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Upgradation in Physio-Chemical Characteristics and Thermal Behaviour of Thar Coal Subjected to Torrefaction

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ABSTRACT

This study explores the improvement in physiochemical attributes, thermal behavior, and combustion characteristics of low-rank Thar coal through torrefaction, a thermal pretreatment process. Thar coal samples were torrefied at five temperatures (200, 225, 250, 275, and 300°C) for two residence times (30 and 60 minutes). The study assessed changes in proximate and ultimate composition, calorific value, energy density and combustion traits of untreated and torrefied coal using thermogravimetric analysis (TGA). Results indicated slight improvement at low torrefaction temperatures (200-225°C), while higher temperatures (275-300°C) significantly enhanced fuel properties. Key findings include increased fixed carbon, reduced moisture and volatile matter, and higher heating value (HHV). The TGA graph demonstrated that torrefied coal has improved fuel properties compared to raw coal samples. The derivative thermogravimetric curve (DTG) shifted upward, signifying a change in peak (Tm) and decomposition temperatures along with an increased torrefaction temperature. The samples torrefied at 300°C for 60 minutes yielded best results, showing improved ignition (Ti) and burnout temperatures (Tb) and reduced emissions. This research focuses on torrefaction as a potential technique for improving the quality of low-grade coals of Pakistan hence opening up opportunities for controlled energy production.

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1. Introduction

With rising population and industrialization, the demand for energy continues to escalate at an unprecedented pace. The International Energy Agency (IEA) projects a 25% surge in global energy needs by 2040 (Kohl, 2019). While different renewable energy options are gaining traction, challenges such as high costs and technological constraints limit their widespread adoption in developing economies like Pakistan (Qadir et al., 2021). Coal remains a critical energy source, contributing to over one-third of global electricity generation in 2022

(Liu, 2023). A positive correlation has been observed between coal development, industrialization, and economic growth in many developing countries (Joshua et al., 2020; Zhang et al., 2017). Nonetheless, growing environmental concerns and the demand for sustainable and cleaner energy sources have induced extensive research into coal's viability as a sustainable energy source.

Pakistan faces an energy shortfall; the country is reliant on imported fossil fuels with the lowest per capita energy utilization worldwide (Naeem et al., 2021). Pakistan is experiencing a demand and supply deficit of about to 4,000–5,000 MW, which necessitates a review of the state's fundamental energy policy (Ahmed et al., 2020; Safeer & Fatima, 2019). Pakistan has the 6^{th} world largest lignite coal reserve of about 175 billion tons in thar coalfield, Sindh. This unmined reserve has the ability to fulfil Pakistan's energy needs for a minimum of 200 years (Kumar et al., 2021; Malik, 2023; Raza et al., 2022). Nevertheless, due to its physical and thermal characteristics like moisture content, heating value, and high volatility, its direct utilization is limited for high-efficiency energy applications (Ali et al., 2015; Kumar et al., 2021; Sarwar et al., 2012). Considering the unstable and expensive supply of imported fossil fuels, it is imperative to responsibly utilize potential indigenous resources and develop value-added chains to strive towards a circular economy. Energy yield at an affordable cost is one of the prevalent challenges that could be structured through assessment of contemporary techniques based on energetic optimization (Sarwar et al., 2012).

Torrefaction, a mild pyrolysis process, has appeared as an operational pretreatment method to enhance the combustion characteristics as well as energy density of low-grade coal. To avoid combustion the process of torrefaction is processed under inert atmosphere at a temperature range between The activity is conducted in an inert atmosphere to prevent combustion at 200-300°C and residence periods varying from a few minutes to hours (Chen et al., 2015; Nhuchhen et al., 2014; Rizvi et al., 2015; Van der Stelt et al., 2011). Raw feedstock's uniformity, grindability, stability, hydrophobicity, and energy density were reported to be significantly improved following the procedure (Phanphanich & Mani, 2011; Repellin et al., 2010). However, the composition of the sample, its physical and chemical characteristics, and its operational circumstances all affect how effectively the quality traits are upgraded (Chen et al., 2019; Parikh et al., 2007). Higher carbon concentrations in the raw material led to enhanced the significant amount of higher heating value (HHV) of the torrefied samples under the same torrefaction setting for varied species of lignocellulosic biomass (Chen & Kuo, 2011; Ivanovski et al., 2022). While extended residence time and increased torrefaction temperatures yielded decreased mass and increased energy content, Almeida et al. (2010) and Park et al. (2017) reported that low-rank coal showed influence of high torrefaction temperature (300°C) in terms of reduced moisture content and coal porosity along with improved calorific value. Kanwal, Munir, et al. (2019) studied the physicochemical characteristics of a lignite coal with corn cob torrefied at five distinctive temperatures and four different residence times. The analysis reported improvement in corn's characteristics could be compared to lignite coal. Guo et al. (2024) studied the ignition and burnout temperature of torrefied samples were observed to shift at higher temperatures, reflecting a boost in combustion efficiency of blends with increased air flow (Asif, 2009; Liu et al., 2016). Another study on lignite coal confirmed that torrefaction not only improves fuel properties but also lowers emissions and reduces power generation costs, supporting sustainable energy goals (Ibeto et al., 2017). With growing global interest, torrefaction has moved beyond research into the commercial sector, with several demonstration plants operating worldwide (Chew & Doshi, 2011; Koppejan et al., 2012; Madanayake et al., 2017). Most existing studies have focused on torrefied biomass as an alternative to low-grade coal (Kanwal, Munir, et al., 2019; Kutlu & Kocar, 2018). However, limited research is available on torrefaction applications specifically for low-rank coals like Thar coal.

This study aims to evaluate whether Thar lignite coal can be upgraded into a more efficient fuel using existing energy optimization methods. Thermogravimetric analysis (TGA) serves as a vital tool in this investigation, helping to track thermal decomposition, weight loss, and the evolution of volatile matter in coal samples under programmed heating (Heydari et al., 2015; Kumar et al., 2021; Speight, 2008). While much literature exists on biomass torrefaction, there is a clear research gap regarding the torrefaction of Thar coal. This study seeks to examine the physicochemical and combustion properties of Thar lignite and assess the potential of torrefaction as a transformative technique for improving local coal resources (Du et al., 2014; Naeem et al., 2021). By understanding how torrefaction and thermal degradation modify combustion behaviour, this work contributes to recognizing coal as a more environmentally acceptable energy source. The outcomes could support innovation in Pakistan's energy sector and promote the sustainable use of indigenous coal reserves (Cardona et al., 2019; Chen et al., 2022; Thomas, 2013).

2. Experimental Methodology

2.1. Sample Collection & Preparation

A representative 5 kg sample of local Thar coal was collected from the Thar coal mines with the assistance of mining engineers. For sample preparation ASTM standard procedure D-2013 was employed. Initially, the coal sample was milled and sieved using 60# and 100# ASTM standard sieves for proximate analysis and subsequent experimental work. To avoid humidity and contamination the prepared samples were stored in airtight plastic containers.

2.2. Pre-Torrefaction Analyses

2.2.1. Proximate, Ultimate and Gross Calorific Analyses of Coal Sample

The proximate analysis was conducted to measure volatile matter, moisture content, fixed carbon and ash content following the standards ASTM D-3173, ASTM D-3175, ASTM D-3174, and ASTM D-3172, respectively. ASTM D-5291 was employed to find out the % of hydrogen, carbon, nitrogen, sulphur and oxygen . The heating value was determined according to ASTM D-5865 standards using an oxygen bomb calorimeter . The PMI Helium Pycnometer was used to find out the true density.

2.2.2. Thermogravimetric Analysis

A thermogravimetric analyzer (TGA LECO-701) was utilized to evaluate the combustion characteristics under an air atmosphere. The samples (0.5 g) were heated at temperatures ranging from 25°C to 950°C at a rate of 20°C/min, with airflow maintained at 3.5 liters per minute. Weight loss and its rate were recorded to assess ignition, peak, and burnout temperatures.

The general kinetic expression of coal degradation is

$$\frac{dx}{dt} = k\,(T)\,f\,(x) \tag{1}$$

where x, the degree of conversion, and denotes the amount of the sample decomposed at time t. The kinetic parameters (A and E) as given in the equation can be determined via TGA experimental results, such as weight loss over time.

$$\text{Rate} = A \cdot e - RTE \tag{2}$$

where: Rate is the reaction rate,

- *A* = pre-exponential factor
- *E* = activation energy
- *R* = gas constant, and
- *T* = absolute temperature

2.3. Torrefaction

A 10 g coal sample was heated in a tube furnace and torrefaction temperatures were fixed at 200°C, 225°C, 250°C, 275°C, and 300°C under a constant nitrogen flow (3 mL/min) to maintain an inert atmosphere while the furnace temperature precision (error bars) was

±2 °C. A residence times of 30 and 60 minutes were employed. While every sample was tested in triplicates to ensure the re-producibility of the results and to obtain adequate portion of torrefied sample for post-treatment analyses. Post-treatment, the mass yield of samples was calculated using:

$$\text{Mass yield } = \frac{mt}{mr} \times 100 \qquad (3)$$

Where m_t is the mass of the torrefied product and m_r is the mass of the raw coal (both on a dry basis). While energy yields on a dry basis were calculated using the given equation:

$$\text{Energy yield } = \text{ Mass yield } \left(\frac{HHVt}{HHVr}\right) \qquad (4)$$

HHVt = Heating value of torrefied sample
HHVr = Heating value of raw sample (before torrefaction)

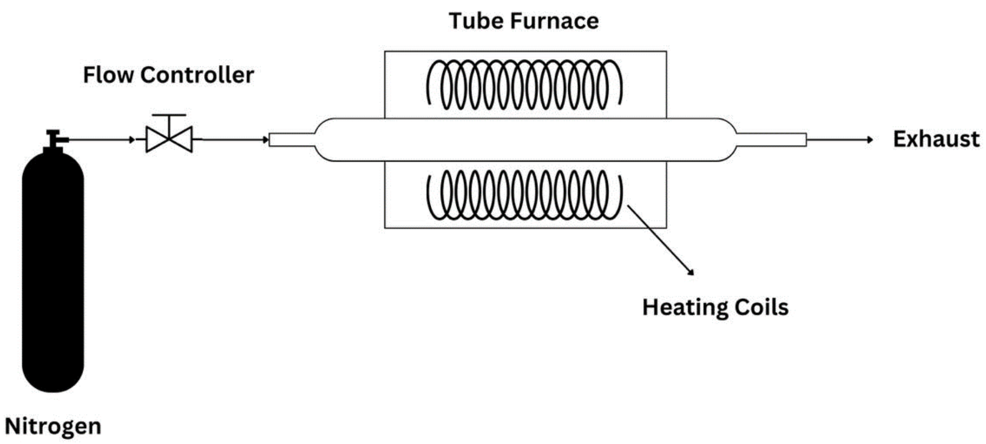

Figure 1: Lab scale Torrefaction setup

2.4. Post-Torrefaction Analysis

All pre-torrefaction tests were repeated for torrefied samples to determine changes in physicochemical characteristics, thermal behavior, and combustion efficiency. The results were compared across temperatures and residence times to identify optimal torrefaction conditions.

3. Results and Discussions

The results of proximate and ultimate analyses and the heating value of raw coal samples are reported here. The proximate analysis presents the profile of raw Thar coal in terms of moisture, FC, VM, ash and HHV. The coal samples were found to have high moisture and volatile content of 28.31% and 32.55% respectively. The percentage of fixed carbon was found to be around 21.23, while the ash content was around 3.0%. The coal contains a low HHV as compared to high quality coal, making it unsuitable for high-efficiency applications. The carbon, hydrogen, nitrogen, sulphur and oxygen content of the samples were found to be 61.30%, 5.51%. 0.75%, 1.02%, and 31.43% respectively.

Table 1
Baseline Proximate and Elemental Properties of Thar Coal Samples

Proximate Analysis of raw Thar Coal						Ultimate analysis of raw Thar Coal				
Mass Yield %	Moisture %	Volatile Matter (VM) %	Ash %	Fixed Carbon (FC) %	HHV MJ/Kg	Carbon %	Hydrogen %	Nitrogen %	Sulfur %	Oxygen %
100	28.31	32.55	3.0	21.23	14.61	61.30	5.51	0.75	1.02	31.43

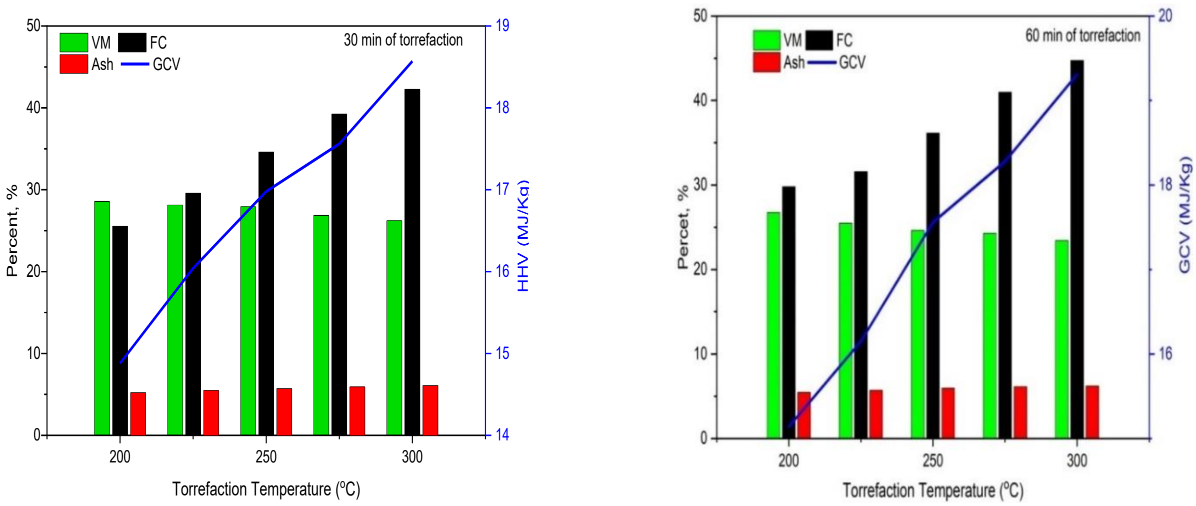

Figure 2 (a &b): Variation in Proximate Characteristics of Thar Coal Under Different Thermal Conditions

Torrefaction is a thermochemical treatment process whereby the structure of coal or biomass is impacted by removing volatile matter to enhance its carbon content and improve the fuel characteristics. The graph depicts that as the torrefaction temperature rises from 200°C to 300°C, the fixed carbon content of sample increases noticeably from around 25.5% to over 42% and up to 44.7% for the residence time of 30 and 60 minutes, respectively. The results indicated a decline in volatile matter in a manner dependent on torrefaction conditions: 28.58% to 26.21% and 26.74 to 23.45% respectively.(Park et al., 2017). It is assumed that torrefaction lowers volatile matter content due to the evaporation of light hydrocarbons and gases, such as CO, CO_2, and CH_4. The ash content does not show a significant change for various torrefaction conditions (Bergman et al., 2005). The ash content shows a slight rise from 5.46% to 6.21% even with higher temperatures and longer residence times. However, this increase in ash content is relatively insignificant. (Kanwal, Chaudhry, et al., 2019; Tumuluru, 2016). The HHV increases steadily as the torrefaction temperature and time increase. The increase in HHV indicates that the energy content of the torrefied coal improves as more volatiles are released and the carbon content becomes more concentrated. This enhancement makes torrefied coal a more energy-efficient fuel source, beneficial for energy related applications.

The graph highlights how torrefaction affects the elemental composition of coal, especially carbon and sulfur. As both temperature and residence time increased, the carbon content also rise from 61.87% to 70.02% at 30 minute, and from 62.09% to 71.27% at 60 minute residence time. This is beneficial because increased carbon translates to a higher energy capacity and improved combustion. Simultaneously, oxygen and hydrogen levels decrease nominally, most likely because of the losing moisture content and gases during heating. Slight losses of hydrogen, nitrogen, and sulfur are also indicative of removal of volatile compounds. This assists in cleaning the coal by reducing the amount of air pollutants such as NOx and SOx produced. The heating value (HHV) also improves steadily with higher torrefaction temperatures. As a guide, it rises as 14.88 to 18.57 MJ/kg in 30 minutes and 15.14 to 19.31 MJ/kg in 60 minutes. This rise can be attributed to increased carbon density and lower moisture levels so that the fuel becomes more effective in energy applications (Park et al., 2017; Rousset et al., 2011).

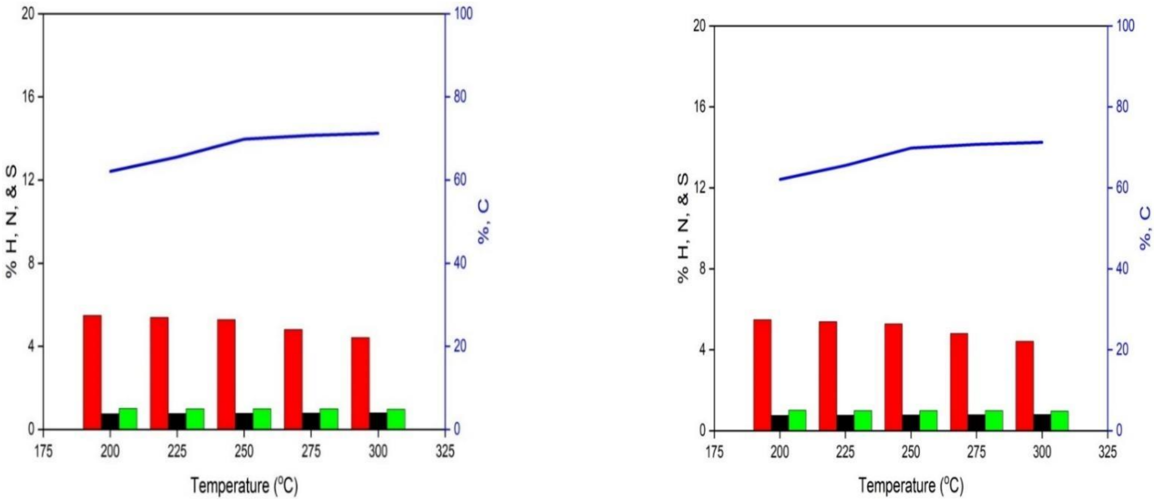

Figure 3 (a &b): Impact of Torrefaction Temperature and Residence Time on C, H, N & S Contents of Coal

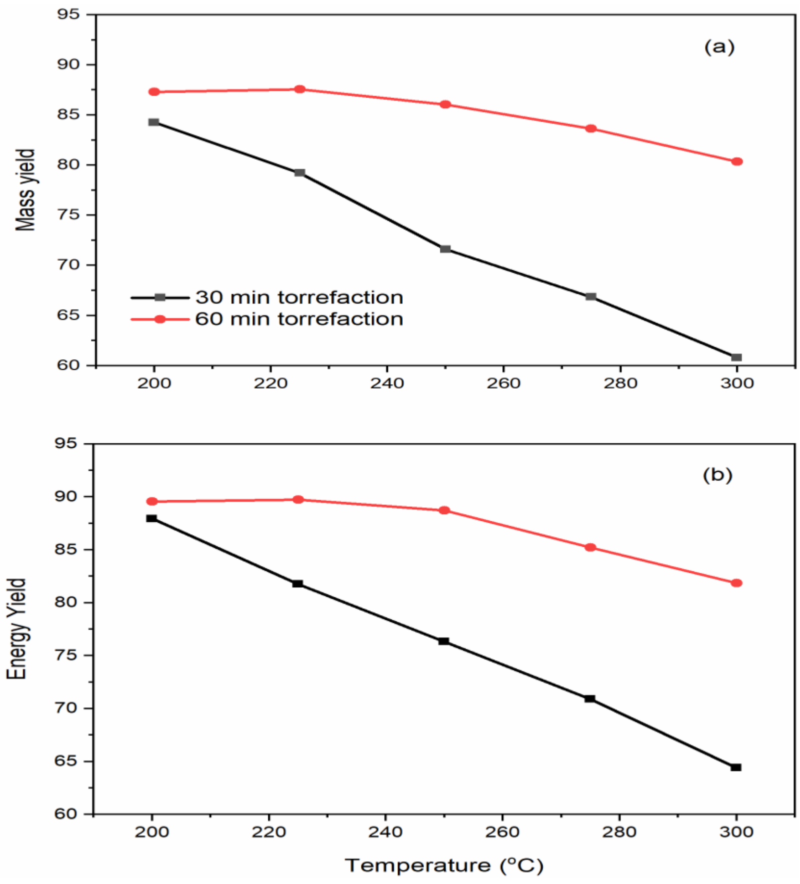

Figure 4 (a & b): Effect of Torrefaction on Mass and Energy Yield% of Coal at Varying Residence Time

The graphs represent the effect of temperature and residence time on the mass yield and energy yield of torrefied coal. Mass yield is the percentage of the original coal mass retained after torrefaction. Mass yield declined with increasing torrefaction temperature and time due to the release of volatiles, falling to 65% at 300°C for 30 minutes and 60.79% for 60 minutes. However, energy yield remained above 80% under optimal conditions, reflecting the retention of energy-dense components. Longer residence times (60 minutes) consistently yielded higher energy retention compared to 30-minute treatments, making them more suitable for high-efficiency applications. For instance, at 300°C and 60 minutes, the mass yield was 60.79%, while the energy yield exceeded 80%. The reductions in mass yield, i.e., 60.79% under severe torrefaction conditions (300°C and 60mins.), may have occurred due to the decomposition of volatile organic compounds, leaving behind a carbon-rich residue (Pahla et al., 2018; Park et al., 2017). Higher torrefaction temperatures reduce both mass and energy yields while longer residence time (60 minutes) retains more mass and energy compared to 30 minutes, suggesting that extending the residence time helps mitigate energy loss while achieving substantial improvements in coal properties (Bridgeman et al., 2008; Ramula, 2023).

3.1. TGA Analysis

TGA analysis of raw and torrefied Thar coal was conducted in an air environment at temperature increasing from ambient to 950°C with a fixed heating rate of 20°C/min.

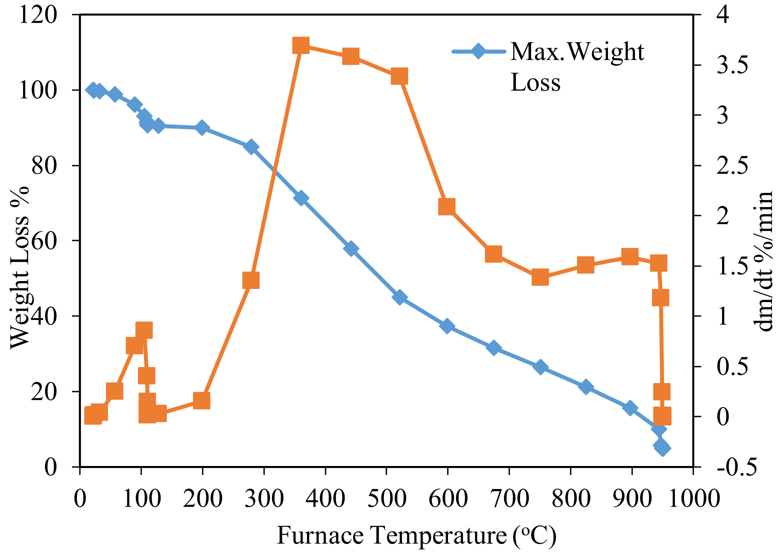

Figure 5: TGA and DTG Curve of Thar Coal (Un-treated Sample)

The combustion behavior of Thar coal was analyzed under an oxygen enriched environment with constant heating rate of 20°C/min to assess its thermal stability and efficiency. The TGA and DTG graphs of coal provide essential insights into its combustion behavior. The TGA curve displays weight loss as a function of temperature. The graph reveals a typical high-volatile coal profile, with substantial volatile matter, followed by steady fixed carbon combustion, and a small amount of ash remains after complete burn-off. The maximum and average weight loss rates of Thar coal were determined to be 3.689 and 0.760 %respectively. This initial weight loss can be attributed to the evaporation of moisture content. The coal moisture is usually released at low temperatures, typically around 100-150°C. As the temperature increases, weight loss between 150°C and 500°C was observed. This region represents the devolatilization phase, where volatile organic compounds, tars, and gases are released due to the thermal decomposition of coal's organic matter. The sharp decline in weight at around 300°C to 500°C is typical for coal and corresponds to the loss of volatile matter, which includes light hydrocarbons, CO, CO_2, and other gases. Between 500°C and 800°C, the weight loss continues but at a slower rate. This phase corresponds to the carbonization process, where the remaining carbon-rich material is undergoing thermal degradation. The weight loss here is primarily due to the breakdown of the remaining carbonaceous material and the release of fixed gases. Beyond 800°C, the weight loss rate significantly slows down, approaching a nearly constant value. This indicates the formation of ash, which is the inorganic residue left after the complete combustion of coal.

The DTG curve provides information on the rate of weight loss. Peaks in the DTG curve indicate the temperatures at which the rate of weight loss is at its maximum. The DTG curve showed three distinct peaks. The first small peak in the DTG curve corresponds to the moisture evaporation rate. The most prominent peak around 300°C to 500°C represents the maximum rate of devolatilization. This is where the bulk of volatile components are released. The smaller peaks in the 600°C to 800°C range indicate the decomposition of more stable organic materials and possibly some mineral decomposition. A sharp decline near 900°C indicates the final stages of the carbonization process, transitioning into ash formation. The DTG curve here approaches zero, reflecting the negligible weight loss as the material becomes mostly inert ash. Raw coal here appears to have moderate thermal stability and efficiency but a broad decomposition range. All this specifies that Thar coal loses material steadily over a wide temperature range with a slow and long-burning process.

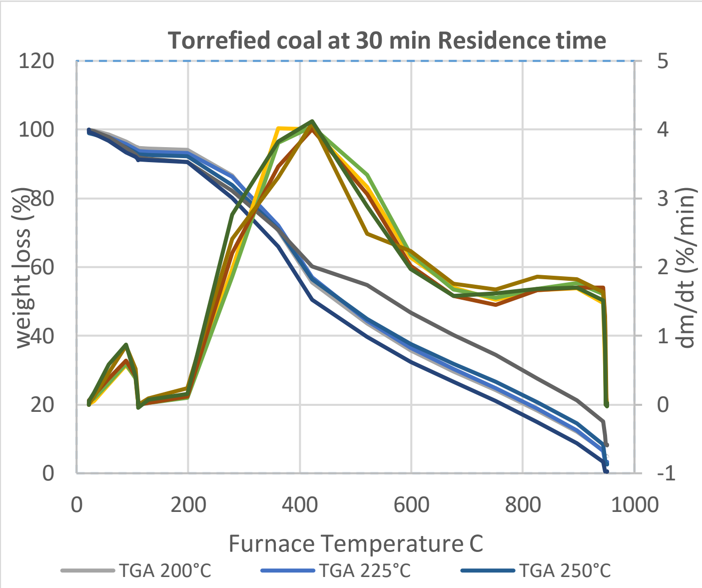

Figure 6: TGA and DTG Curve of Thar Coal Torrefied at 30 Min Residence Time

The graphs for coal torrefied at 200°C, 225°C, 250°C, 275°C, and 300°C with 30 residence times depict some alterations in thermal properties of coal. For coal torrefied at 200°C, the weight loss begins at a relatively low temperature, indicating the presence of residual volatiles. The peak temperature is slightly higher than raw coal, suggesting some increase in thermal stability. The peak decomposition rate shows that the material still has significant volatiles, but its combustion properties have enhanced compared to raw coal. The coal torrefied at 225 and 250C indicates significant improvement in stability, with an increase in ignition and peak temperature. This shows that torrefied coal is becoming more resistant to early decomposition. At 275°C significant thermal stability and efficiency is achieved as more volatiles have been removed. The ignition temperature has risen to 273°C, and the peak temperature is now 421°C. The maximum decomposition rate reaches 4.106, and burnout temperature reaches 946°C, suggesting high fuel efficiency and low residual content with improved combustion behavior. The sample torrefied at 300°C is the most stable sample, with an ignition temperature of 280°C and a peak temperature of 426°C. The coal is resistant to early ignition and has minimal volatiles left. Fuels with higher volatile matter content generally exhibit shorter ignition delay times. This is observed across various studies, indicating that the presence of more volatile components facilitates quicker ignition (Gong & Fu, 2001; Qi et al., 2021; Rybak et al., 2019).

An extended residence time appears to marginally improve stability for torrefied coal samples compared to the shorter residence time. The coal samples torrefied at 200°C and 250°C show behavior comparable to raw coal, with a significant weight loss during devolatilization and a large DTG peak. The peak temperature changes to 417°C and the ignition temperature rises to 264°C for torrefaction at 250°C, indicating considerable thermal stability and volatile removal. Torrefied coal at 275°C and 300°C shows reduced weight loss, delayed decomposition, and sharp DTG peaks. These samples exhibit enhanced thermal stability with a peak temperature of 427°C and 434°C respectively and an ignition temperature of 277°C and 286°C, respectively. The coal has very little volatile content and burns more efficiently, as evidenced by the rise in ignition and the peak shifting to higher temperatures. The stable samples (275°C and 300°C) have longer mass retention with a burnout temperature of 956°C and maximum decomposition rate of 4.51. At greater ranges of torrefaction temperature, DTG peaks shifted slightly toward greater temperatures, indicating that higher-torrefied coal degrades slowly over time, as described by the authors in Yang et al. (2019). The highest points of DTG curves are observed at coal torrefied of temp 250-300 C, where these abrupt spikes signify that, the combustibility is good at elevated torrefaction temperature.

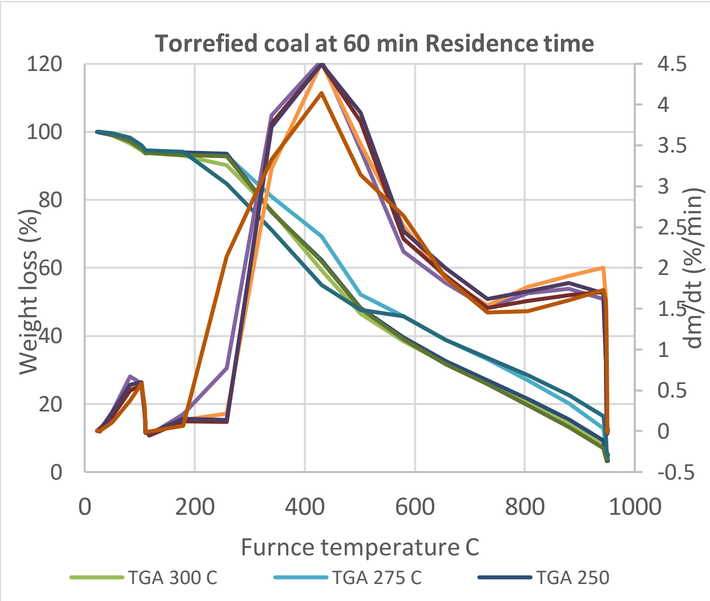

Figure 7: TGA and DTG Curve of Thar Coal Torrefied at 60 Min Residence Time

Table 2

Combustion Characteristics of Raw and Treated Thar Coal

Sample	Torrefaction Temperature	Heating Rate °C/min.	(dw/dt) max	(dw/dt) avg	Ignition Temperature °C	Peak Temperature °C	Burnout Temperature °C
Raw coal		20	3.689	0.760	235	375	870
Torrefied Coal	30 min						
	200°C		4.019	0.762	240	388	905
	225°C		4.044	0.770	251	405	932
	250°C	20	4.061	0.778	260	413	940
	275°C		4.106	0.788	273	421	946
	300°C		4.122	0.790	280	426	952
	60 min						
	200°C		4.141	0.764	243	391	913
	225°C		4.304	0.773	256	408	930
	250°C	20	4.499	0.782	264	417	944
	275°C		4.513	0.787	277	427	956
	300°C		4.546	0.793	286	434	965

Raw and torrefied coal TGA data and DTG graphs were used to obtain the peak temperature, burnout temperature, and ignition temperature. The analysis of the results reveals that as the torrefaction temperature goes up, Ti, Tm, and Tb also increases, possibly attributed to the loss of volatile matter in torrefaction. DTG peaks are shifted to higher temperatures, and breadth decreases, which is evidence of better devolatilization and concentration of energy in the torrefied product (Tian et al., 2016; Wang & Howard, 2017). The conclusions contrast with earlier research studies on torrefied biomass in which peak values in DTG tend to remain broad due to poor devolatilization (Jia, 2021; Park & Jang, 2012; Salehabadi et al., 2020). The findings show that the optimized torrefaction maximizes the fuel properties and reduces combustion risks during storage, which pertain specifically to the commercial scale of use (Kanwal, Munir, et al., 2019). Moreover, increased decomposition rates and burnout temperature are most likely to relate to better combustion and reduced emissions. This leads to a better combustion rate of the fuel, reduced emissions and improved energy returns (Huang et al., 2021; Zhang et al., 2017).

4. Conclusion

This study addresses a significant research gap in the use of low-grade lignite coal to substantially analyze the possibility of torrefaction for upgrading the different physicochemical as well as combustion characteristics of Thar coal. The effect of various torrefaction temperatures and residence time on the suitability of Thar coal as a resource for energy generation is further explored in this work. The studies show that torrefaction cause reduction in moisture and volatile matter and also augmented the fixed carbon content and calorific value of raw coal. Increased efficiency of energy per quantity of fuel means less coal is needed to produce the same level of energ,y which helps for better economic production(Cremers et al., 2015). Combustion analysis revealed enhanced thermal stability of the product owing to higher ignition and burnout temperatures. These improvements make Thar coal a more efficient and cleaner option for energy production.

The findings highlight that coal treated at 300°C for 60 minutes delivers the best results in terms of energy efficiency and thermal stability. Higher ignition and burnout temperatures make it especially suitable for waste-to-energy plants and other combustion-based systems. This means less coal is needed to produce the same amount of energy, which supports cost-effective and cleaner fuel production.

A key strength of this work is its focus on applying torrefaction as a modern solution for upgrading Pakistan's domestic coal. This not only reduces the need for imported fuels but also helps lower greenhouse gas emissions. For companies like Petronas and other energy producers, these findings offer a pathway toward more sustainable and localized energy strategies. Future studies should explore ways to improve torrefaction technology further, especially through better emission control systems and scaling up the process for industrial use.

Authors Contribution

Sumaira Kanwal, Javaid Akhter and Iram Batool conceived and designed the analysis and composed the manuscript.
Haleema Manawar performed the experiment.
Asma Majeed and Hassan Zeb contributed data analysis, manuscript write-up and review of manuscript.

Conflict of Interests/Disclosures

The authors declared no potential conflicts of interest w.r.t. the research, authorship and/or publication of this article.

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This study is a part of M.Phil research guided by Dr Sumaira Kanwal, PhD, (corresponding author). It was postulated in this research that by employing torrefaction treatment the combustion aspect of Thar coal could be enhanced, and, in this manner, the optimum utilization of local coal can reduce the costs that are incurred in importing international coal.

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