

01 Aug 2023

Development And Characterization Of Coal-Based Thermoplastic Composite Material For Sustainable Construction

Haibin Zhang

Wenyu Liao

Missouri University of Science and Technology, wliao@mst.edu

Genda Chen

Missouri University of Science and Technology, gchen@mst.edu

Hongyan Ma

Missouri University of Science and Technology, mahon@mst.edu

Follow this and additional works at: https://scholarsmine.mst.edu/civarc_enveng_facwork



Part of the [Architectural Engineering Commons](#), and the [Structural Engineering Commons](#)

Recommended Citation

H. Zhang et al., "Development And Characterization Of Coal-Based Thermoplastic Composite Material For Sustainable Construction," *Sustainability (Switzerland)*, vol. 15, no. 16, article no. 12446, MDPI, Aug 2023. The definitive version is available at <https://doi.org/10.3390/su151612446>



This work is licensed under a [Creative Commons Attribution 4.0 License](#).

This Article - Journal is brought to you for free and open access by Scholars' Mine. It has been accepted for inclusion in Civil, Architectural and Environmental Engineering Faculty Research & Creative Works by an authorized administrator of Scholars' Mine. This work is protected by U. S. Copyright Law. Unauthorized use including reproduction for redistribution requires the permission of the copyright holder. For more information, please contact scholarsmine@mst.edu.

Article

Development and Characterization of Coal-Based Thermoplastic Composite Material for Sustainable Construction

Haibin Zhang ^{1,2}, Wenyu Liao ^{2,*}, Genda Chen ²  and Hongyan Ma ^{2,*} 

¹ School of Civil Engineering and Architecture, Hainan University, Haikou 570228, China; zhanghb01@hainanu.edu.cn

² Department of Civil, Architectural and Environmental Engineering, Missouri University of Science and Technology, Rolla, MO 65409, USA

* Correspondence: wliao@mst.edu (W.L.); mahon@mst.edu (H.M.)

Abstract: The exploitation of coal and the disposal of waste plastic present significant environmental and economic challenges that require sustainable and profitable solutions. In response, we propose a renewable construction composite material of coal-based thermoplastic composite (CTC) that can be made from low-grade coal and plastic waste. We developed and tested the hot-press fabrication method for this CTC, using coal with a maximum particle size of 4.75 mm and recycled high-density polyethylene (HDPE). The effects of the coal fraction (50–80 wt%) on compressive properties, thermal properties, microstructure, and ecological and economic efficiencies of the CTC were investigated. Test results revealed that the compressive strength and modulus decrease as the coal fraction increases. However, the thermal properties, including thermal conductivity and specific heat, increase with higher coal contents. Compared to concrete, the CTC has about half the thermal conductivity and twice the specific heat, making it a more energy-efficient construction material. Microstructure testing helped to reveal the mechanisms behind the above behaviors of CTC from the observation of binder volume, bonding quality between coal and HDPE, and porosity variation. The life cycle analysis indicated that the CTC production reduced embodied energy, carbon footprint, and cost by up to 84%, 73%, and 14%, respectively. Therefore, we recommend the CTC with 50–70% coal fraction as an innovative construction material with satisfied mechanical and thermal properties, better cost efficiency, and a reduced ecological impact.

Keywords: coal-based thermoplastic composites; high-density polyethylene; mechanical properties; thermal properties; microstructure; ecological and economic efficiencies



Citation: Zhang, H.; Liao, W.; Chen, G.; Ma, H. Development and Characterization of Coal-Based Thermoplastic Composite Material for Sustainable Construction. *Sustainability* **2023**, *15*, 12446. <https://doi.org/10.3390/su151612446>

Academic Editor: Joao Paulo Davim

Received: 22 June 2023

Revised: 14 August 2023

Accepted: 14 August 2023

Published: 16 August 2023



Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (<https://creativecommons.org/licenses/by/4.0/>).

1. Introduction

Coal is widely used as a raw material for electricity, chemical, and heat production worldwide. In the United States, over 70% of electricity consumption in residential and commercial buildings, and more than 40% of associated greenhouse gas emissions, are attributed to thermal loads such as space and water heating [1], which involved 93% of coal consumption [2]. However, the potential climate and health effects of emissions from coal combustion, along with the recent development of shale gas, have significantly reduced coal consumption in the United States. Although the Department of Energy (DOE) [3] has explored and funded innovative approaches to coal use with minimal emissions, such as coal conversion to graphene, carbon nanotubes, and other high-margin solid production, these approaches require complicated pretreatments like high temperatures and pressures or harsh chemicals and are not produced at high enough volumes to impact coal sale at scale.

A more environmentally friendly, cost-effective, and scalable alternative for significant coal consumption should minimize pretreatment steps and maximize the conversion

efficiency of carbon from coal to final production [4]. Drawing from the literature [5], an encouraging approach emerges wherein natural-organic fillers are integrated with environmentally friendly polymer-based materials to fabricate innovative building products. In this context, polymeric composites that mix coal and polymer for construction materials have attracted more attention as a direct use of coal. Prior research mainly focused on blending fine coal powder and polymers to improve the composites' mechanical strength, flammability stability, and thermal properties [6–10]. However, in these composites, coal played the role of filler in construction materials, with a particle size and weight fraction of less than 125 μm and 60%, respectively. The fine particle size of coal usually requires vigorous grinding and sieving, which can be energy-intensive pretreatment procedures and may pose an increasing insidious hazard for finer combustible coal dust. Additionally, there are still limited studies on the performance of composites with a coal weight fraction larger than 60%.

Plastic waste generation in the United States has been increasing rapidly, with 40 million tons of municipal plastic waste generated in 2021. Unfortunately, recycling rates for plastics have been decreasing, from 9.5% in 2014 to 8.7% in 2018, and then to 5–6% in 2021 [11]. Even the most recyclable types of plastics, such as bottles and jugs made of PET #1 and HDPE #2, have a recycling rate of only 19% [12]. These low recycling rates fall well below the threshold of 30% needed to meet the definition of recyclable by the Ellen MacArthur Foundation's New Plastics Economy Initiative [13], and they are also far from the national recycling goal of 50% announced by the U.S. Environmental Protection Agency [14]. To address this issue, the construction industry has an opportunity to play a role in increasing the recycling of plastics, as there is a high demand for alternative construction materials [15]. Although HDPE can be used as a coating material on the steel rebar to mitigate the process of corrosion [16], the demand is still limited.

Therefore, this study endeavors to simplify the pretreatment process and explore higher volumes of coal in the polymeric composite by innovatively harnessing the potential of larger-sized coal and recycled plastics. In this research, coal-based thermoplastic composites (CTC) with 50–80% large-sized coal particles and recycled HDPE were investigated as sustainable construction materials. Thorough investigations were conducted to scrutinize the mechanical properties, thermal properties, microstructural characteristics, and environmental impact of the proposed CTC. By synergistically engaging in the reutilization of discarded plastics and employing coal in an ecologically responsible manner, this research serves as a testament to the immense potential for fabricating construction materials that not only foster enhanced carbon fixation within the final product but also exhibit a marked reduction in carbon dioxide emissions.

2. Materials and Methods

2.1. Raw Material

The proposed coal-based thermoplastic composite (CTC) is composed of two kinds of material: coal aggregate and plastic binder. This study used bituminous coal, the most abundant rank of coal found in the United States. The coal aggregate, which had a particle size smaller than 4.75 mm, was supplied by Arch Resources Inc. of St. Louis, MO, USA. Recycled HDPE granules were used as the binder in the CTC and presented alongside the bituminous coal sample in Figure 1.

2.2. Composite Manufacturing Process and Principle

The proposed CTC formulae are designed to include coal weight percentages ranging from 50 to 80%, as shown in Table 1. A coal content larger than 80% would make the mixture too harsh to manufacture due to a lack of binder, while a coal content lower than 50% was not considered because we aimed to maintain percolation of the filler phases to utilize the superior thermal properties of coal fillers.



Figure 1. Sample of (a) bituminous coal and (b) recycled HDPE granules.

Table 1. Typical composite formulae and carbon contents.

Property		Coal	HDPE	Carbon Weight (%)
Density (kg/L)		1.3–1.5	0.97	–
Carbon content (%)		65–92	86	–
Weight percentages (%)	Case 1	50	50	75.5–89.0
	Case 2	60	40	73.4–89.6
	Case 3	70	30	71.3–90.2
	Case 4	80	20	69.2–90.8

Manufacturing sustainable building material is essential for ecological safety [17]. In this light, the CTC is produced using a hot-press manufacturing process with a relatively low temperature and pressure. Figure 2 shows that coal particles (<4.75 mm or passing ASTM #4 sieve) and HDPE were uniformly dry-mixed to the desired component ratio and then heated to 170 °C for 20 min. To prevent a rapid reduction in the mixture's temperature during the compression procedure, the steel mold used for shaping the CTC was also placed in the oven. Next, the mixture was moved into the mold with a dimension of 50 × 50 × 100 mm and compressed through a hydraulic jack at 10 MPa for 5 min. This compressive pressure allows for the penetration of the partially melted binder phase into the inter-particle voids and surface pores of the coal particles. Finally, the resulting CTC was demolded and cut into a specific dimension for the thermal tests.

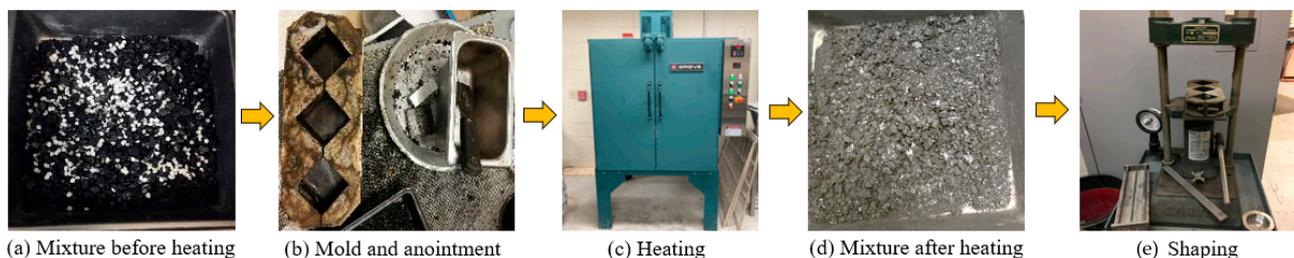


Figure 2. CTC fabrication process.

The proposed fabrication technology offers a sustainability advantage in that the CTC is easily recyclable. These durable composites can be reused after disassembly from existing buildings, and if the original shape is no longer desirable, they can be easily heated and pressed into a new configuration. After multiple cycles, when the binder performance degrades severely, the material can be repurposed for waste-to-energy streams, such as incineration, pyrolysis/gasification, or other future methods of generating electricity and/or heat energy. Such waste-to-energy streams will align with policy and industry trends and add more value to coal and its derivatives.

Simultaneous heating and compacting of coal and HDPE in a rigid mold produce high-performance composites through three mechanisms, as schematically shown in Figure 3. Firstly, the mesostructure features beneficial entangled structures as various filler phases rotate and slide against each other. Secondly, driven by the pressure and large capillary forces in narrow micro-gaps between filler particles, the partially melted binder phase is squeezed in and self-assembled into micro-agglomerations at the most critical load-carrying microstructural sites, before filling defects. Lastly, following cooling, the filler particles are permanently compressed and strongly bonded together. These theories have been previously supported by relevant publications [18–20]. A sample of CTC composite with 70% bituminous coal and 30% HDPE binder by weight fraction is shown in Figure 4.

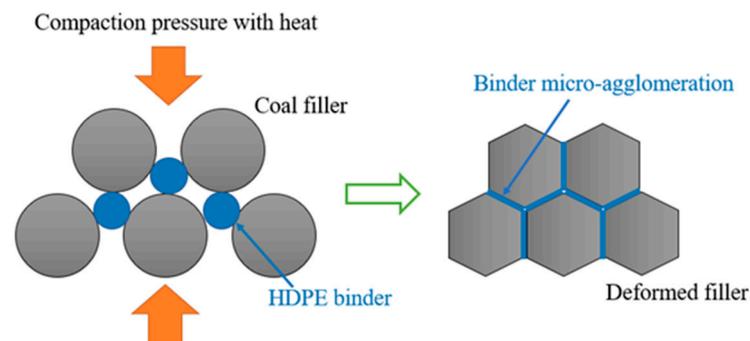


Figure 3. Principle of hot-press.

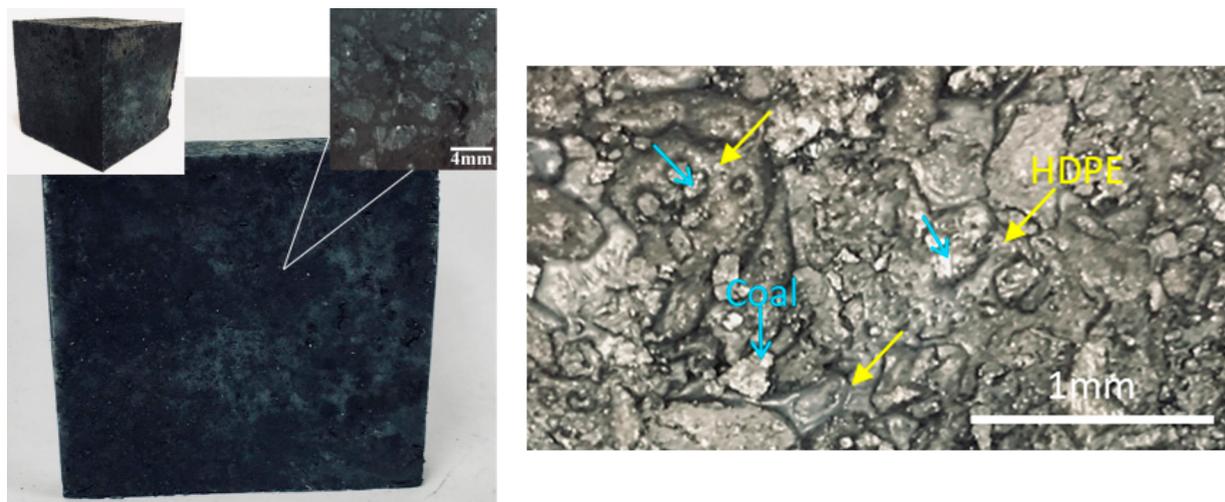


Figure 4. Cube sample and panel sample of the CTC with 70% weight fraction of coal.

2.3. Experimental Methodologies

To comprehensively investigate the mechanical and thermal properties of the CTC, as well as its micro characteristics, various methodologies are employed. A flow diagram of the overall experimental or analytical methodologies is presented in Figure 5.

2.3.1. Testing Methods of Mechanical Properties

The mechanical properties of the CTC, including compressive strength and elastic modulus, were evaluated using the equipment shown in Figure 5. The compressive strength test was evaluated by applying monotonic loading to four types of cube specimens with a side length of 50 mm using a 200,000 lbs servo-controlled Tinius-Olsen universal compression/tension machine with a loading rate of 0.9 kN/s (200 lbs/s), according to the ASTM C109/C109M-20 standard [21]. Four specimens of each CTC type were tested, resulting in a total of sixteen cube specimens.

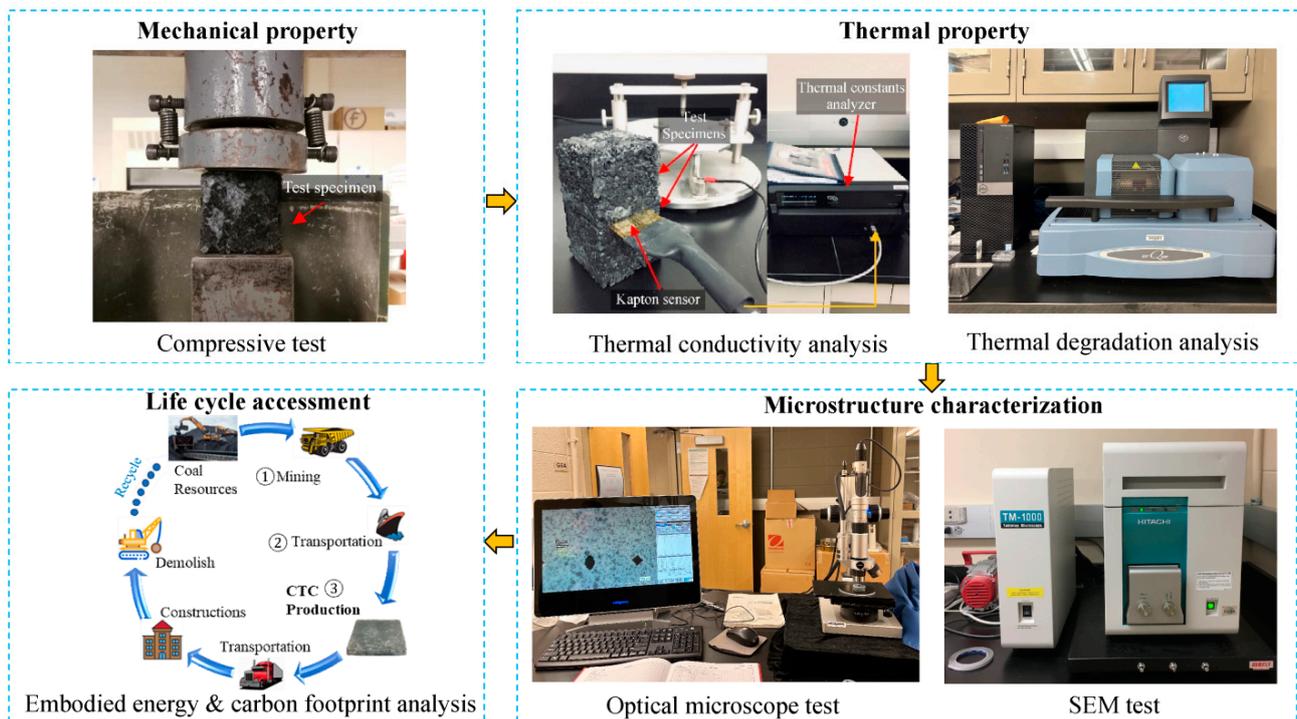


Figure 5. A flow diagram of the overall methodologies.

2.3.2. Testing Methods of Thermal Properties

As CTC has potential as a construction material, it is essential to investigate its thermal properties, including thermal conductivity and specific heat capability, which determine the rate of heat dissipation within the bulk material. The Hot Disk Thermal Constant Analyzer TPS 500 S (Hot Disk Ltd., Göteborg, Sweden) was used to measure thermal conductivity and specific heat capability using the Transient Plane Source (TPS) method [22], in accordance with ISO 22007-2. Two 50 mm cube specimens were used in each experiment, with a thin hot disk sensor sandwiched between them. During the transient recording, the sensor was electrically heated, and the resistance, i.e., the sensor's temperature variation, was monitored. The thermal constant analyzer provides absolute values of thermal conductivity and specific heat from a single transient recording. Each experiment was repeated at least 10 times to calculate the mean value of the experimental data, with an accuracy better than 5% and a reproducibility of approximately 2%.

For the TGA-DSC simultaneous thermal analysis, the TA Instruments SDT 600 was employed. The analysis was initiated by subjecting the sample to a controlled temperature ramp, spanning from room temperature to 800 °C, utilizing a heating rate of 20 °C per minute. This analysis was conducted under an air atmosphere, maintaining a consistent flow of 100 mL/min. The aim of this test was to reveal valuable insights into the thermal degradation and combustion behaviors of the sample.

2.3.3. Microstructural Characterization Techniques

To investigate the relationship between the mechanical properties of CTC and its microstructure, a digital optical microscope and scanning electron microscope (SEM) were utilized to examine the particle morphology, pore distribution, fracture front, and interfacial bondage of the material. The natural cross sections of CTC were observed using an optical microscope (Hirox RX-100, Hirox-USA Inc., Hackensack, NJ, USA) and an SEM (Hitachi TM-1000, Hitachi Ltd., Tokyo, Japan).

2.3.4. Life Cycle Assessment of the CTC

The evaluation of environmental impact and cost-effectiveness of the CTC is crucial in assessing its potential benefits as an innovative material. To achieve this, it is indispensable to quantify the resources, emissions, and environmental impacts, as well as the cost efficiency throughout the life cycle of the CTC. Therefore, the ecological and economic evaluations of the CTC's production are essential. The parameters, including the embodied energy (EE, the energy it takes to create them, MJ/kg) and carbon footprint (CF, the CO₂ or CO₂-equivalent that creating them releases, kg CO₂/kg material), were used to proceed with the ecological evaluation of CTC, whilst the cost analysis was leveraged in the economic efficiency evaluation of the CTC. This study is a cradle-to-gate life cycle assessment (LCA), which covers the three stages of raw material extraction, transportation, and manufacturing for a product or system.

3. Results and Discussion

3.1. Mechanical Properties

The effect of coal filler content on the mean compressive stress–strain curves is depicted in Figure 6a. The results indicate that compressive strength, elasticity modulus, and ductility increased as coal filler decreased. This is due to a weaker binding in the system caused by the decrease in plastic binder content, as well as the vulnerability of the coal filler as a result of its porous microstructure discussed in Section 3.4. Additionally, note that there is an increasing or almost constant stress level at the end of the loading stage. This is attributed to the unignorable/significant increments of the cross-section areas of the specimens along with compression deformations during their failure processes. Figure 6b presents the details of the variation of compressive strength and elastic modulus with the weight fraction of coal. The compressive strength and elastic modulus exhibited a decrease as the coal fraction increased, in contrast to the trend observed within the coal fraction range of 30% to 40%, as reported by [23]. This divergence could potentially be attributed to variations in filler properties and fraction, which modified the bonding strength and failure mode. Note that the compressive strength was higher than 40 MPa when the coal filler was no larger than 70%. This value is higher than that of commonly used normal-strength concrete, indicating the potential application of the CTC in civil engineering as an innovative construction material. Figure 7 illustrates the different destruction states of CTC after subjecting the specimens to axial compression. It was observed that the damage to CTC with 80% wt coal was particularly severe, leading to slip damage caused by the higher dosage of coal and resulting in a lower bonding strength between coal and HDPE.

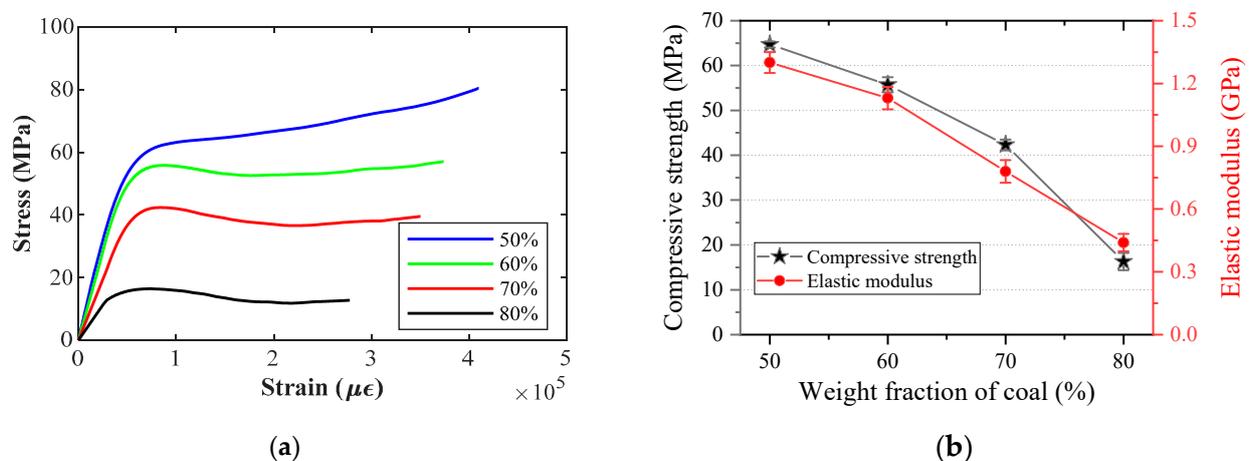


Figure 6. Compressive strength test: (a) stress–strain curves; (b) strength and modulus vs. weight fraction of coal.

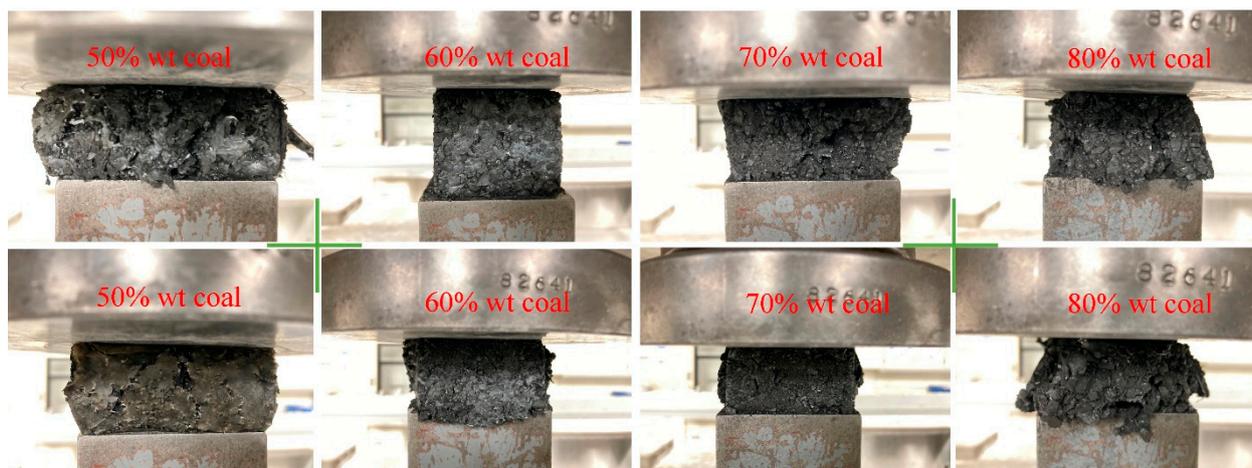


Figure 7. Typical compressive failure patterns of specimens.

Based on above experimental results, the mechanical properties of CTC are summarized in Table 2.

Table 2. Summary of the mechanical properties of CTC.

Coal Fraction	Compressive Strength (MPa)		Elastic Modulus (GPa)	
	Mean	SD	Mean	SD
50%	64.7	0.71	1.30	0.051
60%	55.7	1.70	1.13	0.055
70%	42.3	1.20	0.78	0.054
80%	16.3	1.92	0.44	0.040

3.2. Thermal Properties

In addition to the mechanical properties, the coal filler content also has an impact on its thermal properties, which are important for the thermal performance of construction materials. Figure 8 illustrates the effects of coal filler content on the thermal conductivity and specific heat capability of the CTC. It was observed that the thermal conductivity of CTC increased with rising coal filler content, reaching $0.41 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$ for 80% coal content, which is an 11% increment compared to 50% coal content ($0.37 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$). Despite this increase, the overall thermal conductivity of CTC ($0.37\text{--}0.41 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) is still much lower than that of concrete ($0.8 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) or common brick ($0.6 \text{ W}\cdot\text{m}^{-1}\cdot\text{K}^{-1}$) [24], indicating that the CTC is a more efficient thermal insulation material. Furthermore, the specific heat capacity of CTC was found to increase with an increase in coal content, ranging from 1.20 to $1.45 \text{ J}\cdot\text{g}^{-1}\cdot\text{K}^{-1}$ for coal content of 50–80%. This capacity is 1.5 times larger than that of normal concrete ($0.75\text{--}1 \text{ J}\cdot\text{g}^{-1}\cdot\text{K}^{-1}$) [25,26], making CTC a better material for sensible heat storage preferred for buildings with high thermal mass. These findings demonstrate the potential of CTC as an effective and energy-efficient construction material, particularly for use in insulation and heat storage applications. The summary of the thermal properties test results is presented in Table 3. However, when HDPE was infused with graphite fillers up to 40% and particles as large as 50 mm, an opposing trend emerged [27], indicating the substantial impact of filler fraction and particle size on the thermal properties of the composites.

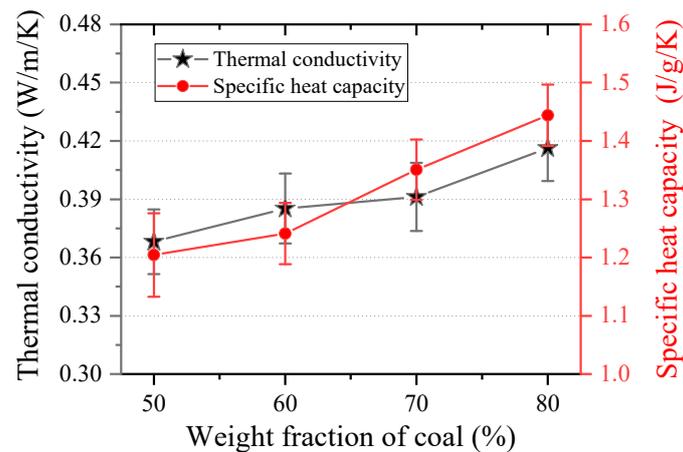


Figure 8. Variations of thermal conductivity and specific heat capacity with respect to the weight fraction of coal.

Table 3. Summary of thermal properties of CTC.

Coal Fraction	Thermal Conductivity (W/m/K)		Specific Heat Capacity (J/g/K)	
	Mean	SD	Mean	SD
50%	0.37	0.017	1204.3	71.6
60%	0.39	0.018	1241.2	52.5
70%	0.39	0.018	1350.7	51.9
80%	0.42	0.017	1443.9	52.8

The TGA-DSC simultaneous thermal analysis of the CTC was carried out for an analysis of its thermal degradation and combustion processes. As shown in Figure 9, the CTC is quite stable before 250 °C, evidenced by the absence of weight loss. This stability coincides with the occurrence of an endothermic peak between 120 °C and 200 °C, attributed to the phase change of HDPE within the composite. Beyond this point, the CTC initiates an exothermic combustion and oxidation process, resulting in weight loss and the release of heat. This transition is accompanied by a discernible decline in weight, aligning with the exothermic reactions occurring as the temperature exceeds 250 °C.

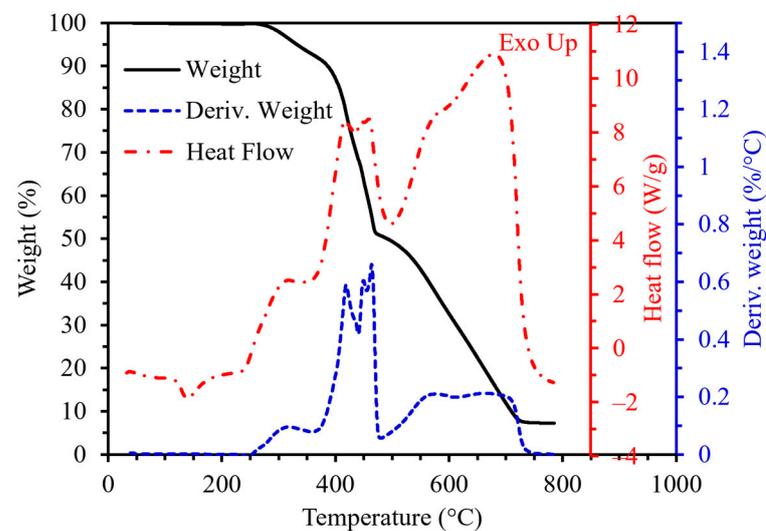


Figure 9. TGA-DSC simultaneous thermal analysis of the CTC.

3.3. Microstructural Analysis

The microstructure of composite materials plays a crucial role in determining their mechanical and thermal properties. To gain insights into the behavior of CTC, the mesostructure of CTC with varying coal weight fractions was examined using microscopes. Figure 10 shows the SEM image of the interface between coal and HDPE, which reveals a strong bond between them, demonstrated by the absence of gaps. The microstructure of coal exhibited more fractures and pores, which accounted for its compressive strength compared to HDPE. Consequently, the weaker coal is more likely to be the defect of the CTC under tension or compression, which explains the failure modes revealed in the compressive tests in Section 3.1.

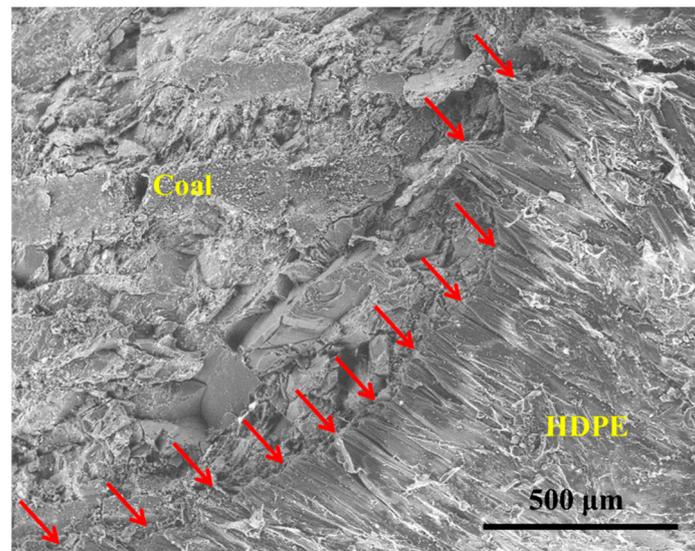


Figure 10. Microstructure of the CTC and the interfacial zone between coal and HDPE (marked with red arrow).

The effectiveness of achieving a good bond can be attributed primarily to the hot-pressing technique, which introduces enhanced adherence between coal and HDPE—an essential factor influencing the overall strength of the composite. This is evident through the SEM observations, where the interface width is notably small, corresponding to commendable mechanical strength. Additionally, the incorporation of small coal particles (powder fraction) into the plastic matrix may activate stiffening, reinforcement, and toughening mechanisms within the HDPE, as previously indicated by [28,29]. Regarding the chemical interaction, coal molecules often contain numerous hydrogen bonds, particularly in lower-ranked or weathered coal, which can be disrupted by pretreatment (e.g., acid treatment) and subsequently bond with polymers to bolster bonding strength—a phenomenon corroborated by the Fourier Transform Infrared (FTIR) spectroscopy and X-ray Photoelectron Spectroscopy (XPS) test results, as proposed by [6]. It is acknowledged that high pressure and temperature lead to the possibility of chemical introduction, which necessitates meticulous examination of the interaction zone and has not yet been explored within this study.

Figure 11 displays optical microscope images of CTC with varying coal contents. Upon closer examination of areas with the same dimension for CTC samples with different coal fractions, it becomes evident that as the coal content increases, more coal particles appear in the images, and the volume of HDPE binding material decreases. Consequently, more coal particles are loosely contacting each other, and more pores are observed, leading to lower compressive strengths in CTC samples with higher coal fractions. This finding supports the results presented in Section 3.1. However, the effect of higher porosity in the CTC has a dual nature. While the higher porosity of CTC with larger coal content is associated with poorer mechanical performance, it enables the CTC to effectively absorb (from solar

radiation) and emit (to heat air) heat, more so than concrete and masonry. In conjunction with the larger heat capacity presented in Section 3.3, CTC with a higher coal content is advantageous for energy-efficient building elements, like Trombe walls [30].

Note that the distribution of the coal is not perfectly uniform as schematically presented in Figure 3. In pursuit of the material structure depicted in Figure 3, two methods can be adopted: firstly, elevating the applied pressure and temperature on the composite to obtain thinner and more evenly distributed binding; secondly, reducing the particle size of coal to improve the mixing efficacy. Implementing these methods has the potential to improve composite uniformity, leading to enhanced CTC performance.

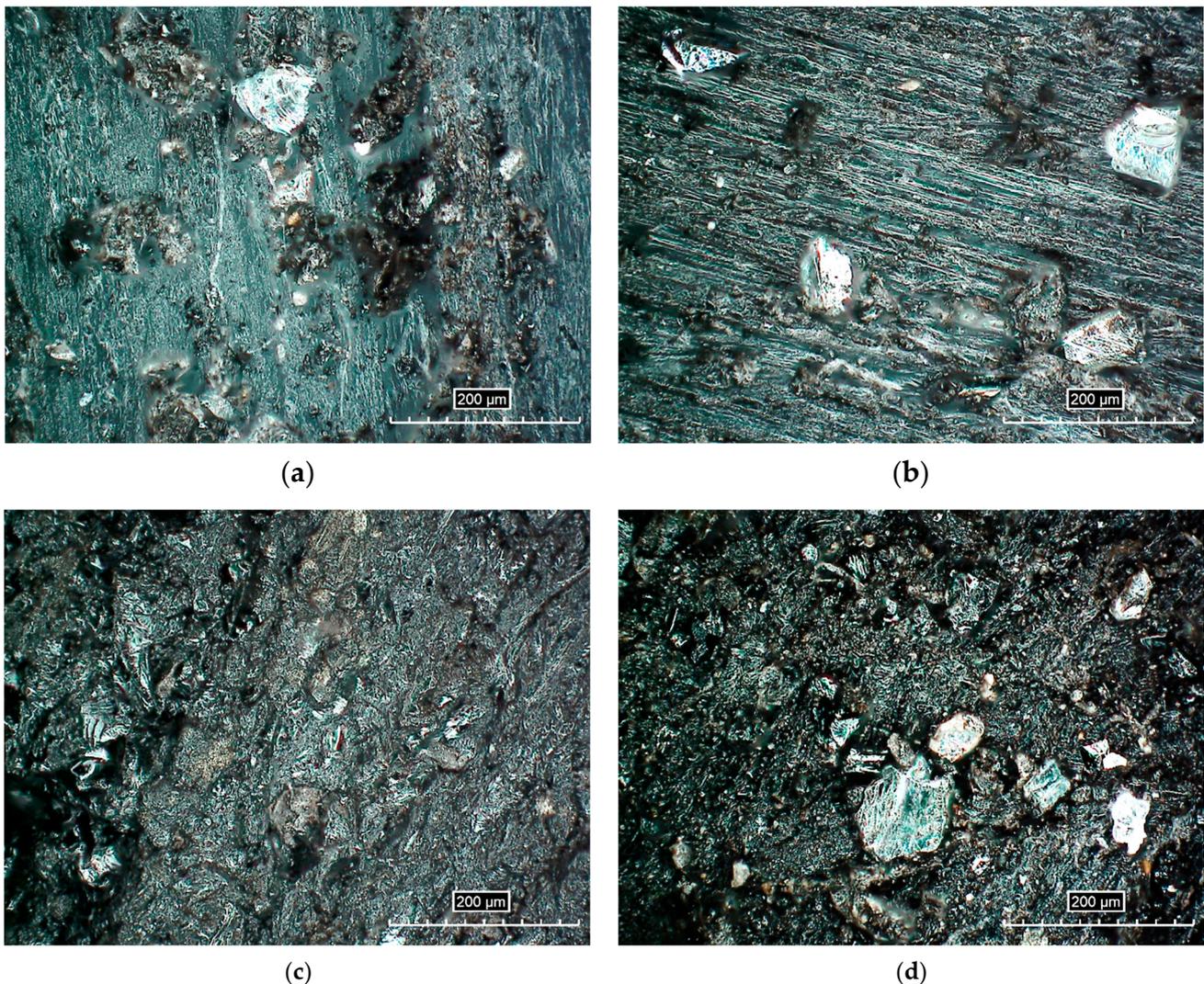


Figure 11. Optical microscope images of the composite with different weight fractions of coal (bright/brown particles in the images): (a) 50%; (b) 60%; (c) 70%; and (d) 80%.

3.4. Life Cycle Assessment

In addition to the mechanical and thermal properties, the CTC offers potential benefits in terms of environmental impact and cost-effectiveness. Figure 12 provides conceptual drawings of the LCA systems for coal in two scenarios: one for conventional use in power generation, and the other for innovative use in producing CTC. As shown in the figure, the application of CTC diverts the life cycle of coal from the conventional loop (a) to the alternative loop (b). Coal is derived from plant life and contains carbon that was once in the atmosphere and not captured, resulting in a positive carbon footprint. In contrast, the use

of coal in CTC does not involve burning and releasing the sequestered carbon, potentially allowing it to have a negative carbon footprint and receive carbon credits.

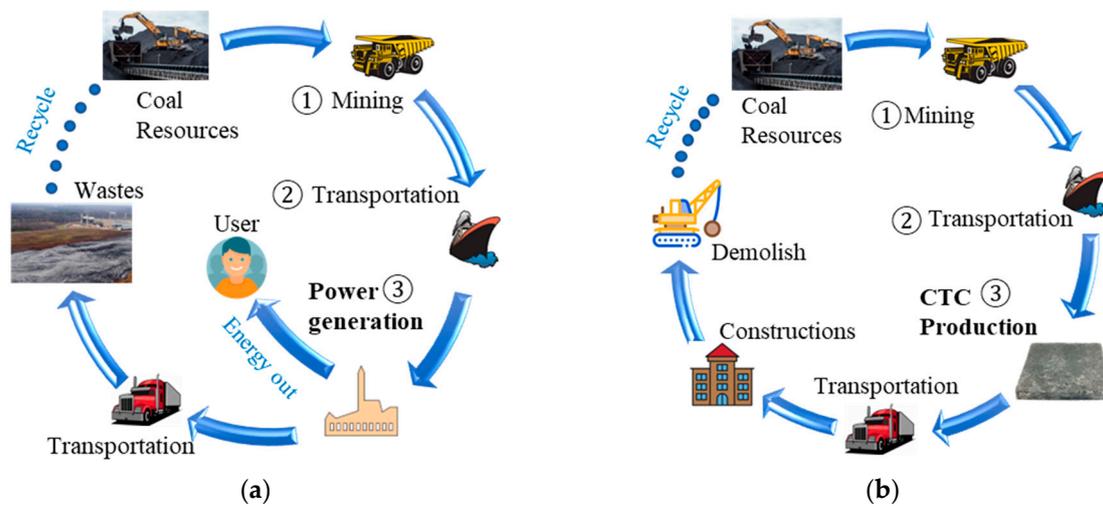


Figure 12. Conceptual drawings of the coal LCA systems in two different scenarios: (a) coal used in the conventional way of power generation; (b) coal used in the production of CTC (system boundaries include the processes from 1 to 3).

Figure 13 illustrates the plastic LCA systems for HDPE and recycled HDPE (rHDPE) in the production of CTC. The physical binding mechanism of the plastic binder in CTC allows for high tolerance in the chemical composition of the recycled plastic, making it possible to recycle more waste plastics. Therefore, it is preferable to obtain the plastic binder component from the material recovery facilities for CTC production. The rHDPE only needs to be shredded or melted into pellets, requiring less energy and emitting less CO₂ than the production of HDPE. Another significant difference between the LCA systems for rHDPE and HDPE is that the production of rHDPE involves feeding sorted waste HDPE instead of oil derivatives, making it more eco-friendly. Additionally, recycling HDPE reduces the amount of plastic waste sent to landfills, thereby reducing greenhouse gas emissions. The thermoplastic property of CTC provides another end-of-life option of being reengineered for other constructions via hot pressing.

The cradle-to-gate life cycle analysis (LCA) of the CTC system involves evaluating the environmental impact (EE) and carbon footprint (CF) of each component, namely coal and plastic. This analysis is carried out by analyzing the system boundary and life cycle inventory at each stage. The EcoInvent database [31] and the Association of Plastic Recyclers report [2] were used for EE and CF evaluations of coal and plastic, respectively. The cost data were obtained from the EcoInvent database [31] and the Recycling Market report [32]. Table 4 presents the LCA results of the CTC components in various scenarios [2,31,33], including EE, CF, and cost analyses of coal and plastic. The table shows that using coal for CTC production instead of burning it reduces energy consumption by 92.5%, CO₂ by 97.7%, and cost by 36%. Recycling plastic also has significant benefits; the EE of rHDPE is 8.7 MJ/kg, representing a dramatic 88.4% reduction in energy consumption compared to virgin HDPE. Additionally, rHDPE production has a CF of 0.56 kg CO₂/kg, which is 70.4% less than virgin HDPE's CF of 1.89 kg CO₂/kg. Moreover, the rHDPE is 18.3% less expensive than virgin HDPE. These findings demonstrate the potential of the two-component CTC system to achieve both ecological and economic efficiency, making it a promising solution for sustainable production and recycling practices.

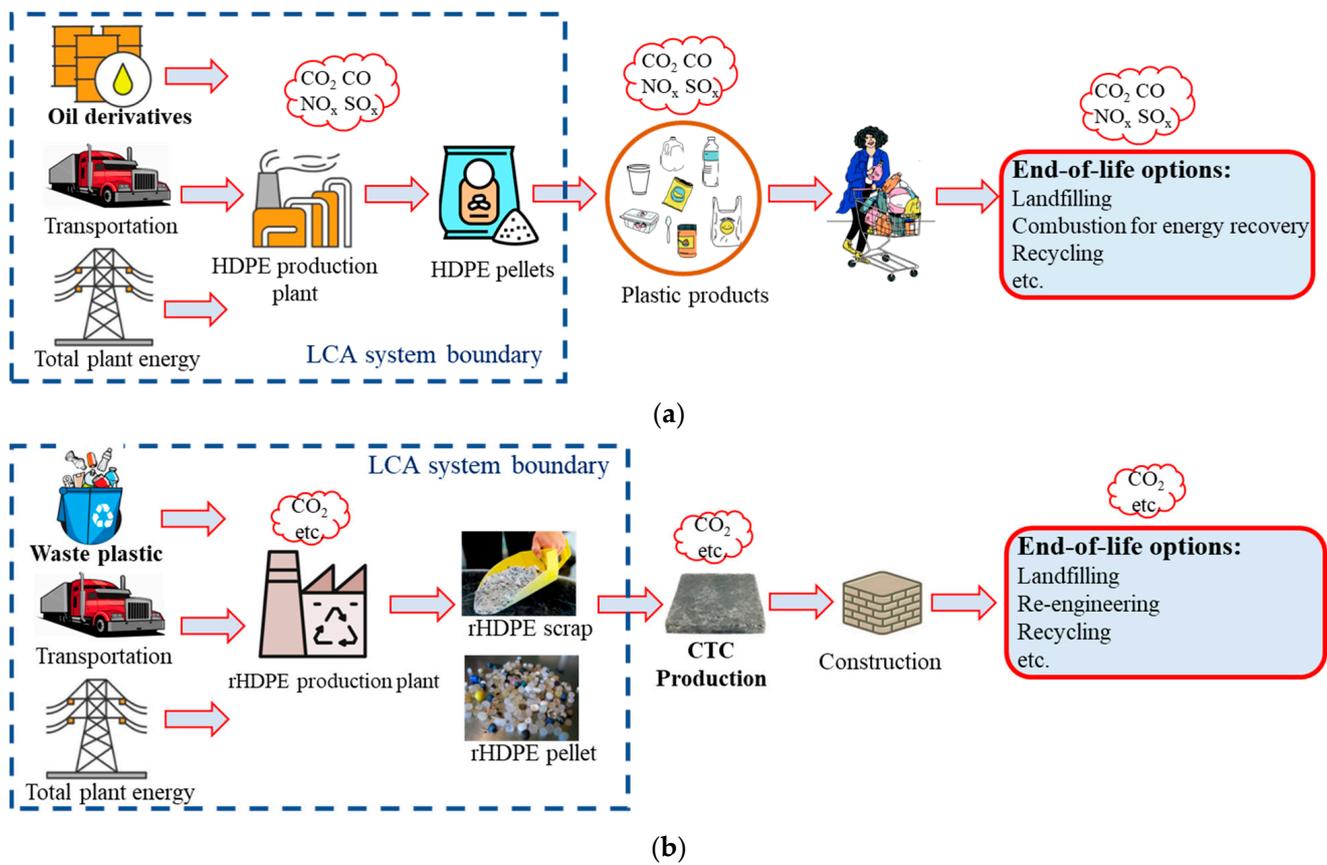


Figure 13. Conceptual drawings of plastic LCA systems for HDPE and recycled HPDE (rHDPE): (a) HDPE LCA system; (b) rHDPE LCA system (system boundaries are marked with dashed boxes).

Table 4. The EE, CF, and cost analysis results of coal and plastic.

Index	Coal		HDPE	Recycled HDPE (rHDPE)
	Conventional Use as Fuel for Generating Electricity	Creative Use in the CTC Production		
Embodied energy (EE, MJ/kg)	11.35	0.85	75.30	8.70
CO ₂ footprint (CF, kg CO ₂ /kg)	1.04	0.02	1.89	0.56
Cost (USD/kg)	0.03	0.02	1.20	0.98

Figure 14 depicts the LCA system for the CTC, which involves raw material input, transportation, and manufacture. The EE, CF, and cost of raw materials (i.e., coal and plastic) can be found in Table 4. The transportation and manufacturing stages have estimated values of 2.2 MJ/kg, 0.2 kg CO₂/kg, and 0.03 USD/kg, respectively [31,34]. By summing up these inputs, the EE, CF, and cost of CTC with various mix proportions (i.e., coal fractions of 50, 60, 70, and 80%) can be calculated, and the results are shown in bar charts in Figure 15.

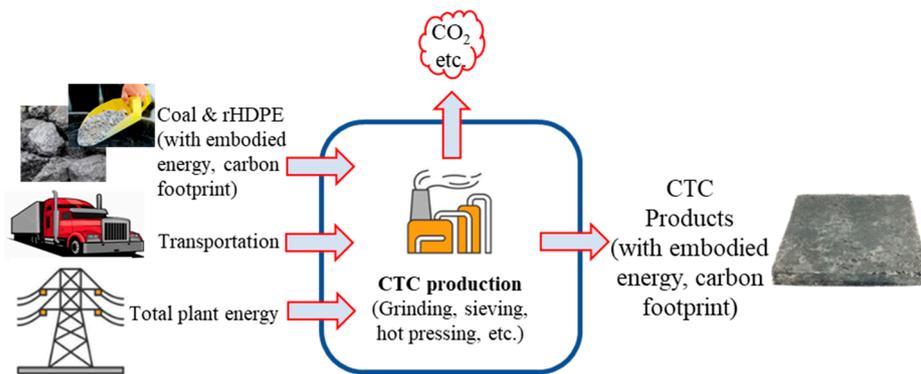


Figure 14. Conceptual drawings of the cradle-to-gate LCA system for the CTC.

As shown in Figure 15, the EE, CF, and cost for both systems decrease with the increase in coal fraction. This is due to the lower EE, CF, and cost of coal compared to plastic. When the coal fraction rises from 50% to 80%, the EE reduces from 43.33 to 24.14 MJ/kg (a 44% reduction) for conventional use and from 6.98 to 4.62 MJ/kg (a 34% reduction) for CTC production; the CF declines from 1.47 to 1.21 kg CO₂/kg (a 17% reduction) for conventional use and from 0.49 to 0.33 kg CO₂/kg (a 33% reduction) for CTC production; and the cost drops from 0.62 USD/kg to 0.27 USD/kg (a 57% reduction) for conventional use and from 0.53 USD/kg to 0.24 USD/kg (a 54% reduction) for CTC production. It is noticeable that there are remarkable drops in EE and CF by producing CTC instead of using the conventional method, as revealed in Table 5.

Table 5. Percentage reductions in the EE, CF, and cost through the application of CTC based on the LCA study.

Coal Fraction	Strength (MPa)	Embodied Energy (MJ/kg)	CO ₂ Footprint (kg CO _{2e} /kg)	Cost (USD/kg)
50%	64.7	83.9%	66.4%	13.9%
60%	55.7	83.2%	68.3%	13.0%
70%	42.3	82.3%	70.3%	11.5%
80%	16.3	80.9%	72.7%	8.7%

In Table 5, the percentages of improvements in the EE, CF, and cost from conventional use to CTC production are presented. It is impressive to see significant improvements in the EE, CF, and cost efficiency from the CTC production, as shown in the table. When coal and plastic are used to produce the CTC, EE plummets by 80–84%, while CF is reduced by 66–73%. The cost efficiency has a 9–14% improvement, which is positive but relatively insignificant compared to the improvement of ecological efficiency. This is due to the overestimation of low-rank coal cost by averaging prices of all types of coal and the soaring price of rHDPE during the pandemic period. Using low-rank coal, such as sub-bituminous coal or lignite coal, can decrease the cost of coal. Additionally, the price of rHDPE is expected to decline in the post-pandemic period [35], and other types of cheaper recycled plastics, such as PE or PET, can also be used as binders in the CTC. By optimizing the specific application case, a more significant improvement in cost efficiency can be achieved. The LCA study demonstrates that the CTC is much more environmentally friendly and economically efficient than conventional uses of raw materials. Therefore, the extensive application of CTC has the potential to become a profit-driven incentive for businesses and have a significant impact on promoting ecological sustainability and the circular economy.

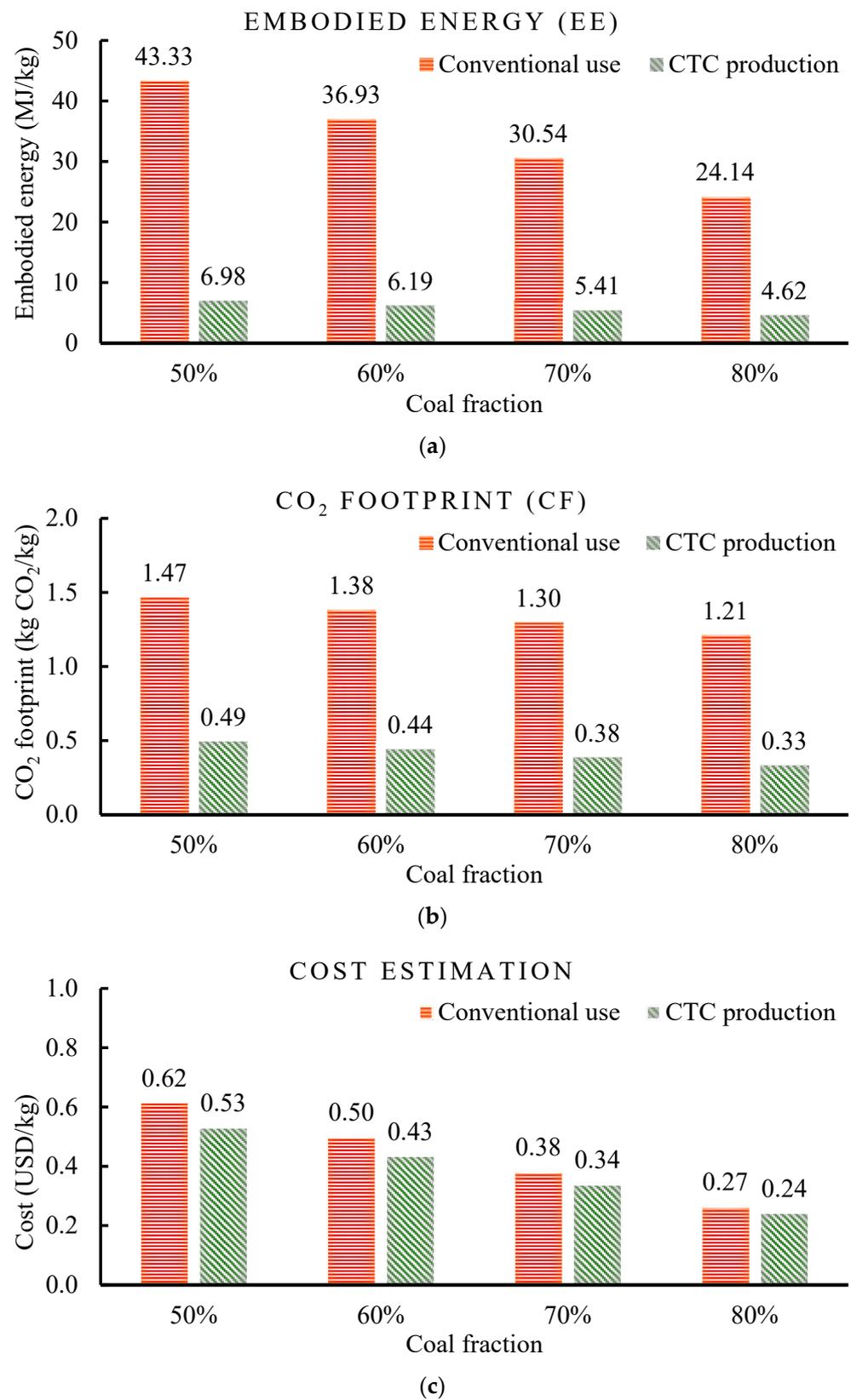


Figure 15. LCA results of the CTC production in comparison to the conventional use of CTC's all components: (a) embodied energy, EE; (b) CO₂ footprint, CF; and (c) cost analysis.

4. Conclusions

To facilitate the practical application of coal-based thermo-plastic composite (CTC) material made from low-grade coal and recycled high-density polyethylene (HDPE) as a building material, this study investigated the effect of large coal fractions (50–80 wt%) and coal particle sizes (up to 4.75 mm) on the mechanical and thermal properties of the CTC. A hot-press fabrication method was developed utilizing a heated temperature of 170 °C and a pressure of 100 MPa for a duration of 5 min. The effects of the coal fraction on the mechanical and thermal properties, microstructure, and ecological and economic efficiencies of the CTC were investigated. The following conclusions can be drawn:

- The compressive strength and related moduli decrease with an increase in the coal fraction. The compressive strength varied between 17 and 65 MPa. The good ductility of CTC results in relatively low compressive moduli compared to normal concrete, which is 1.35 GPa.
- The CTC is quite stable below 250 °C, evidenced by the absence of weight loss. The thermal conductivity and specific heat capability increase as the coal content increases. Compared to concrete, CTC has approximately half the thermal conductivity and twice the specific heat capacity. These properties make CTC a superior material to concrete as an energy-efficient construction material.
- Fractures and pores are found in the microstructure of coal, indicating that coal is more vulnerable than HDPE. CTC with higher coal content has lower compressive strengths due to a lower binder volume, more loosely contacting coal particles, and more pores. However, the higher porosity of CTC allows for more effective absorption and emission of heat than concrete and masonry.
- Based on the LCA study, the use of coal and plastic to produce CTC results in significant improvements in ecological and economic efficiency. The embodied energy drops by 80–84%, the carbon footprint is reduced by 66–73%, and cost efficiency is improved by 9–14%.
- These findings demonstrate the promising potential of CTC technology in promoting sustainable ecology and economy as a building material.

Author Contributions: H.Z.: experimental design, writing—original draft, writing—review and editing. W.L.: experimental design, supervision, writing—original draft, writing—review and editing. G.C.: supervision, writing—review and editing. H.M.: supervision, writing—review and editing. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by [U.S. Department of Transportation, Office of the Assistant Secretary for Research and Technology (USDOT/OST-R)] under grant number [69A3551747126].

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: All the test data was presented in the article.

Acknowledgments: Financial support for this study is provided by USDOT/OST-R through INSPIRE University Transportation Center (<http://inspire-utc.mst.edu>, accessed on 1 January 2023) at Missouri University of Science and Technology. The views, opinions, findings, and conclusions reflected in this publication are solely those of the authors and do not represent the official policy or position of the USDOT/OST-R, or any State or other entity. The authors would like to thank Yanping Zhu for his input during mechanical tests.

Conflicts of Interest: The authors declare that there is no conflict of interest with respect to the research, authorship, and/or publication of this article.

References

1. Energy Information Administration (EIA). Annual Energy Review. 2014. Available online: <http://www.eia.gov/forecasts/archive/aeo14/> (accessed on 12 January 2023).
2. The Association of Plastic Recyclers. *Virgin vs. Recycled Plastic Life Cycle Assessment Energy Profile and Life Cycle Assessment Environmental Burdens*; The Association of Plastic Recyclers: Washington, DC, USA, 2020.
3. Nemtsov, D.; Sawyer, K.; Mumme, S.; James, N. *Novel Materials in Thermal Energy Storage for Buildings*; Thermal Energy Storage Webinar Series; US Department of Energy: Washington, DC, USA, 2020.
4. Zhang, T.; Yuchi, W.; Bai, Z.; Hou, R.; Feng, Z.; Guo, Z.; Kong, L.; Bai, J.; Meyer, B.; Li, W. Insight into the charging methods effects during clean recycling of plastic by co-pyrolysis with low-rank coal. *J. Clean. Prod.* **2022**, *333*, 130168. [[CrossRef](#)]
5. Davim, J.P. *Green Composites: Materials, Manufacturing and Engineering*; De Gruyter: Berlin, Germany; Boston, MA, USA, 2017.
6. Hu, G.; Bian, Z.; Xue, R.; Huang, W.; Komarneni, S. Polymer-coal composite as a novel plastic material. *Mater. Lett.* **2017**, *197*, 31–34. [[CrossRef](#)]
7. Wang, G.H.; Zhou, A.N.; Hu, X.B. Effect of coal filler on the properties of soy protein plastics. *J. Appl. Polym. Sci.* **2006**, *102*, 3134–3143.
8. Zhu, X.; Pan, Q.; Xu, H.; Lu, J. Effects of coal and ammonium polyphosphate on thermal degradation and flame retardancy of polyethylene terephthalate. *J. Polym. Res.* **2010**, *17*, 621–629. [[CrossRef](#)]
9. Al-majali, Y.A.; Chirume, C.T.; Marcum, E.P.; Daramola, D.A.; Kappagantula, K.S.; Tremblay, J.P. Coal-filler-based thermoplastic composites as construction materials: A new sustainable end-use application. *ACS Sustain. Chem. Eng.* **2019**, *7*, 16870–16878. [[CrossRef](#)]
10. Phillips, L.N.; Kappagantula, K.S.; Tremblay, J.P. Mechanical performance of thermoplastic composites using bituminous coal as filler: Study of a potentially sustainable end-use application for Appalachian coal. *Polym. Compos.* **2019**, *40*, 591–599. [[CrossRef](#)]
11. Bennington College. *The Real Truth About the U.S. Plastics Recycling Rate*; Bennington College: Bennington, VT, USA, 2022.
12. The Association of Plastic Recyclers. *Recommit, Reimagine, Rework Recycling*; The Association of Plastic Recyclers: Washington, DC, USA, 2022.
13. Ellen Macarthur Foundation. *Global Commitment 2021 Signatory Report*; Ellen Macarthur Foundation: Isle of Wight, UK, 2021.
14. Fact Sheet about the National Recycling Goal: 50 Percent by 2030. 2023. Available online: <https://www.epa.gov/recyclingstrategy/fact-sheet-about-national-recycling-goal-50-percent-2030> (accessed on 12 January 2023).
15. Kulkarni, P.; Ravekar, V.; Rao, P.R.; Waigokar, S.; Hingankar, S. Recycling of waste HDPE and PP plastic in preparation of plastic brick and its mechanical properties. *Clean. Mater.* **2022**, *5*, 100113. [[CrossRef](#)]
16. Kumar, K.; Babu, B.S.; Davim, J.P. *Coatings: Materials, Processes, Characterization and Optimization*; Springer Nature: Berlin/Heidelberg, Germany, 2021.
17. Davim, J.P. *Sustainable Manufacturing*; Wiley: Hoboken, NJ, USA, 2010.
18. Dhakate, S.R.; Sharma, S.; Borah, M.; Mathu, R.R.B.; Dhami, T.L. Expanded graphite-based electrically conductive composites as bipolar plate for PEM fuel cell. *Int. J. Hydrogen Energy* **2008**, *33*, 7146–7152. [[CrossRef](#)]
19. Hao, W.; Ma, H.; Lu, Z.; Sun, G.; Li, Z. Design of magnesium phosphate cement based composite for high performance bipolar plate of fuel cells. *RSC Adv.* **2016**, *6*, 56711–56720. [[CrossRef](#)]
20. Luo, Z.; Ma, Y.; He, H.; Mu, W.; Zhou, X.; Liao, W.; Ma, H. Preparation and characterization of ferrous oxalate cement—A novel acid-base cement. *J. Am. Ceram. Soc.* **2021**, *104*, 1120–1131. [[CrossRef](#)]
21. *ASTM C109/C109M-20*; Standard Test Method for Compressive Strength of Hydraulic Cement Mortars (Using 2-in. or 50 mm cube Specimens). ASTM International: West Conshohocken, PA, USA, 2020.
22. Gustafsson, S.E. Transient plane source techniques for thermal conductivity and thermal diffusivity measurements of solid materials. *Rev. Sci. Instrum.* **1991**, *62*, 797–804. [[CrossRef](#)]
23. Das, P.; Satapathy, A.; Mishra, M.K. Development and sliding wear response of epoxy composites filled with coal mine overburden material. *IOP Conf. Ser. Mater. Sci. Eng.* **2018**, *338*, 012032. [[CrossRef](#)]
24. Asadi, I.; Shafiq, P.; Hassan, Z.F.B.A.; Mahyuddi, N.N.B. Thermal conductivity of concrete—A review. *J. Build. Eng.* **2018**, *20*, 81–93. [[CrossRef](#)]
25. Mydin, M.O.; Rozlan, N.A.; Sani, N.M.; Ganesan, S. Analysis of micro-morphology, thermal conductivity, thermal diffusivity and specific heat capacity of coconut fibre reinforced foamed concrete. MATEC Web of Conferences. *EDP Sci.* **2014**, *17*, 1020.
26. Liao, W.; Zeng, C.; Zhuang, Y.; Ma, H.; Deng, W.; Huang, J. Mitigation of thermal curling of concrete slab using phase change material: A feasibility study. *Cem. Concr. Compos.* **2021**, *120*, 104021. [[CrossRef](#)]
27. Tavman, I.; Aydogdu, Y.; Kök, M.; Turgut, A.; Ezan, A.T.I.F. Measurement of heat capacity and thermal conductivity of HDPE/expanded graphite nanocomposites by differential scanning calorimetry. *Arch. Mater. Sci.* **2011**, *50*, 56–60.
28. Fu, S.; Feng, X.; Lauke, B.; Mai, Y. Effects of particle size, particle/matrix interface adhesion and particle loading on mechanical properties of particulate–polymer composites. *Compos. Part B Eng.* **2008**, *39*, 933–961. [[CrossRef](#)]
29. Ahmed, S.; Jones, F.R. A Review of Particulate Reinforcement Theories for Polymer Composites. *J. Mater. Sci.* **1990**, *25*, 4933–4942. [[CrossRef](#)]
30. Serge, I.K.; Shen, C.; Jiang, Y. A review of the current work potential of a trombe wall. *Renew. Sustain. Energy Rev.* **2020**, *130*, 109947. [[CrossRef](#)]
31. Ashby, M. *Materials and the Environment: Eco-Informed Material Choice*; Elsevier: Amsterdam, The Netherlands, 2012.

32. Österle, L. Recycled HDPE with an excellent carbon footprint. *Renewable Carbon News*, 5 May 2022.
33. U.S. Energy Information Administration. Coal Data Browser. 2022. Available online: <https://www.eia.gov/coal/data/browser/> (accessed on 12 January 2023).
34. Istrate, I.; Juan, R.; Martin-gamboa, M.; Domínguez, C.; García-muñoz, R.A.; Dufour, J. Environmental life cycle assessment of the incorporation of recycled high-density polyethylene to polyethylene pipe grade resins. *J. Clean. Prod.* **2021**, *319*, 128580. [[CrossRef](#)]
35. Li, D. *US Plastics Prices Seen Extending Decline in US as Glut Emerges*; Bloomberg: New York, NY, USA, 2022.

Disclaimer/Publisher's Note: The statements, opinions and data contained in all publications are solely those of the individual author(s) and contributor(s) and not of MDPI and/or the editor(s). MDPI and/or the editor(s) disclaim responsibility for any injury to people or property resulting from any ideas, methods, instructions or products referred to in the content.