrasive resistance, and water uptake resistances, were investigated to understand the effect of torrefaction on the fuel properties of the prepared pellets.

2. Material and methods

In the study, PS and RP were maintained respectively from a local sawmill plant and a rose oil factory in Isparta, Türkiye. Elbistan Lignite (L), a local lignite, was also used to prepare fuel mixtures. While the RP had high moisture content (65 % $\pm$ 1.2 %) and was bulky, it was dried at 70 °C for 3 h in the oven to eliminate microbial activity before the experiments. Furthermore, biomass and lignite were crushed and sieved to 2 mm at room temperature for pelletization. In the study, a custom-made pellet machine (3 kW) was used to perform the pelleting process. In light of the data obtained in the preliminary experiments, the pellets were prepared using PS, RP, and L at different weight ratios using optimum moisture and binder ratio (50 % moisture and 4 % starch), which was determined in the previous study [26]. In addition, the torrefaction of the prepared pellets using optimum torrefaction temperature and duration was also carried out in the study. Accordingly, 14 raw and torrefied pellets were produced by mixing the biomass and lignite in different weight ratios (Table 1).

Elemental analysis of biomass and lignite were performed using a LECO CHNS-932 elemental analyzer in the study. The ASTM E−872 standard was applied to determine the volatile matter (VM) in the samples. The ash content of the samples was calculated with ASTM D-1102 and ASTM E−1755-01 for wood and biomass, respectively. Moisture content was determined by the ASTM standards D-871-82 and ASTM D317213 standards for biomass and lignite samples, respectively. Fixed carbon (FC) was obtained from the difference [27]. For the calculation of HHV of the samples, correlations that were developed for raw (Equation (1)) and torrefied biomasses (Equation (2)) and lignite samples (Equation (3)) given in the literature were used [28–30];

$$HHV = -10.8146 + 0.3133\,(VM + FC) \tag{1}$$

$$HHV = 0.1846\,VM + 0.3525\,FC \tag{2}$$

$$HHV = 144.54C + 610.2H + 40.5S - \left(65.88 - \left(\frac{30.96O}{100 - A}\right)\right)O \tag{3}$$

where VM and FC are the volatile matter and the fixed carbon content of the samples. C, H, S, O, and A are carbon, hydrogen, sulfur, oxygen, and ash contents of the lignite.

Torrefaction experiments were performed in an ash furnace (Nüve MF 5000). Each biomass sample (about 10 g) was put into a sealed crucible (50 cm^3), and experiments were performed at three temperatures (250, 270, and 290 °C) and three holding times (15, 30, and 60 min) [31,32]. These holding times have been chosen in this study considering both the literature and electricity consumption [33]. As better fuel properties were obtained in the torrefaction process performed between 250 and 300 °C compared to the raw fuel, this temperature interval was also selected in this study [15,16,34], In general, pelleting followed by torrefaction is a more energy-efficient process than a conventional pelleting process and results in pellets with higher HHV and lower moisture content [35]. Accordingly, in this study, firstly pellets were prepared and then torrefied at selected torrefaction temperature and duration.

Mass yield, energy yield, and energy density ratio of the thermally treated samples were obtained using Equations (4)–(6), respectively. The mass yield gives the ratio of the initial mass of the biomass remaining after the torrefaction [36].

$$Mass\ Yield = \frac{M_t}{M_b} \tag{4}$$

where M_t is the mass of torrefied biomass at time t (g), and M_b is the biomass before the torrefaction. The energy yield of the samples was calculated by using Equation (5). Energy yield gives the ratio of the chemical energy in the original dry biomass that remained in the torrefied biomass [36]. Accordingly, it determines how much energy remains after the combustion of samples.

$$Energy\ Yield = \frac{M_t H_2}{M_b H_1} \tag{5}$$

where H_2 is the HHV of the torrefied biomass, H_1 is the HHV of the raw biomass.

The energy density ratio of the samples, which is the fraction of energy emitted from the torrefied product when fully combusted to its original energy level, was also calculated in the study using Equation (5) [36].

$$Energy\ density\ ratio = \frac{H_2}{H_1} \tag{6}$$

The study also examined various fuel characteristics of the prepared pellets. The details of the determination of bulk density, particle density, tensile strength, impact, and water intake resistance of the pellets were given in the previous study [26].

The data sets were analyzed by using the IBM SPSS Statistics 29.0.0.0 software, and the statistical analysis was done following two-way ANOVA and Tukey Test and independent samples t-test depend on the data set for the identification of significant differences among samples. The level of significance was established at p = 0.05.

3. Results and discussion

3.1. Proximate and elemental analysis of raw biomasses

In cases where biomass is used as a fuel for energy supply, the primary chemical components of the biomass need to be identified. The energy released during the combustion process is positively related to the carbon and hydrogen content in the biomass [37]. On the other hand, high oxygen and nitrogen values reduce the calorific value of the biomass, thereby reducing its energy potential [38, 39]. However, the biomass should have low nitrogen and sulfur content because when the biomass is burned, the nitrogen and sulfur cause atmospheric pollution [40,41]. The proximate and elemental analysis results of the biomasses and lignite used in this study are given in Table 2. It should also be noted that oxygen is an essential element in the combustion of the material [37]. In this study, carbon and oxygen content of the PS sample was higher than the RP sample. Furthermore, the nitrogen content of the RP sample (4.14 %) was found to be also higher than the nitrogen content of various lignocellulosic biomass which has been reported in the literature, such as wood, twigs, bark, and leaves (0.69–3.10 %) [37], poplar, rice husk, cotton and straw sunflower corns (0.30–0.70 %) [42], tobacco, maize, oat, barley and sunflower stalks and corn cobs and straw (0.41%–1.65 %) [43] and coffee bark and eucalyptus (0.48–2.74 %) [44]. In a study where 26 Turkish lignites were examined, the S content of lignite samples varied between 0.7 % (Ilgın) and 9.1 % (Beypazari) [45]. In the same study, the C contents of local lignites changed between 60 % (Beyşehir) and 74 % (Yeniçeltik), H contents of 4.6 % (Elbistan) and 5.8 % (Yeniçeltik), N of 1.4 % (Söke) and 2.8 % (Yeniçeltik) [45]. Accordingly, the C and H contents of Elbistan lignite used in this study were lower than the Turkish lignites studied in the literature [45]. The HHV of Turkish lignites was reported to vary between 22.4 MJ/kg (Beyşehir) and 29.5 MJ/kg (Yeniçeltik) in the literature [45]. The HHV of Elbistan lignite used in this study was found to be 23.36 MJ/kg, which is between the HHV given in the literature but close to the HHV given in the literature.

3.2. Torrefaction of raw biomasses

The proximate analysis results of the RP and PS samples for the various torrefaction parameters are given in Tables 3 and 4, respectively. Three samples were used for each analysis, and average values (avg ± stdev) are presented in these tables. As shown in Tables 3 and 4, moisture loss from all samples was substantial at all temperatures and holding times. At 250 °C and 15 min, the moisture loss of the RP and PS samples was 85.92 % and 49.71 % compared to their raw RP and PS, respectively. With the increase in holding time, the moisture loss reached 89.92 % and 73.02 % for the RP and PS samples at the same temperature at 60 min, respectively. Although moisture loss increased with temperature and holding time, this increase could not be found statistically significant ($p > 0.05$) for both RP and PS samples. The highest moisture loss was observed for the RP and PS samples at 270 °C and 60 min (99.68 %) and 290 °C and 60 min (96.04 %), respectively.

Increases in temperature and holding time caused lower VM ($p > 0.05$) in the samples. Accordingly, the average VM of the RP samples was 74.56 ± 1.70 % (torrefied at 250 °C), 71.83 ± 6.08 % (torrefied at 270 °C), and 68.84 ± 7.93 % (torrefied at 290 °C). The same trend was also observed for the PS samples, and the average VM of the PS samples were 76.37 ± 1.07 % (torrefied at 250 °C), 75.37 ± 1.60 % (torrefied at 270 °C) and 72.04 ± 4.69 % (torrefied at 290 °C). Although the thermal degradation of hemicellulose and cellulose starts at 150 °C and 275 °C, respectively, lignin decomposition occurs at higher temperatures [46]. Gaseous and liquid byproducts increase when the torrefaction temperature rises, owing to a reduction in solid byproducts [47,48]. Accordingly, lower VM was observed with the increasing torrefaction temperature for the biomass samples in the study. The findings were consistent with the literature data [49–51].

The FC of the RP and PS samples was 9.56 ± 2.33 % and 13.9 ± 2.42 %, respectively. With increasing torrefaction temperature, FC of the RP samples increased ($p > 0.05$), and the average values were 16.99 ± 1.17 % (torrefied at 250 °C), 19.39 ± 5.07 % (torrefied at 270 °C), and 21.67 ± 6.35 % (torrefied at 290 °C). Although the FC contents of both RP and PS increased with temperature, the increase in PS samples was statistically significant ($p < 0.05$). The average FC of the PS samples was found to be 18.59 ± 1.62 % (torrefied at 250 °C), 19.77 ± 0.07 % (torrefied at 270 °C), and 21.60 ± 2.88 % (torrefied at 290 °C). Accordingly, the FC of the RP and PS doubled at 290 °C. In the literature, similar results were also found by Tumuluru (2015) [50]. On the other hand, during the torrefaction process, the ash content of both the RP and PS samples increased with the temperature; however, it was statistically not significant ($p > 0.05$). The increase in the ash content of the PS samples varied between 1.39 % and 365 %, and the highest ash content was observed at 290 °C. The increase in the ash content of the RP samples continued with torrefaction temperature, and it was found in

Table 2
Proximate and elemental composition of the samples.

	Moisture (%)	VM (%)	FC*** (%)	Ash (%)	HHV**** (MJ/kg)
PS[a]	6.82 ± 2.93	77.1 ± 4.28	13.9 ± 2.42	2.15 ± 0.57	19.8
RP**	6.25 ± 2.17	77.4 ± 4.85	9.56 ± 2.33	6.75 ± 0.46	18.3
L	20.93 ± 1.5	43.3 ± 3.24	15.63 ± 2.23	19.71 ± 0.4	23.35
	% C	% H	% N	% S	% O***
PS[a]	48.2	6.40	–	–	44.2
RP**	45.6	6.02	4.14	0.29	37.2
L	35.14	3.82	1.99	2.55	36.79

[a] as received, **pre-dried, ***calculated by using the difference, ash free, **** calculated on a dry basis, and since average values are used, only one HHV value is given.

Table 3
Effect of torrefaction parameters on PS samples.

Torrefaction						
Temperature (°C)	Holding time (min)	Moisture (%)	VM (%)	FC (%)	Ash (%)	HHV[a] (MJ/kg)
Raw PS		6.82 ± 2.93	77.1 ± 4.28	13.9 ± 2.42	2.15 ± 0.57	19.8
250	15	3.43 ± 1.87	77.0 ± 3.95	17.4 ± 1.48	2.21 ± 0.52	21.1
	30	2.85 ± 1.21	76.9 ± 4.15	18.0 ± 0.46	2.18 ± 0.18	21.1
	60	1.84 ± 0.15	75.1 ± 3.92	20.4 ± 2.47	2.59 ± 0.10	21.5
270	15	3.04 ± 1.19	77.1 ± 4.34	17.7 ± 1.54	2.18 ± 0.11	21.1
	30	2.74 ± 0.92	75.1 ± 3.21	20.0 ± 1.21	2.26 ± 0.19	21.5
	60	2.04 ± 0.05	74.0 ± 2.89	21.7 ± 1.23	2.32 ± 0.21	21.7
290	15	1.47 ± 0.95	76.2 ± 3.14	19.7 ± 1.37	2.61 ± 0.07	21.3
	30	2.49 ± 0.07	73.0 ± 2.93	20.2 ± 1.20	4.37 ± 0.14	21.1
	60	0.27 ± 0.03	67.0 ± 1.76	24.9 ± 2.34	7.86 ± 2.11	21.2

[a] calculated on a dry basis, and since average values are used only one HHV value is given.

Table 4
Effect of torrefaction variables on RP samples.

Torrefaction						
Temperature (°C)	Holding time (min)	Moisture (%)	VM (%)	FC (%)	Ash (%)	HHV[a] (MJ/kg)
Raw RP		6.25 ± 2.17	77.4 ± 4.85	9.56 ± 2.33	6.75 ± 0.46	18.3
250	15	0.88 ± 0.63	76.4 ± 4.06	15.7 ± 3.63	6.98 ± 0.21	19.9
	30	0.27 ± 0.27	74.1 ± 8.12	17.5 ± 7.66	7.84 ± 0.74	19.9
	60	0.63 ± 0,80	76.01 ± 6,11	17.75 ± 6.30	8.70 ± 0.62	19.9
270	15	0.40 ± 0.31	77.4 ± 6.39	14.68 ± 6.07	7.53 ± 0.63	19.5
	30	0.67 ± 0.06	72.8 ± 7.37	18.72 ± 7.14	7.83 ± 0.17	20.2
	60	0.02 ± 0.02	65.3 ± 6.54	24.8 ± 6.33	9.87 ± 0.19	20.8
290	15	0.42 ± 0.54	74.0 ± 5.65	17.6 ± 5.80	7.97 ± 0.01	20.0
	30	0.16 ± 0.16	72.8 ± 1.68	18.4 ± 0.52	8.63 ± 1.31	20.0
	60	0.06 ± 0.04	59.7 ± 5.04	29.0 ± 5.08	11.2 ± 0.08	21.3

[a] calculated on a dry basis, and since average values are used, only one HHV value is given.

the range of 11.5%–46.2 % and 18.1%–66.7 % at 270 °C and 290 °C, respectively. The RP samples in this study had greater ash content, likely attributable to a decrease in VM [49–51]. Since FC of the samples increased, and VM of the samples with lower energy content decreased, HHV values of the biomass samples displayed higher values [34]. After torrefaction, the average HHV of the RP samples was increased with the increasing torrefaction temperature ($p < 0.05$) and found to be 19.9 ± 0.05 MJ/kg, 20.2 ± 0.63 MJ/kg, and 20.4 ± 0.75 MJ/kg for the 250, 270, and 290 °C torrefaction temperatures, respectively. On the other hand, although, the average HHVs of the PS samples were increased with the temperature, the increase could not be found significant ($p > 0.05$) and the values were 21.2 ± 0.21 MJ/kg, 21.4 ± 0.19 MJ/kg, and 21.4 ± 0.21 MJ/kg for the 250 °C, 270 °C, and 290 °C, respectively.

Mass yield, energy yield, and energy density ratios of the biomass samples were also calculated to obtain the ideal torrefaction temperature of the RP and PS samples (Fig. 1, Fig. 2, and Fig. 3). At 250 °C at 15 min, the mass yield of the RP and PS samples were 89 % and 90 %, respectively (Fig. 1). With the increasing holding time, the mass yield of the biomasses decreased to 75 % and 82 % at the same temperature for the RP and PS samples, respectively. Therefore, mass yields decreased with the both torrefaction temperature and holding time ($p < 0.05$), and the lowest mass yield was observed at 290 °C at 60 min for both the RP (57 %) and PS (63 %). At higher temperatures, due to an increase in moisture loss and thermal decomposition (i.e., depolymerization of the wood components), the mass yield of lignocellulosic samples decreased [34,52,53]. As the degradation rate of agricultural residues is relatively higher than that of woody biomass due to their higher hemicellulose contents, lower mass yields were obtained for the RP samples as compared to the PS samples in this study. Like the mass yield, energy yields of the biomass samples also decreased ($p < 0.05$) with both torrefaction
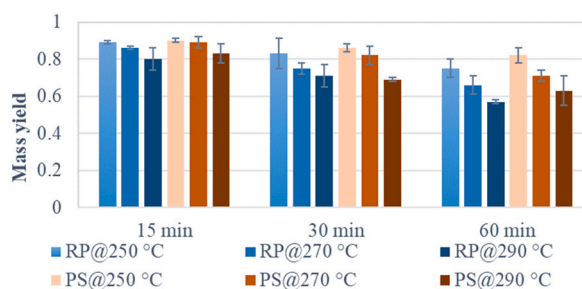

Fig. 1. The effect of holding time and temperature on the mass yield of the RP and PS samples.
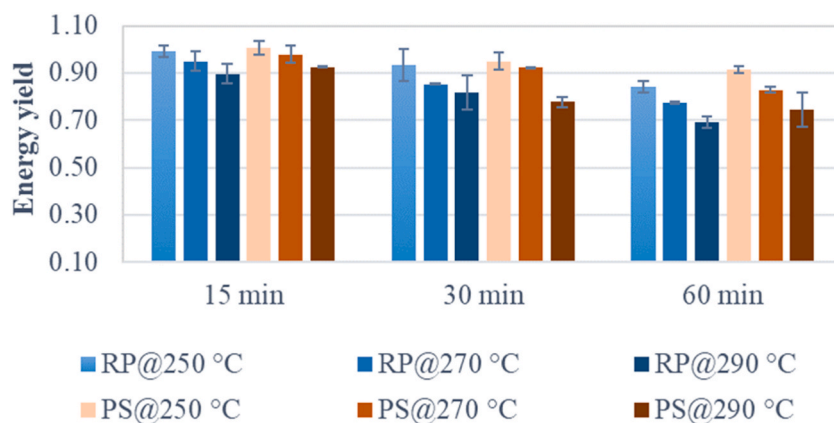
Fig. 2. The effect of holding time and temperature on the energy yield of the RP and PS samples.
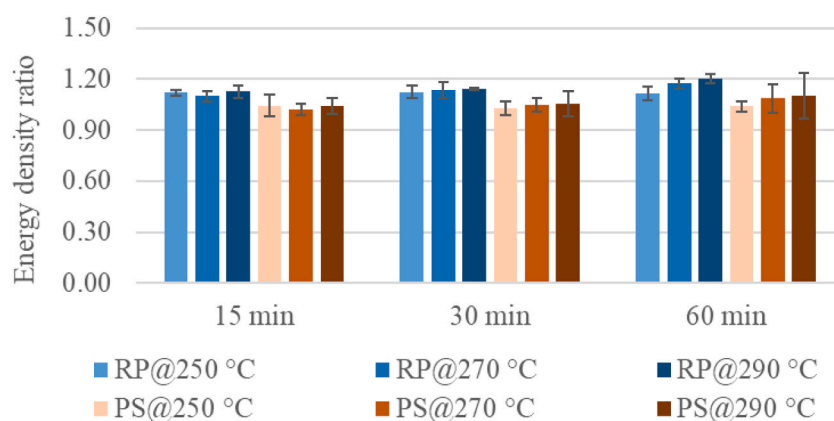

Fig. 3. The effect of holding time and temperature on the energy density ratio of the RP and PS samples.

temperature and holding time. It was in the range of 99 % (at 250 °C and 15 min) - 69 % (at 290 °C and 60 min), and 101 % (at 250 °C and 15 min) - 75 % (at 290 °C and 60 min) for the RP and PS samples, respectively (Fig. 2). Fig. 3 shows that at lower torrefaction temperatures (250 °C), the effect of the holding time on energy density ratios was found to be low for both the RP and PS samples. On the other hand, at higher temperatures at 60 min, energy density ratios increased from 1.12 to 1.21 and from 1.04 to 1.10 for the RP and PS samples, respectively. However, these increases were statistically insignificant ($p > 0.05$), probably due to small increases in HHV of the biomasses. When Figs. 1–3 were examined, the results obtained at 270 °C and 290 °C were close to each other.

Table 5
The results of proximate analysis of the raw pellets.

Pellets	Moisture (%)	VM (%)	FC (%)	Ash (%)
100PS-0RP-0L	7.18 ± 0.03	78.46 ± 1.23	12.29 ± 0.32	2.07 ± 0.06
0 PS-100RP-0L	9.14 ± 0.12	68.74 ± 1.14	15.76 ± 0.27	6.36 ± 0.03
0 PS-98RP-2L	6.03 ± 0.39	69.45 ± 1.22	16.95 ± 1.68	7.57 ± 0.07
0 PS-96RP-4L	6.46 ± 0.19	69.02 ± 0.85	16.93 ± 0.82	7.59 ± 0.22
0 PS-90RP-10L	7.01 ± 0.55	67.63 ± 1.35	17.76 ± 2.02	7.59 ± 0.13
0 PS-75RP-25L	8.11 ± 0.14	65.10 ± 3.33	18.25 ± 3.75	8.54 ± 0.29
0 PS-50RP-50L	8.55 ± 0.45	63.43 ± 0.52	18.43 ± 1.06	9.59 ± 0.09
98PS-0RP-2L	6.84 ± 0.16	78.31 ± 2.55	12.87 ± 1.18	2.09 ± 1.22
96PS-0RP-4L	6.53 ± 0.09	78.06 ± 0.41	13.76 ± 0.41	1.65 ± 0.09
90PS-0RP-10L	6.22 ± 0.18	77.65 ± 0.37	13.86 ± 0.56	2.27 ± 0.01
75PS-25RP-0L	6.14 ± 0.35	75.37 ± 1.82	15.60 ± 2.09	2.89 ± 0.08
50PS-50RP-0L	5.88 ± 0.34	76.84 ± 1.75	13.32 ± 2.03	3.96 ± 0.05
25PS-75RP-0L	6.67 ± 0.32	70.52 ± 2.84	16.93 ± 2.53	5.88 ± 0.01
49PS-49RP-2L	5.82 ± 0.21	76.64 ± 3.48	13.28 ± 3.12	4.26 ± 0.15
48PS-48RP-4L	5.60 ± 0.25	75.91 ± 2.0	13.90 ± 2.24	4.59 ± 0.01
45PS-45RP-10L	6.21 ± 0.25	74.23 ± 1.40	14.34 ± 1.21	5.21 ± 0.05

Nevertheless, due to energy consumption and similar higher energy density ratios, 270 °C and 1 h were chosen as the optimum temperature and holding time for both samples.

3.3. Fuel properties of the raw and torrefied pellets

3.3.1. Proximate analysis results of the prepared pellets

The results proximate analysis of raw and torrefied pellets are given in Tables 5 and 6, respectively. Moisture content is a factor that negatively affects pellet properties, such as bulk density and durability during storage and transportation. Furthermore, it reduces energy consumption during pelletization [54]. The moisture content of the raw pellets was found to be between 5.60 % and 9.14 %. This range is below 15 %, which is the limit value according to ISO 18122 (2015) and ISO 18134-2 (2017) standards, and below 10 % according to EN-14961-2 (2013) European pellet standards. Furthermore [55], also reported that a moisture content of 5%–12 % is required to obtain good-quality pellets. Similar results were also reported by Ungureanu et al. (2018) [54]. They stated in their study that the durability of pellets with 5 % moisture content is low. In addition, it was stated in the same study that higher moisture contents (>15 %) give damage the pellets during storage. Shankar Tumuluru et al. (2011) [56] stated in their study that the moisture content of the wood pellets was between 7 % and 10 %, while the moisture content of the torrefied pellets decreased to 1%–5%. Similarly, in the study, moisture contents of the raw pellets were significantly higher than all torrefied pellets (p < 0.05) (Table 6).

The VM of the raw pellets varied between 63.43 % (0 PS-50RP-50L) and 78.46 % (100PS-0RP-0L). After torrefaction, it was observed that the amount of VM in all mixtures decreased (p < 0.05) in parallel with the decrease in hemicellulose ratio [57] and changed between 55.27 % (0 PS-50RP-50L) and 77.74 % (100PS-0RP-0L). Shankar Tumuluru et al. (2011) [56] reported that while the VM of wood pellets varied between 70% and 75 %, the VM of torrefied wood pellets decreased and ranged between 55% and 65 %. The decrease in VM in the pellets due to torrefaction was between 0.18 % (98PS-0RP-2L) and 14.71 % (48PS-48RP-2L). It was observed that as the amount of lignite in the mixtures increased, VM decreased; on the other hand, as the amount of the RP and PS increased, in other words, as the amount of biomass increased, VM of the pellets increased due to high VM of the biomass.

In general, the calorific value of a carbonaceous fuel is a function of FC, and the HHV increases as the FC of the fuel increases [45]. As a result of the degradation of organic matter (e.g., hemicellulose) in the pellet, the content of lignin, which is rich in carbon, increases in biomass. Lignin generally contains C–C bonds, and since the energy densities of these bonds are higher than C–H and C–O bonds, higher HHV is obtained due to torrefaction [58]. Generally, it is understood that as the RP and lignite content increased, FC of the prepared fuel mixtures also increased. The highest FC was obtained in pellet mixtures prepared from 0 PS-50RP-50L (18.43 %) fuel mixture and 0 PS-75RP-25L (18.25 %). Therefore, FC of the prepared pellets increased significantly (p < 0.05) in all blends with the torrefaction process. Shankar Tumuluru et al. (2011) [56] also found similar results, and it was stated that the FC of the raw wood pellets was between 20% and 25 %, while the FC of the torrefied wood pellets increased and reached 28%–35 %. In general, the moisture content of the pellets was decreased. On the other hand, FC increased with torrefaction. Therefore, the quality of the prepared pellets was improved with the torrefaction [59].

In the study, the ash content of the raw pellets varied between 1.65 % and 9.59 %. The ash content of all the pellet mixtures was found to be below the limit value of 10 %, which provides the standard ISO 18122. After torrefaction, the amount of ash increased (p > 0.05) and varied between 1.56 % (98PS-0RP-2L) and 12.01 % (0 PS-50RP-50L). The ash contents of torrefied 0 PS-50RP-50L and 0 PS-75RP-25L mixtures with high lignite content were 10.47 % and 12.01 %, respectively, and remained above the limit value.

3.3.2. Mass yields, energy yields, energy density ratios, and higher heating values of the prepared pellets

Mass and energy efficiency are the most critical qualitative and quantitative parameters in the torrefaction process [60]. Table 7 gives the mass yields, energy yields, energy density ratios, and HHVs of the raw and torrefied pellets. The literature states that the mass yield varies between 50 % and 97 % in general [61,62]. During torrefaction, the loss of carbonyl and carboxyl groups from cellulose,

Table 6
The results of proximate analysis of the torrefied pellets.

Pellets	Moisture (%)	VM (%)	FC (%)	Ash (%)
100PS-0RP-0L	0.55 ± 0.13	77.74 ± 2.12	18.95 ± 0.87	2.76 ± 0.02
0 PS-100RP-0L	0.05 ± 0.18	65.18 ± 1.32	21.88 ± 0.34	9.59 ± 0.04
0 PS-98RP-2L	3.08 ± 0.21	63.66 ± 1.52	23.59 ± 0.91	9.68 ± 0.82
0 PS-96RP-4L	3.08 ± 0.46	62.76 ± 0.42	24.24 ± 0.21	9.92 ± 1.09
0 PS-90RP-10L	3.31 ± 0.13	66.08 ± 2.57	21.17 ± 2.38	9.44 ± 0.32
0 PS-75RP-25L	3.80 ± 0.07	60.58 ± 1.08	25.15 ± 0.92	10.47 ± 0.08
0 PS-50RP-50L	4.30 ± 0.22	55.27 ± 1.88	28.43 ± 2.04	12.01 ± 0.05
98PS-0RP-2L	2.41 ± 0.16	78.17 ± 3.41	17.75 ± 3.36	1.56 ± 0.12
96PS-0RP-4L	2.63 ± 0.19	75.50 ± 4.50	20.00 ± 4.70	1.87 ± 0.01
90PS-0RP-10L	2.39 ± 0.05	73.28 ± 2.21	21.76 ± 1.78	2.57 ± 0.37
75PS-25RP-0L	2.67 ± 0.23	72.20 ± 1.12	21.60 ± 0.92	3.53 ± 0.03
50PS-50RP-0L	2.71 ± 0.26	68.68 ± 0.54	23.31 ± 0.18	5.30 ± 0.62
25PS-75RP-0L	3.04 ± 0.08	63.56 ± 2.84	25.41 ± 2.73	7.99 ± 0.19
49PS-49RP-2L	2.66 ± 0.20	65.79 ± 4.70	25.39 ± 4.93	6.16 ± 0.03
48PS-48RP-4L	2.28 ± 0.48	64.74 ± 9.43	26.46 ± 9.97	6.51 ± 0.07
45PS-45RP-10L	2.69 ± 0.45	65.35 ± 3.45	26.55 ± 3.86	5.41 ± 0.04

Table 7
Mass yield, energy yield, and energy density ratios of torrefied pellets.

Pellets	Mass yield (%)	Energy yield (%)	Energy density ratio	HHV for torrefied pellets (MJ/kg)	HHV for raw pellets (MJ/kg)
100PS-0RP-0L	71.10 ± 0.32	81.24 ± 3.21	1.14 ± 0.01	19.48 ± 0.32	16.99 ± 0.13
0 PS-100RP-0L	71.49 ± 0.43	73.12 ± 2.56	1.03 ± 0.02	16.46 ± 0.17	15.66 ± 0.23
0 PS-98RP-2L	73.02 ± 0.80	88.17 ± 0.97	1.19 ± 0.03	16.52 ± 0.40	16.26 ± 0.34
0 PS-96RP-4L	71.32 ± 2.31	86.74 ± 2.81	1.22 ± 0.23	16.44 ± 0.15	16.11 ± 0.23
0 PS-90RP-10L	75.38 ± 2.49	90.31 ± 2.89	1.17 ± 0.01	16.52 ± 0.50	15.94 ± 0.17
0 PS-75RP-25L	76.37 ± 0.93	92.60 ± 1.13	1.21 ± 0.07	16.05 ± 0.81	15.30 ± 0.34
0 PS-50RP-50L	76.26 ± 1.04	90.13 ± 1.22	1.24 ± 0.01	15.41 ± 0.29	14.83 ± 0.36
98PS-0RP-2L	74.87 ± 2.91	92.76 ± 3.60	1.24 ± 0.03	19.27 ± 0.01	17.71 ± 0.15
96PS-0RP-4L	78.07 ± 1.02	94.53 ± 1.23	1.21 ± 0.05	19.11 ± 0.07	17.95 ± 0.12
90PS-0RP-10L	77.28 ± 2.34	95.87 ± 2.90	1.25 ± 0.02	18.96 ± 0.14	17.86 ± 0.09
75PS-25RP-0L	78.21 ± 0.73	96.80 ± 0.90	1.21 ± 0.04	18.57 ± 0.45	17.69 ± 0.32
50PS-50RP-0L	75.27 ± 0.51	96.05 ± 0.65	1.28 ± 0.04	18.01 ± 0.44	17.43 ± 0.14
25PS-75RP-0L	70.69 ± 2.21	85.06 ± 2.65	1.22 ± 0.06	17.06 ± 0.45	16.58 ± 0.19
49PS-49RP-2L	74.96 ± 2.07	96.18 ± 2.65	1.29 ± 0.10	17.75 ± 0.55	17.36 ± 0.23
48PS-48RP-4L	72.36 ± 7.26	94.06 ± 9.44	1.27 ± 0.07	17.76 ± 0.48	17.32 ± 0.28
45PS-45RP-10L	78.46 ± 2.27	102.13 ± 2.95	1.27 ± 0.06	17.98 ± 0.21	16.93 ± 0.15

carboxyl groups from hemicellulose, and aromatic and methoxyl groups from lignin are the reasons for mass loss [63]. In this study, as seen in Table 7, the mass yields of pellets decreased due to partial devolatilization and destruction of cellulose in the sample during torrefaction. They ranged from 70.69 % (25PS-75RP-0L) to 78.46 % (45PS-45RP-10L) by weight. It is thought that a lower mass yield was obtained in this study since the degradation rate of agricultural residues is relatively higher than that of woody biomass due to their higher hemicellulose content [64,65]. The energy yield of the samples varied between 73.12 % (0 PS-100RP-0L) and 102.12 (45PS-45RP-0L). In general, it is stated in the literature that biomasses preserve approximately 90 % of their energy density [62]. However, depending on the biomass types, the energy yield of the biomass differs during the torrefaction process. The energy yield of biomasses with higher lignin content is also higher. Accordingly, pellets with high RP, 25PS-75RP-0L, 0PS-98RP-2L, and 0 PS-96RP-4L, were found to have the lowest energy yield. The lignin contents of RP and conifers are given as 20 % and 18–26 %, respectively [66, 67]. Therefore, since the lignin content of RP is lower than that of PS, the energy yield of pellets with high RP was lower ($p > 0.05$) than other pellets [60]. An energy yield of more than 100 % is ideal due to the energy efficiency process. Therefore, it can be said that the net useable energy of the 45PS-45RP-0L pellets increased with the torrefaction process. Since volatile substances decrease during torrefaction, the O/C ratio of the sample decreases, which increases the energy density of the material [63]. In this study, it is seen that the energy densities of all torrefied samples were greater than one; that is, there is an energy gain per unit mass. In addition, in the study, the pellets prepared by adding the RP and PS samples in close proportions by weight; the energy densities of 49PS-49RP-2L, 50PS-50RP-0L, 48PS-48RP-4L, and 45PS-45RP-10L were found to be higher than the other pellets and were 1.29; 1.28; 1.27 and 1.27, respectively. This situation probably creates a synergy between them. In other words, it is thought that during the torrefaction of mixtures prepared by using different biomasses in similar amounts by weight, the interaction of biomass with each other occurs in a way that increases the energy density of the material more than in its singular form.

While HHV increases with the increase in the hydrogen-to-carbon content ratio in the biomass, it decreases with the increase in moisture content [68]. When HHV values of all the mixtures are examined, it is said that the HHV of fuel mixtures increased with the increase in lignite content in the mixtures. Another remarkable point in torrefied pellets was that the HHV of the mixtures approached each other due to torrefaction, and the difference between RP and PS mixtures decreased. In this study, the calculated HHV of the raw pellets also increased ($p < 0.05$) in parallel with the increased energy density values due to the torrefaction process. Accordingly, the

Table 8
Fuel properties of the raw pellets.

Pellets	Mass (g)	Diameter (mm)	Length (cm)	Particle density (kg/m³)	Bulk density (kg/m³)
100PS-0RP-0L	0.681 ± 0.070	6.00 ± 0.01	2.62 ± 0.12	1400 ± 0.07	450 ± 0.000009
0 PS-100RP-0L	0.626 ± 0.047	5.10 ± 0.31	2.59 ± 0.13	1620 ± 0.24	460 ± 0.000021
0 PS-98RP-2L	0.517 ± 0.036	5.47 ± 0.05	2.53 ± 0.14	1110 ± 0.05	520 ± 0.00016
0 PS-96RP-4L	0.519 ± 0.054	5.41 ± 0.10	2.52 ± 0.18	1130 ± 0.12	520 ± 0.00014
0 PS-90RP-10L	0.522 ± 0.031	5.32 ± 0.07	2.51 ± 0.09	1170 ± 0.05	540 ± 3.8E-06
0 PS-75RP-25L	0.497 ± 0.026	5.37 ± 0.10	2.59 ± 0.12	1060 ± 0.03	490 ± 5.8E-06
0 PS-50RP-50L	0.529 ± 0.088	5.51 ± 0.11	2.66 ± 0.15	1050 ± 0.16	440 ± 4.4E-06
98PS-0RP-2L	0.568 ± 0.052	6.66 ± 0.13	2.66 ± 0.17	850 ± 0.04	380 ± 7.6E-06
96PS-0RP-4L	0.590 ± 0.030	6.52 ± 0.13	2.61 ± 0.10	870 ± 0.04	380±1E-06
90PS-0RP-10L	0.656 ± 0.039	6.38 ± 0.18	2.57 ± 0.17	890 ± 0.01	420 ± 2.5E-06
75PS-25RP-0L	0.536 ± 0.035	6.42 ± 0.10	2.69 ± 0.16	780 ± 0.03	330 ± 1.5E-06
50PS-50RP-0L	0.531 ± 0.031	5.87 ± 0.04	2.62 ± 0.14	940 ± 0.03	370 ± 1.38E-06
25PS-75RP-0L	0.557 ± 0.037	5.49 ± 0.05	2.60 ± 0.11	1120 ± 0.02	500 ± 0.00013
49PS-49RP-2L	0.517 ± 0.035	5.79 ± 0.09	2.61 ± 0.13	950 ± 0.05	410 ± 1.1E-06
48PS-48RP-4L	0.518 ± 0.026	5.88 ± 0.04	2.62 ± 0.11	890 ± 0.03	410 ± 1.9E-06
45PS-45RP-10L	0.534 ± 0.032	5.81 ± 0.05	2.62 ± 0.13	960 ± 0.04	420±2E-06

HHV of the raw pellets ranged from 14.83 MJ/kg (50PS-50RP-0L) to 17.95 MJ/kg (96PS-0RP-4L), while the HHV of the torrefied pellets was 15.41 MJ/kg (50PS-50RP-0L) to 19.48 MJ/kg (100PS-0RP-0L). The increase in HHV of the raw pellets made with different biomasses was found to be between 19.7 % and 50 % [56,59,69]. Wang et al. (2020) [57] found that HHVs of the raw pellets obtained from the trunk, bark, and remains of the spruce tree were 19.9 MJ/kg, 20.6 MJ/kg, and 21.1 MJ/kg, respectively, with an increase in the torrefaction temperature and holding time, and HHV, respectively. They stated that the values varied between 20.1 and 22.7 MJ/kg, 20.6–23.4 MJ/kg, and 21.1–23.4 MJ/kg. Therefore, in this study, the increase in HHV of the raw pellets as a result of torrefaction was found to be between 1.64 % (0 PS-98RP-2L) and 14.64 % (100PS-0RP-0L). Therefore, although the HHV obtained in this study showed parallelism with HHV given in the literature, however, it is seen that the increase was relatively lower.

3.3.3. Mass, diameter, length, bulk density, particle density, and tensile strength of the prepared pellets

Knowledge of changes in volume and density after torrefaction is important for the transport of biomass and the design of biomass processing plants [60]. Due to devolatilization, voids form in the biomass and cause a decrease in its density. There is also a decrease in the physical dimensions of the biomass due to devolatilization, resulting in a reduction in volume. Dehydration of the biomass, changes in chemical bonds, and coalescence of graphite cores in the solid structure are the reasons for the reduction in the size of the biomass [70].

The properties of raw and torrefied pellets are given in Tables 8 and 9, respectively. The results showed that the mass of the pellets increased with the addition of lignite. In the study, the masses of the raw pellets varied between 0.497 ± 0.026g (0 PS-75RP-25L) and 0.626 ± 0.047g (0 PS-100RP-0L). Mass decreased with the removal of volatile substances from the fuel mixtures with torrefaction, and the masses of the torrefied pellets varied between 0.346 ± 0.039 (0 PS-50RP-50L) and 0.554 ± 0.051g (100PS-0RP-0L). Therefore, statistically a significant decrease ($p < 0.05$) between 9.80 % and 18.30 % was observed in the mass of the pellets with torrefaction. Işık (2019) [71] also stated a decrease between 11.3 % and 43.8 % in the weight of the pellets prepared with ash, hazelnut, and waste-derived fuel (RDF) after torrefaction in his study.

According to the ISO 17829 standard for determining the lengths and diameters of solid biofuels-pellets, the lengths of pellets can be between 3.15 mm and 50 mm. The lengths of the raw pellets were between 2.51 ± 0.09 cm (0 PS-90RP-10L) and 2.69 ± 0.16 cm (75PS-25RP-0L), whereas for the torrefied pellets were 2.22 ± 0.27 cm (0 PS-50RP-50L) to 2.60 ± 0.12 cm (75PS-25RP-0L). It was determined that there was a decrease in the length ($p < 0.05$) of the pellets with torrefaction. According to the DIN 51731 standard, it is stated that the length of the pellets should be less than 5 cm, and the lengths of both torrefied and raw pellets met this standard. According to the ISO 17225-2 standard, it is pointed out that the length of the pellets should be greater than 3.15 cm. Therefore, it is thought that the lengths of the pellets prepared in this study could not meet this standard, probably due to the structural features of the pellet machine.

In this study, the diameters of the raw pellets varied between 5.10 ± 0.31 mm (0 PS-100RP-0L) and 6.66 ± 0.13 mm (98PS-0RP-2L), but the diameters of the pellets decreased with torrefaction and changed between 4.40 ± 0.51 mm (0 PS-100RP-0L) and 6.40 ± 0.12 mm (98PS-0RP-2L). However, a decrease in the diameter of the torrefied pellets was statistically insignificant ($p > 0.05$). The diameters of the pellets should be between 4 and 12 mm according to the DIN 51731 standard, and the diameter of the pellets should be between 4 and 10 mm according to the ÖNORM 7135 standard. On the other hand, according to the EN14961 standard, the pellets' diameters should be between 6 and 25 mm. It can be seen that the diameter of the raw pellets cannot exceed 7 mm in this study. Furthermore, the diameters of the prepared pellets decreased further with the addition of lignite.

As the voids in the biomass increase due to torrefaction, a decrease in bulk density is expected; this decrease in density varies depending on the biomass type and operating parameters during torrefaction [60]. Similar to literature, the densities of the pellets were also decreased ($p < 0.05$) after the torrefaction and varied between 330 kg/m^3 (75PS-25RP-0L) and 540 kg/m^3 (0 PS-90RP-10L), while the bulk densities of the torrefied pellets were changed between 280 kg/m^3 (75PS-25RP-0L) and 540 kg/m^3 (0 PS-90RP-10L) in this study. According to the standard for determination of solid biofuels-particle density for pellets and briquettes (EN ISO 18847

Table 9
Fuel properties of the torrefied pellets.

Pellets	Mass (g)	Diameter (mm)	Length (cm)	Particle density (kg/m^3)	Bulk density (kg/m^3)
100PS-0RP-0L	0.554 ± 0.051	6.00 ± 0.00	2.59 ± 0.12	950 ± 0.087	350 ± 0.00000
0 PS-100RP-0L	0.479 ± 0.036	4.40 ± 0.51	2.50 ± 0.18	1530 ± 0.346	450 ± 0.00000
0 PS-98RP-2L	0.401 ± 0.053	5.10 ± 0.36	2.45 ± 0.18	1020 ± 0.18	510 ± 0.00002
0 PS-96RP-4L	0.391 ± 0.056	4.90 ± 0.00	2.42 ± 0.10	1070 ± 0.15	430 ± 5.4E-06
0 PS-90RP-10L	0.396 ± 0.041	4.96 ± 0.08	2.43 ± 0.15	1020 ± 0.17	530 ± 6.7E-06
0 PS-75RP-25L	0.399 ± 0.039	5.37 ± 0.09	2.41 ± 0.21	910 ± 0.04	330 ± 8.3E-07
0 PS-50RP-50L	0.346 ± 0.039	5.35 ± 0.08	2.22 ± 0.27	870 ± 0.02	330 ± 1.9E-06
98PS-0RP-2L	0.512 ± 0.031	6.40 ± 0.12	2.44 ± 0.14	820 ± 0.04	330 ± 2.2E-06
96PS-0RP-4L	0.492 ± 0.035	6.34 ± 0.12	2.43 ± 0.15	800 ± 0.04	350 ± 2.1E-06
90PS-0RP-10L	0.430 ± 0.042	6.22 ± 0.11	2.39 ± 0.21	740 ± 0.05	310 ± 2.8E-06
75PS-25RP-0L	0.421 ± 0.037	6.09 ± 0.03	2.60 ± 0.12	710 ± 0.05	280 ± 1.5E-06
50PS-50RP-0L	0.405 ± 0.024	5.73 ± 0.11	2.57 ± 0.14	740 ± 0.04	320 ± 3.5E-06
25PS-75RP-0L	0.407 ± 0.030	5.21 ± 0.10	2.55 ± 0.23	920 ± 0.05	500 ± 2.8E-06
49PS-49RP-2L	0.406 ± 0.017	5.71 ± 0.05	2.59 ± 0.09	780 ± 0.05	410 ± 2.0E-06
48PS-48RP-4L	0.405 ± 0.034	5.62 ± 0.13	2.56 ± 0.15	670 ± 0.04	420 ± 2.8E-06
45PS-45RP-10L	0.406 ± 0.019	5.59 ± 0.09	2.44 ± 0.20	890 ± 0.05	420 ± 2.8E-06

(2016)), the particle density of solid biofuels should be above 600 kg/m^3. Accordingly, the particle densities of all the raw and torrefied pellets varied between 670 and 1620 kg/m^3 (Tables 8 and 9).

Tensile strength is one of the other important properties of pellets and is defined as the force required to break the pellets. Furthermore, the decrease in strength means that pellets can crumble more easily and will be more amenable to grinding during transportation and storage [57]. In this study, the tensile strength of the prepared pellets was also investigated (Table 10), and it was observed that the strength of the pellets obtained by mixing RP and PS with each other and lignite decreased as compared to their unmixed forms. The highest strength belongs to 100 % PS pellet (2.38 MPa), and the strength of the raw pellets obtained from mixing PS with lignite was also higher ($p < 0.05$) than that of raw RP pellets mixed with lignite at the same amount. However, this effect was not found statistically significant ($p > 0.05$) for the torrefied pellets. The lowest strength among the raw pellets was obtained for 0 PS-50RP-50L (0.58 MPa). As the strengths of the torrefied pellets were compared with the strengths of their raw form, a decrease ($p < 0.05$) was observed in the strength of the pellets after torrefaction. The decrease varied between 0 PS-50RP-50L (7.49 %) and 90PS-0RP-10L (51.94 %); the average decrease was 35 ± 12.48 %. It was stated that the decomposition rate of agricultural wastes is higher compared to woody biomass due to the higher hemicellulose content, and as a result, the mass yield is lower [64,65]. Therefore, since the hemicellulose content of RP was higher than that of PS, it can be said that the strength decreased in RP pellets less ($p > 0.05$). Wang et al. (2020) [57] also found the tensile strength of woody pellets between 1.58 MPa and 2.52 MPa in their study. However, the strength of the pellets decreased after the torrefaction and was found to be between 1.31 MPa and 2.41 MPa.

3.3.4. Abrasive resistance and durability of the prepared pellets

Table 11 gives abrasive resistances of both raw and torrefied pellets. The losses of the pellets were minuscule, and the raw pellets preserved 99.98 %–99.88 % of their integrity. That is, while raw pellets showed high resistance to disintegration, the strength of the torrefied pellets decreased ($p < 0.05$) and preserved only 99.92 %–99.55 % of their integrity. The deficiencies in both the hydroxyl groups and the cellulose content of the torrefied pellets, together with the decreasing moisture, cause the forces holding the particles together in the torrefied pellets to weaken; in this case, it causes the torrefied pellets to be weaker against breakage compared to the raw pellets [72].

The literature has opposing views regarding the durability of the pellets measured because of torrefaction. Dutta and Mathias (2011) [62] stated in their study that the durability of pellets increased (1.5–2 times) with torrefaction. However, in many studies, it is said that there is a decrease in the durability of the pellets with torrefaction temperature and torrefaction time [57,69,72–74]. A durability of 95 % is sufficient for the safe transportation and storage of the [57]. In the study, the durability was obtained above 95 % for all prepared pellet types therefore, the durability of the pellets was found to be sufficient. Furthermore, although a statistically significant decrease ($p < 0.05$) was observed for the abrasive resistance of the pellets due to torrefaction, a decrease in the pellets' durability due to torrefaction was not found statistically significant ($p > 0.05$). In the study, the durability of the raw pellets ranged from 97.31 % to 99.71 %, while the durability of the torrefied pellets ranged from 97.39 % to 99.69 % (Table 12). Similarly, the durability of the raw woody pellets and pellets with agricultural wastes was between 92.9 % and 99.71 %. However, after the torrefaction, durability varied from 95.45 % to 98.19 % in the literature [57,75,76].

3.3.5. Water intake resistance of both the raw and torrefied pellets

Pellets have the potential to absorb moisture due to the structure of biomass, and hygroscopic pellets may undergo microbial degradation during storage. The hydroxyl group, which can form ions and attract water molecules, is responsible for the hydrophilic behavior of the biomass. However, the hydrophilic nature of the biomass decreases during the torrefaction process. Removal of hydroxyl groups also reduces the ability of the biomass to form hydrogen bonds, which reduces its moisture absorption capacity. Therefore, the hydrophilic structure of the biomass is converted to a hydrophobic structure by the torrefaction process [60].

The study showed a structural deterioration due to reabsorption after the pellets were kept in water for 2 h. In contrast, the torrefied pellets retained their shape much better than the raw pellets. Similar results also reported in the literature [77]. Due to the removal of hydroxyl groups in the torrefied pellets, it can be said that the torrefied pellets cannot form a bond with water molecules compared to the raw pellets. As a result, torrefied pellets show more resistance to moisture absorption [78].

Re-absorption of moisture increases physicochemical degradation and accelerates microbial degradation of biomass [79]. In this context, increasing the moisture resistance of the pellets by using them as raw material in terms of their storage by being torrefied is very important. The water intake resistance of the raw and torrefied pellets is given in Figs. 4 and 5. According to the Figures, the holding time for the torrefied pellets to reach constant weight was lower ($p < 0.05$) than the raw pellets. Therefore, the water intake resistance of the torrefied pellets was found to be higher as compared to their raw forms. This situation will likely affect the moisture-absorbing properties of the materials obtained by torrefaction processes positively, and the humidity will be less of a problem in the torrefied pellets compared to the raw pellets during storage. Similarly, the water absorption of the raw pellets varied between 2 and 3 times their weight at the end of 2 h, while the water absorption rates of the torrefied materials changed between 0.2 and 0.4 times their weight at the end of 2 h.

4. Conclusion

Torrefaction after the pelletization is an encouraging method to improve the fuel properties of pellets made from woody biomass. With co-pelletization of agricultural residues with lignite, the highest FC contents were obtained. Because the calculated HHV of the raw pellets significantly increased after torrefaction in parallel with the increased energy density values of the studied pellets. However, the fuel properties of the pellets displayed different findings depending on the biomass type in terms of energy yield. For

Table 10
Tensile strengths of both the raw and torrefied pellets.

Pellets	Raw pellets (MPa)	Torrefied pellets (MPa)
100PS-0RP-0L	2.38	1.20
0 PS-100RP-0L	1.29	0.88
0 PS-98RP-2L	1.04	0.69
0 PS-96RP-4L	0.92	0.66
0 PS-90RP-10L	0.86	0.57
0 PS-75RP-25L	0.82	0.67
0 PS-50RP-50L	0.58	0.53
98PS-0RP-2L	1.40	0.99
96PS-0RP-4L	1.38	0.70
90PS-0RP-10L	1.01	0.49
75PS-25RP-0L	0.69	0.43
50PS-50RP-0L	0.88	0.46
25PS-75RP-0L	0.89	0.71
49PS-49RP-2L	1.00	0.62
48PS-48RP-4L	0.83	0.45
45PS-45RP-10L	0.89	0.58

Table 11
Abrasive resistance of both the raw and torrefied pellets.

Pellets	Abrasive resistance of raw pellets (% loss)	Abrasive resistance of torrefied pellets (% loss)
100PS-0RP-0L	0.08 ± 0.04	0.14 ± 0.01
0 PS-100RP-0L	0.08 ± 0.03	0.25 ± 0.11
0 PS-98RP-2L	0.11 ± 0.07	0.31 ± 0.16
0 PS-96RP-4L	0.08 ± 0.08	0.43 ± 0.44
0 PS-90RP-10L	0.08 ± 0.05	0.45 ± 0.25
0 PS-75RP-25L	0.02 ± 0.03	0.15 ± 0.11
0 PS-50RP-50L	0.05 ± 0.04	0.21 ± 0.10
98PS-0RP-2L	0.03 ± 0.03	0.16 ± 0.04
96PS-0RP-4L	0.04 ± 0.04	0.08 ± 0.02
90PS-0RP-10L	0.12 ± 0.23	0.19 ± 0.08
75PS-25RP-0L	0.02 ± 0.02	0.11 ± 0.06
50PS-50RP-0L	0.06 ± 0.01	0.15 ± 0.08
25PS-75RP-0L	0.07 ± 0.03	0.12 ± 0.04
49PS-49RP-2L	0.06 ± 0.04	0.13 ± 0.05
48PS-48RP-4L	0.06 ± 0.05	0.17 ± 0.07
45PS-45RP-10L	0.09 ± 0.12	0.18 ± 0.15

Table 12
The durability of both the raw and torrefied pellets.

Pellets	The durability of raw pellets (%)	The durability of torrefied pellets (%)
100PS-0RP-0L	99.50	99.19
0 PS-100RP-0L	98.41	98.20
0 PS-98RP-2L	98.61	97.39
0 PS-96RP-4L	98.36	98.00
0 PS-90RP-10L	98.38	97.73
0 PS-75RP-25L	99.36	99.43
0 PS-50RP-50L	99.58	99.56
98PS-0RP-2L	99.61	99.55
96PS-0RP-4L	99.33	99.00
90PS-0RP-10L	99.59	99.32
75PS-25RP-0L	99.50	99.44
50PS-50RP-0L	99.51	99.62
25PS-75RP-0L	99.57	99.41
49PS-49RP-2L	99.79	99.69
48PS-48RP-4L	99.69	98.43
45PS-45RP-10L	99.36	98.27

example, the energy yield of pellets containing high amounts of RP was found to be lower than other studied pellet mixtures. Furthermore, the energy densities of the pellets having equal amounts of RP and PS were obtained high, and it was thought that due to the synergy between rose and pine biomass. The mass, length, and densities of the pellets were decreased after torrefaction. However, a decrease in the diameter of the pellets was found to be statistically insignificant. The strength of the raw pellets obtained from mixing
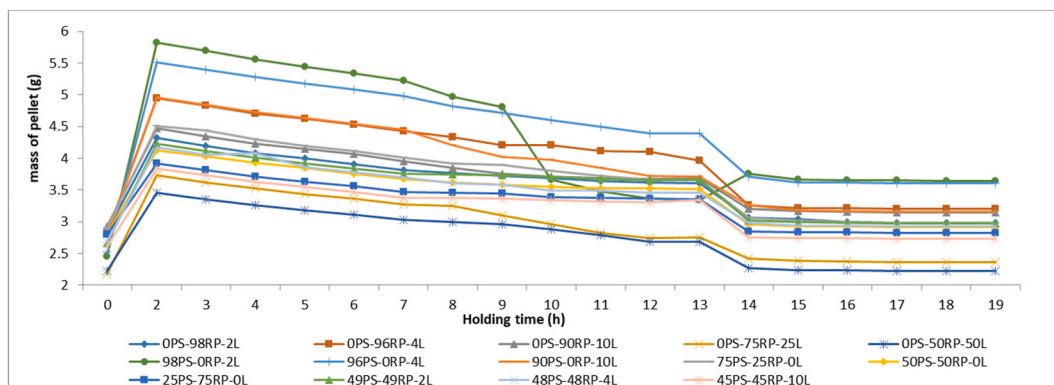
Fig. 4. The change of water intake resistance of the raw pellets concerning holding time.
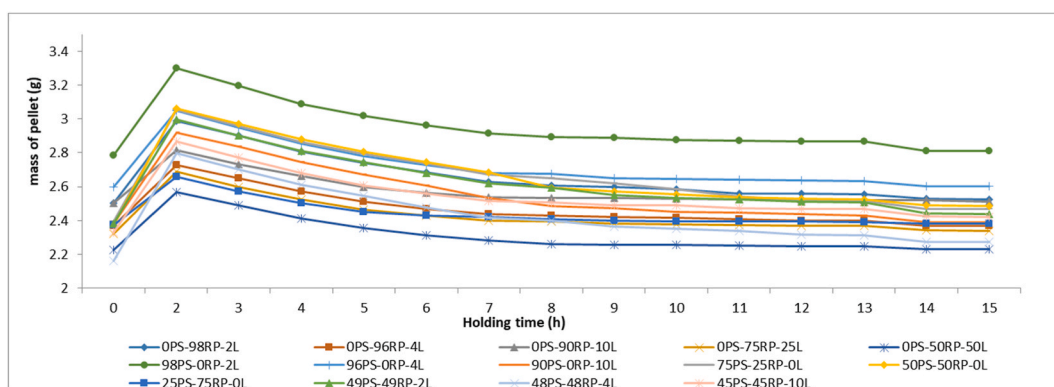

Fig. 5. The change of water intake resistance of the torrefied pellets concerning holding time.

PS with lignite was found to be higher than that of raw RP pellets mixed with lignite at the same amount. However, this effect was not found statistically significant for torrefied pellets. Although, upon torrefaction treatment, the strength of the pellets decreased, the torrefied pellets' durability did not statistically significantly decrease compared to that of untreated pellets. Finally, torrefied pellets displayed high hydrophobicity, and the water intake resistance of the treated pellets was found to be significantly higher in comparison to that of their raw forms. In conclusion, compared to single-content biopellets, co-pelletization and torrefaction can produce pellets with higher energy content, hydrophobicity, and energy densities.

Data availability statement

Data can be shared upon request.

CRediT authorship contribution statement

Büşra Çetinkaya: Formal analysis. **Sena Erkent:** Formal analysis. **Kamil Ekinci:** Writing – review & editing, Formal analysis. **Mihriban Civan:** Methodology. **Mehmet Emin Bilgili:** Writing – review & editing. **Sema Yurdakul:** Writing – original draft.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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