Research Article GU J Sci, Part A, 11(4): 801-813 (2024) 10.54287/gujsa.1552835

Gazi University
Journal of Science
PART A: ENGINEERING AND INNOVATION
http://dergipark.org.tr/gujsa

Experimental Study on Gasification of Banknote Waste: Effects of Torrefaction Pre-Treatment and Co-Gasification on Producer Gas Composition

Hakan KAVUŞTU[1] Emir Hüseyin ŞIMŞEK[1*]

[1] *Ankara University, Department of Chemical Engineering, Ankara, Türkiye*

Keywords

End-of-Life Banknote
Banknote Waste
Gasification
Co-Gasification
Torrefaction
Fluidized Bed

Abstract

There is approximately 500,000 tonnes of potential end-of-life banknote waste worldwide, which is increasing by 2-3% per year. This waste consists of cotton and polymer-based banknotes printed on substrates whose raw materials are cotton and polypropylene, respectively. The vast majority of banknotes in circulation are cotton-based banknotes. End-of-life cotton banknotes, which are lignocellulosic biomass, are generally disposed of by landfill and incineration. Studies to reduce the environmental impact of these wastes to find more effective ways of using them is becoming increasingly important. Syngas, which can be used for the production of electricity, energy and chemicals is obtained by gasification of end-of-life cotton banknotes. In this study, DSC and FTIR analysis were performed as part of the characterization tests of the cotton-based banknote sample. As a result of the analysis, the sample was found to have characteristics similar to those of cotton. Within the scope of the investigation of thermal decomposition kinetics, activation energies were calculated as 134-171 kJ/mol by the Flynn-Wall-Ozawa (FWO) and Kissinger-Akahira-Sunose (KAS) methods. Experiments were performed in a fluidized-bed reactor at 800°C with an inlet H_2O/O_2 ratio of 25. The content of the producer gas formed during gasification was examined according to the maximum mole fraction achieved. In order to facilitate handling, storage and transportation and to improve fuel quality, the effect of torrefaction pre-treatment on the producer gas content was studied by conducting torrefaction to the cotton-based banknote sample at 250°C for 10 min. To overcome the disadvantages of plastic gasification in terms of operational sustainability, the cotton and polymer-based banknote samples were co-gasified. With the torrefaction pre-treatment, the mole fractions of H_2, CO and CH_4 increased, while the mole fraction of CO_2 decreased. This finding revealed the effects of Boudouard, hydrogasification, water-gas and steam reformation reactions. With the co-gasification of cotton and polymer-based banknote samples, H_2, CO and CO_2 mole fractions decreased while CH_4 mole fraction increased. This result showed that as the proportion of polymer-based banknote samples in the feedstock increased, the conversion efficiency decreased and the hydrogasification reaction became dominant.

Cite

Kavuştu, H., & Şimşek, E. H. (2024). Experimental Study on Gasification of Banknote Waste: Effects of Torrefaction Pre-Treatment and Co-Gasification on Producer Gas Composition. *GU J Sci, Part A*, *11*(4), 801-813. doi:10.54287/gujsa.1552835

Author ID (ORCID Number)		Article Process	
0000-0002-3492-2754	Hakan KAVUŞTU	**Submission Date**	19.09.2024
0000-0001-7945-8222	Emir Hüseyin ŞIMŞEK	**Revision Date**	30.10.2024
		Accepted Date	04.11.2024
		Published Date	30.12.2024

1. INTRODUCTION

Energy production is one of the most important economic factors that provides development and increases efficiency. In the context of energy sustainability, the importance of renewable energy sources is increasing due to (i) rapidly growing demand for energy, (ii) the fact that most of this demand is met by fossil fuels, and (iii) increasing concerns about global warming and environmental pollution caused by the use of fossil fuels (Jahromi et al., 2021; Zhang et al., 2024). The total energy supply increased by 15% between 2000 and 2021, and oil (29.5%), coal (27.2%), natural gas (23.6%), biofuels and waste (9.5%) were the sources with the largest share in the total energy supply in 2021 (IEA, 2021). Modern bioenergy sources, which provide more than half of the renewable energy supply, can exist in solid, liquid and gaseous forms. According to the

---

*Corresponding Author, e-mail: simsek@eng.ankara.edu.tr

International Energy Agency (IEA) Energy Policy Scenario, the modern solid bioenergy supply will reach 57 Exajoules (EJ) in 2050 (IEA, 2023).

Biomass, which is a carbon-neutral and renewable energy source, has an important role in sustainable energy policies (George et al., 2019). Biomass can be classified according to the different raw material types such as agricultural, forest, municipal, industrial wastes and wood (Hussain et al., 2023). Biomass differs in terms of their properties such as molecular morphology and thermal stability, and the type and structure of biomass directly affect the product quality and yield to be obtained as a result of thermochemical conversion. Lignocellulosic biomass is basically composed of cellulose, hemicellulose and lignin (Chan et al., 2019).

In principle, banknotes are printed on cotton and polymer (polypropylene) based materials (Luján-Ornelas et al., 2018). The vast majority of banknotes produced worldwide are cotton-based banknotes with a high cotton content (DLR, 2022). Cotton-based banknote production generally includes (i) the production of banknote paper containing cotton, security thread, foil and (ii) the printing on banknote paper with inks. The Euro banknote consists of an average 0.815 g of cotton, 0.082 g of ink, 0.010 g of security thread, and 0.049 g of foil (Hanegraaf et al., 2019).

Banknotes in circulation are routinely sorted by central banks. At the end of banknote processing, worn or damaged banknotes which are not fit for circulation in terms of quality standards are considered as end-of-life banknotes (BoI, 2023). End-of-life cotton banknotes can be classified as lignocellulosic biomass due to their high cotton content (Yousef et al., 2020; Kavuştu & Şimşek, 2023). Banknote demand is basically determined by (i) the increase in the amount of banknotes in circulation, (ii) the issuance of new series of banknotes, and (iii) the renewal of end-of-life banknotes (DLR, 2023). It is estimated that there are approximately 500 billion banknotes in circulation worldwide and more than 150 billion banknotes are produced each year (G+D, 2024). The amount of banknote production worldwide tends to increase by 2-3% per year (K&B, 2022). These production data mean that (i) the potential amount of end-of-life banknote waste is approximately 500,000 tonnes assuming an average banknote weight of 1 g, (ii) nearly 150,000 tonnes of potential waste will be generated annually, and (iii) this amount will continue to increase by 2-3% each year. End-of-life banknotes are mostly disposed of by landfill and incineration (RDK, 2022). The European Central Bank (ECB) has increased the use of incineration and recycling with the ban of the disposal of the waste banknotes by landfill and combustion in order to reduce their environmental impact (ECB, 2023)

In gasification, biomass is converted into a gas mixture (synthesis gas/syngas/producer gas), which can be used for the production of liquid fuels, chemicals and energy, with the gasification agent at the gasifier temperature of 600-1400°C (Ng et al., 2013). Air, oxygen, steam and CO_2 are generally used as gasification agents. The high nitrogen content in the gasification agent reduces the calorific value of the syngas, while the use of pure oxygen is costly. In addition to providing a higher gasification rate compared to the use of CO_2 with gasification by feeding steam, a product with high calorific value and low tar content can be obtained (Xie et al., 2021). The gasification process basically consists of the following stages: (i) drying, where the moisture in the biomass is removed, (ii) pyrolysis, where volatile components are vaporized, (iii) oxidation (combustion) of combustible materials with oxygen (O_2), and (iv) reduction (char gasification) occurring as a result of the contact of coal formed by pyrolysis with carbon dioxide (CO_2) and water (H_2O) (Baruah & Baruah, 2014).

In gasification, syngas is produced, which contains combustible gases such as hydrogen (H_2), carbon monoxide (CO), methane (CH_4) and non-combustible gases such as nitrogen (N_2) and carbon dioxide (CO_2). Operating conditions, the types of feedstock and gasifier affect the composition of the syngas (Das et al., 2020). Transition metal catalysts contribute to the reduction of tar formation and the increase of the quality of the syngas (Meng et al., 2023). Banknotes can contain many of the elements that can be used as catalysts, such as zeolites that can be synthesized at different Si/Al ratios (Yousef et al., 2020; Kavuştu & Şimşek, 2023). Bubbling fluidized-bed gasifier has advantages such as (i) ease of operation, (ii) good mixing and high-quality gas-solid contact, (iii) increased mass, heat transfer and chemical conversion efficiency, and (iv) ability to operate under different operating conditions (Parrillo et al., 2021). Kinetic studies for the devolatilisation of lignocellulosic biomass contribute to understanding biomass decomposition behaviour.

Non-isothermal methods such as Flynn-Wall-Ozawa (FWO) and Kissinger-Akahira-Sunose (KAS) are widely used in estimating kinetic parameters and extent of conversion (Mohammed et al., 2022).

With the torrefaction pre-treatment, which is pyrolysis that occurs at relatively low temperatures, the fuel quality of biomass and the syngas yield can be increased in addition to facilitating handling, storage and transportation of biomass (Cabuk et al., 2020). In the study of Sadaka (2013), as a result of torrefaction of cotton gin waste, the moisture and volatile solid content decreased, while the bulk density and calorific value increased, and generally more carbon conversion and gasification efficiency were obtained compared to raw waste by gasification. With co-gasification, where two or more feedstocks are mixed and gasified, the mass transfer, gasification rate, reactivity and efficiency can be increased and the amount of pollutant gases can be reduced (Hanchate et al., 2021).

Polymer banknotes are printed on substrates made of biaxially-oriented polypropylene film (BOPP) (BoC, 2011). Using only plastic as feedstock in gasification has some disadvantages such as (i) having a very high calorific value, making gasifier temperature control difficult, (ii) its sticky structure making operation difficult, and (iii) increasing tar formation. In order to overcome these disadvantages, the product quality and yield can be increased by co-gasification of plastic and biomass in a bubbling fluidized-bed gasifier (Parrillo et al., 2023). One of the most suitable plastic types for co-gasification with biomass is polypropylene (PP) (Block et al., 2018). Co-gasification of biomass and plastic has advantages such as accelerating plastic degradation with the catalytic effect of the biomass char, and reducing char formation due to the increase in syngas yield (Ajorloo et al., 2024).

There are studies examining the end-of-life cotton-rich banknotes by (i) biochemical processes such as methane production by “anaerobic digestion” (Mancilla-Leytón et al., 2024), glucose production by “fermentation” (Sheikh et al., 2013; Yousef et al., 2021) and (ii) thermochemical processes such as bio-oil, bio-gas and char formation by “pyrolysis” (Yousef et al., 2020), producer gas production by “gasification” (Kavuştu & Şimşek, 2023).

In the present study, Differential Scanning Calorimetry (DSC) and Fourier Transform Infrared Spectroscopy (FTIR) analysis were performed within the scope of characterization tests of the end-of-life cotton-rich samples. Thermal decomposition kinetics were examined by Flynn-Wall-Ozawa (FWO) and Kissinger-Akahira-Sunose (KAS) methods. The effects of (i) torrefaction of cotton-based banknote sample and (ii) co-gasification of cotton and polymer-based banknote samples on the producer gas content as maximum mole fractions were investigated within the scope of experiments on gasification in a fluidized-bed reactor. The main novelty of this study is to investigate the effects of torrefaction pre-treatment and co-gasification on producer gas composition with gasification of end-of-life banknotes.

2. MATERIAL AND METHOD

2.1. Materials

In this study, the following samples were used: (i) 5 Turkish Lira (TL) banknote samples, which were put into circulation in 2009 and were printed on a substrate made of cotton fibers and (ii) 1 Romanian Leu (RON) banknote samples, which entered into circulation in 2005 and were printed on a substrate made of polypropylene resin. The physical properties of banknote samples are given in Table 1.

Table 1. *Physical properties of 5 TL and 1 RON samples*

Sample	**Physical Properties**		
	Thickness (µm)	Grammage (g/m^2)	Particle Density (g/cm^3)
5 TL (Kavuştu & Şimşek, 2023)	116	98.5	0.85
1 RON	96	90.8	0.95

2.2. Preparation of Samples

The samples were sieved from a mesh after turning into small particles. 5 TL and 1 RON samples with a mean particle size of 890 µm were prepared. Additionally, the 5 TL sample with a mean size of 890 µm was torrefied with inert gas (N_2: 99.9%) at 250°C for 10 min in the gasifier. The prepared samples are demonstrated in Figure 1.

Figure 1. *Prepared banknote samples:* ***a)*** *raw and* ***b)*** *torrefied cotton-based;* ***c)*** *polymer-based*

2.3. Characterization Techniques

Cotton-based raw sample shown in Figure 1a was used in characterization tests. Differential Scanning Calorimetry (DSC) analysis was performed using a TA Instruments SDT 650 Simultaneous DSC/TGA, at six different heating rates between 5-30°C/min and a temperature range of 25-1000°C in nitrogen. Fourier Transformed Infrared Spectroscopy (FTIR) analysis was performed using a Perkin Elmer Pyris STA 6000 Simultaneous TGA/FTIR at six different heating rates between 5-30°C/min and a temperature range of 30-950°C in nitrogen.

2.4. Activation Energy Determination

The thermal analysis data used to investigate the thermal decomposition kinetics were obtained from our previous study (Kavuştu & Şimşek, 2023) in which thermogravimetric analysis was performed on 5TL banknote sample at six different heating rates in the range of 5-30°C/min. The formula used to calculate the activation energy with FWO and KAS methods is given in Table 2 (Yousef et al., 2020).

Table 2. *FWO and KAS methods*

Method	**Formula**	**x**	**y**	**Slope**
Flynn-Wall-Ozawa (FWO)	$\ln\beta = \left(\frac{\ln AEa}{Rf(y)}\right) - 5.335 - \frac{1.051Ea}{RT}$	1/T	ln(β)	-1.0516 Ea/R
Kissinger-Akahira-Sunose (KAS)	$\ln\left(\frac{\beta}{T^2}\right) = \ln\left(\frac{AR}{Eaf(y)}\right) - \frac{-Ea}{RT}$	1/T	$\ln(\beta/T^2)$	-Ea/R

2.5. Gasification Experiments

An experimental setup consisting of a gas preparation unit, a fluidized-bed reactor and a gas analyzer was used (Figure 2). In the gas preparation unit consisting of PID controlled flow meters (MKS), the mixture of steam and air was prepared at the desired rates by adjusting the bubble column temperature. The mixture of steam-air was fed to the fluidized-bed reactor at the minimum fluidization velocity in order to prevent the elutriation of sample particles in the reactor. 3 g of sample was fed to the furnace-reactor combined system consisting of a gas distributor, a ceramic furnace and an atmospheric pressure cylindrical fluidized-bed reactor. Experiments were performed for 600 s of operation time. After the undesired components such as sulfur oxides (SO_x) and nitrogen oxides (NO_x) in the gas formed in the reactor were trapped in the conditioning unit, the CO, CO_2, CH_4, H_2 and O_2 mole fraction (% y_i) values were determined with the gas analyzer (GASBOARD-3100P).

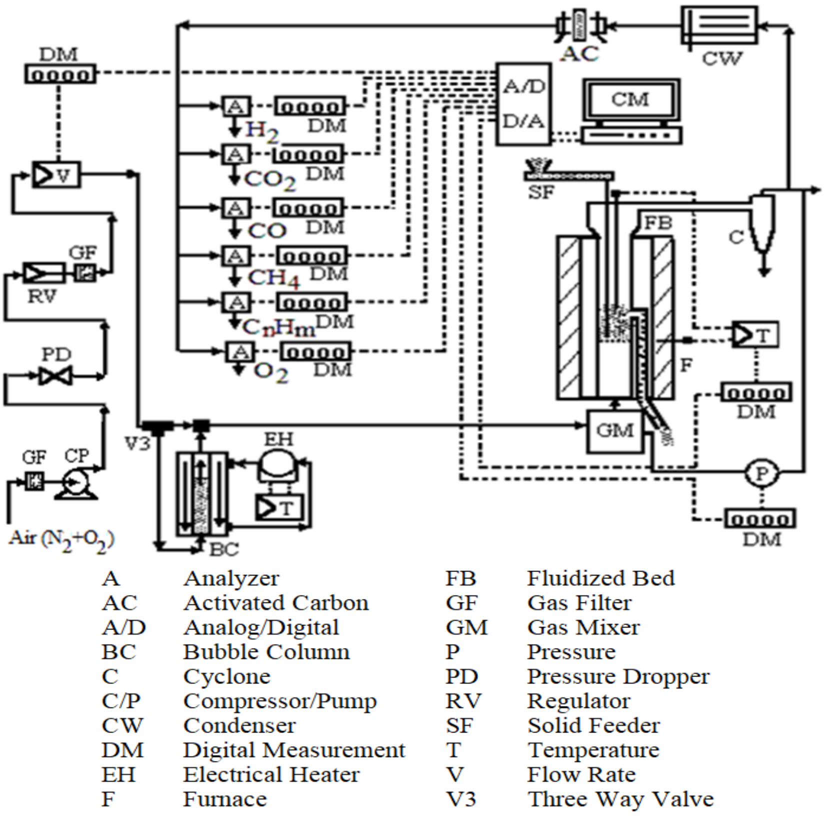

Figure 2. *The experimental set-up*

2.6. Gasification Reactions

Gasification reactions that may affect the producer gas content obtained from gasification experiments were investigated. Water-gas, water-gas shift, steam reforming and hydrogasification reactions occur in the presence of steam (Kumar et al., 2009). With the Boudouard reaction, which includes the reaction of C with CO_2, the syngas with a high CO content can be produced. The need for a gas purification process is reduced by converting of CO_2, which has a greenhouse effect, into CO (Huang et al., 2021). Main gasification reactions are shown in Table 3 (Block et al., 2018).

Table 3. *Main gasification reactions*

Gasification Reactions		**ΔH^0_{298K} (kJ/mol)**
Complete Oxidation	$C + 0,5O_2 \Leftrightarrow CO$	-111
Partial Oxidation	$C + 0,5O_2 \Leftrightarrow CO_2$	-283
Water-gas Reaction	$C + H_2O \Leftrightarrow CO + H_2$	+131
Water-gas Shift Reaction	$CO + H_2O \Leftrightarrow CO_2 + H_2$	-42
Steam Reforming Reaction	$CH_4 + H_2O \Leftrightarrow CO + 3H_2$	+206
Hydrogasification Reaction	$C + 2H_2 \Leftrightarrow CH_4$	-75
Boudouard Reaction	$C + CO_2 \Leftrightarrow 2CO$	+173

3. RESULTS AND DISCUSSION

The cotton-based banknote sample has a similar structure to cotton gin waste and cotton fibers in terms of characterization test results (Table 4).

Table 4. *Comparison of characterization analysis of cotton and cotton-based banknote sample*

Sample	**Proximate Analysis (wt.%)**		**Calorific Value (MJ/kg)**	**Elemental Analysis (wt.%)**					**Lignocellulosic Analysis (wt.%)**		
	Volatile Solid Matter	Ash	Higher Calorific Value	C	H	N	S	O	Cellulose	Hemicellulose	Lignin
Cotton-based Banknote (5 TL) (Kavuştu & Şimşek, 2023)	81.12	4.83	17.42	38.95	6.70	0.65	-	53.70	71.82	2.83	17.64
Cotton Gin Waste (Raveendran et al., 1995)	88.00	5.40	17.48	42.70	6.00	0.10	-	49.50	77.80	16.00	-
Cotton Fiber (Corradini et al., 2009)									88.3±0.3	8.0±0.3	4.8±0.5

The higher calorific value of the cotton-based banknote sample is higher than that of oil palm empty fruit bunch (17.02 MJ/kg), beef cattle manure (15.93 MJ/kg), rice straw (16.28 MJ/kg) and rice hulls (16.14 MJ/kg) (Widjaya et al., 2018). The effect of lignin content of biomass on energy content is greater than cellulose and hemicellulose (Elbersen et al., 2017). The lignin content in the cotton-based banknote sample is higher than that in the wheat straw (17 %), rice straw (12 %) and poultry litter (11 %) (Ren et al., 2019).

3.1. DSC Analysis Results

The DSC curves obtained at six different heating rates in the range of 5-30°C/min are shown in Figure 3. Endothermic peaks observed in the range of 93.51-179.91°C at different heating rates indicated physical changes and the removal of moisture from the fibers. Exothermic peaks observed in the range of 357.04-398.11°C represented radical and char formation reactions of small molecules such as CO, CO_2 and acetaldehyde, which were converted into intermediate products with the increase in temperature (Smith et al., 2024). Afterwards, the endothermic values obtained indicated the ongoing decomposition, which was more prominent at the heating rate of 10°C/min. In the study of Smith et al. (2024), where DSC analysis was performed on cotton, an endothermic peak was observed in the range of 120-150°C and an exothermic peak at 372°C, similar to the cotton-based banknote sample.

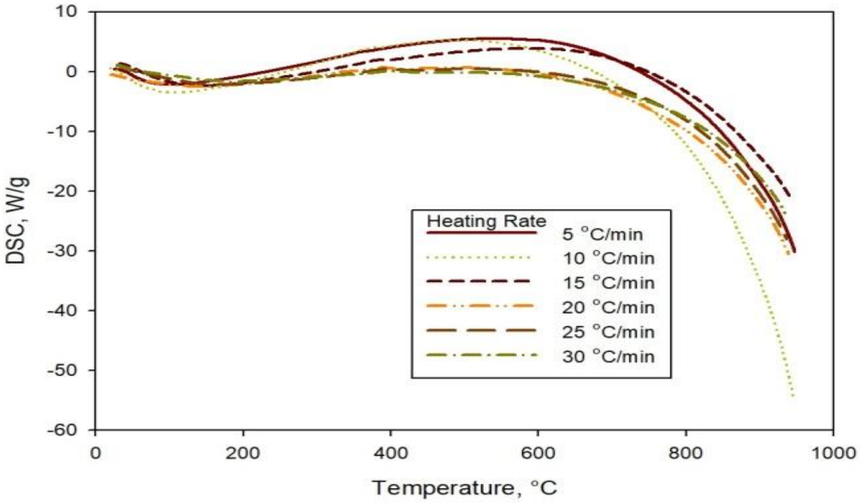

Figure 3. *DSC curves of cotton-based banknote sample*

3.2. FTIR Analysis Results

The FTIR spectrum obtained at six different heating rates in the range of 5-30°C/min are shown in Figure 4. The bands from 3500-3700 cm^{-1} represented the O–H or N–H stretching of carbohydrates, proteins and adsorbed water. The peaks in the 2800-3200 cm^{-1} presented the C-H stretching in cellulose, hemicellulose and hydrocarbons. The peaks in the 1300-1800 cm^{-1} indicated the C=O stretching modes of carbonyl groups originating from esters, carboxylic acids, ketones and aldehydes. The C-O stretching in the bands of 1000-1200 cm^{-1} indicated the presence of ethers, alcohols and phenols (Portella et al., 2016; He et al., 2022; Pang et al., 2024). With FTIR analysis of cotton-based banknote sample, similar peaks were obtained with the cotton organs and tissues in the study of He et al. (2022) and cotton fibers in the study of Portella et al. (2016).

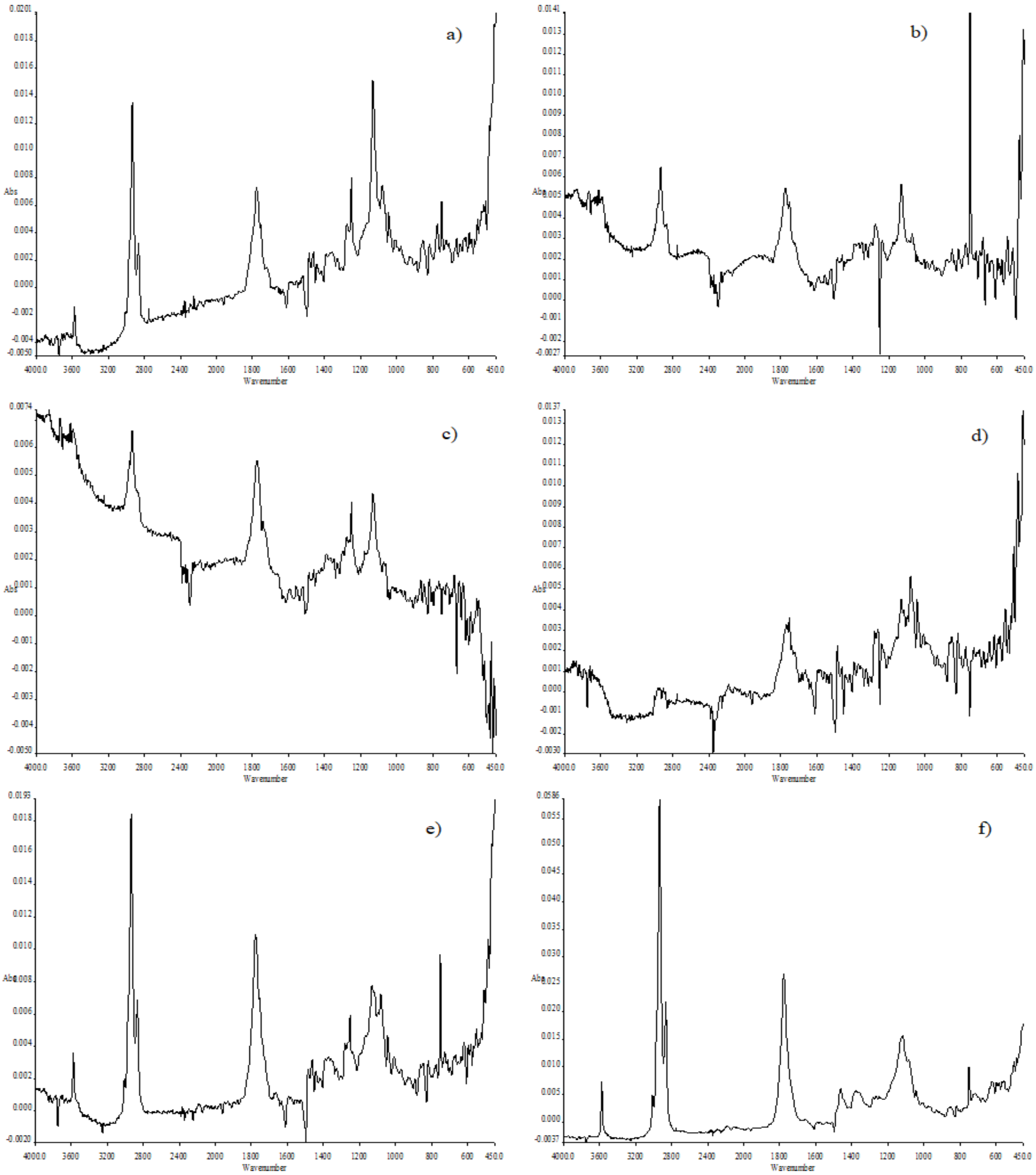

Figure 4. *FTIR spectrum of cotton-based banknote sample at heating rates (°C/min) of* ***a)*** *5,* ***b)*** *10,* ***c)*** *15,* ***d)*** *20,* ***e)*** *25, and* ***f)*** *30*

3.3. Activation Energy Results

The plots obtained for different extent of conversions using the KAS and FWO methods are shown in Figure 5, and the calculated activation energies are given in Table 5. The regression lines and calculated activation energies obtained from the KAS and FWO methods were compatible with each other. The activation energies of the cotton-based banknote sample were calculated in the range of 134-171 kJ/mol, and these values continued to increase until the x=0.6 conversion and then started to decrease. In the study of Yousef et al. (2020), the activation energies of the end-of-life Egyptian Pound (EGP) cotton-rich banknote were calculated in the range of 157-276 kJ/mol by FWO and KAS methods, and these values continued to increase until the x=0.7 conversion and then started to decrease. Multi-step reactions occurred due to the ink, hemicellulose, lignin and other materials contained in the sample, making it difficult to examine thermal degradation (Yousef et al., 2020). The difference in activation energies of TL and EGP banknotes is due to the difference in their chemical composition and structure. In the study of Corradini et al. (2009), the average activation energy for cotton fibers was calculated as 150.4±3.6 kJ/mol by FWO method. This value was nearly the same as the value calculated with FWO as 154.25 kJ/mol for the cotton-based banknote sample.

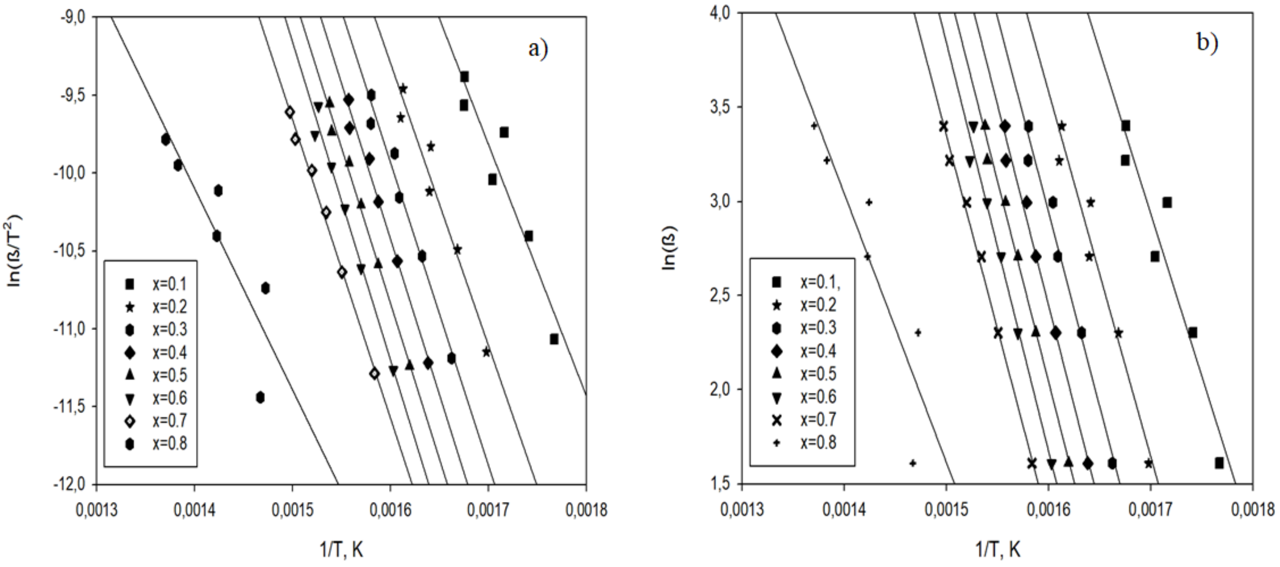

***Figure 5. a)** KAS and **b)** FWO plots of cotton-based banknote sample*

***Table 5.** Activation energies for cotton-based banknote sample*

Ea (kJ/mol)	**Extent of Conversion (x)**								
	0.1	0.2	0.3	0.4	0.5	0.6	0.7	0.8	Mean
Flynn-Wall-Ozawa (FWO)	136.638	152.946	162.841	167.563	167.689	171.419	161.734	113.168	154.250
Kissinger-Akahira-Sunose (KAS)	134.011	150.767	160.970	165.793	165.800	169.612	159.269	107.308	151.691

3.4. Effect of Torrefaction Pre-Treatment on Experimental Mole Fractions

Raw and torrefied (250°C for 10 min) cotton-based banknote samples were used as feedstocks. Gasification experiments were carried out at 800°C with an inlet H_2O/O_2 ratio of 25. Mole fraction-time curves obtained with gasification are given in Figure 6a and 6b. Considering the maximum mole fractions achieved, with torrefaction pre-treatment, H_2 mole fraction increased from 4.44% to 4.64%; CO mole fraction increased from 9.32% to 10.71% and CH_4 mole fraction increased from 3.47% to 3.65%, while CO_2 mole fraction decreased from 6.28% to 5.48%. The decrease in CO_2 mole fraction showed the effects of Boudouard reaction, the increase in CH_4 mole fraction showed the effects of hydrogasification reaction, and the increase in mole fractions of H_2 and CO showed the effects of water-gas and steam reforming reactions. The increase in CO mole fraction and the decrease in CO_2 mole fraction also showed that the effect of water-gas shift reaction was limited. The experimental results were consistent with the studies of Singh and Yadav (2021) and Sarker et al. (2022) in which the syngas yield increased as a result of gasification with torrefied biomass.

3.5. Effect of Co-Gasification Pre-Treatment on Experimental Mole Fractions

Samples prepared by mixing cotton and polymer-based banknote samples with the ratios of 50:50 and 75:25 (wt.%) were used as feedstocks. Gasification experiments were carried out at 800°C with an inlet H_2O/O_2 ratio of 25. Mole fraction-time curves obtained with gasification are given in Figure 6c and 6d. Considering the maximum mole fractions achieved, with the co-gasification of cotton and polymer-based samples at 75:25 ratio, H_2 mole fraction decreased from 4.44% to 3.71%; CO mole fraction decreased from 9.32% to 8.02% and CO_2 mole fraction decreased from 6.28% to 4.77%, while CH_4 mole fraction increased from 3.47% to 4.69%, compared to the raw cotton-based sample. With the co-gasification of cotton and polymer-based samples at 50:50 ratio compared to 75:25 ratio, H_2 mole fraction decreased from 3.71% to 3.68%; CO mole fraction decreased from 8.02% to 6.68% and CO_2 mole fraction from decreased 4.77% to 4.74%, while CH_4 mole fraction increased from 4.69% to 5.65%. Producer gas production was generally achieved efficiently and the decreasing maximum mole fractions indicated that the conversion efficiency of the gasification reactions and therefore the energy content of the gas decreased with the increasing ratio of the polymer-based sample in the feedstock. The increase in only the CH_4 mole fraction indicated that the hydrogasification reaction was effective and the effect of the steam reforming reaction was reduced. The conversion of feedstock primarily to tar with low char yield with the gasification of cellulose-rich biomass with plastic may have limited gas efficiency (Ajorloo et al., 2024). On the other hand, the difference in materials and inks contained in cotton and polymer-based samples may have also caused this result.

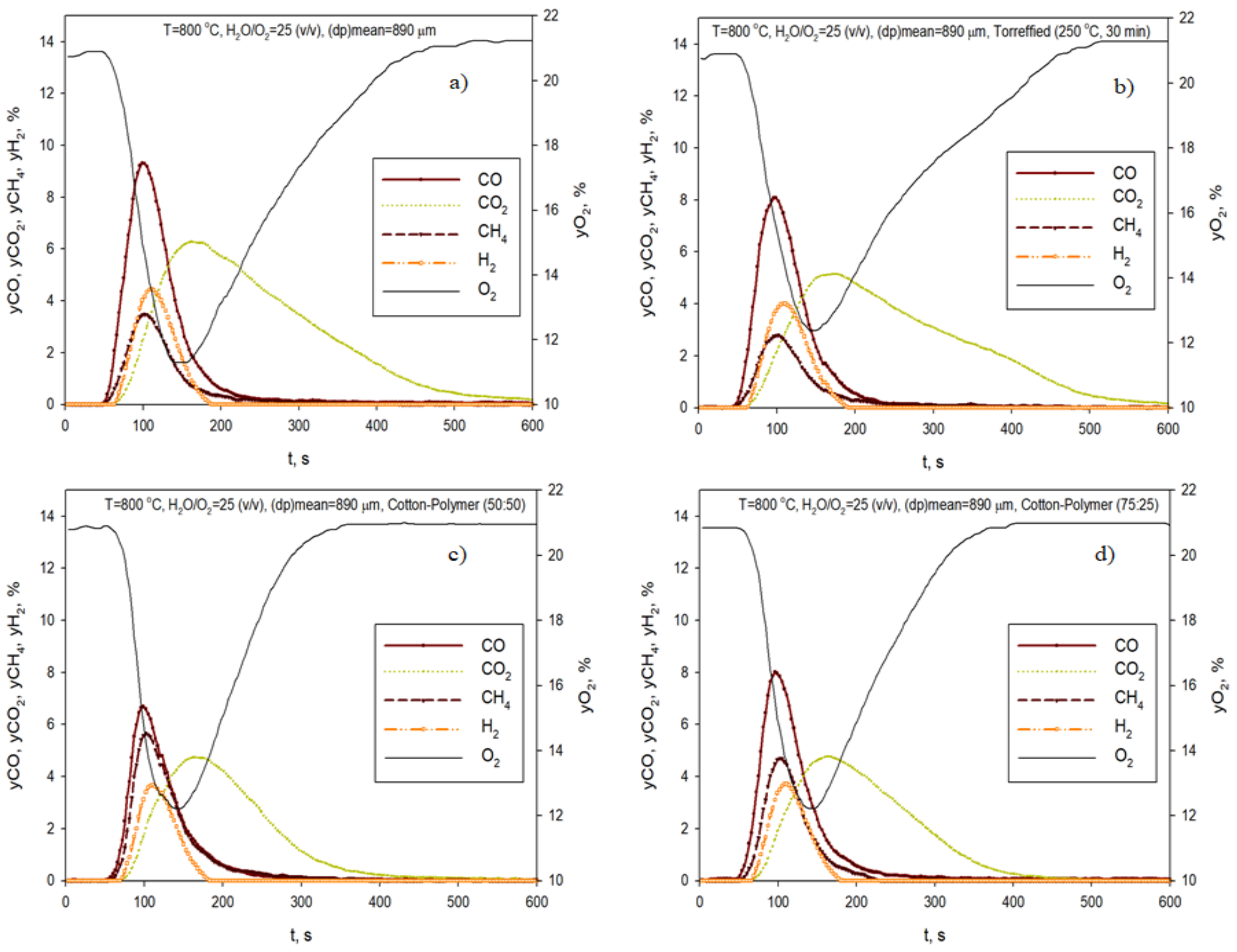

Figure 6. *Mole fraction-time curves for **a)** raw, **b)** torrefied cotton-based banknote samples; **c)** 50:50, **d)** 75:25 cotton:polymer-based banknote samples*

4. CONCLUSION

The results of DSC and FTIR analysis performed on the cotton-rich banknote sample and the calculated mean activation energy by FWO method showed that the cotton-based banknote sample had characteristic structures similar to those of cotton and thus end-of-life cotton banknotes could be used as a biomass source. With the non-isothermal kinetic methods of Flynn-Wall-Ozawa (FWO) and Kissinger-Akahira-Sunose (KAS), the activation energies were calculated as 134-171 kJ/mol at different conversions and the activation energy started to decrease after the x=0.6 conversion. This situation showed that the multi-step reactions occurring due to the complex structure of the sample had made it difficult to study the thermal decomposition.

In the gasification experiments performed at 800°C with an inlet H_2O/O_2 ratio of 25, the effects of torrefaction pre-treatment and co-gasification on the producer gas content were investigated with mole fraction- time curves. With the gasification torrefied (250°C for 10 min) cotton-based banknote sample, the H_2, CO, CH_4 mole fractions and product yield increased, while the CO_2 mole fraction decreased. With the increase in the polymer-based sample ratio in the feedstock, the gas product yield and H_2, CO, CO_2 mole fractions decreased, while the CH_4 mole fraction increased. In the gasification of torrefied cotton-based banknote sample, the water-gas, water-gas shift, steam reforming and hydrogasification reactions, which took place in the steam environment, were effective; while in the co-gasification of the cotton and polymer-based banknote samples, the hydrogasification reaction was effective.

In conclusion, the results of the study showed that (i) end-of-life cotton banknotes can be effectively used as a biomass source in thermochemical processes, (ii) torrefaction can facilitate the storage and transportation of dispersed waste banknotes, and thus improved fuel properties can increase producer gas efficiency, (iii) producer gas can be efficiently obtained by co-gasification of end-of-life cotton and polymer-based banknotes considering the diversity of these wastes.

AUTHOR CONTRIBUTIONS

Methodology, H.K. and E.H.Ş.; laboratory work, H.K.; manuscript-original draft, H.K.; manuscript-review and editing, H.K. and E.H.Ş. All authors have read and legally accepted the final version of the article published in the journal.

ACKNOWLEDGEMENT

This study is a part of PhD thesis of H.K. under the supervision of E.H.Ş. (Kavuştu, 2024). Gasification experiments were carried out in Ankara University Chemical Engineering Environmental Research Laboratory. DSC and FTIR analysis were performed in METU Central Laboratory.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

REFERENCES

Ajorloo, M., Ghodrat, M., Scott, J., & Strezov, V. (2024). Experimental analysis of the effects of feedstock composition on the plastic and biomass co-gasification process. *Renewable Energy*, *231*, 120960. https://doi.org/10.1016/j.renene.2024.120960

Baruah, D., & Baruah, D. C. (2014). Modeling of biomass gasification: A review. *Renewable and Sustainable Energy Reviews*, *39*, 806-815. https://doi.org/10.1016/j.rser.2014.07.129

Block, C., Ephraim, A., Weiss-Hortala, E., Minh, D. P., Nzihou, A., & Vandecasteele, C. (2018). Co-pyrogasification of plastics and biomass, a Review. *Waste and Biomass Valorization*, *10*(3), 483-509. https://doi.org/10.1007/s12649-018-0219-8

BoC. (2011). *Life cycle assessment of Canada's polymer bank notes and cotton-paper bank notes final report*. Bank of Canada.

BoI. (2023). *Environment Report 2023*. Bank of Italy.

Cabuk, B., Duman, G., Yanik, J., & Olgun, H. (2020). Effect of fuel blend composition on hydrogen yield in co-gasification of coal and non-woody biomass. *International Journal of Hydrogen Energy*, *45*(5), 3435-3443. https://doi.org/10.1016/j.ijhydene.2019.02.130

Chan, Y. H., Cheah, K. W., How, B. S., Loy, A. C. M., Shahbaz, M., Singh, H. K. G., Yusuf, N. R., Shuhaili, A. F. A., Yusup, S., Ghani, W. A. W. A. K. G., Rambli, J., Kansha, Y., Lam, H. L., Hong, B. H., & Ngan, S. L. (2019). An overview of biomass thermochemical conversion technologies in Malaysia. *The Science of the Total Environment*, *680*, 105-123. https://doi.org/10.1016/j.scitotenv.2019.04.211

Corradini, E., Teixeira, E. M., Paladin, P. D., Agnelli, J. A., Silva, O. R. R. F., & Mattoso, L. H. C. (2009). Thermal stability and degradation kinetic study of white and colored cotton fibers by thermogravimetric analysis. *Journal of Thermal Analysis and Calorimetry*, *97*(2), 415-419. https://doi.org/10.1007/s10973-008-9693-8

Das, B., Bhattacharya, A., & Datta, A. (2020). Kinetic modeling of biomass gasification and tar formation in a fluidized bed gasifier using equivalent reactor network (ERN). *Fuel*, *280*, 118582. https://doi.org/10.1016/j.fuel.2020.118582

DLR. (2022). *Annual Report 2022*. De La Rue plc.

DLR. (2023). *Annual Report 2023*. De La Rue plc.

ECB. (2023). *Product Environmental Footprint study of euro banknotes as a payment instrument*. European Central Bank.

Elbersen, W., Lammens, T. M., Alakangas, E. A., Annevelink, B., Harmsen, P., & Elbersen, B. (2017). Lignocellulosic Biomass Quality: Matching Characteristics With Biomass Conversion Requirements. In: C. Panoutsou (Eds.), *Modeling and Optimization of Biomass Supply Chains*, (pp. 55-78). https://doi.org/10.1016/b978-0-12-812303-4.00003-3

George, J., Arun, P., & Muraleedharan, C. (2019). Experimental investigation on co-gasification of coffee husk and sawdust in a bubbling fluidised bed gasifier. *Journal of the Energy Institute*, *92*(6), 1977-1986. https://doi.org/10.1016/j.joei.2018.10.014

G+D (2024). Number of Banknotes in circulation worldwide (Accessed:01/09/2024) https://www.gi-de.com/en/currency-technology/banknote-solutions/banknote-production/banknote-printing

Hanchate, N., Ramani, S., Mathpati, C., & Dalvi, V. H. (2021). Biomass gasification using dual fluidized bed gasification systems: A review. *Journal of Cleaner Production*, *280*, 123148. https://doi.org/10.1016/j.jclepro.2020.123148

Hanegraaf, R., Larçin, A., Jonker, N., Mandley, S., & Miedema, J. (2019). Life cycle assessment of cash payments in the Netherlands. *The International Journal of Life Cycle Assessment*, *25*(1), 120-140. https://doi.org/10.1007/s11367-019-01637-3

He, Z., Liu, Y., Kim, H. J., Tewolde, H., & Zhang, H. (2022). Fourier transform infrared spectral features of plant biomass components during cotton organ development and their biological implications. *Journal of Cotton Research*, *5*(1). https://doi.org/10.1186/s42397-022-00117-8

Huang, J., Zhang, H., Tan, Q., Li, L., Xu, R., Xu, Z., & Li, X. (2021). Enhanced conversion of CO_2 into O_2-free fuel gas via the Boudouard reaction with biochar in an atmospheric plasmatron. *Journal of CO_2 Utilization*, *45*, 101429. https://doi.org/10.1016/j.jcou.2020.101429

Hussain, M., Ali, O., Raza, N., Zabiri, H., Ahmed, A., & Ali, I. (2023). Recent advances in dynamic modeling and control studies of biomass gasification for production of hydrogen rich syngas. *RSC Advances*, *13*(34), 23796-23811. https://doi.org/10.1039/d3ra01219k

IEA. (2021). Energy supply (Accessed:01/09/2024) https://www.iea.org/world/energy-mix

IEA. (2023). *World Energy Outlook 2023*. International Energy Agency.

Jahromi, R., Rezaei, M., Samadi, S. H., & Jahromi, H. (2021). Biomass gasification in a downdraft fixed-bed gasifier: Optimization of operating conditions. *Chemical Engineering Science*, *231*, 116249. https://doi.org/10.1016/j.ces.2020.116249

K&B (2022). Annual increase trend in banknote production (Accessed:01/09/2024) https://www.koenig-bauer.com/en/news/details/article/90-of-the-worlds-banknotes-are-swiss-1

Kavuştu, H., & Şimşek, E. H. (2023). Characterization and gasification of end-of-life banknotes rich in cotton content. *Waste Management*, *171*, 473-481. https://doi.org/10.1016/j.wasman.2023.09.034

Kavuştu, H. (2024). *Gasification of end-of-life banknotes rich in cotton content in a fluidized bed reactor and modelling (in Turkish)*. PhD Thesis, Ankara University.

Kumar, A., Jones, D., & Hanna, M. (2009). Thermochemical Biomass Gasification: A Review of the Current Status of the Technology. *Energies*, *2*(3), 556-581. https://doi.org/10.3390/en20300556

Luján-Ornelas, C., del C Sternenfels, U. M., & Güereca, L. P. (2018). Life cycle assessment of Mexican polymer and high-durability cotton paper banknotes. *The Science of the Total Environment*, *630*, 409-421. https://doi.org/10.1016/j.scitotenv.2018.02.177

Mancilla-Leytón, J., Fernández-Rodríguez, M., De La Lama-Calvente, D., & Borja, R. (2024). Evaluation of batch mesophilic anaerobic digestion of waste Euro banknotes for methane Production: Preliminary studies and kinetic approach. *Waste Management*, *173*, 22-28. https://doi.org/10.1016/j.wasman.2023.11.003

Meng, S., Li, W., Li, Z., & Song, H. (2023). Recent progress of the transition metal-based catalysts in the catalytic biomass gasification: A mini-review. *Fuel*, *353*, 129169. https://doi.org/10.1016/j.fuel.2023.129169

Mohammed, H. I., Garba, K., Ahmed, S. I., & Abubakar, L. G. (2022). Thermodynamics and kinetics of Doum (Hyphaene thebaica) shell using thermogravimetric analysis: A study on pyrolysis pathway to produce bioenergy. *Renewable Energy*, *200*, 1275-1285. https://doi.org/10.1016/j.renene.2022.10.042

Ng, R. T., Tay, D. H., Ghani, W. a. W. a. K., & Ng, D. K. (2013). Modelling and optimisation of biomass fluidised bed gasifier. *Applied Thermal Engineering*, *61*(1), 98-105. https://doi.org/10.1016/j.applthermaleng.2013.03.048

Pang, Y., Zhu, X., Li, N., & Wang, Z. (2024). Study on CO_2 co-gasification of cellulose and high-density polyethylene via TG-FTIR and ReaxFF MD. *Process Safety and Environmental Protection*. https://doi.org/10.1016/j.psep.2024.04.119

Parrillo, F., Ardolino, F., Calì, G., Marotto, D., Pettinau, A., & Arena, U. (2021). Fluidized bed gasification of eucalyptus chips: Axial profiles of syngas composition in a pilot scale reactor. *Energy*, *219*, 119604. https://doi.org/10.1016/j.energy.2020.119604

Parrillo, F., Ardolino, F., Boccia, C., Calì, G., Marotto, D., Pettinau, A., & Arena, U. (2023). Co-gasification of plastics waste and biomass in a pilot scale fluidized bed reactor. *Energy*, *273*, 127220. https://doi.org/10.1016/j.energy.2023.127220

Portella, E. H., Romanzini, D., Angrizani, C. C., Amico, S. C., & Zattera, A. J. (2016). Influence of Stacking Sequence on the mechanical and dynamic mechanical properties of cotton/glass fiber reinforced polyester composites. *Materials Research*, *19*(3), 542-547. https://doi.org/10.1590/1980-5373-mr-2016-0058

Raveendran, K., Ganesh, A., & Khilar, K. C. (1995). Influence of mineral matter on biomass pyrolysis characteristics. *Fuel*, *74*(12), 1812-1822. https://doi.org/10.1016/0016-2361(95)80013-8

Ren, J., Cao, J., Zhao, X., Yang, F., & Wei, X. (2019). Recent advances in syngas production from biomass catalytic gasification: A critical review on reactors, catalysts, catalytic mechanisms and mathematical models. *Renewable and Sustainable Energy Reviews*, *116*, 109426. https://doi.org/10.1016/j.rser.2019.109426

RDK. (2022). Summary banknote recycling study November 2022 (Accessed:01/09/2024) https://www.royaldutchkusters.com/blog/summary-banknote-recycling-study-2022

Sadaka, S. S. (2013). Gasification of raw and torrefied cotton gin wastes in an auger system. *Applied Engineering in Agriculture*, 405-414. https://doi.org/10.13031/aea.29.9919

Sarker, T. R., Nanda, S., Meda, V., & Dalai, A. K. (2022). Process optimization and investigating the effects of torrefaction and pelletization on steam gasification of canola residue. *Fuel*, *323*, 124239. https://doi.org/10.1016/j.fuel.2022.124239

Sheikh, M. M. I., Kim, C. H., Park, H. J., Kim, S. H., Kim, G. C., Lee, J. Y., Sim, S., & Kim, J. W. (2013). Alkaline pretreatment ımproves saccharification and ethanol yield from waste money bills. *Bioscience Biotechnology and Biochemistry*, *77*(7), 1397-1402. https://doi.org/10.1271/bbb.130002

Singh, D., & Yadav, S. (2021). Steam gasification with torrefaction as pretreatment to enhance syngas production from mixed food waste. *Journal of Environmental Chemical Engineering*, *9*(1), 104722. https://doi.org/10.1016/j.jece.2020.104722

Smith, D. L., Montemayor, M. D., Carosio, F., & Grunlan, J. C. (2024). Universal intumescent polyelectrolyte complex treatment for cotton, polyester, and blends thereof. *Polymer Degradation and Stability*, *228*, 110936. https://doi.org/10.1016/j.polymdegradstab.2024.110936

Widjaya, E. R., Chen, G., Bowtell, L., & Hills, C. (2018). Gasification of non-woody biomass: A literature review. *Renewable and Sustainable Energy Reviews*, 89, 184-193. https://doi.org/10.1016/j.rser.2018.03.023

Xie, J., Zhong, W., Shao, Y., & Zhou, G. (2021). Simulation of co-gasification of coal and wood in a dual fluidized bed system. *Advanced Powder Technology*, *32*(1), 52-71. https://doi.org/10.1016/j.apt.2020.11.017

Yousef, S., Eimontas, J., Striūgas, N., Trofimov, E., Hamdy, M., & Abdelnaby, M. A. (2020). Conversion of end-of-life cotton banknotes into liquid fuel using mini-pyrolysis plant. *Journal of Cleaner Production*, *267*, 121612. https://doi.org/10.1016/j.jclepro.2020.121612

Yousef, S., Kuliešienė, N., Sakalauskaitė, S., Nenartavičius, T., & Daugelavičius, R. (2021). Sustainable green strategy for recovery of glucose from end-of-life euro banknotes. *Waste Management*, *123*, 23-32. https://doi.org/10.1016/j.wasman.2021.01.007

Zhang, X., Xu, W., Rauf, A., & Ozturk, I. (2024). Transitioning from conventional energy to clean renewable energy in G7 countries: A signed network approach. *Energy*, *307*, 132655. https://doi.org/10.1016/j.energy.2024.132655