Edited by Carlos Miguel Costa

Renato Gonçalves | Senentxu Lanceros-Méndez

Sustainable Energy Storage in the Scope of Circular Economy

Advanced Materials and Device Design

WILEY

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Preface

"Energy and persistence conquer all things."

Benjamin Franklin (1706–1790)

Considering the United Nations Agenda 2030 for Sustainable Development and the urgent need to reduce the environmental impact of current and emerging technologies, the development of sustainable smart and multifunctional materials and processes is essential for an increasing number of application areas.

One of the most relevant areas is energy, based both on the increasing energy demand that must be supported by a growing share of renewable energy sources and energy storage systems and in the electrification and digitalization of the economy in the scope of the Industry 4.0 and Internet of Things (IoT) paradigms, demanding ubiquitous sensing, data communication systems, and mobile electronic devices, also relying in energy storage systems.

In this book, the current state of the art on energy storage devices based on sustainable materials is addressed within the framework of the circular economy.

Further, the main applications being developed for these energy storage devices as well as the main advantages and remaining challenges in this research field are presented.

The first chapter provides an overview on circular economy, including the general concepts, definition, benefits, and origins. The principles of the circular economy are presented and their relevance in the context of energy is discussed. Also, the approach based on the sustainability for energy field, with special focus on battery devices, is described.

Chapter two presents the energy carrier properties of reactive metals and their application in energy storage applications. The economic and environmental implications of reactive metals are also discussed.

Chapters three, four and five focuses on sustainable battery materials, covering materials for lithium-ion batteries, beyond lithium, and biodegradable batteries, respectively.

Chapter six is dedicated to sustainable materials for supercapacitor devices, with special attention to electrodes and electrolyte materials. Recent advances on nanomaterials for electrodes are presented and discussed.

Chapters seven and eight focuses on sustainable approaches for fuel cells, describing the materials for fuel cell devices and the recent advances in microbial fuel cells, respectively.

Chapter nine presents multifunctional sustainable materials for energy storage based on biomass-derived electrodes and traditional carbon electrodes for redox flow batteries.

Chapters ten and eleven focuses on sustainable devices and their corresponding design, covering sensors/actuators and devices in the scope of Internet of Things (IoT), respectively.

Chapter twelve reports on waste prevention for energy storage devices based on second-life use of lithium-ion batteries.

Chapter thirteen describes the recycling procedures for energy storage devices with a particular focus on the implementation of the electric vehicle.

Finally, chapter fourteen summarizes some of the open questions, needs, and future trends in the scope of the principles of sustainability of materials and processes applied to energy storage systems.

This book was only possible by the excellent contributions of the authors who accepted the invitation to write different chapters in this relevant, needed, and timely research and technological field that encompasses sustainability and energy storage systems. The editors are grateful for the time and effort to write these magnificent chapters that will serve as a reference for the state of the art and as a guide for further developments in this area. For us, it was real pleasure and an honor to have contacted, to have worked together, and to provide all together the important milestone that this book represents!

Also, this book would not have been possible without the support, understanding, and dedication of our research group and also of the research centers at the University of Minho, Braga, Portugal and BCMateriais, Basque Center for Materials, Applications and Nanostructures, Leioa, Spain, to which the different editors belong. Pushing science and technology forward through the idealization, writing and materialization of a book is one of the most beautiful and relevant efforts to share with you all!

Last but not least, we truly thank the excellent support from the team from Wiley: from the first contacts with Sarah Higginbotham and Sakeena Quraishi to the latter support from Stefanie Volk and Jenny Cossham. The continuous support, technical expertise, patience, and kindness were essential to make this book come true. It has been a pleasure working with you all!

Finally, being a book that covers sustainable materials for batteries, supercapacitors, and fuel cells; energy storage devices for sensors, biomedical, and wearable applications, as well as recycling and utilization, the book can be used on a variety of different scopes: from researcher and professionals aiming to enter any of the different fields covered by the book to courses at universities focusing on sustainable energy. It is also suitable for political organizations, companies, and NGOs focusing on this subject and/or organizing specific funding schemes and/or development strategies in the area. The editors truly believe that this book will become a milestone to further foster increasing scientific and engineering efforts based on sustainability and circular economy paradigms applied to energy storage devices, urgently needed to successfully tackle some of the main challenges society is facing nowadays: energy and environment.

Carlos Miguel Costa, Renato Gonçalves, and
Senentxu Lanceros-Méndez
Braga, Portugal
10 November 2022

Part I

Introduction

1

The Central Role of Energy in the Scope of Circular Economy and Sustainable Approaches in Energy Generation and Storage

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1.1 Introduction

Since the last century, it is observed that fast economic growth worldwide has also led to a strong environmental impact. Thus, efforts from different world countries are being directed toward the development of more sustainable technologies. In this context, the attention to the circular economy concept increased, where the waste of resources and environmental pollution matters have been intensively addressed in a variety of ways. Different countries have been developing approaches to increase the implementation of the circular economy paradigm in their own country to substitute the current linear economy model, which leads to a strong environmental impact [1]. Different countries and geographical areas such as China, the United States, the European Union, and India have implemented targets and conditions to improve the circular economy not only for their individual country but also to interrelate which other countries create a world circular economy based on exchange and product valorization [2].

The general concept of circular economy is based on the fact that economic activity contributes to the overall sustainability of the system. The circular economy concept recognizes how important the functioning of the economy is at any level – large and small businesses, organizations and individuals, and globally and locally. Thinking about the subject, it is realized that nature has no room for waste or trash. That is, all the elements in nature are related in a delicate balance. Thus, the circular economy must be a system of use of resources where the reduction, reuse, and recycling of elements predominate. Considering this, the circular economy model is based on consumption and production in relation to concepts such as sharing, leasing, reusing, repairing, refurbishing, and

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recycling, preserving the materials and products as long as possible and extending their life cycle. Through the reduction of waste and extending the material's life cycle, the circular economy can also provide further value.

Unlike the traditional model of linear economy, where natural resources and energy are turned into products that are ultimately destined to become waste based on a take-make-consume-throw-away pattern [3], a circular economy employs reuse, sharing, repair, refurbishment, remanufacturing, and recycling to create a closed-loop system, minimizing the use of resource inputs and the creation of waste, pollution, and carbon emissions [4]. In this sense, the circular economy model relies on large quantities of cheaper, easily accessible materials and energy, and aims to keep products, materials, equipment, and infrastructure [5] in use for longer time, thus improving the productivity of the resources. Waste materials and energy should become input for other processes through waste valorization: either as a component for another industrial process or as regenerative resources for nature (Figure 1.1).

A circular economy approach aims to preserve the value of products, materials, and resources for as long as possible. Following the procedures of fewer products discarded and fewer materials extracted, least energy is lost and the environment is preserved. Thus, the circular economy, in order to tackle global challenges like climate change, biodiversity loss, waste, and pollution, is a framework of three principles, driven by design: eliminate waste and pollution, keep products and materials in use, and regenerate natural systems [6].

The circular economy proposes an opportunity to restructure our economy. This comprehensive strategy that aims to reduce both the input of raw materials as well as the production of waste closes the cycles or economic and ecological flows of resources, providing the benefits summarized in Table 1.1 for the economy, the environment, and the community.

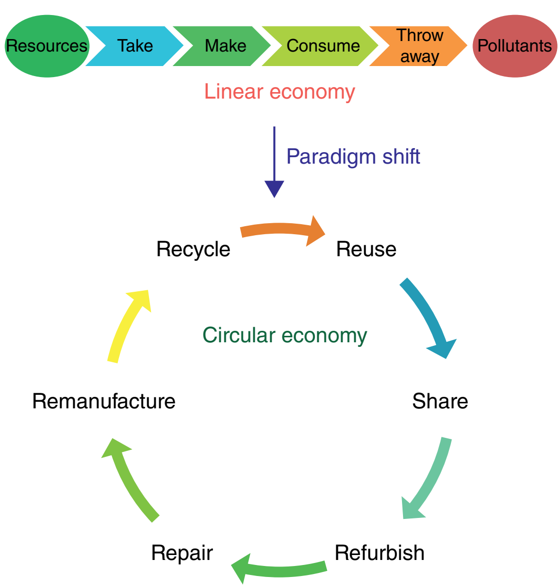

Figure 1.1 An illustration showing the paradigm shift from the take-make-consume-throw-away pattern of the linear economy to the circular economy approach.

Table 1.1 Summary of the benefits provided by the circular economy in the three main areas of action.

Benefit area	
Economy	Creates balanced wealth
	Opportunities for local jobs and social integration
	Reduces expenses and investments. More innovative and efficient ways of producing and consuming
	Reorients the production of the countries, protecting businesses against scarcity of resources and volatile prices
Environment	Reduces the use of resources
	Optimization of waste management which boosts recycling and reduces landfill
	Limits energy consumption, fewer production processes require less energy
	Benefits for the environment in terms of climate and biodiversity, air, soil, and water preservation
Community	Allows the change of consumption habits.
	Creates awareness
	Balances society with the economy and the environment

1.2 Circular Economy and the Central Role of Energy

The notion of circularity has its origin in the historical as well as in the philosophical field. The idea of feedback and cycles in world systems is old and arises in various philosophical schools. This concept regained interest at the end of World War II, when computer studies of nonlinear systems revealed the complex, connected, and unpredictable nature of our world. Those studies led to the conclusion that the circular economy concept is more like a metabolism than a machine. In this scope, recent technological advances have the power to support the transition to a circular economy through modeling and computer simulations, allied to the new materials and production technologies. Finally, the term *circular economy* was first used in Western literature in 1980 [7] to describe a closed system of interactions between the economy and the environment. Therefore, the circular economy is part of the study of feedback from nonlinear systems, including living systems.

More recently, the idea and concept of circular economy have been introduced in the guidelines and action plans of institutions worldwide. In 2015, the European Commission adopted an ambitious "Circular Economy Package." An EU Action Plan for the circular economy establishes a specific program of actions outlining measures that cover the entire product life cycle: from production and consumption to waste management and the market for secondary raw materials. On 4 March 2019, the European Commission adopted a comprehensive report on the implementation of the Circular Economy Action Plan.

On 11 March 2020, the European Commission adopted a new Circular Economy Action Plan – one of the main building blocks of the European Green Deal, Europe's new agenda for sustainable growth. The new Action Plan announces initiatives along the entire life cycle of products, targeting for example their design, promoting circular economy processes, fostering sustainable consumption, and aiming to ensure that the resources used are kept in the EU economy for as long as possible. It introduces legislative and non-legislative measures targeting areas where action at the EU level brings real added value.

In addition, the circular economy has strong synergies with the EU's objectives on climate and energy and with the Commission's package on "Clean Energy for all Europeans." The circular economy is also instrumental in supporting the EU's commitments on sustainability, as outlined in the communication "Next steps for a sustainable European future" and in particular to reach Sustainable Development Goal 12 "Responsible consumption and production" [8]. China has established guidelines and regulations at a multilevel scale from micro to macro in a top-down structure. The *Circular Economy Promotion Law,* published in 2008, is one of the many efforts that promote the 3R policy (reduce, reuse, and recycle). With the "National Development and Reform Commission (NDRC)" in 2016, digital solutions have into account the circular economy to design products and new business models. In 2018, the "*Circular Economy Promotion Law*" was revised highlighting the recycling processes until 2020, by the domestic–international circulation [9]. Latin American countries are organizing primary work on circular economy roadmap since 2020 based on policies related to landfill bans, extended producer responsibility, and renewable energy end-of-use [10]. The US *National Recycling Strategy* implements a national recycling rate goal of 50% by 2030. This goal is based on five strategies: support recycling, standardize measurements, reduce contamination from recycling, improve markets for recycling commodities, and increase materials recycling collection and infrastructures [11].

Although some circular economy practices are already well established in some places, most definitions and actions relate to perspective possibilities and strategies, rather than to what actually exists. Multiple interrelated concepts such as loop closing, ecodesign, industrial ecology, industrial symbiosis, life cycle analysis, and performance economy have contributed to the concept of a circular economy. Nonetheless, a strong policy direction is likely needed for effective implementation [12].

The general objective of the circular economy is to obtain products and manufacturing processes that consume little energy and do not generate waste or garbage that negatively affect society and the environment. In order to meet this important challenge, there are different components or principles that define how the circular economy should be implemented:

1) **The waste becomes a resource:** it is the main characteristic. All biodegradable materials return to nature and the non-biodegradable materials are reused.
2) **The second use:** to reintroduce into the economic circuit those products that no longer correspond to the initial needs of the consumers.
3) **Reuse:** reuse certain residues or certain parts of them, which can still work for the production of new products.
4) **Repair:** find a second life for damaged products.
5) **Recycling:** use the materials found in waste.
6) **Valorization:** take advantage of waste that cannot be recycled.
7) **The economy of functionality:** the circular economy proposes to eliminate the sale of products in many cases to implement a system of renting goods. When the product completes its main function, it returns to the company, which will disassemble it to reuse its valid parts.

8) **Energy from renewable sources:** elimination of fossil fuels to produce the product, reuse, and recycle.
9) **Eco-conception:** considers the environmental impacts throughout the life cycle of a product and integrates them from its conception.
10) **Industrial and territorial ecology:** establishment of a mode of industrial organization in the same territory characterized by optimized management of stocks and flows of materials, energy, and services.

These ten principles are interrelated in the concept of circular economy as represented in Figure 1.2.

As a consequence of the 10 principles, oil and gas energy resources are incompatible with the idea of a circular economy, since they represent "development that meets the needs of the present while compromising the ability of future generations to meet their own needs" [13]. A sustainable circular economy can only be powered by renewable energies, such as wind, solar, hydropower, and geothermal, among others [14].

What gives entities the ability to achieve "net zero" carbon emissions, is that they can offset their fossil fuel consumption by removing carbon from the atmosphere. While this is a necessary first step, it has been also stated that in order to create a truly circular economy we should adopt the concept of "true zero" as opposed to "net zero," which is eliminating fossil fuel consumption entirely so that all energy is produced from renewable sources [15, 16].

Current growth projections in the renewable energy industry expect a significant amount of energy and raw materials to manufacture and maintain these renewable systems. Nevertheless, "due to the emissions attributed to fossil-fuel electricity generation, the overall carbon footprint of renewable energy technologies is significantly lower than for fossil-fuel generation over the respective systems lifespan" [17]. However, there are still

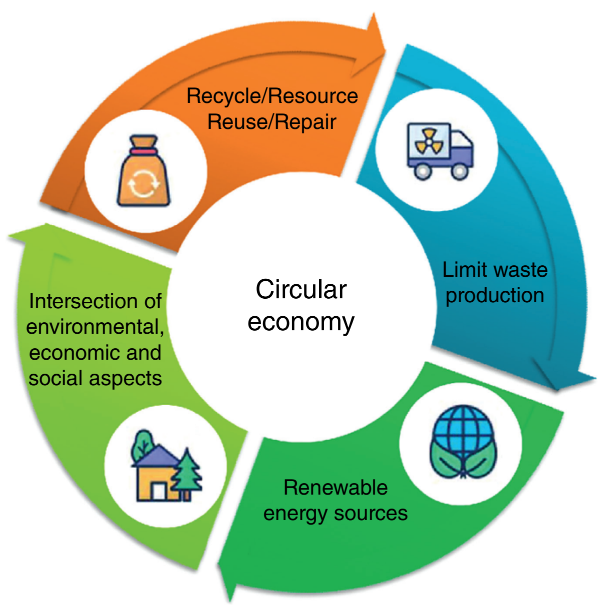

Figure 1.2 Illustration of the relationship between the basic principles of the circular economy.

linear trajectories when establishing renewable energy systems that should be assessed in order to fully transition to a circular economy.

In addition to the perspective of renewable energies, different regulations across the world have been applied, such as the European Commission, in the field of batteries and accumulators, which aims to ensure that batteries and accumulators placed in the EU market are sustainable and safe throughout their entire life cycle. This regulation focuses on the importance of sustainable energy in the area of batteries and accumulators, as they will play an essential role to ensure that many daily-used products, appliances, and services work properly anywhere and anytime, constituting an indispensable energy source in our society. In the last couple of years, approximately 800 000 tons of automotive batteries, 190 000 tons of industrial batteries, and 160 000 tons of consumer batteries enter the European Union every year, so that, in order to achieve the Green Deal, sustainable batteries for a circular and climate neutral economy are necessary.

1.3 The Central Role of Energy in the Scope of Sustainability

Companies are playing a key role in energy transition by contributing to energy security and decarbonisation. In the face of growing political pressure and increasing levels of regulation, investors on the one hand and citizens on the other are demanding energy supply organizations to address climate change and sustainability. These trends continue to compel energy companies to systematically pursue their transition to affordable, reliable, and more sustainable energy and to set ambitious targets in terms of reducing CO_2 emissions. In this sense, circular approaches decouple economic activity from the consumption of materials and energy by building closed cycles in which waste is minimized or even eliminated, and in which resources are reused, including carbon. They do this by using resources efficiently, prioritizing renewable inputs, maximizing the effective life of a product, and capturing and reusing what was previously considered waste. Obviously, not all actors in the energy sector have the opportunity to aim for full circularity. But sticking to circular thinking as the core approach will lead to powerful new insights and levers for efficiency and costing. The increasing use of renewable energy and energy storage can bring new perspectives for a sustainable approach (Figure 1.3) [18].

1.3.1 Energy Generation

Industries with operational models centered on the extraction and use of fossil fuels are inherently linear and hard to be compatible with the idea of a sustainable circular economy. However, companies in these industries are able to incorporate cyclic elements into their operations and, in some cases, have adopted technological advances to reduce the impact of fossil fuel use. For example, several initiatives are underway to explore the potential for reusing oil fields and offshore assets after drilling operations cease. One of the most common initiatives is the reuse of platforms to support offshore wind projects [19]. In the Mediterranean, a system of turbines is being developed that could be used to turn decommissioned oil platforms into islands of renewable energy [20]. Further, there are different

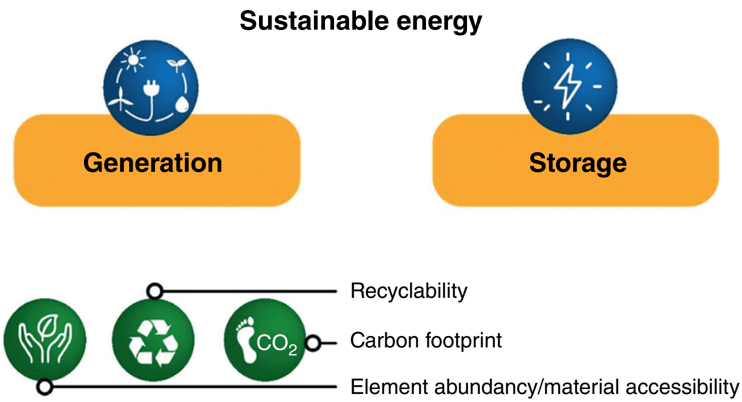

Figure 1.3 Sustainable energy insights for energy generation and storage.

projects intending to use discontinued offshore oil and gas fields to store CO_2 permanently. It is estimated to store approximately 3.5 million tons of CO_2 per year by 2030 [21]. In this regard, other initiatives are experimenting with ways to capture greenhouse gases that would otherwise be released into the atmosphere from factories and power plants. These gases are being used for enhanced oil recovery, injecting these gases into aging oil fields in an attempt to pump out the remaining supply [21, 22].

On the other hand, different organizations are focusing on technologies to generate energy based on gas with low carbon emissions. To do this, natural gas is burned with pure oxygen instead of air to generate electricity without emitting CO_2. This process also avoids generating nitrogen oxides, the main air and health pollutant emitted by gas plants [23]. Early developments point to a 20% increase in power output compared to a conventional plant using the same amount of fuel [24].

Circularity provides a strategic and effective way to identify opportunities for both cost savings and value forming. In some cases, switching to renewable energy as an input is the first step. As technologies advance, the adoption of zero-emission energy is becoming more widespread. This shift to renewable energy is a key driver of the circular economy, and a growing number of organizations are seeking to minimize prices, increase sustainability, and create long-term strategic costs through the shift. Several aspects are driving renewable energy adoption, and the economic dimension is also becoming less of a barrier not only in the power generation sector but also for end users [25]. Examples of organizations continuing to move toward renewable energy in order to achieve a sustainable approach span continents and sectors: in the mining industry, which has generally relied on natural gas or diesel to power its operations, significant investments in solar energy and battery technology have been announced for ore extraction operations. The integration of photovoltaic solar energy combined with large-scale battery storage will eventually meet 25–30% of stationary energy needs from solar generation [26, 27]. More ambitious plans aim to achieve 100% energy from renewable sources by mid-2020s [28]. Similarly, many refineries and chemical plants are moving to solar power to satisfy the energy demand [29, 30].

Although the change to solar energy and batteries is one of the key steps to achieve sustainable energy production in all dimensions, without a commitment to better recycling, the adoption of renewable energy will come with ever-increasing material footprints. Solar panel and battery technology present challenges and opportunities. In 2016, the International Renewable Energy Agency (IRENA) estimated that there was around 250 000 tons of solar panel waste on the planet. The IRENA study shows that many technical obstacles need to be overcome before closed circularity for solar panels is feasible. But the raw materials already have the opportunity to be recycled at a rate of 65–70% in weight [31].

According to the International Energy Agency, the energy obtained from wind renewable sources in 2020 is responsible for around 1500 TWh of the total world energy production and it is estimated that in 2030 this production reaches 8000 TWh. This technology increased significantly over the last few years contributing to the reduction of carbon emissions. The lifetime of a wind turbine is around 25 years reaching in some cases 35 years. Nevertheless, this increase leads to higher consumption of wind turbines that require a huge number of different components. To prevent these components from going to landfills, it is urgent to develop new components capable of being incorporated into the circular economy. The circular economy for this system is focused on recycling, upcycling, extending life cycles, and a combination of new and fewer materials. To overcome these obstacles, some initiatives have been started as increasing funding for resources and development (R&D) and incentives to use recycled composite materials [25].

1.3.2 Energy Storage

In the case of battery technologies, the increasing use of battery technology is at the forefront of renewable energy storage and transportation decarbonisation strategies. But battery production itself has a significant carbon footprint. The CO_2 footprint from electric vehicle fabrication, for example, is higher than for an internal combustion engine vehicle fabrication. Nevertheless, the lower direct/indirect emissions during the usage of the electric vehicle result in a lower overall CO_2 footprint [32]. Battery circular economic chain should be considered in the context of the end market and assessed against industry benefits for power generation and transportation.

The World Economic Forum and the Global Battery Alliance emphasize the value of such cross-sector coupling. However, they also recognize broader concerns about the battery cost chain (economic dimension). For example, the production of basic raw materials for batteries has been linked to hazardous working conditions, child labor, poverty, and other problems related to social and environmental sustainability dimensions [33]. To overcome these obstacles, the European Union, for example, will propose by 2023 a new regulation to update the regulation of 2006. This new regulation will restrict not only battery producers but also companies that produce electric vehicles (EVs).

The main battery system used in EVs is lithium-ion batteries, mainly due to the relatively high-energy density and the high diversity of power/energy ratios. This system is complex and by each battery component (electrodes, separator, electrolyte, current collector, and others) there exists a large number of different materials that can be used [34]. Once that, the recycling process of this system experiences many difficulties. Different techniques (metallurgical processes, including pyrometallurgy and hydrometallurgy) have been developed and improved to recycle these materials. The use of eutectic solvents to extract active materials from the cathodes, the use of natural materials, and the use of biodegradable precursors and solvents to allow their reutilization are examples of the different approaches used to increase the life cycle of these systems and to reduce the environmental impact of materials, devices, and fabrication processes [35].

The cathode electrode is the main costly battery component and is mainly responsible for battery energy density. Cathode-active materials such as lithium nickel cobalt aluminum oxides and lithium nickel manganese oxides are used in majority of EVs. To achieve the sustainability of lithium-ion batteries, one of the possible routes is the use of more sustainable materials with less toxicity (cobalt-free) as lithium iron phosphate. Furthermore, research has been focusing on the development of other systems, such as sodium, sulfur, and others, to develop batteries without lithium to overcome the problem associated with lithium extraction [36]. Together with direct recycling, different approaches, such as second-use applications, can avoid battery accumulation in landfills. Second-use application is based on giving another use to a battery that no longer has the needed capacity in the application that requires less capacity. This approach will not only prolong the life of spent batteries but also avoid the extraction, synthesis, purification, and modification of materials to fabricate a new battery, increasing the sustainability of battery systems [37].

Hydrogen (H_2) is the most exciting example of a circular economy across industries, a clean fuel with no direct emissions of harmful pollutants or greenhouse gases. However, the industrial demand for hydrogen is currently almost completely covered by fossil fuels and is therefore one of the most important causes of CO_2 emissions. The perspective of a hydrogen-powered future is based on the production of hydrogen from low-carbon energy sources such as renewable generation (green hydrogen) or from natural gas (blue hydrogen) [38].

Hydrogen could play an important role in a sector-coupled circular economy, offering a solution to decarbonize a wide range of sectors, including transport, heating, chemicals and iron and steel, and production/transformation [39]. Hydrogen and its derivatives such as synthetic fuels or chemicals offer an opportunity to replace fossil hydrocarbons, which can reduce emissions [39]. In this scope, electricity and hydrogen are complementary and both will be needed to achieve climate neutrality and contribute to climate protection and sustainability. Since hydrogen can be generated by capturing excess production from solar and wind power, it is also a leading contender for a storage solution for renewable energy generation.

1.4 Conclusions and Outlook

Energy sustainability approaches and circular economy are nowadays topics with increasing relevance since energy consumption increases year after year. Thus, it is essential to be aware in which way this growing energy consumption will impact the ecological footprint. The transition to clean energy generation/storage and urgent financial/environmental considerations are taking place toward a sustainable energy system to guarantee the future quality of life.

Environmental sustainability is the responsibility to preserve natural resources and protect global ecosystems to support health and well-being, now and in the future. Life cycle assessment is a commonly used tool for evaluating the environmental impact. The less damage is caused to the environment, the more positively this criterion is fulfilled. Opportunities and challenges of the different energy technologies must be faced and explored. Some energy technologies, such as photovoltaic, have the possibility of recycling up to 95% of their materials; however, challenges such as the presence of hazardous substances including cadmium, arsenic, lead, and antimony should be overcome. In wind turbine technology, 90% of components could be recycled, although the huge size makes transportation costs prohibitive for long-distance hauls. Energy storage systems have the opportunity for all metals from a battery to be recycled. Nevertheless, increasing the economic profitability and economic efficiency of the recycling process can be challenging, due to fluctuating material values.

Raw materials recovery is essential, although recovering these materials and reintroducing them into production cycles present some challenges. The first is the processing obstacles due to the use of composite materials, the presence of hazardous substances, and low concentrations of valuable/scarce elements. Furthermore, the devices and systems are typically not designed to facilitate end-of-life/recyclability aspects. Nowadays, market conditions do not properly price using virgin materials vs. recycled ones, associated with logistical issues due to the remote locations, size, and safety requirements of energy infrastructure. The inclusion of materials with high recycling characteristics and less harmful to nature must be the way to overcome such obstacles. The standardization of devices, materials, and processing will simplify the later stages of recycling allowing the interconnection of raw materials to different applications.

Efforts to overcome these barriers are being implemented by different governments/authorities in different countries of the world. Sustainability is ruled by three main dimensions: economic, social, and environmental. These three dimensions are strongly interlinked and equally interdependent. It is important to keep the balance between these dimensions in order to maintain, for example, economic growth without compromising the natural environment, resources, and community health. Life cycle costing, social life cycle assessment, and life cycle assessment are necessary tools for a better assessment of these dimensions.

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