

Carbon-based Materials for Sustainable Energy Innovations: Recent Advances and Future Challenges

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Abstract

Sustainable energy is essential to reduce fossil fuel consumption. However, sustainable energy development still faces many problems that make its progress insignificant in the last decades. Alternative energy sources are needed to minimize fossil fuels (coal, natural gas, and petroleum). Biomass and carbon fullerenes are a choice, but their innovation technologies are still restricted to pilot-scale production in the laboratory. Biomass is a climate-neutral energy source, but the CO₂ emission of biomass combustion is inevitable, indicating the importance of innovative methods in processing biomass to energy. Different methods (thermochemical or biochemical) will produce different carbon footprints. Besides, biomass development also requires innovative small-scale technology to generate affordable energy for citizens and communities. Carbon fullerenes have been applied for energy applications, especially capacitor and lithium-ion (Li-ion) batteries. Up today, progress in Li-ion battery manufacturing moves at a slower pace and not much progress. The significant problems of Li-ion batteries are volume expansion, low electric conductivity, and inappropriate coulombic efficiency. Carbon-based fibers such as carbon nanofibers, CNT-based fibers, and graphene-based fibers have been used for capacitors, but there are still many obstacles, such as a lack of knowledge to solve capacitor self-discharging. Therefore, research and development of carbon-based materials are urgent energy innovation developments.

Keywords:

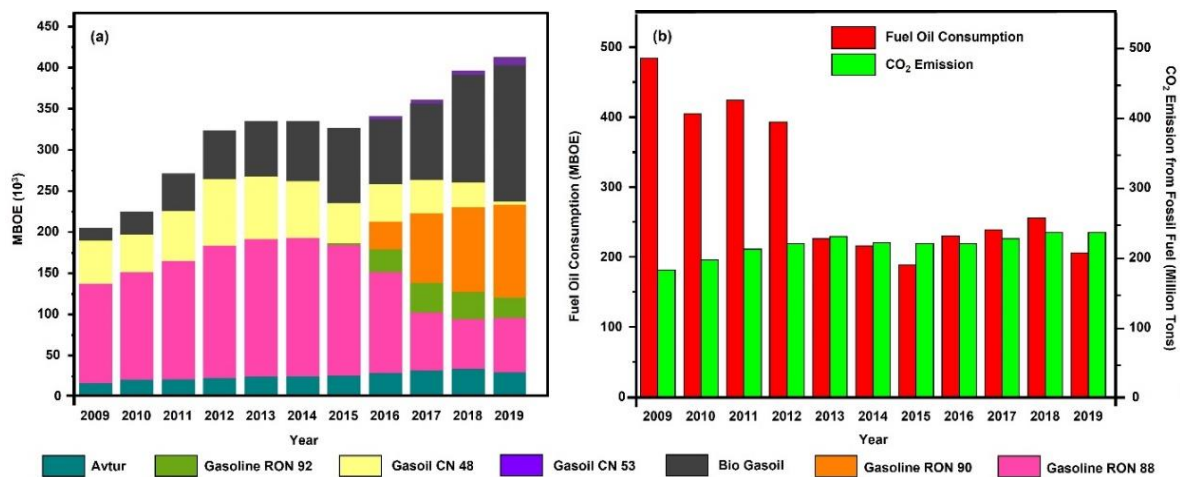
Carbon materials; biomass; sustainability; energy innovations

Introduction

Up to date, clean energy is facing many barriers such as energy policies, finances, and massive coal reserves, which restrict the advancements of clean energy technologies. For example, the use of fossil fuels such as oil, gas, and coal has always caused environmental pollution problems in carbon dioxide (CO₂) emissions and global warming. Greenhouse gases such as CO₂, methane (CH₄), and nitrogen dioxide (NO₂) form a layer in the atmosphere that can withstand the heat leaking from the Earth, thus warming the Earth's

atmosphere (Kamal 2012). Unfortunately, Indonesia still relies heavily on fossil fuel energy. As a result, fuel consumption in the transportation sector sharply increases yearly, increasing greenhouse gas emissions, which are depicted in Fig. 1(a) and 1(b), respectively.

Figure 1.



(a) Fuel consumption in the transportation sector and (b) The correlation between fuel oil consumption and its produced CO₂ emission in Indonesia (source: PYC Data Center).

Besides, a high proportion of fossil energy in a country's energy mix can negatively affect the country's development. As a result, it becomes more dependent on imports for its energy supply and increases greenhouse gas emissions. In Indonesia, there is a gap between oil production and demand. Energy demand is continually increasing to support growth and economic development. At the same time, the capacity of oil production tends to decline. The demand-production gap tends to widen (Haryana 2016). Thacker (2014) estimated that Indonesia may have to import more than 70% of the country's oil demand in 2025 due to lower domestic oil production and increased energy demand (Thacker 2013). Unfortunately, the availability of fossil fuels, especially oil, natural gas, and coal, is also currently declining. Therefore, as part of efforts to save energy by increasing energy efficiency, human needs to find alternative energy sources to replace oil like carbon-based materials.

Carbon-based materials are composed of carbon elements naturally or synthesized in a laboratory. One natural type of carbon-based material is biomass, used as an energy source before people became aware of fossil fuels. However, biomass has been expelled from human

life since people started using oil, gas, or coal to generate electricity. Biomass is a renewable and sustainable energy source. Increasing the use of biomass from waste can reduce pollutions. Biomass as an energy source is a more environmentally friendly option and helps reduce overall greenhouse gas emissions. Biomass sources can be found in all countries of the world, including Indonesia (Parinduri and Parinduri 2020). The significant biomass components are carbon, hydrogen, and nitrogen, which affect the fuel efficacy and the produced pollutant of biomass (Cavalaglio et al. 2020). Biomass as an energy source is acceptable as it is a net energy source that does not contain CO₂ and does not contribute to increasing greenhouse gas emissions, indicating that biomass is climate-neutral.

There are many types of carbon-based materials synthesized in the laboratory, such as graphite (Kim et al. 2020), activated carbon (Feng et al. 2020), fullerene (Collavini and Delgado 2018), porous carbon (Hamouda et al. 2020), graphene, carbon aerogel (Haghighi Poudeh et al. 2020), carbon fiber (Chen, Qiu, and Cheng 2020), carbon dots (CDs) (Hu et al. 2019), and carbon nanotube (CNT) (Iqbal et al. 2019). Carbon-based materials have been applied in many applications such as a capacitor, sensor, medicine, and energy storage because of their abundance and excellent properties like high electrochemical activity (X. Wang, Liu, and Niu 2019), large surface area, excellent conductivity, various morphology, and good chemical properties (Haghighi Poudeh et al. 2020). The different dimensions (macro, micro, or nano size) and characteristics of carbon-based materials depend on their raw materials, chemical treatments, and synthesis methods, producing sp¹, sp², or sp³ carbon hybridizations. This review will summarize the recent use of carbon-based materials for sustainable energy innovations, especially the application of biomass as a sustainable energy source and the utilization of carbon fullerenes for energy applications. At the end of this review, some future outlooks like the advantages and challenges of using carbon-based materials for sustainable technology innovations will be presented to consider the next research plan.

Biomass as a sustainable energy source

Biomass is a resource found in various materials such as wood, sawdust, straw, seed waste, fertilizer, waste paper, household waste, and sewage. These are traditionally used as biomass

resources, increasing their use. Its economic potential is substantial because the annual agricultural production and its by-products can be used as an energy source, especially promoted as so-called energy crops for this purpose (Perea-Moreno, Samerón-Manzano, and Perea-Moreno 2019). Biomass as an energy carrier has not yet been commercialized, but it will be one of the most promising energy carriers. Biomass is very diverse and differs in chemistry, physical characteristics, moisture content, mechanical strength, etc. The conversion of technology to materials and energy is also different (Herlambang et al. 2011).

Many studies suggest that biomass energy will significantly contribute to the total energy supply if fossil fuel prices rise over the next decade. Research is underway to develop affordable and environmentally friendly conversion technologies to reduce reliance on fossil fuels, reduce carbon emissions, and stimulate rural economies. Political decision-makers, scientists, environmentalists, and the biomass industry are discussing whether carbon emissions from biomass combustion should be included in the greenhouse gas balance of bioenergy systems. This discussion is related to the timing of capture, release, and recovery of carbon emissions. In addition, the consideration of biomass to be climate-neutral at the time of production is necessary to allocate CO₂ credits to the carbon captured during the growth of the biomass or assume that the combustion of the biomass will generate carbon emissions immediately (Parinduri and Parinduri 2020).

Based on the primary carbon cycle of the photosynthetic process, multiple and unlimited biomass resources are available. The combustion process of biomass will produce CO₂, but many people assume that biomass treatment equals the net amount of zero CO₂ because new trees and other newly planted plants provide O₂ and CO₂ is absorbed by plants during the growth process. It can be said that CO₂ emission is equal to the CO₂ binding through the growth process. Coal can also be obtained from biomass, but its carbon reach is long-term, millions of years (Herlambang et al. 2011). The bland of research provided a great type of biomass and potential for its utility for energy generation. It is possible to seize power dedicated harvest for specific climate and soil types, in addition to proper waste use. The increase of electricity in the food-agriculture manufacturing process is a simple attitude towards clean energy production (Drożyner et al. 2013). A new phenomenon that is very important for the development of biomass use for sustainable energy is innovative small-scale

technology to generate energy from renewable energy sources, providing the growth of the civil energy sector based on the initiative of citizens and their communities. Regional energy production from biomass fuel is an element of environmental protection (Roman et al. 2021). The total carbon footprint of biomass power depends on many factors, including emissions from biomass sources, biomass transport, power generation methods, and biomass waste sources. The emissions from alternative disposal methods will be used for power generation. In terms of biomass sources, it is important to distinguish between biomass energy crops, biomass waste sources, and other woody biomass sources. Different methods of producing electricity from biomass, such as burning wood chips in a boiler, pyrolysis of biomass (biochar), and conversion of biomass to liquid fuel, lead to different carbon footprints. The timing and rate of CO₂ emissions and forest absorption can also affect the overall CO₂ emissions of biomass power (Zeller-Powell 2011). Biomass, after partial processing, can be in solid-state (briquettes, pellets), liquid (biodiesel, biomethanol, and bioethanol), or gas (biogas, syngas, and hydrogen) (Dodić et al. 2012), and their uses for energy applications are shown in Figure 2. To be applied as an energy source, biomass can be processed by several methods such as thermochemical conversion (combustion, pyrolysis, gasification, and liquefaction) and biochemical conversion (anaerobic digestion and fermentation) (Benti et al. 2021). Besides, biomass can also be processed by a biotechnological process through oily plant fermentation, producing biodiesel that can be used in standard diesel and gas engines after some modifications. Biogas with excessive methane content can be obtained through fermentation of vegetable waste and cattle manure (Zagorskis et al. 2012).

Figure 2.



Applications of biomass for sustainable energy: (a) use of charcoal briquettes for cooking; (b)

charcoal briquettes from sugarcane bagasse, cassava rhizomes, and water hyacinth (Suttibak and Loengbudnark 2018); (c) pellets from bamboo and pine particles (Liu et al. 2016); (d) biodiesel from soybean oil and methanol; (e) biogas from organic or food waste (Al-Wahaibi et al. 2020); and (f) syngas from the mixture of CO₂ and water molecules to substitute fossil-based fuels ((Luciani, Landi, and Di Benedetto 2020).

Bioethanol as a fuel for automobiles can reduce the contribution of CO₂ to the atmosphere because the production of bioethanol from biomass can be considered a closed cycle. As a result of this entire process, greenhouse gases are not considered for CO₂ liquefied gas emissions into the atmosphere. Bioethanol-cellulose can reduce greenhouse gas emissions by 80%. In addition, bioethanol can be used as a solvent to overcome the problem of waxy crude oil. The process of bioethanol products is carried out by hydrolysis, fermentation, distillation, and characterization using gas chromatography-mass spectrometry (GCMS) (Afdhol, Lubis, and Siregar n.d.). The three major technologies which are classified as third-generation bioethanol production technologies are (i) fermentation of processed biomass, (ii) dark fermentation of stored carbohydrates, and (iii) direct photofermentation of CO₂ into bioethanol using light energy. However, traditional techniques for biomass fermentation using these microorganisms are still of great interest (Lakatos et al. 2019).

Biogas manufacturing from agricultural wastes, animal excrements, and business wastes can supply electricity for many countries. For example, one cubic meter of biogas can produce electric power of 2.1 kWh and 2.9 kWh warmness electricity equivalent (Frac and Ziemiński 2012). The process of biogas production leads to the development of significant opportunities, including the use of waste and biomass, the energy generated from biogas in plant self-sufficiency, and improving the quality of environmental conditions (environmental protection and climate). Reducing the use of non-renewable resources will generate energy and reduce the amount of landfill waste. The big picture of strategic factors related to opportunities and risks clearly shows that the biogas production process offers opportunities for change that are more favorable than the potential risks that represent future threats.

Syngas from the pyrolysis and gasification of biomass is an essential intermediate product for synthesizing many industrial products. Examples include hydrocarbons using ammonia synthesis or Fischer-Tropsch synthesis. Syngas is also a raw material for methanol (D. Li et al. 2013). Maximizing the yield of syngas from biomass will significantly facilitate the efficient

use of biomass. Biofuels and chemicals can be produced from high-quality syngas (mainly H_2 and CO). With or without catalyst, syngas is obtained from biomass and waste through thermochemical processes (Skoulou and Zabaniotou 2013). Because of that, syngas can be used in an energy-efficient and environmentally friendly way by integrating with the existing agriculture, forestry, energy, or chemical industries in the concept of biorefinery (Peres, Lunelli, and Filho 2013).

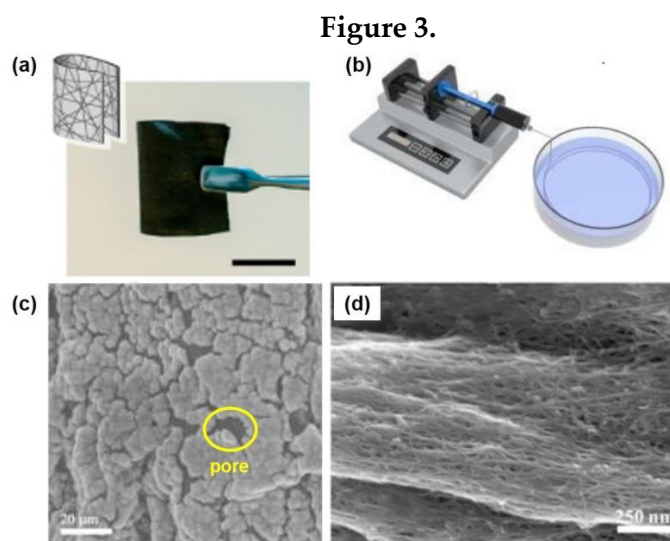
Biomass gasification is an attractive thermochemical conversion process that enables the production of synthetic gas (syngas) that can be used to generate electricity in an internal combustion engine or for other applications such as the production of liquid fuels (Gallucci et al. 2019). The gasification process can be applied to biomass with less than 35% moisture content. Direct gasification (or self-heat gasification) occurs when the oxidative gasifier partially oxidizes the raw material and provides heat to the process. Low-temperature direct gasification (less than $900^{\circ}C$) can be performed on fixed beds, fluidized beds (FB), or circulating fluidized beds (CFB), and high-temperature direct gasification ($1300^{\circ}C$ and above) can be performed on companion flow gasifiers (T. Song et al. 2012). Nipattummakul et al. (2012) investigated the production of synthetic gas from palm oil-trunk waste vapor gasification and observed that the high initial synthetic gas flow rate was mainly due to the thermal decomposition of volatile substances from palm oil samples. Almost 50% of syngas is produced in the first five minutes. The results showed that steam gasification increased hydrogen production by more than 60% compared to pyrolysis. Increasing the steam flow rate shortened the gasification time and promoted the steam reforming reaction, increasing hydrogen yield. The increase in steam flow had little effect on the apparent thermal efficiency (Nipattummakul et al. 2012).

Carbon fullerenes for energy applications

One of the most popular energy applications is energy storage because it is a vital component of the development of the energy technology conversation. Energy storage stores electricity in many different forms, such as chemical, thermal, electrical, and electrochemical energy until converted back to electrical energy. Advanced energy storage technology provides economic benefits and protects our environment from pollution. Besides, energy storage can keep a market equilibrium, stabilize electrical parameters, prevent reductions during high energy

demand, and enhance grid efficiency through stabilizing grid power to reduce energy losses. The energy storage applications have tremendous advancements, from micro batteries to wide-ranging electric vehicles. However, the development of energy storage is necessary because our gadgets are impossible without modern energy storage which possesses high performance, smaller, lighter, versatile, and affordable.

Carbon-based fibers, especially carbon nanofibers, CNT-based fibers, and graphene-based fibers, have been widely used for advanced electrochemical energy storage devices because of their outstanding properties such as low density, good conductivity, elasticity, excellent mechanical strength, and tunable electrochemical performance (Chen, Qiu, and Cheng 2020). A free-standing and flexible carbon nanofiber has been successfully synthesized from lignin/polyvinyl alcohol (PVA) solution using electrospinning technique, producing good flexibility and electrochemical performance which enables it to be applied for supercapacitor electrodes. The carbonization process could affect the electrochemical properties of nanofiber. The increased carbonization temperature enhanced its hydrophobic property, which restricted electrolyte ions from being absorbed on the electrode surface (Wei et al. 2020). A hybrid CNT fiber-based supercapacitor was made from manganese dioxide (MnO_2) flakes and CNT fiber through the electrodeposition process to form elastic nanocomposite fiber. CNT fibers were produced by using a conventional wet-spinning process. The electrodeposition of MnO_2 flakes onto CNT fibers could enhance fiber's elasticity and porosity, which is depicted in Figure 3c, contributing to electrons and ions transport necessary for supercapacitor (Lu et al. 2017). In another report, the addition of graphene to CNT fiber could enhance the elasticity of CNT, which was suitable for wearable energy storage. Graphene would reduce the dense density of stacking CNT fibers, increasing the inner porosity, which possessed ions to access the electrode easily (Lu et al. 2018). However, there are still many challenges to advancing carbon-based materials for energy storage, such as lack of knowledge about surface chemistry, significantly when the size of carbon-based materials contributes to the self-discharging of capacitors (Jeerapan and Ma 2019).



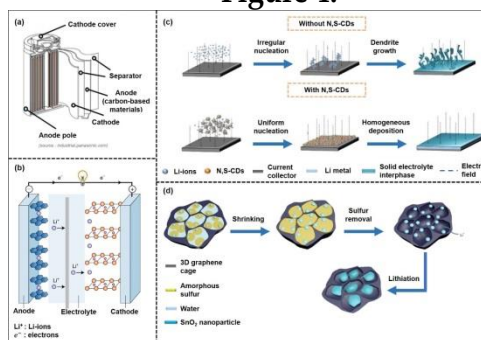
Carbon fullerenes for energy applications: (a) carbon nanofiber networks from lignin/PVA solution for supercapacitor electrode (Wei et al. 2020); (b) preparation of CNT fibers from liquid crystal dispersion of single-walled CNT for wearable energy storage; (c) SEM image of hybrid CNT fiber and MnO₂ flake (Lu et al. 2017) and (d) SEM image of CNT/graphene fiber (Lu et al. 2018).

Depending on the battery's ability to be re-used, the battery is classified into non-rechargeable (primary) and rechargeable (secondary) batteries. Rechargeable batteries can be better for the environment because they can reduce carbon footprint and waste. One of the most popular rechargeable batteries for electronics is the Li-ion battery. Generally, a Li-ion battery has four essential components: a negative electrode (anode), a positive electrode (cathode), electrolyte, and separator, which is made from insulator and porous material (Figure 4a). Since being introduced in 1991 by Sony Corporations, Li-ion batteries have been significantly improved, like using carbon-based materials in various functions. The hybrid structure of highly active graphene quantum dot wrapped conductive CNT as an anode of Li-ion battery could enhance battery performance. Graphene functional groups massively provided storage sites for Li-ions and CNT inhibited graphene agglomeration (Zhao et al. 2020). Li-ion batteries can convert chemical energy to electrical energy through electrochemical oxidation-reduction reactions. To produce an electrical current, electrons in the Li-ion battery will flow from anode to cathode through an external circuit (physical process) and in the electrolyte solution (chemical process), which is presented schematically in Figure 4b (Goodenough 2018).

Different electrodes and electrolytes materials provide different schemes of chemical reactions, influencing the Li-battery mechanism and its capacity to store energy.

CDs have been used for anode and electrolyte of Li-ion battery. The combination of CDs and graphene quantum dot as battery anode could increase the battery's electrochemical performance, such as preventing volume expansion, increasing electric conductivity, and improving coulombic efficiency (T. B. Song et al. 2020). However, CDs for anode still require high-temperature treatment, which restricts large-scale production because of the cost. As an electrolyte additive, CDs could inhibit the dendrite growth and play a role as hosts to facilitate volume expansion (Guo et al. 2021). In addition, the dendrite can lead to anode destruction, which reduces battery efficiency. The addition of nitrogen, sulfur-codoped (N,S-CDs) to the Li-battery electrolyte could make lithium metal anode more stable because N,S-CDs could absorb lithium-ion in the surrounding current collector, which enabled the formation of a uniform deposition layer on the electrode. The comparison between electrodes without N,S-CDs and electrodes with N,S-CDs is graphically presented in Figure 4c (S. Li et al. 2021). Wang et al. (2019) proposed that carbon cages can be applied to improve the capacity of Li-ion battery anode because its strength will provide enough space for the volume expansion (L. Wang et al. 2019). It is relevant to another report that graphene-caged tin oxide could prevent the volume change. In Figure 4d, the graphene cage was prepared using sulfur as a template. When the solution evaporated, a shrinkage would be produced. If graphene did not have enough space for noncarbon expansion, the graphene cage would crack during lithiation. Therefore a template is needed to prevent noncarbon expansion (Han et al. 2018), but the appropriate design is still advanced.

Figure 4.



Carbon-based materials for Li-ion battery: (a) components of Li-ion battery; (b) scheme

of Li-ion battery when charging and discharging (Goodenough 2018); (c) N,S-CDs for anode Li-ion battery (S. Li et al. 2021); and (e) graphene cage to overcome volume expansion problem (Han et al. 2018).

Challenges and future outlooks

Some benefits of using biomass as an energy source are abundant, carbon-neutral, eco-friendly, and easy synthesis process. Besides, hydrogen can also be produced from biomass through photocatalytic biomass conversion. Hydrogen is an energy carrier that can be converted to electricity by a fuel cell or hydrogen-fueled engine. Bioeconomy has to be involved in measuring how sustainable biomass can be harvested and the types of products made from it because biomass also produces CO₂ emission during its energy production process. The use of woody biomass is significantly increasing, but the challenge is not enough space to plant trees because lack of human attention to nurture forest provides inadequate biomass energy for society. A high production cost is also a significant problem because petroleum is cheap, which provides difficulties in selling low-cost sustainable energy. Therefore, the search for innovative methods and low-cost technology is essential for processing biomass to energy. In addition, the lack of tools to store biogas before consumption and its conversion into electricity is still a problem for the population, especially in developing countries.

Up today, progress in Li-ion battery manufacturing moves slower, and not much progress has been made because it is not easy to bring a pilot-scale production in the laboratory to be industrialized. Several issues of Li-ion batteries are high cost, low coulombic efficiency, limited energy density, and complicated synthesis process. An appropriate material structure design is needed to produce a long-life battery. Besides, the recycling process of Li-ion batteries has to be tackled because of the shortage of raw materials and producing chemical waste to the environment due to the short-term battery life cycle. Consequently, advancement in battery life is crucial to minimize chemical waste. One of the best solutions is carbon fullerenes, increasing the battery life cycle. However, its cost production is still high, especially for carbon-based nanomaterials such as CNT and CDs. For a capacitor, the primary keys are mechanical properties and accessible surface area of electrodes. Highly porous carbon-based

materials are suitable for a high-capacity capacitor. Pores in the electrodes will provide ion diffusion channels in a capacitor. The electrochemical performance of capacitors depends on their specific surface area (micropores, mesopores, or macropores), which leads to finding an appropriate material design for capacitors.

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