Journal of
Composites Science

MDPI

Review

Towpreg—An Advanced Composite Material with a Potential for Pressurized Hydrogen Storage Vessels

Anka Trajkovska Petkoska [1,2,*], Blagoja Samakoski [3,4], Bisera Samardjioska Azmanoska [3] and Viktorija Velkovska [3]

[1] Faculty of Technology and Technical Sciences, St. Kliment Ohridski University-Bitola, Dimitar Vlahov, 1400 Veles, North Macedonia
[2] Department of Materials Science and Engineering, Korea University, 145 Anam-no, Seongbuk-gu, Seoul 02841, Republic of Korea
[3] Mikrosam DOO, Krushevski Pat bb, 7500 Prilep, North Macedonia; blagojas@gmail.com (B.S.); biseras@mikrosam.com (B.S.A.); viktorijav@mikrosam.com (V.V.)
[4] Institutte for Advanced Composites and Robotics, Krushevski Pat bb, 7500 Prilep, North Macedonia
* Correspondence: anka.trajkovska@uklo.edu.mk or ankatp@korea.ac.kr

Abstract: Hydrogen is one of the critical components to address global challenges such as climate change, environmental pollution and global warming. It is a renewable source of energy that has many advantages compared to other renewables. Even though it may not be a "silver bullet" solution for the polluted world, there is still a big expectation that it can solve some of the energy crisis and challenges in the transportation, domestic and industry sectors. This study reviews the latest advancements in materials science, especially in the composite materials used for energy storage/transportation tanks. Special attention is given to towpreg material structures as the most promising ones for hydrogen storage. Various types of storage vessels are reviewed with emphasis on the most advanced type IV and type V vessels for energy (hydrogen) storage. The manufacturing processes, mainly filament winding (FW) and automatic fiber placement (AFP), are reviewed with their pros and cons. The sustainability aspects for the most promising hydrogen technologies, limitations and future challenges are also discussed.

Keywords: towpreg material; hydrogen; pressurized storage vessels; filament winding; automatic fiber placement; sustainability aspects

check for updates

Citation: Trajkovska Petkoska, A.; Samakoski, B.; Samardjioska Azmanoska, B.; Velkovska, V. Towpreg—An Advanced Composite Material with a Potential for Pressurized Hydrogen Storage Vessels. *J. Compos. Sci.* **2024**, *8*, 374. https://doi.org/10.3390/jcs8090374

Academic Editor: Hu Liu

Received: 13 August 2024
Revised: 10 September 2024
Accepted: 18 September 2024
Published: 21 September 2024

Copyright: © 2024 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

Efforts to develop clean energy technologies using advanced materials, such as novel composite materials with enhanced properties, reduced costs and increased sustainability, have been intensified in the past few decades. Namely, advanced materials with enhanced performance are essential for the development of renewable energy technologies and low (to zero) carbon energy solutions with high efficiencies in energy storage and management. This is in the context of novel approaches toward preservation of the environment and reaching carbon neutrality by 2050. There is an urgent need for novel high-performing materials to support efficient energy production, storage and distribution; the interplay of advanced materials' technologies and green energy alternatives fosters innovation in industries and academia and consequently contributes toward enhancing the EU and national competitiveness and growth [1]. It is also in line with the Materials 2030 Manifesto:

> "*A strong European Materials ecosystem drives the green and digital transition as well as a sustainable inclusive European society through a systemic collaboration of upstream developers, downstream users and citizens and all stakeholders in between*" [2]

Today, we live in an era of materials' revolution—an era of advanced materials that removes the boundaries of the known conventional materials, offering solutions to global

challenges; we witness daily innovation, leading to materials that are stronger than steel, but lighter than aluminum—materials that offer new possibilities in design, engineering, and sustainability. This could be the era of composite revolution. However, composite materials are not new—they have been around for thousands of years—but through the centuries, they have been developed and advanced by the use of a variety of engineering designs and tools. Composite materials are in high demand owing to their advantages over traditional materials, including flexibility of design, enhanced mechanical properties, and less susceptibility to weathering, among others. Due to their excellent mechanical and electrical properties as well as being low density and lightweight, composites are building blocks for a variety of technical products in many industrial applications, from cars to spacecraft, reservoirs for gasses or liquids, sporting goods, civil engineering structures, armor plates, etc. However, they can show poor resistance to damage if the properties of the individual elements in the composite are unmatching, poorly manufactured and/or are not in a functional relationship. In such cases, for example, microcracks and delamination in the composite could appear and grow until the part loses its properties and eventually breaks [3–6].

In terms of green and clean energy sources needed for future exploitation, many of the renewables are intensively discussed, and all fingers point to 2030/2050 targets for carbon neutrality and how to shorten the time to decarbonization. One of the attractive renewable energy sources is hydrogen—it has attracted renown and rapidly growing attention around the world—considered by many governments, important stakeholders and policy makers worldwide because there is a strong belief that hydrogen has immense potential to develop a cleaner sustainable world. If used from now through 2050, hydrogen can avoid 80 gigatons (GT) of cumulative CO_2 emissions [7–12].

In this context, Europe also reserves a special place in the hydrogen partition in the next projects; specifically, an extract of the newest Hydrogen Manifesto declares the following:

> "*Hydrogen is one of the vectors of this necessary change contributing with 15–20% of the final energy demand by 2050 (as recently presented by the European Commission in the 2040 Climate strategy). It can transform industries and drastically reduce their CO_2 emissions. It can revolutionise air, maritime and road transportation. It can store variable renewable electricity and bring stability and flexibility to an electricity grid transporting ever increasing quantities of energy. And it can help with heating and cooling buildings without carbon emissions. . .*"

Europe has tremendous potential for hydrogen [13]. The Future of Hydrogen report (published by the International Energy Agency) urged that international hydrogen trade needs to start soon to make an impact on the global energy system [9]. Therefore, many are trying to solve the problem of the energy crisis with novel types of renewable energy sources, such as hydrogen, a fuel of the future, but at the same time, besides the concept of its production, many are trying to solve the challenges related to storage, transport and distribution of the fuel to those who need it.

This study reviews the novel trends and the latest accomplishments in the science of advanced composite materials intended for energy storage and, more particularly, towpreg materials for high-pressure vessels for fuel storage. Therefore, an overview of towpreg materials will be presented, along with their properties and applications. A discussion of hydrogen as a potential future green fuel and its beneficial properties compared to other energy sources is also covered by the study, as well as the storage of it. In addition, sustainability aspects and the limitations of hydrogen technology are also presented. It is our belief that the study could also initiate novel ideas and research to overcome the present challenges and to trace new routes to better utilization of materials and resources—all for better planetary health and human wellbeing.

2. Advanced Composite Materials: The Case of Towpreg

2.1. Basics of Composite Materials—The Building Blocks

Composite materials have many advantages over conventional materials, such as metals, alloys, polymers and ceramics. The main advantages of composite materials are their high strength-to-weight ratio, enhanced anisotropic properties (if needed), fatigue and corrosion resistance, efficient vibration damping, high fracture toughness, design flexibility and composite part integration, among others. On the other hand, composites have some limitations, such as the potential for raw materials to be costly, complex failure modes, their environmental degradation and issues with recycling, etc. [3]. In general, composite materials represent compositions of two or more immiscible materials combined together at a macroscopic level, with the qualities of each constituent complementing those of the other constituent(s) to form a composition with improved properties that overcome those of the single component, while still retaining their own chemical identities. In most of the cases, constituents play synergistic roles in the composite material and hence contribute toward the enhanced composite's performance in a variety of applications.

The building blocks of polymer-based composites are polymer matrix and reinforcement components. For industrial and technical applications, synthetic or natural fibers are being used as reinforcements, among which carbon, glass, aramid and others in the form of continuous or discontinuous fibers, aligned or randomly distributed in the polymer matrix, are often utilized [5,6].

Carbon fibers are the most used ones; they have very good mechanical characteristics while having low density and excellent resistance to high temperatures. They are able to retain their mechanical characteristics up to temperatures of ~1500 °C in a non-oxidizing atmosphere. According to Bhardwaj (1992), carbon fiber reinforced composite materials are lighter than aluminum and stronger than steel. Apart from having the highest specific modulus and specific strength, carbon fibers have high thermal resistance, excellent electrical and thermal conductivity, low co-efficient of friction and thermal expansion, vibration damping and many other useful characteristics. They are usually supplied in the form of *tows*, commercially available in a continuous form as "*bundles*" of filaments, which widely vary in the number of filaments per tow. In addition, they can be pre-impregnated with a resin matrix during their production to obtain *towpregs*, consisting of thousands of fibers/filaments embedded in a continuous matrix, in order to promote wetting and adhesion [14–16].

The matrix, the fibers and the fiber–matrix interface are directly related to the final properties of the composite materials and, in particular, to their mechanical properties. The reinforcement is embedded in the *resin matrix* that penetrates between the fibers, isolating them from each other, and, as a result, the individual fibers can act separately, i.e., transfer the mechanical forces from fiber to fiber; therefore, they help to stop or slow the propagation of a crack. The rigidity of a composite is ensured mainly by the fibers, which have better mechanical characteristics than the polymer matrix; therefore, the mechanical strength and stiffness of the composite are mainly contributed by them. On the other hand, the matrix has a role of transferring the load to the fibers, protecting them against environmental stresses, and defines the final shape of the structure. However, it needs to be taken into account that the mechanical and other characteristics of composite products depend not only on the building blocks of the composite, but also on the manufacturing process, the process conditions used, their geometry, etc. Several manufacturing methods for composite materials are known, and the frequently used ones are hand layup, vacuum molding, resin transfer molding, pultrusion, etc. [5,14,17]. For the hollow structures, such as pipes and pressure vessels, the most preferred manufacturing methods are filament winding and automated fiber placement [3,18–20]. The selection of manufacturing processes for composite products depends on the material components used, the product design, the cost and the end-use application.

Two types of resins, *thermoset* and *thermoplastic*, are usually used in composites based on polymers. The most commonly used is a thermoset resin, which once cured cannot be re-heated and re-shaped, while the thermoplastic resin is gaining more interest, as it can be reheated and reshaped multiple times. Moreover, thermoplastic composites can be recycled, thus having more environmentally friendly profiles.

Among the resins, the epoxy resins are the most used representatives of thermoset resins for development of advanced composites due to their excellent properties—high electrical insulation, superior heat and chemical resistance, high mechanical resistance, low cost, low toxicity, good dimensional stability and ease of manufacturing. On the other hand, polyester resins are low cost and chemically resistant and provide lower strength/stiffness, while vinyl esters are corrosion resistant and have intermediate cost and performance [3,5,21].

Thermoplastic composites manufactured by additive manufacturing and automated fiber placement are gaining more popularity in recent years because of the material system's inherent advantages over thermosets. Thermoplastic polymers present a formidable alternative to the traditional matrix materials due to shorter processing cycles, easier handling, indefinite shelf life at room temperature and superior energy absorption characteristics. However, the high melt viscosity of these materials remains an obstacle to obtaining void-free high-quality composites with similar production methods employed for thermoset matrices. The high viscosity of these resins protects the impregnation of the resin into fiber preforms and releases the air/voids from the fiber bundles; therefore, the thoroughly impregnated, well-consolidated, void-free composite structures are hard to achieve [5]. On the other hand, the high material costs and limited production flexibility are hindering more widespread use of thermoplastics. They are used for highly engineered parts and structures where performance matters, such as in the aerospace, automotive, construction, infrastructure, marine, defense and materials handling industries. Growth in the use of thermoplastic resins compared to thermosets is attributable to favorable mechanical and physical properties, such as higher impact resistance and damage tolerance, malleability at increased temperatures allowing them to be reshaped, better recyclability and reparability options, and higher specific mechanical properties. The real benefit of thermoplastics for structural applications is attributed to the ones with continuous fibers due to the order-of-magnitude higher strength and stiffness and more tailorable properties as compared to engineering composites. There are a variety of conventional processes that are used to manufacture thermoplastic parts, including automated fiber placement (AFP), automated tape layup (ATL), compression molding, pultrusion, filament winding (FW) or injection molding with short fibers [22,23].

2.2. Towpreg—Material Structure with a Lot of Potential

Towpregs are continuous fiber bundles (*tows*) pre-impregnated with a resin. After partial curing, these fiber bundles could be stored and used later on in many composite manufacturing techniques, such as fiber placement processes, filament winding, and so on. When the process of impregnation is controlled, towpreg rovings contain a homogenous fiber/resin ratio—therefore, they can provide composite final products with targeted and uniform properties. In addition, towpregs have the potential to solve issues related to reproducibility and homogeneity usually occurring in the traditional wet winding process [19]. Therefore, towpreg material is a continuous prepreg composite with a particularly high filament ratio, making it a good choice for diverse applications for automotive parts, cylindrical components such as pressure storage vessels, aircraft wings, etc., mostly in applications, which require robust mechanical performance at low cost by use of numerous manufacturing processes including pultrusion, automated tape placement, filament winding, etc. Towpreg is consider as a potential competitor to the traditional wet winding processes [24,25].

In most of the technical applications, tows are impregnated with a thermoset or a thermoplastic resin and produce towpreg, which in fact means tows that have been impregnated with resin and consolidated but that are still uncured and can be cured later on during the final processing step. It is a high-performance material used in manufacturing of high-end products in industries such as aerospace, automotive, sports equipment and so on.

Thermoset resins, in most of the cases, dominate the thermoplastics in towpreg production; more particularly, the epoxy resins are the most considered for making pressurized vessels. Thermoplastic materials are difficult to manufacture due to their higher molecular weights and higher viscosities, consequently making the impregnation process more difficult than that with thermosets. However, a lot of efforts have been made to overcome the challenges and successfully bring the thermoplastics to the stage of their useful application in the towpreg production. For this purpose, the manufacturing of the towpreg was modified by replacing the in-process impregnation of the fibers with a separate preparation of pre-impregnated fibers with a unidirectional arrangement. This enables the use of a semi-product that is characterized by high-fiber and low-void content. Moreover, such products are already available on the market, like those composed of carbon fibers/PEEK, carbon/PS, or carbon/PA-6. The trend of switching to thermoplastic resins is also due to the global trends for greener materials that could provide use of their waste (e.g., upcycling) or recycling possibilities. In this context, thermoplastics are preferred over the thermoset materials because there are already known recycling schemes for them—mechanical, chemical and thermal recycling options [21].

Technical fibers like carbon, glass and aramid are prevalent in towpregs; carbon fiber towpregs are the most used ones for pressure storage reservoirs. More particularly, carbon fibers are attractive due to their mechanical properties like tensile strength, outstanding fatigue performance and resistance to creep, as well as chemical resistance to most of the chemicals (the main properties are presented in Table 1). However, in most of the cases, they are main cost drivers in the processes of composite materials and especially in high-pressurized vessels. Many of the other types of fibers could replace the carbon fibers in certain applications, for instance, glass fibers for compressed gas vessels that operate at lower pressures or an outer layer in hydrogen high-pressure vessels as a protective layer against galvanic corrosion and damage resistance. On the other hand, basalt fibers are an eco-friendly alternative to other types of fibers, especially when they are compared to glass fibers—they are cost-efficient and possess better mechanical properties as well as improved thermo-chemical stability. However, compared to carbon fibers, basalt fibers are less attractive in terms of their mechanical properties but are a much more cost-effective alternative. They could find applications for compressed natural gas (CNG) storage. In addition, ultra thin steel wires have been suggested as a replacement of carbon fibers; however, this option adds weight more than any other of the previously mentioned fibers. It is still possible for this option to find application for stationary storage facilities and fueling stations, for example [21]. The trend of using carbon fiber towpreg over the traditional wet production method has grown in recent years, and many companies, like TCR Composites (Ogden, UT, USA), JXTG Nippon Oil & Energy Corp (Tokyo, Japan), Kümpers GmbH (Rheine, Germany), SGL Carbon SE (Wiesbaden, Germany) and Teijin Carbon (Tokyo, Japan) have adopted this trend. Towpregs from different manufacturers can vary in their resin content; the content mainly depends on their final applications [25].

Table 1. Characteristics of carbon (C-) fibers mostly used in towpreg process.

C-Fibers	Tensile Strength [GPa]	Young's Modulus [GPa]	Elongation at Break [%]	Ref.
T700	4.90	230	1.8	
M40	3.65	230	1.4	
IM7	4.50	275	1.5	[26]
T800	4.60	240	1.6	
T800S	4.95	230	2.0	
Case study:				
C-fiber/epoxy composite	**Tensile strength** 4.4/2.4 GPa	**Young's modulus** 250/170 GPa	**Elongation at break [%]** 1.65/n.d.	[18]
	Resin mass context 35%	**Towpreg bandwidth** 6.4 mm	**ILSS** 75 MPa	**Tex** 1600 g/1000 m

Note: Epoxy pre-impregnated carbon (C-) fiber towpreg material produced by SGL Carbon Inc. (Wiesbaden, Germany) with the trade name of SIGRAPREG C [TP24/6-5.0/270-E910/35%] is used in ref. [18].

2.2.1. Manufacturing of Towpreg

(a) *Wet Process*

The production process of towpregs involves resin impregnation systems based on the open bath with drum tray or dip tray principle. Fibers run down into and out of the resin bath. In the drum system, the tows are compacted on a roller on which there is a resin film controlled by a scraper. In the dip-type units, the tow is guided below the resin level and passes over a series of pins to complete the impregnation. After the impregnation process, the resin-impregnated fibers are subsequently wound on a mandrel [16].

The open bath impregnation process has some limitations, which affect the quality of the towpreg and the process productivity, such as poor quality of the tow impregnation due to the resin viscosity fluctuations over time and due to the potential temperature or moisture variations, as well as variable fiber/resin weight ratio and poor geometric and dimensional control of the towpreg cross section. In this process, the organic solvent may remain trapped in the towpreg material, and voids may be generated that deteriorate the final properties of the composite material. The equipment cost increases due to the step of drying the organic solvent, which is not the case in the dry process. In addition, the limitations of the wet impregnation techniques are extensive time and labor consumption, which lead to high costs and sometimes difficulty achieving the desired quality standards [16,19,27].

(b) *Dry Process*

Significant efforts are being made to replace the impregnation open bath with new concepts, such as hot melt and solution-dip processes. In particular, the hot melt prepreg process utilizes heat and pressure to transfer the resin film from the carrier paper into the fiber bundle and then impregnate the fibers to produce the towpreg. On the other hand, the solution-dip systems use a solvent to dissolve the resin, reduce the viscosity of the system and improve its wetting characteristics; after the soaking of fibers in the solution, the solvent is evaporated away, and the resin is partially polymerized, leaving only resin and fiber in the final towpreg.

In most of the cases, the tow of the fibers is pressed flat (with rollers) and passes through a resin bath; by adjusting the pressure, one can control the wetting of the fibers as well as the resin content in the tow. At the same time, the temperature plays an important role for the resin's viscosity—by adjusting the resin temperature, the wetting of the fibers and the resin content on the tow can be controlled as well, but very low resin viscosity could be difficult to manage due to proper resin intake on the tow and the possibility to start earlier with the curing process with the towpreg at elevated temperatures. After a good wetting of the tows in the resin, the towpreg goes to a partial heating stage known as B-stage, which in general reduces the tackiness level of the semi-product. At this B-stage, the degree of cure of the resin in the towpreg is ~30% (100% is a fully cured resin), and it should be stored properly, because the process of curing has started and by managing the storing temperature, one can manage the duration of storage. Namely, by storing at

sub-zero temperatures, the shelf life of towpregs could be extended to several months, but besides the extended curing time/shelf life of towpregs, the curing process also decreases tackiness, which allows the towpreg to be stored on spools. Due to the partially cured resin systems, towpregs require shorter final curing periods. In case the tackiness causes problems for winding/unwinding operations, the towpreg could be placed on release film or paper that will protect the sticking effect between towpreg layers; but this operation involves an extra paper material that later causes additional waste [18,25].

On the other hand, the tackiness of towpreg results in better adhesion between the bundle and mandrel surface, which reduces slippage possibility, and, therefore, complex-shaped structures with complicated patterns or low winding angles become possible to manufacture.

Dry processing of towpregs offers many advantages over the wet production method, including improved consistency, more efficient manufacturing and reduced environmental impact; Table 2 summarizes the respective advantages [14,21,25,28]. In addition, increased produced part quality and homogeneity have been observed, and process durations have been significantly reduced. as the resin preparation, resin bath adjustment and its cleaning steps are excluded [18,19]. Automated fiber impregnation technology as a dry process is a good example—it ensures the repeatability of the process, the dimensional stability of towpregs and the accuracy of fiber impregnation [16]. Moreover, another advantage of dry towpregs compared to the wet filament winding technique is that the load transfer mechanism from the matrix to the fibers is more effective in the dry process; in other words, the degree of the effective fiber strength in the composite structure is higher (this is known as "*fiber strength translation ratio*"). This is a significant advantage, especially for the structures where weight saving is critical.

Table 2. Characteristics of wet and dry processes of towpregs.

	Wet Process	**Dry Process**
Advantages	Better set-up and clean-up time Higher laminate quality (with lower weight) Lower production pollution Winding speed does not depend on fiber wetour requirements Cost effective (for epoxy-carbon composites) Curing process is easy	Dimensional stability Determined fiber/resin ratio Improved consistency Efficient manufacturing/easy handling Reduced environmental impact Increased produced part quality/homogeneity Resin bath adjustment/resin bath cleaning steps are excluded Fiber strength translation ratio Better labor health and safety
Disadvantages	Desired quality standards are difficult to achieve Labor efforts Resin dripping/accumulation Time consumption Less control over resin content variability Poor external finish	High viscosity usually requiring high processing temperatures High material costs Lower winding speeds

2.2.2. Properties of Carbon Fiber Towpreg

The demand for lightweight, strong and high-performance structures and products continues to be a top priority. Carbon fiber towpreg is well-positioned to meet most of these requirements; therefore, many industries are adopting the use of carbon-fiber towpregs in the manufacturing processes in the race for products with advanced performance. Some of the most impressive characteristics of carbon-fiber towpreg materials are summarized as follows:

- *Exceptional strength-to-weight ratio*—towpreg weighs significantly less, e.g., than steels; it is stronger than steel, making it ideal for applications that require high strength and low weight.
- *Controlled consistency of pre-impregnation of fibers with resin*—it leads to consistent results; therefore, the final product can easily meet the required specifications.
- *Improved manufacturing efficiency*—the pre-impregnated fibers with resin reduce the time and effort (usually required to apply the resin to the fibers during the final product manufacturing).
- *Minimal waste production*—compared to the traditional (wet) production method.
- *Environmental consciousness*—the controlled application of resin to the fibers results in minimal waste, consequently lowering the environmental impact during the manufacturing process.
- *Reduced fuel consumption*—the lighter final products can reduce fuel consumption during their transportation; therefore, less pollution and better sustainable aspects are envisioned.

2.2.3. Applications of Towpreg Materials

The potential applications of towpregs are numerous. They can be suitable to produce tanks for storage, transport or distribution of hydrogen, pressure vessels, dynamically loaded components like springs, shafts, pipes, various sporting goods and goods for the energy sector, etc. (some of these applications are given in Figure 1). As the technology continues to overcome the current barriers and meets future challenges, it is likely that even more advanced and innovative applications of carbon fiber towpreg will be seen in the near future. More particularly, carbon fiber towpreg, as a high-performance material, has revolutionized the manufacturing industry, providing high-tech performance and lightweight and durable material that can be used in a wide range of applications—in the aerospace, maritime, automotive, sporting goods and other sectors.



Figure 1. Various applications of towpreg composite materials.

One of the most targeted applications of carbon fiber towpreg is for high pressure automotive reservoirs (explained in more detail in Section 3), as well as in the aerospace industry for high-performance aircraft components. The lightweight durable carbon fiber towpreg makes it an ideal material for weight reduction of the aircraft and improved fuel efficiency. In addition, carbon towpreg can provide excellent resistance to extreme temperatures and environmental conditions, making it an ideal material for space applications as well.

Another significant application of carbon fiber towpreg is in the automotive industry, where it is again used to manufacture lightweight, high-strength components like body panels, suspension systems and engine parts; these components offer significant weight savings, which can improve fuel efficiency and reduce emissions of pollutants. Moreover, carbon fiber towpreg-based parts can help improve safety by providing superior crash resistance and impact protection. In the sporting goods industry, to manufacture lightweight, high-performance equipment like bicycle frames, tennis rackets and golf clubs, carbon fiber towpreg is also highly appreciated. It has led to significant improvements in athletic performance, allowing athletes to perform better by using light-weight sporting goods, for example.

3. Pressurized Vessels for Gas and Liquid Storage

Pressure vessels (or pressurized tanks) are vessels designed for storage of gas or liquid under high pressure. In general, carbon fiber composite pressure vessels are used to hold liquids and gases at very high pressures when low weight is a priority, like in automobile or aerospace applications. There are five types of pressure vessels, of which three of them use (carbon) fiber composite materials: type III pressure vessels have a metal liner, with carbon fiber wrapped around to hold the pressure, while type IV (with polymer liner) and type V (without liner) are all-composite types of tanks. Due to the impressive weight savings, many metallic pressure vessels have been replaced with filament wound composite pressure vessels like types IV and V. (More details will be provided in Section 3.1.) [29,30].

The *liner* is the core element of the pressurized vessel (except for type V, which is linerless). The liner acts as a physical barrier between the content/fluid and the overwrap; it should have low permeability parameters that suppress leaking of the stored content. Therefore, the liner materials should be properly chosen—to be safe, cost-effective and chemically stable in order to not react with the stored fluid inside. The design of the liner depends on many parameters, e.g., the application, the size and the mass of the reservoirs, among others. In the currently used pressurized vessels, liners are made from metals (steel or aluminum alloys) and plastics (polyamide, high density polyethylene). In general, plastic liners are more permeable than metallic ones, so their use in the hydrogen pressurized vessels is becoming questionable [21].

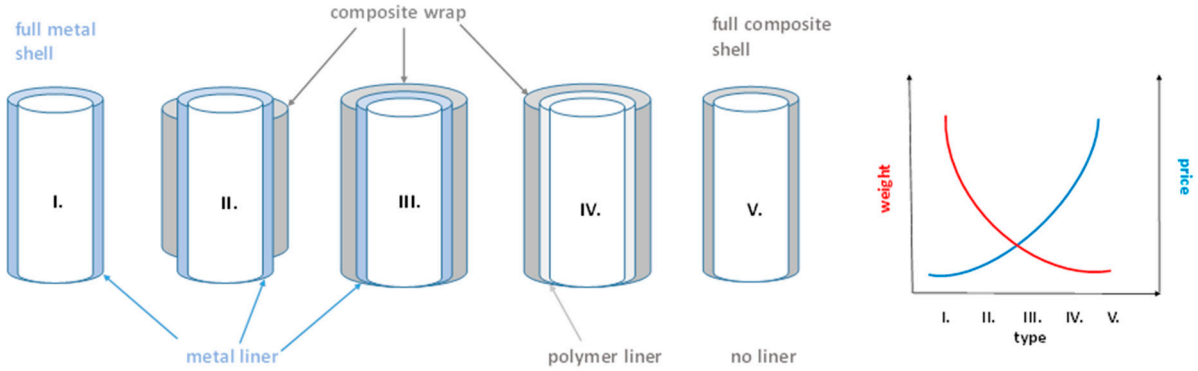
3.1. Types of Pressure Vessels

There are five types of pressure vessels that are known today (presented in Figure 2) [19,20]:

i. Type I is full metallic pressure tanks;
ii. Type II contains metallic liner material and composite hoop layers;
iii. Type III describes tanks with metallic liners and fully composite overwrapped layers;
iv. Type IV vessels consist of a polymer-based liner and composite overwrapped shell;
v. Type V represents composite overwrapped vessels without any liner.

The most frequently used composite vessel type for pressurized storage of gasses is type IV, which contains a polymer liner to act as a non-permeable barrier to prevent leakage and a composite overwrapped shell that carries internal pressure and all other loads. Moreover, for high-pressure gaseous hydrogen storage, the last two types (IV and V) are generally considered.

Type I vessels are the simplest to design and manufacture and make up most of the vessels in use today. They are all-metallic vessels. Types II and III vessels are known as *composite overwrapped pressure vessels* (COPVs) and often utilize carbon fiber to withstand part or all of the structural load and a metal liner. Type IV vessels contain plastic liners for gas containment. Many ground transport applications use type IV vessels, as they offer the lightest weight and highest storage densities. *Composite pressure vessels* (CPVs) can affect fuel gas tank weight savings up to 75% compared to metallic vessels, for example. Namely, it has been estimated that a 1 kg weight drop saves ~2900 L of fuel per year compared to metallic vessels, or another study presents weight and cost reductions of 25% and 30%, respectively, compared to a traditional aluminum-lithium tank [20,31,32].



(**a**) (**b**)

Figure 2. (**a**) Classification of pressure vessels with different shell/liner configurations. (**b**) Weight and price aspects vs. vessel type.

In continuation, types IV and V pressure vessels will be elaborated on in more details due to their importance in hydrogen storage technology.

Type IV and Type V Pressure Vessels

In general, type IV COPVs are based on a thermoplastic inner tank (liner) reaching the necessary pressure stability by fiber reinforcement using the filament winding process. They are characterized by their lightness and safety. The state of the art in composite pressure vessel technology is known as the type V—liner-less vessel, which does not have an internal liner, so the composite acts as both the gas barrier and load-bearing structure. As the composite is not overwrapped on a liner in type V vessels, they can be referred to as CPVs. In most of the cases, they rely on the carbon fibers and the resin to fully seal the tank; therefore, these CPVs are among several pressure vessel solutions in consideration for high-pressure hydrogen storage, particularly in vehicles where the weight savings are considerable [20,33].

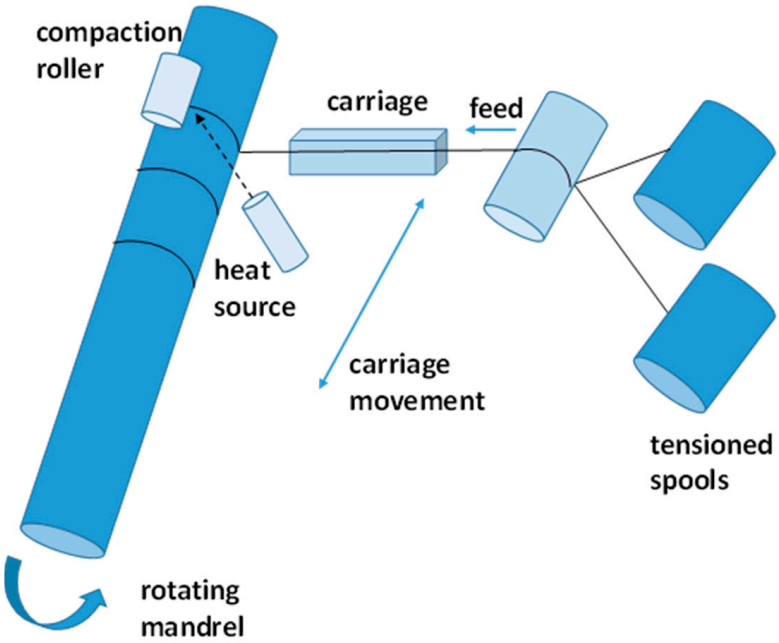
Development of type V vessels began in the 1980s in the aerospace industry, but later on, many research groups in other application fields reached the maturity level of development of type V tanks for commercial applications. However, the potential operating pressures of type V tanks have still not yet been fully explored for wide exploitation [20].

COPVs/CPVs are usually manufactured using filament winding (FW), a technique that winds tensioned fiber bands around a rotating mandrel. This technique is capable and mature, but it still has several inherent limitations that could be overcome by automated fiber placement (AFP). Namely, the design of COPVs/CPVs can be improved with the use of more advanced manufacturing methods like AFP, which is an evolution of the traditional filament winding technique that provides high accuracy, better quality and less waste. Both the type V design and AFP technology have high potential for increasing the gravimetric and volumetric density of hydrogen gas storage; however, several engineering challenges must be overcome before they can be adopted commercially. Type V technology is a state-of-the-art technology that offers a reliable alternative to type IV tanks [20].

3.2. Manufacturing of COPV/CPVs

3.2.1. Filament Winding (FW)

Filament winding is a technique primarily used to manufacture hollow, circular parts such as pipes and tanks. It is performed by winding continuous fiber tows onto a rotating mandrel using a specialized winding machine (Figure 3) [18,19]. FW is a particularly suitable technology for producing axially asymmetrical shapes. Filament wound parts are commonly used in the aerospace, energy and consumer product industries [20,28,32,34].



(**a**)



(**b**)

Figure 3. (**a**) Principle of filament winding (FW) technology. (**b**) Case of FW equipment: process of pressurized vessels (Mikrosam, Prilep, N. Macedonia, https://www.mikrosam.com/, accessed 19 September 2024).

The composite overwrap functions as the backbone of the storage system and provides strength for pressurized gas storage. The composite thickness distribution, mandrel geometry, winding pattern, winding angle and manufacturing technique affect the load-bearing characteristics of the reinforcing fibers supported by proper load transfer in the resin medium. Unidirectional fiber reinforced composites are orthotropic materials that mainly bear loads in longitudinal directions. The winding angles, sequence of layers and individual layer thicknesses at different dome locations are arranged to target proper structural strength in all directions. The winding angles/layer sequence are predetermined based on designed pressure and layer thicknesses. The orientation of fibers and their location in the composite strongly allows shaping its strength for a given set of possible

loads, while the failure of the vessel depends on the material properties, the vessel design, the manufacturing and the loading conditions. Due to its fast speed, it is suitable for large-scale production with applications in several industries, providing products with high fiber content (e.g., 60% or more) and excellent mechanical properties [35]. To overcome many shortcomings of traditional wet filament winding (presented in Section 2.2.1, Table 2), significant progress in the "towpreg" technology today has been made. Namely, "dry filament winding," which uses towpregs is an alternative to wet technologies is novel and high-tech because towpregs are the materials that solve the reproducibility, reliability and homogeneity issues occurring in the traditional wet winding process [18].

Another significant advantage in the use of towpregs is "the load transfer mechanism from the matrix to the fibers," which is much more effective in the dry filament winding process; thus, the degree of effective fiber strength in the composite structure is significantly higher. This is simply named as the "fiber strength translation ratio." This higher translation ratio is a significant advantage, especially for the structures, where weight-saving is critical. Thus, it would be possible to obtain equal performance vessels by winding fewer materials. Then, in terms of handling, process speed and simplicity, cleaning and operators' health and safety, dry winding becomes very advantageous compared to the traditional wet winding. In this case, there are important parameters that should be taken into account for the final structures, like the winding tension, the laminate stacking sequence, and the winding time. For instance, it is known that the winding tension directly increases the performance of wet-wound pressure vessels, because increasing the winding tensions increases the carbon fiber content with lower possibility of void formation. It is also known that a strong relationship exists between the fiber volume fraction and the hoop strength of the composite pressure vessels. Thus, the use of dry filament winding via towpregs could significantly improve the vessel performance due to higher fiber content [18].

In summary, the benefits of the towpreg (dry) winding process could be expressed as follows:

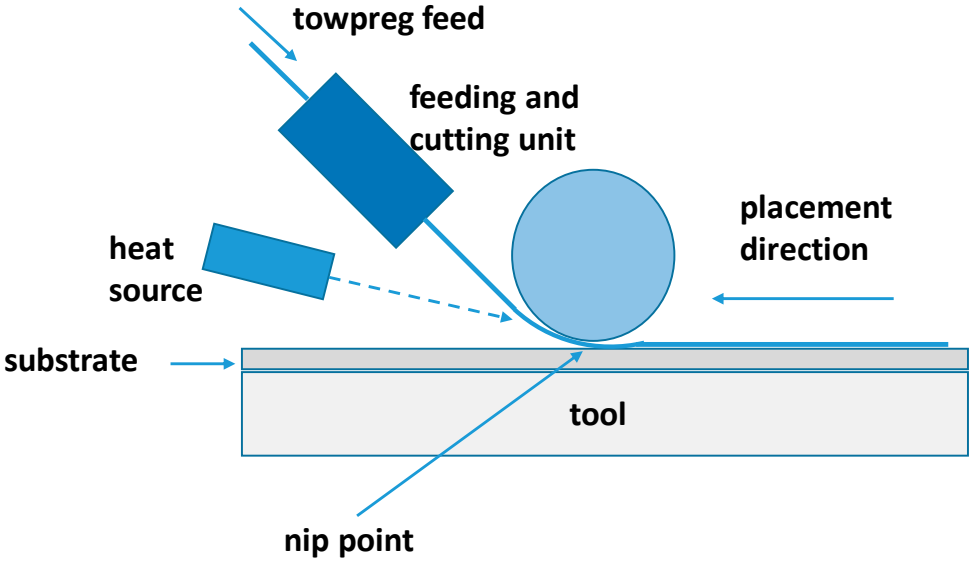
- The towpreg winding process is based on the use of industrialized raw materials that have, in most cases, previously passed rigorous quality control.
- Higher deposition rates and high production efficiency.
- Cleaner machine operations.
- Better work environment and operators' health/safety.
- The parts have precise resin content and greater uniformity.
- The parts in general have better mechanical performance.

In filament winding with thermoplastic tapes, complete in situ consolidation happens during the winding, and no oven is necessary. Therefore, thermoplastic-wound parts can be processed in a single production step. In addition, thermoplastic consolidation avoids fiber slippage; therefore, stability for different winding paths besides geodesic trajectories can be achieved [21].

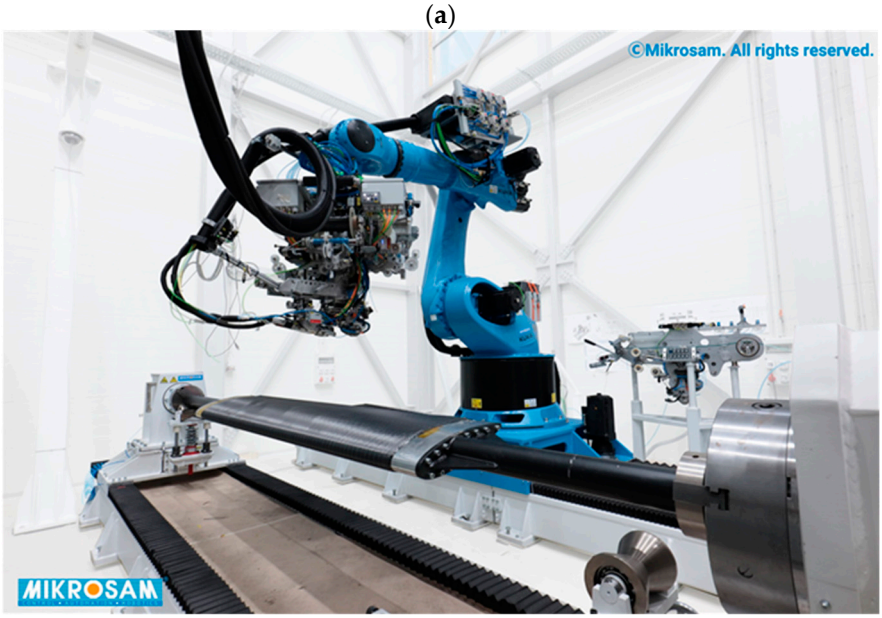
3.2.2. Automated Fiber Placement (AFP)

Techniques such as AFP use either pure rovings or already pre-impregnated towpreg materials. AFP is a highly flexible robotic technology for composite materials (Figure 4). Towpreg features higher filament numbers and is frequently used in automotive parts, which require particularly great robustness—large volumes of preform output while minimizing the risk of human error [4,20,36].

The main benefits of AFP for production of pressure vessels, among others, are variable angle tow and fiber steering and selective reinforcement; however, there are some challenges in applying AFP to pressure vessels that need to be solved, such as defect control and physical limitations.



(**a**)



(**b**)

Figure 4. (**a**) Automated fiber placement technology (AFP). (**b**) AFP equipment in process of customized part (Mikrosam, Prilep, N. Macedonia, https://www.mikrosam.com/product/automated-fiber-and-tape-placement/, accessed on 19 September 2024).

3.2.3. Production of Pressurized Vessel

The schematic presentation of the whole process of pressurized vessel production is presented in Figure 5.

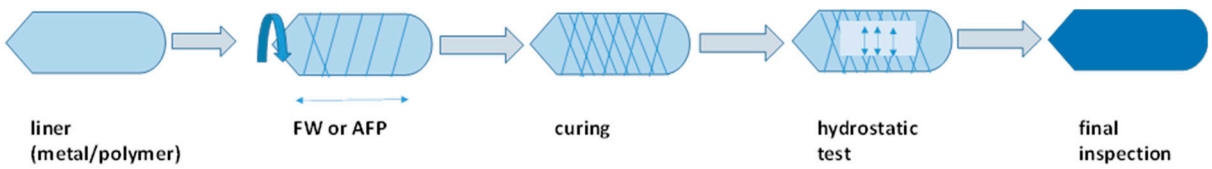


Figure 5. Schematic presentation of the process of production of high-pressurized vessel.

4. Hydrogen—Potentials, Challenges and Perspectives

4.1. Potential for Clean Energy

The global environment and human society should be in a harmonious relation with the Earth, where the global cycles of materials are of crucial importance in order to have a planetary balance and better human health and wellbeing for everyone. However, today we are witnessing that global warming, climate changes and urbanization are some of the reasons for a big imbalance on the planet—not just with depletion of energy resources and the energy crisis, but also with material cycle imbalance that is caused by improper utilization of materials. In this context, climate changes and environment pollution are two critical problems that can be resolved by use of clean or "green" energy such as hydrogen that can be produced and consumed without environmental consequences. Reducing carbon emissions is both urgent and hard to achieve, but hydrogen is a promising energy medium that could also be a solution for the energy crisis. As a clean energy source, it could be used in a number of industrial and economic sectors. Compared to non-renewable fossil fuels, hydrogen has the advantage of zero emission of greenhouse gases (GHGs); it retains a high calorific value, high reserve abundance and low production requirements when compared to other alternative energy systems.

In general, hydrogen can be used as a feedstock, an energy carrier and a storage in many applications across the industry, transport, power and buildings sectors. It does not emit CO_2 and causes almost no air pollution; therefore, it offers a solution to decarbonize industrial processes and economic sectors. Moreover, hydrogen is an energy vector, which can be used to convert, store and release energy, like electricity [37]. It could be an essential solution to contribute to the global effort of implementation of the Paris Agreement while working towards zero pollution and to support the EU's commitment to reach carbon neutrality by 2050 [10].

Currently, hydrogen use is very modest; it is not naturally available as ready-to-use fuel. Its production is still mainly dependent on fossil fuels (coal, natural gas), which means that for its production, huge amounts of CO_2 are still released; therefore, this type of hydrogen is declared as *gray* hydrogen. In order for hydrogen to contribute to the initiatives for climate neutrality, its production should be decarbonized—to be produced from sources like solar or wind energy or from hydro-sources; then, it will be declared as *green* hydrogen. There are many on-going new projects on developments for hydrogen production that could mean green and clean energy for many sectors of daily life [38–45].

4.2. Properties of Hydrogen

According to the H2 Manifesto,

> "*. . . Hydrogen is not a miracle molecule—but it can do what electricity can't and it is therefore complementary to a renewables-based energy system. Electricity and hydrogen are both urgent and vital prerequisites for the achievement of climate goals, for the retention of Europe's industrial base and competitiveness, and for European energy security. Without both in large quantities, there will be no transition away from fossil fuels and dependencies on third countries. Yet with both of them, Europe can position itself as the global lead, doing good not only for climate and economic sustainability, but also for European research, industrial development, and jobs. . .*" [13]

Hydrogen is the most abundant element around us; it forms 0.15% of the Earth's crust, and it is a major constituent of water; 14% (by weight) hydrogen is in the biomass as one of the components, while 0.5 ppm can be found in the air. Hydrogen is a non-toxic, colorless, odorless and tasteless gas with the highest energy density per mass compared to standard fuels. For example, its energy density is 120 MJ/kg compared to 44 MJ/kg for gasoline, while liquid hydrogen has an energy density of 8 MJ/L compared to 32 MJ/L gasoline. In other words, 1 kg of hydrogen is equivalent to 33.3 kWh, which is three times more energy than conventional fuel. However, low ambient temperature density of hydrogen results in a low energy per unit volume. This is a big challenge for finding a solution

for hydrogen storage and transportation that has the potential for higher energy density. On the other hand, hydrogen is a very efficient fuel because the mass of hydrogen for automotive applications is approx. 4 kg for 400 km transport range, or 3 min refueling time/500 km driving, and reduced weight is key, since every 10 kg removed from a vehicle usually means a reduction in 1 g/km CO_2 emissions from the vehicle on the road [9,46,47].

Other important properties of hydrogen include its diffusivity, which is higher compared to other gases due to the small size of its molecules; for example, hydrogen diffusivity is greater than that of helium and approx. three times the diffusivity of nitrogen in air at ambient conditions. The low viscosity of hydrogen and the small size of the molecule cause a comparatively high flow rate, if gas leaks through fittings, seals or porous material are present. Moreover, the heat capacity of H_2 gas is similar to that of other diatomic gases despite its low molecular mass, while the thermal conductivity of hydrogen is significantly higher than that of other gases. It is more soluble in organic solvents than in water. Although hydrogen is a non-corrosive element and does not react with the containers where it is stored, at certain conditions at elevated temperatures and pressures, it can diffuse into a metal lattice, causing an effect known as "hydrogen embrittlement" or hydrogen-assisted cracking. In the case of steel, for example, steel adsorbs hydrogen and becomes more brittle, which can cause failure.

At normal temperature and pressure (NTP), hydrogen is not a very reactive substance, but it is a combustible gas. Pure hydrogen burns with a pale-blue flame invisible in daylight. Hydrogen is not more (or less) dangerous than any other conventional fuel. On the other hand, hydrogen can be ignited easily, as its minimum ignition energy is 0.017 mJ, which is 10 times lower compared to other fuels. The main characteristics of hydrogen are given in Table 3.

Table 3. Properties of hydrogen.

Property	Value/Units
Molecular weight	2.016 g/mol
Density gas/liquid	0.089 g/L/0.07 g/mL
Color/odor/taste	Colorless/odorless/tasteless
Toxicity	None
Boiling point	20.27 K
Melting point	14.01 K
Triple point	13.8 K; 7.2 kPa
Critical point	33.25 K; 1.297 MPa
Diffusion coefficient	0.61×10^{-4} m^2/s
Viscosity (gas)	89.48 μP
Thermal conductivity (gas)	0.187 W/m.K
Specific Heat (gas)	14.85 kJ/kg.K (at NTP)
Flame temperature	2318 K
Liquid-to-gas expansion ratio	1:848

Hydrogen can be obtained by different methods, but those with low carbon routes or green hydrogen production are preferable nowadays, such as electrolysis of water (low- and high-temperature processes), methane splitting (pyrolysis), utilization of biowaste, etc. [8,48,49]. Namely, hydrogen can be extracted from various substances where it is in the form of a compound; it is a universal fuel in unlimited quantities across the planet. This offers potential for localized hydrogen production. It is the fuel of the future that provides reduced harmful gases and better safety aspects compared to other energy carriers.

In terms of the energy density properties of hydrogen, Table 4 provides basic data for different forms of hydrogen at certain conditions (pressures) [9,14,50].

Table 4. Basic energy density properties of hydrogen.

Form of H_2	Energy Density MJ/kg	MJ/m^3	Density kg/m^3
Hydrogen gas [0.1 MPa]	120	10	0.090
Hydrogen gas [20 MPa]	120	1900	15.90
Hydrogen gas [30 MPa]	120	2700	22.50
Hydrogen gas [liquid]	120	8700	71.90
Natural gas/liquid	50	/	0.65/423
Gasoline/liquid	45	/	4.40/700

4.3. Hydrogen Storage

There is no universal solution for storing hydrogen; the solution should be carefully selected to address specific system requirements. Many scientific studies are conducted in order to solve the challenge of improving the reliability and efficiency of hydrogen storage and transportation. Currently, there are various known ways to store hydrogen, as presented in Figure 6 [47,51–54]. However, other technologies also exist like H_2 storage underground [55] and large-scale storage options [56], but they are not the focus of this study and will be not elaborated on in detail.

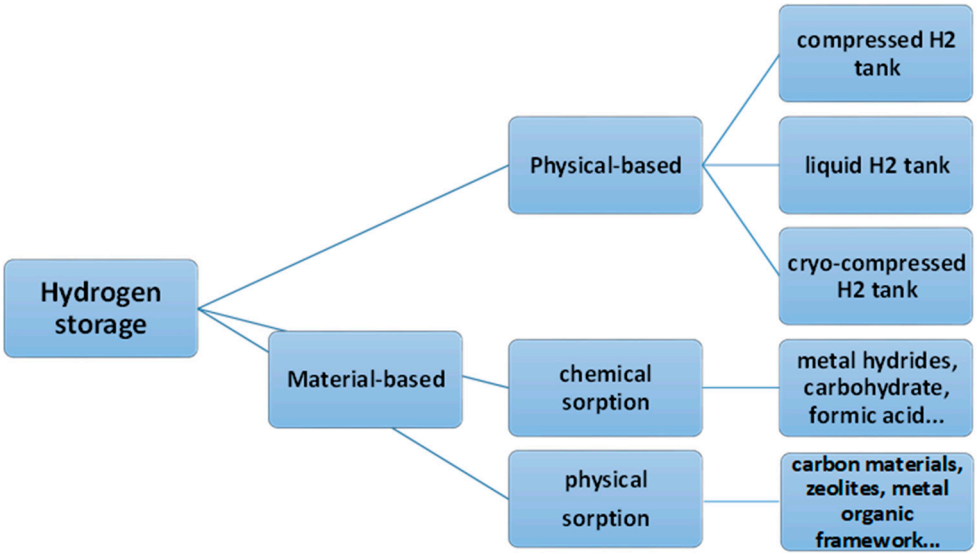


Figure 6. Storage options for hydrogen.

Hydrogen storage requires something more than a bucket; the main goal in hydrogen storage is to pack the hydrogen molecules as close as possible—to reach the highest volumetric density by using as little additional material as possible. Hydrogen storage implies the reduction of an enormous volume of hydrogen gas. At ambient temperature and atmospheric pressure (NTP), 1 kg of the gas has a volume of 11 m^3. To increase hydrogen density, work must either be applied to compress the gas, decrease the temperature below the critical temperature or reduce the repulsion by the interaction of hydrogen with another material [57,58].

Different options for hydrogen storage, both physical and chemical ways, are known (Figure 6). [9,42,46,59,60] Three physical-based techniques for onboard hydrogen storage in transport vehicles have been considered, i.e., storage in compressed gas (35 MPa or 70 MPa), storage as cryogenic liquid (20 K) and solid storage [47]. Due to their efficacy, high-pressure hydrogen storage vessels are the most promising and mature ones. Table 5 provides a comparative analysis of different modes of hydrogen storage along with their pros and cons [20,53,60–63].

Table 5. Characteristics of different modes of hydrogen storage.

Storage Method	Temperature [K]	Pressure [MPa]	Volumetric Capacity [kg H_2/m^3]	Pros	Cons
Compressed H_2 gas	room temperature	70	<40	Convenient/simple, mature technology, efficient method, short refilling time	Expensive storage tanks, high internal pressure/thick walls, low density/large size of tanks
Liquid H_2	20	0.1	70	Efficient method, high liquid density, short refilling time	Low temperatures, high energy/time consumption, evaporation/leakage of H_2, high-cost refrigeration unit is needed
Cryo-compressed H_2	20	30	40–70	Low boil off	High costs
Physical sorption	77	10	20	High porosity, high uptake of H_2, high specific area, reversible	Low storage temperature
Chemical sorption	room temperature	2	150	Large volume of H_2 density, high safety, good reversible cycle	Absorbing impurities, reducing H_2 capacity and tank's lifetime

4.3.1. Pressurized Vessels for H_2 Storage

Composite pressure vessel demands led to the development of more reliable storage systems with higher working pressure capability. There are many studies and on-going efforts for the optimization of composite pressure vessels. The strength variables of composite hydrogen storage vessels can be designed and optimized by changing stacking sequences of the laminate; thus, the manufacturing process and the used materials affect the final properties of the vessels the most. At the same time, the main requirements for the vessel are reduction of the high costs and weight of the product, as well as increase in the burst strength and load capacities [46]. Therefore, it remains a privilege and a prime challenge for future scientists to provide better versions of light-weight and cost-effective storage for hydrogen. Significant advances have been made, with composite vessels—type IV vessels—having exceptionally higher hydrogen storage density and lighter weight than previous generations of storage vessels, yet better storage options are needed [62]. Moreover, type V vessels could be an even better option if they overcome the challenges that the hydrogen storage technology faces at the moment.

4.3.2. Standard Policies for Hydrogen Storage and Production

Standardization plays a crucial role in the development of the hydrogen industry; it is essential for innovation, safety and integration into global markets. As hydrogen emerges as a clean energy source, setting the standards is paramount to guide its development and ensure compatibility with existing systems. International and national standardization bodies such as the following are central to this effort:

(1) ISO (e.g., "ISO/TC 197 Hydrogen technologies"; "ISO/TC 220 Cryogenic vessels"; "ISO/TC 58 Gas cylinders; "ISO/TC 22/SC 41 Gaseous fuels-specific issues") [64–67].
(2) IEC (International Electro-Technical Commission).
(3) CEN (European Committee for Standardization; "Hydrogen-CEN/CLC/TC 6"; "CEN/TC 23 Transportable gas cylinders"; "CEN/TC 69 Industrial valves"; "CEN/TC 185 Fasteners"; "CEN/TC 197 Pumps"; "CEN/TC 234 Gas infrastructure"; etc.) [68–73].

(4) European Clean Hydrogen Alliance's (ECHA).
(5) Economic Commission for Europe (ECE; ECE 146 (Hydrogen and Fuel Cell 2&3 wheelers), 134 (Hydrogen and Fuel Cell Vehicles) [74].
(6) EU 2021/535 (LHSS and CHSS, liquid and compressed storage systems, respectively) [75].
(7) Global Technical Regulations (GTR) like GTR13 (Hydrogen and Fuel Cell Vehicles), etc. [76].

Their work ensures that hydrogen technologies meet the safety, efficiency and interoperability standards that are essential for global acceptance and innovation [77–81]. However, sufficient regulatory frameworks for hydrogen technologies are still lacking across many countries and are still far from being harmonized. In fact, hydrogen displays a unique set of properties, which inhibit the extrapolation of existing standards from conventional fuels; this also includes its requirements for storage at high pressure and extremely low temperatures, distinctive thermodynamic properties, etc. [9].

Hydrogen storage equipment must be designed and maintained to high safety standards to ensure the integrity of the container. All types of vessels for hydrogen storage should be designed, manufactured, tested and maintained in accordance with relevant codes and standards at national and international levels. Briefly, some known standard tests related to hydrogen storage will be elaborated on here: bonfire, hydrostatic, ambient pressure cycling, penetration and leak-before-break tests. Namely, bonfire tests consider the fact that the tank shall not fail when exposed to a bonfire of 20 min duration.

In terms of hydrostatic tests, gaseous hydrogen vessels are subjected to suitable tests due to the elevated nominal working pressure (NWP) of 35 MPa to 70 MPa that they should hold with maximum filling and dispensing pressures of 125% of NWP. (NWP is a gauge pressure, which characterizes the typical operation of a system). However, in view of the applicable strict safety regulations, a minimum hydrostatic burst pressure (BP) of 225%, their NWP is prescribed for pressurized hydrogen vessels (international safety standard, ISO11439) [82]. Standards like ISO 16111 [83] and ISO/TC 197 [64] for gas storage devices have led to the development of safe, high-pressure containers, showcasing the scalability of hydrogen storage solutions [63,77,78,84–88]. More particularly, the pressure holding ability, or the burst pressure, is one of the most important concerns of a type IV vessel, which is mainly determined by the winding parameters and always related to the failure behaviors of the composite layers. Therefore, the design of the ultimate strength of the composite layers should be properly addressed in order to survive the strict conditions for these products [18,62]. For comparison purposes, type I vessels survive low- to medium-pressure storage, which is approx. 20–40 MPa, compared to type IV, which should hold up to 70 MPa [15,21].

For the ambient pressure cycling tests, hydrogen tanks shall not fail before reaching 11,250 fill cycles; this represents a 15-year life of use, for example, in commercial heavy-duty vehicles. Penetration tests refer to the tanks' rupture when an armor-piercing bullet/an impactor with a diameter of 7.62 mm or greater fully penetrates its wall, while the leak-before-fail test is the test of the tank to see if it will fail by leakage or exceed the number of filling cycles (11,250) [89].

In terms of permeation properties, according to the EU regulation, the permeation rate of hydrogen (at 20 °C) in cars should not exceed 6 NmL/h/L in order to avoid formation of flammable composition (in a worst credible scenario of a private garage with a ventilation rate 0.03 air-changes per hour). Moreover, hydrogen permeation through the polymeric liner can lead to its accumulation in the space between the liner and wrapped composite layers, forming a "blister" that can cause partial or full collapse of the liner when the pressure of the accumulated hydrogen becomes higher than the internal pressure of the liner [89].

The US Department of Energy (DOE) Hydrogen Program has specified an ultimate leakage target of 0.05 (g/h)/kg of stored hydrogen gas to help or guide researchers in their work towards minimizing permeability. This permeation is dominated by two phenomena:

(i) *diffusion* causes the pressurized gas to adsorb into the vessel wall, diffuse through the material and desorb into the atmosphere, and (ii) *interconnected* microcracking in the matrix opens "channels" for the gas to escape through [20,26]. Knowing this, it is an open call to work on and investigate these issues and be ready for better storage solutions for hydrogen fuel.

As a summary, hydrogen technologies became a key interest in many national strategies/roadmaps around the world. For example, a few years ago, the US DOE at second Hydrogen Energy Earthshot (2021) announced its "*Hydrogen Shot*" or "*111*," which means "*1 dollar for 1 kg clean hydrogen in 1 decade*," in order to unlock the market potential, or, in other words, by 2026, to reach the target of a cost of <2 dollars/kg H_2. This is an example that should be followed or overcome by others as well [90].

4.4. Trends in Development of Novel Vessel Types

Type IV pressure vessel technology is mature and actively used in commercial applications. The current state-of-the-art pressure vessel technology known as type V eliminates the internal polymer liner that serves as a gas barrier in the type IV vessels. Type V vessels have to instead rely on carbon fiber laminate to provide structural properties and prevent gas leakage, which is a real engineering challenge and a main reason why type V vessels are not yet widely used in commercial applications. This is of special importance in the case of storing hydrogen as a gas with small molecules; it is a real challenge to avoid its permeation, especially at elevated pressures [21,26,91].

In addition to this, the traditional manufacturing process for composite overwrapped vessels, like filament winding, has several constraints that limit the design space, but automated fiber placement has the potential to replace it for composite pressure vessel manufacturing and provide pathways for further vessel optimization. There is a strong belief that a combination of both AFP and type V technology could provide a venue for a new generation of high-performance vessels—an upgraded version of type V or even a new type VI generation vessel. There are many ongoing research studies on this topic, like authors who proposed novel ideas and who presented a two-piece model of vessel manufactured with AFP as a novel type V vessel. However, defect-free vessels should be considered seriously, because defects in practice can cause catastrophic damages [20,92,93].

At the moment, the technology readiness level of type V pressure vessels is low. Two challenges have been identified that must be addressed before mass adoption can be achieved: manufacturing and permeation control [20]:

- *Manufacturing*—the mandrel must be removed to achieve the liner-less design. For example, a traditional approach is to use a soluble mandrel that can be removed from the tank using water and has been reported for this purpose. Sand, plaster and polyvinyl alcohol (PVA) are some examples for manufacturing of these mandrels.
- *Permeation*—fiber-reinforced laminates are known to exhibit high permeability to numerous gases and in this case to hydrogen due to its small molecular diameter/high kinematic viscosity. This should be solved with strict norms, especially in the case of type V (liner-less) vessels.

Improving the permeation process through the composite materials could be done with a proper design and by enhancing the barrier properties of the matrix system, using a variety of nano-reinforcements like nanoroads, nanotubes, nano-silica, etc., as dopants. For this purpose, there is ongoing research by many scientists worldwide in the field of nanocomposites intended for this application [79,80,94–98].

4.5. Limitations and Future Challenges

Multiple and often opposing forces are shaping the energy transition worldwide, more particularly in Europe. Namely, some positive and negative factors, which could influence the public acceptance of hydrogen found throughout the literature, are mixed and sometimes not consistent. As positive factors, the following are mostly mentioned: environmental benefits, ability to meet modern heating demands compared to traditional

ones, economic opportunities in terms of creating new jobs and growing businesses, etc.. The negative factors divided into social, economic or technical ones. For example, lack of information and public awareness are critical for consideration for deployment of new technologies like hydrogen technology; this affects the encouragement or slow-down of the innovation processes. Economic issues are costly hydrogen systems and transportation and shortage of willingness to pay for hydrogen vehicles, for example. On the other hand, technical issues are of a different origin, e.g., in terms of refueling station availability, installations in domestic homes or industry sectors and familiarity with or readiness to tackle potential hydrogen issues, like flammability, leakage, detection, etc. Hydrogen technologies might be perceived as being "most" sustainable, but still there is a missing link to the local environment and the use of traditional materials for H_2 storage [9,99].

Previous studies have indicated that people have positive beliefs and attitudes towards hydrogen and accept its development as a fuel; for instance, in a recent UK study, 50% of participants supported the conversion of home heating to hydrogen. Another study in Japan, in 2008, explored participants' awareness of hydrogen—only 32% claimed that they knew about hydrogen, which was lower than results observed for alternative forms of energy such as solar, nuclear or wind. However, seven years later when asked, this value increased by 12%. In Spain, one study observed high levels of awareness concerning the existence of hydrogen as a transportation fuel, while another study in the US indicated that only 35% of a total sample were able to correctly answer questions about hydrogen technologies. Increased knowledge and understanding can lead to greater support for hydrogen technologies; otherwise, it can lead to greater opposition and indifference. Therefore, more public information and scientific evidence are needed in order to positively influence people's choices in future projects, decisions, transitions and suitable applications [9].

Interest in hydrogen's potential as a great low-carbon energy carrier is not new. The future of hydrogen technologies could spread in different directions—from finding costly ways of hydrogen production to optimal solutions for hydrogen storage or its distribution. However, clean, widespread use of hydrogen in global energy transitions faces many challenges. One of them is the policy and technology uncertainty mainly dictated by the national or governmental politics—how strong they push the process of energy transitions, for example. In addition, the infrastructure needs, the value chain complexity and regulations, standards and acceptance of the new energy sources are also important challenges. Governments play a crucial role in this process of enabling hydrogen to achieve its potential. In this context, internationalization of hydrogen usage is very important; the Future of Hydrogen report by the International Energy Agency (IEA) urged that international hydrogen trade needs to start soon to make an impact on the global energy system. It mainly focusses on developing international standards for H_2, launching international shipping routes, establishment of coordinated actions and international collaboration for the decarbonization of the world by 2050 [100].

5. Sustainability Aspects

5.1. Sustainability of Hydrogen Technology

Among the 17 interrelated SDGs, SDG 7 (affordable and clean energy), SDG 8 (decent work and economic growth), SDG 9 (industry, innovation and infrastructure), SDG 10 (reduced inequalities), SDG 11 (sustainable cities and communities), SDG 12 (responsible consumption and production), SDG 13 (climate action) and SDG 17 (partnership for the goals) are the ones that are directly affected by the hydrogen technology (UN SDG 2015). The remaining SDGs are also affected, but indirectly through their relationship with the mentioned SDGs. Figure 7 provides a scheme of the relationship between hydrogen technology, basic sustainable pillars and the SDGs directly affected by the adoption of hydrogen technology.



Figure 7. Basic pillars of sustainable development—economic prosperity, social responsibility and environmental stewardship in the context of hydrogen storage technology.

The basic pillars of sustainable development—*economy*, *society* and *environment*—are particularly stressed here; which means that by adopting the hydrogen technology, good planetary health and wellbeing are achievable. In other words, the 3P's principle—people, planet and prosperity—is on the horizon:

Good for the planet + Good for the people = Prosperity.

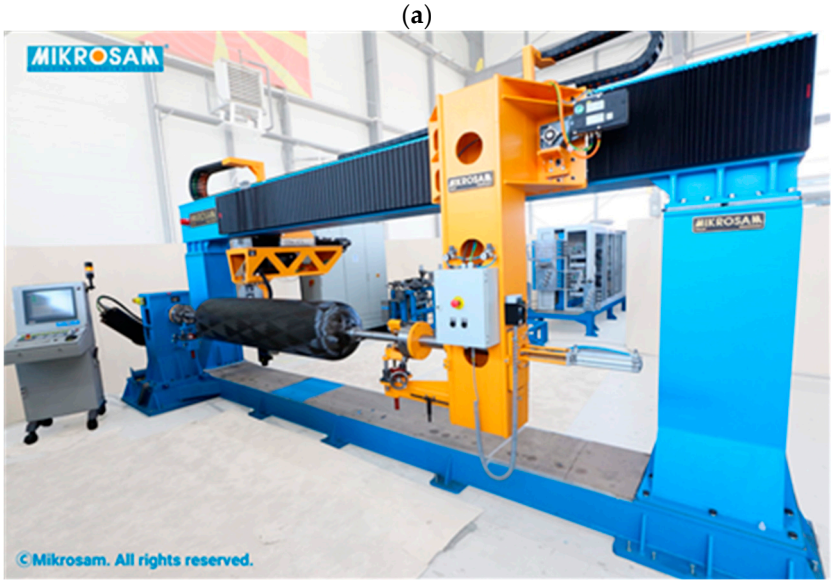
By adoption of this technology, innovation and economic growth are possible and aligned with new jobs openings (*socio-economic* benefits; in good agreement with SDGs #8, 9, 10, 11, 12). The proposed clean "green" H_2 energy (taking it from nature and returning it safely back to nature) is beneficial for future sustainable cities and well aligned with positive *environmental* aspects (in agreement with SDGs #7, 11, 12, 13 as well). The achievement of safe hydrogen technology is only possible with partnerships between different sectors: those related to advanced materials, more particularly composite materials (e.g., towpregs) and those responsible for generation and utilization of green energy (well aligned with SDGs #7, 9, 10, 17).

The environmentally friendly hydrogen fuel is used in a wide range of industrial or transport sectors, contributing to less polluted communities; it also supports the carbon neutrality vision by 2050. Therefore, all eyes are on hydrogen nowadays—many countries and governments worldwide propose ambitious plans and hydrogen strategies in order to meet the targets for decarbonization, lower pollution and GHG emissions. Moreover, hydrogen is being promoted in many global initiatives, such as the above-mentioned SDGs (UN 2015), the Green Deal (European Commission) and many other programs. They all are set with strong ambitions, and for sure, all promise a better future for the next generations. However, there are many expectations for this technology, too—the hydrogen production cost should significantly drop, while production capacity should increase in order to have

a better adoption of hydrogen fuel on a daily basis. It is a good source for cost-efficient fuel with zero emission, but at the same time, finding ways for fast charging times and runs on this fuel for long-range periods should be on the top of the to-do list. Simultaneously, finding a better storage capacity for hydrogen is also a top priority in this sector, which is at the crossroads of the energy sector and advanced materials research; in particular, attempts are directed towards pressurized vessels based on thermoplastic materials that are recyclable and with more sustainable end-of-life aspects [5,21,101,102]. The photographs in Figure 8 present some of the first industrial attempts to make hydrogen storage equipment.



(**a**)



(**b**)

Figure 8. (**a**) Production line for towpreg composite material. (**b**) FW line for dry towpreg processing. [Photographs are taken in Mikrosam, Prilep, N. Macedonia; https://www.mikrosam.com/, accessed on 19 September 2024].

5.2. Hydrogen—The Key Priority for the European Green Deal

Hydrogen is the key priority to achieve the European Green Deal and clean energy transition [10]. In 2022, hydrogen accounted for less than 2% of Europe's energy consumption, and 96% of this hydrogen was produced with natural gas, resulting in significant amounts of CO_2 emissions. Today renewable and low-carbon hydrogen are not yet cost-competitive compared to fossil-based hydrogen [10,37]. However, in the near future, the

CO_2 content of energy carriers will serve as the "new currency" of the energy system. Therefore, the priority for the EU is to develop renewable and green hydrogen. Maybe this is the fuel that will resolve the energy trilemma: this term describes the attempt to balance energy security, equity (accessible and affordable) and environmental sustainability [103].

The best way to finalize this section is to include a part of the EU Hydrogen Manifesto (2024), which says the following:

> "*Across the world, our life chances—any individual's opportunities to improve their quality of life—are being threatened. Conflict and climate change threaten the life chances of many people. Supporting technologies and concepts that can mitigate or solve the current problems is a top priority. Hydrogen is one such technology. Hydrogen is the smallest molecule in the universe but has huge potential to change our world. The way in which we manufacture goods, move people, power our energy needs, and grow crops can be made cleaner and more sustainable. Hydrogen can bring strong industrial development globally and can save jobs in Europe that would otherwise be lost*." [13]

6. Conclusions

As an energy carrier, hydrogen is becoming more and more interesting for the carbon footprint reduction. It is a clean, non-polluting energy source that will positively contribute to carbon neutrality by 2050, especially when it comes from green routes. It is one of the lightest elements that possesses high energy density. There are several technologies for H_2 storage, characterized by pros and cons that make each of them attractive for a specific scenario. Therefore, hydrogen storage systems are becoming the key enabling technologies ensuring good safety performance, cost competitiveness and weight efficiency. In this study, special focus was directed to physical storage systems—pressurized vessels for hydrogen. The main characteristics for these systems should be lightweight and cost-effective storage; this remains a privilege but also a prime challenge to be solved. The weight of the storage tanks mainly depends on the application, while the cost depends on the used materials and manufacturing procedures. The trade-off between the weight and cost still remains a puzzle that many are trying to solve; therefore, partnerships between the manufacturing companies of the raw materials (polymer matrices and reinforcements) and the companies for manufacturing of pressurized vessels are necessary [101–103]. In addition to this partnership, close cooperation with hydrogen producers is required as well.

Furthermore, incorporating sustainable materials into energy-transition technologies could help efforts to decarbonize society. Thermoplastic composite materials are high performance materials, and thanks to their excellent properties and low weight, these materials are becoming very popular and widely used compared to traditional materials in the case of energy solutions such as hydrogen technology [104]. For the purpose of hydrogen pressurized vessels, filament winding technology has experienced a great evolution in recent years; therefore, more material systems in the forms of towpregs are now commercially available at a lower cost. Initial applications of filament winding were restricted to the use of thermoset resins, with impregnation conducted using a resin bath, a compound of uncured polymers in a low-viscosity state. However, towpreg filament winding (using a pre-impregnated fiber tow) is gaining more popularity for mass production of hydrogen tanks, mainly due to clean processing, improved part quality, fast winding speeds, short cure cycles, etc. Many academic and industrial partners are working on the challenge to make suitable hydrogen storage. A lot of concepts have been proposed and tested. Materials challenges have been solved and tested against the strict tests and requirements for certain types of storage not just from a safety point of view but also for making them cost-effective, appropriate to store suitable amounts of hydrogen at satisfactory volume/mass ratios. Certain composite vessel designs (type IV) have performed the best for storage vessels of hydrogen; they exhibit technological maturity and customer satisfaction, while type V vessels need further improvement. They still have not reached the appropriate technology readiness level. However, the authors strongly believe that both AFP and type V pressurized vessels will accommodate the future solution of hydrogen technology in terms

of safe, efficient and low-cost storage/delivery of the future energy carrier that has a lot of promise to solve the energy trilemma (safety, affordability/accessibility and sustainability).

Author Contributions: Conceptualization, A.T.P.; methodology, A.T.P. and B.S.; investigation, B.S.A. and V.V.; resources, A.T.P. and B.S.; writing—original draft preparation, A.T.P.; writing—review and editing, A.T.P. and B.S.; visualization, A.T.P.; supervision, B.S.; project administration, B.S.A. and V.V.; funding acquisition, B.S. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the Ministry of Economy, the Government of R. N. Macedonia (Contract: ME 18-4422/3 from 21 December 2023).

Data Availability Statement: No new data were created or analyzed in this study.

Acknowledgments: The authors are thankful to the Mikrosam company (Prilep, N. Macedonia) and its team for supporting this study. The authors also extend special thanks to the support provided by the Ministry of Economy (the Government of R. N. Macedonia).

Conflicts of Interest: Authors Bisera Samardjioska Azmanoska and Viktorija Velkovska were employed by the company Mikrosam DOO, Krushevski Pat bb, Prilep, North Macedonia. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as potential conflicts of interest.

References

1. Shamoon, A.; Haleem, A.; Bahl, S.; Javaid, M.; Prakash, C.; Budhhi, D. Understanding the role of advanced materials for energy infrastructure and transmission. *Mater. Today Proc.* **2022**, *62*, 4260–4266. [CrossRef]
2. MATERIALS 2030 MANIFESTO—Systemic Approach of Advanced Materials for Prosperity—A 2030 Perspective. 2022. Available online: https://www.ami2030.eu/wp-content/uploads/2022/06/advanced-materials-2030-manifesto-Published-on-7-Feb-2022.pdf (accessed on 19 September 2024).
3. Fan, Q.; Duan, H.; Xing, X. A review of composite materials for enhancing support, flexibility and strength in exercise. *Alex. Eng. J.* **2024**, *94*, 90–103. [CrossRef]
4. Khan, F.; Hossain, N.; Mim, J.J.; Rahman, S.M.M.; Iqbal, M.J.; Billah, M.; Chowdhury, M.A. Advances of composite materials in automobile applications—A review. *J. Eng. Res.* **2024**. [CrossRef]
5. Maiti, S.; Islam, M.R.; Uddin, M.A.; Afroj, S.; Eichhorn, S.J.; Karim, N. Sustainable Fiber-Reinforced Composites: A Review. *Adv. Sustain. Syst.* **2022**, *6*, 2200258. [CrossRef]
6. Ngo, T.-D. (Ed.) Introduction to Composite Materials. In *Composite and Nanocomposite Materials—From Knowledge to Industrial Applications*; IntechOpen: London, UK, 2020. [CrossRef]
7. Nnabuife, S.G.; Oko, E.; Kuang, B.; Bello, A.; Onwualu, A.P.; Oyagha, S.; Whidborne, J. The prospects of hydrogen in achieving net zero emissions by 2050: A critical review. *Sustain. Chem. Clim. Action* **2023**, *2*, 100024. [CrossRef]
8. Agrawal, D.; Mahajan, N.; Singh, S.A.; Sreedhar, I. Green hydrogen production pathways for sustainable future with net zero emissions. *Fuel* **2024**, *359*, 130131. [CrossRef]
9. Edwards, R.L.; Font-Palma, C.; Howe, J. The status of hydrogen technologies in the UK: A multi-disciplinary review. *Sustain. Energy Technol. Assess.* **2021**, *43*, 100901. [CrossRef]
10. *Communication from the Commission to the European Parliament, the Council, the European Economic and Social Committee and the Committee of the Regions—A Hydrogen Strategy for a Climate-Neutral Europe*; European Union: Brussels, Belgium, 2020.
11. IRENA. *Global Hydrogen Trade to Meet the 1.5 °C Climate Goal: Part I—Trade Outlook for 2050 and Way Forward*; International Renewable Energy Agency: Abu Dhabi, United Arab Emirates, 2022.
12. Hydrogen Council & McKinsey Company. *Hydrogen for Net-Zero: A Critical Cost-Competitive Energy Vector*; Hydrogen Council: Brussels, Belgium, 2021. Available online: www.hydrogencouncil.com (accessed on 19 September 2024).
13. Hydrogen Europe Manifesto—For the 2024 European Elections. Available online: www.hydrogeneurope.eu (accessed on 5 August 2024).
14. Slamani, M.; Chatelain, F. A review on the machining of polymer composites reinforced with carbon (CFRP), glass (GFRP), and natural fbers (NFRP). *Discov. Mech. Eng.* **2023**, *2*, 4. [CrossRef]
15. Nagakalyan, S.; Raghukumar, B. Preparation and characterization of carbon/epoxy towpreg composite material. *Mater. Today Proc.* **2020**, *23*, 499–506. [CrossRef]
16. Postacchini, L.; Simoncini, M.; Forcellese, A.; Bevilacqua, M.; Ciarapica, F.E.; Andreassi, G.; Russo, A.C. Environmental assessment of an automated impregnation process of carbon fiber tows. *Procedia CIRP* **2020**, *88*, 445–450. [CrossRef]
17. Jabbar, M.; Nasreen, A. Composite fabrication and joining. In *Composite Solutions for Ballistics*; Nawib, Y., Sapuan, S.M., Shaker, K., Eds.; Woodhead Publishing: Sawston, UK, 2021; ISBN 978-0-12-821984. [CrossRef]
18. Okten, Y.K.; Kaynak, C. Mechanical performance of composite flat specimens and pressure vessels produced by carbon/epoxy towpreg dry winding. *J. Reinf. Plast. Compos.* **2023**, *42*, 558–576. [CrossRef]

19. Ökten, Y.K. Use of Carbon/Epoxy Towpregs during Dry Filament Winding of Composite Flat Specimens and Pressure Vessels. Master's Thesis, Middle East Technical University, Ankara, Türkiye, 2022.
20. Air, A.; Shamsuddoha, M.; Prusty, B.G. A review of Type V composite pressure vessels and automated fibre placement based manufacturing. *Compos. Part B* **2023**, *253*, 110573. [CrossRef]
21. Alves, M.P.; Gul, W.; Cimini Junior, C.A.; Ha, S.K. A Review on Industrial Perspectives and Challenges on Material, Manufacturing, Design and Development of Compressed Hydrogen Storage Tanks for the Transportation Sector. *Energies* **2022**, *15*, 5152. [CrossRef]
22. Garofalo, J.; Walczyk, D. In situ impregnation of continuous thermoplastic composite prepreg for additive manufacturing and automated fiber placement. *Compos. Part A* **2021**, *147*, 106446. [CrossRef]
23. Alagirusamy, R. *Flexible Towpregs and Their Thermoplastic Composites*; Taylor & Francis Group, LLC.: Abingdon, UK, 2023; ISBN 978-0-367-50380-2.
24. Robert, C.; Pecur, T.; Maguire, J.; Lafferty, A.; McCarthy, E.; O Brádaigh, C. A novel powder-epoxy towpregging line for wind and tidal turbine blades. *Compos. Part B Eng.* **2020**, *203*, 108443. [CrossRef]
25. Jois, K.C.; Mölling, T.; Schuster, J.; Grigat, N.; Gries, T. Towpreg manufacturing and characterization for filament winding application. *Polym. Compos.* **2024**, *45*, 7893–7905. [CrossRef]
26. Kadri, K.; Abdallah, A.B.; Ballut, S. Hydrogen Storage Vessels of Type 4 and Type 5. In *Hydrogen Technologies—Recent Advances, New Perspectives and Applications*; Abdallah, Z., Aldoumani, N., Eds.; IntechOpen: London, UK, 2024.
27. Masahiro, T.; Kentaro, S.; Toshiya, K. Towpreg, Method for Manufacturing Same, and Method for Manufacturing Pressure Container. WO2019/225442, 28 November 2019.
28. Shin, H.K.; Ha, S.K. A Review on the Cost Analysis of Hydrogen Gas Storage Tanks for Fuel Cell Vehicles. *Energies* **2023**, *16*, 5233. [CrossRef]
29. Ayakdas, O.; Aydin, L. Hydrogen storage technologies. In *Researches in Science and Art in 21st Century Turkey*; Arapgirlioglu, H., Atik, A., Elliott, R.L., Turgeon, E., Eds.; Bilosfet: Ankara, Turkey, 2017.
30. Ayakdas, O.; Aydin, L.; Savran, M.; Kucukdogan, N.; Ozturk, S. Optimal design of the type III hydrogen storage tank for different carbon/epoxy materials by modified differential evolution method. *Res. Eng. Struct. Mater.* **2019**, *5*, 189–201. [CrossRef]
31. Siddique, A.; Iqbal, Z.; Nawab, Y.; Shaker, K. A review of joining techniques for thermoplastic composite materials. *J. Thermoplast. Compos. Mater.* **2022**, *36*, 3417–3454. [CrossRef]
32. Srebrenkoska, S.; Kochoski, F.; Srebrenkoska, V.; Risteska, S.; Kotynia, R. Effect of Process Parameters on Thermal and Mechanical Properties of Filament Wound Polymer-Based Composite Pipes. *Polymers* **2023**, *15*, 2829. [CrossRef]
33. *Hydrogen Tanks by Filament Winding Process for the Mobility of the Future. Machine Concepts of Roth Composite Machinery*; Roth Composite Machinery GmbH: Steffenberg, Germany, 2021. Available online: https://www.roth-composite-machinery.com/en/company/news/detail/hydrogen-tanks-by-filament-winding-process-for-the-mobility-of-the-future (accessed on 19 September 2024).
34. Purnomo, H.; Soemardi, T.P.; Budiono, H.D.S.; Wibowo, H.B.; Ibadi, M. The filament winding method's finishing process impact on high-fidelity specimens: Homogenity of density, fiber volume fraction, outer surface roughness and tensile strength. *East.-Eur. J. Enterp. Technol.* **2023**, *6*, 43–51. [CrossRef]
35. Sharma, P.; Neogi, S. Performance-based design and manufacturing of filament wound Type-4 cylinders for compressed gas storage. *Compos. Struct.* **2023**, *309*, 116710. [CrossRef]
36. Boon, Y.D.; Joshi, S.C.; Bhudolia, S.K. Review: Filament Winding and Automated Fiber Placement with In Situ Consolidation for Fiber Reinforced Thermoplastic Polymer Composites. *Polymers* **2021**, *13*, 1951. [CrossRef] [PubMed]
37. Ajanovic, A.; Sayer, M.; Haas, R. The economics and the environmental benignity of different colors of hydrogen. *Int. J. Hydrogen Energy* **2022**, *47*, 24136–24154. [CrossRef]
38. Ahmad, A.; Khan, S.; Chhabra, T.; Tariq, S.; Javed, M.S.; Li, H.; Naqvi, S.R.; Rajendran, S.; Luque, R.; Ahmad, I. Synergic impact of renewable resources and advanced technologies for green hydrogen production: Trends and perspectives. *Int. J. Hydrogen Energy* **2024**, *67*, 788.e806. [CrossRef]
39. Arsad, A.Z.; Hannan, M.A.; Al-Shetwi, A.Q.; Begum, R.A.; Hossain, M.J.; Ker, P.J.; Mahlia, T.M.I. Hydrogen electrolyser technologies and their modelling for sustainable energy production: A comprehensive review and suggestions. *Int. J. Hydrogen Energy* **2023**, *48*, 27841–27871. [CrossRef]
40. Liponi; Pasini, G.; Baccioli, A.; Ferrari, L. Hydrogen from renewables: Is it always green? The Italian scenario. *Energy Convers. Manag.* **2023**, *276*, 116525. [CrossRef]
41. Hosseini, S.E.; Wahid, M.A. Hydrogen production from renewable and sustainable energy resources: Promising green energy carrier for clean development. *Renew. Sustain. Energy Rev.* **2016**, *57*, 850–866. [CrossRef]
42. Sikiru, S.; Oladosu, T.L.; Amosa, T.I.; Olutoki, J.O.; Ansari, M.N.M.; Abioye, K.J.; Rehman, Z.U.; Soleimani, H. Hydrogen-powered horizons: Transformative technologies in clean energy generation, distribution, and storage for sustainable innovation. *Int. J. Hydrogen Energy* **2024**, *56*, 1152–1182. [CrossRef]
43. Zainal, B.S.; Ker, P.J.; Mohamed, H.; Ong, H.C.; Fattah, I.M.R.; Rahman, S.M.A.; Nghiem, L.D.; Mahlia, T.M.I. Recent advancement and assessment of green hydrogen production technologies. *Renew. Sustain. Energy Rev.* **2024**, *189*, 113941. [CrossRef]
44. Hassan, Q.; Abdulateef, A.M.; Hafedh, S.A.; Al-samari, A.; Abdulateef, J.; Sameen, A.Z.; Salman, H.M.; Al-Jiboory, A.K.; Wieteska, S.; Jaszczur, M. Renewable energy-to-green hydrogen: A review of main resources routes, processes and evaluation. *Int. J. Hydrogen Energy* **2023**, *48*, 17383–17408. [CrossRef]

45. Mani, S.S.; Rajendran, S.; Mathew, T.; Gopinath, C.S. A review on the recent advances in the design and structure–activity relationship of TiO_2-based photocatalysts for solar hydrogen production. *Energy Adv.* **2024**, *3*, 1472. [CrossRef]
46. Usman, M.R. Hydrogen storage methods: Review and current status. *Renew. Sustain. Energy Rev.* **2022**, *167*, 112743. [CrossRef]
47. Nachtane, M.; Tarfaoui, M.; Abichou, M.a.; Vetcher, A.; Rouway, M.; Aâmir, A.; Mouadili, H.; Laaouidi, H.; Naanani, H. An Overview of the Recent Advances in Composite Materials and Artificial Intelligence for Hydrogen Storage Vessels Design. *J. Compos. Sci.* **2023**, *7*, 119. [CrossRef]
48. Berna-Escriche, C.; Rivera-Duran, Y.; Cordova-Chavez, Y.; Munoz-Cobo, J.L. Maximizing the use of hydrogen as energy vector to cover the final energy demand for stand-alone systems, application and sensitivity analysis for the Canary Archipelago by 2040. *Prog. Nucl. Energy* **2023**, *163*, 104791. [CrossRef]
49. Bird & Bird. International Green Hydrogen Report 2024. Available online: https://www.twobirds.com/en/insights/2024/global/international-green-hydrogen-report-2024 (accessed on 19 September 2024).
50. Idriss, H.; Scott, M.; Subramani, V. Introduction to hydrogen and its properties. *Compend. Hydrog. Energy* **2015**, 3–19. [CrossRef]
51. Zhang, Y.G.; Jia, Z.-C.; Yuan, Z.G.; Yang, T.; Qi, Y.; Zhao, D.G. Development and Application of Hydrogen Storage. *J. Iron Steel Res. Int.* **2015**, *22*, 757–770. [CrossRef]
52. Moran, C.; Deane, P.; Yousefian, S.; Monaghan, R.F.D. The hydrogen storage challenge: Does storage method and size affect the cost and operational flexibility of hydrogen supply chains? *Int. J. Hydrogen Energy* **2024**, *52*, 1090–1100. [CrossRef]
53. Cheng, Q.; Zhang, R.; Shi, Z.; Lin, J. Review of common hydrogen storage tanks and current manufacturing methods for aluminium alloy tank liners. *Int. J. Lightweight Mater. Manuf.* **2024**, *7*, 269–284. [CrossRef]
54. Machado, H.; Ferreira, A.C.; Teixeira, S.F.; Teixeira, J.C. Green Hydrogen Value Chain: Modelling of a PV Power Plant Integrated with H2 Production for Industry Application. *Energies* **2024**, *17*, 1414. [CrossRef]
55. Talukdar, M.; Blum, P.; Heinemann, N.; Miocic, J. Techno-economic analysis of underground hydrogen storage in Europe. *iScience* **2024**, *27*, 108771. [CrossRef] [PubMed]
56. Muthukumar, P.; Kumar, A.; Afzal, M.; Bhogilla, S.; Sharma, P.; Parida, A.; Jana, S.; Kumar, E.A.; Pai, R.K.; Jain, I.P. Review on large-scale hydrogen storage systems for better sustainability. *Int. J. Hydrogen Energy* **2023**, *48*, 33223–33259. [CrossRef]
57. Zuttel, A. Materials for hydrogen storage. *Mater. Today* **2003**, *6*, 24–33. [CrossRef]
58. Kaura, M.; Pal, K. Review on hydrogen storage materials and methods from an electrochemical viewpoint. *J. Energy Storage* **2019**, *23*, 234–249. [CrossRef]
59. Kovac, A.; Paranos, M.; Marcius, D. Hydrogen in energy transition: A review. *Int. J. Hydrogen Energy* **2021**, *46*, 10016–10035. [CrossRef]
60. Stelitano, S.; Rullo, A.; Piredda, L.; Mecozzi, E.; Di Vito, L.; Agostino, R.G.; Filosa, R.; Formoso, V.; Conte, G.; Policicchio, A. The Deltah Lab, a New Multidisciplinary European Facility to Support the H2 Distribution & Storage Economy. *Appl. Sci.* **2021**, *11*, 3272. [CrossRef]
61. Lebrouhi, B.E.; Djoupo, J.J.; Lamrani, B.; Benabdelaziz, K.; Kousksou, T. Global hydrogen development—A technological and geopolitical overview. *Int. J. Hydrogen Energy* **2022**, *47*, 7016–7048. [CrossRef]
62. Li, F.; Chen, X.; Xu, P.; Fan, Z.; Wang, Q.; Lyu, C.; Zhang, Q.; Yu, H.; Wu, H. Optimal design of thin-layered composites for type IV vessels: Finite element analysis enhanced by ANN. *Thin-Walled Struct.* **2023**, *187*, 110752. [CrossRef]
63. Alcántar, V.; Aceves, S.M.; Ledesma, E.; Ledesma, S.; Aguilera, E. Optimization of Type 4 composite pressure vessels using genetic algorithms and simulated annealing. *Int. J. Hydrogen Energy* **2017**, *42*, 15770–15781. [CrossRef]
64. *ISO/TC 197*; Hydrogen Technologies. ISO: Geneva, Switzerland, 1990. Available online: https://www.iso.org/committee/54560.html (accessed on 19 September 2024).
65. *ISO TC 220*; Cryogenic Vessels. ISO: Geneva, Switzerland, 1999. Available online: https://www.iso.org/committee/54990.html (accessed on 19 September 2024).
66. *ISO TC 58*; Gas Cylinders. ISO: Geneva, Switzerland, 1947. Available online: https://www.iso.org/committee/49008.html (accessed on 19 September 2024).
67. *ISO/TC 22/SC 41*; Specific Aspects for Gaseous Fuels. ISO: Geneva, Switzerland, 2014. Available online: https://www.iso.org/committee/5391370.html (accessed on 19 September 2024).
68. *CEN/CLC/TC 6*; Hydrogen. CEN: Brussels, Belgium, 2001. Available online: https://standards.iteh.ai/catalog/tc/cen/a852b9cf-c888-4752-b21d-4ce9b393b262/cen-clc-tc-6 (accessed on 19 September 2024).
69. *CEN/TC 23*; Transportable Gas Cylinders. CEN: Brussels, Belgium, 1989. Available online: https://standards.iteh.ai/catalog/tc/cen/3ab44d2a-f490-4a16-956c-6387134ff622/cen-tc-23?srsltid=AfmBOopWmW1IatRWcD3PGrywp3OTBdqucT0eglEpk-wCgLRj4d0vvnyx (accessed on 19 September 2024).
70. *CEN/TC 69*; Industrial Valves. CEN: Brussels, Belgium, 2003. Available online: https://standards.iteh.ai/catalog/tc/cen/07259daf-c938-4ef6-8454-96428bc28031/cen-tc-69?srsltid=AfmBOor8GJpVH2AS14O5W32RTD9ANuZ-61rd_RMxqZ5Yphtx0FdNL56- (accessed on 19 September 2024).
71. *CEN/TC 185*; Fasteners. CEN: Brussels, Belgium, 2021. Available online: https://standards.iteh.ai/catalog/tc/cen/1ac199f2-b809-4c0d-bbf5-d19a86f1ea0b/cen-tc-185?srsltid=AfmBOor2NcnhzveyEoyCePLTqJYas_RRxdeRhyV4DVgMfgNmg9aKX4Dd (accessed on 19 September 2024).

72. *CEN/TC 197*; Pumps. CEN: Brussels, Belgium, 2023. Available online: https://standards.iteh.ai/catalog/tc/cen/2a913307-9de4-486d-8a84-f56e75d9c5ef/cen-tc-197?srsltid=AfmBOooPUUw88_pPkQAJUkfioJqtD2w1BLCat4ui9JiXZehRx5iNy808 (accessed on 19 September 2024).
73. *CEN/TC 234*; Gas Infrastructure. CEN: Brussels, Belgium, 2023. Available online: https://standards.iteh.ai/catalog/tc/cen/cc25e361-921b-4ba1-822b-21062c37a4c0/cen-tc-234?srsltid=AfmBOoqgIFjef2j5TlbB_xHa0hHOhEUP88PF7HEridRqXh6ueIUteWhr (accessed on 19 September 2024).
74. Economic Commission for Europe (ECE). Available online: https://unece.org/ev-hev-and-fc (accessed on 19 September 2024).
75. EU Type Approval for Hydrogen Powered Vehicles. Available online: https://www.tuvsud.com/en/resource-centre/case-studies/eu-type-approval-hydrogen-vehicles (accessed on 19 September 2024).
76. *GTR 13*; Hydrogen and Fuel Cell Vehicles. GTR: Geneva, Switzerland, 1998. Available online: https://unece.org/transport/standards/transport/vehicle-regulations-wp29/global-technical-regulations-gtrs (accessed on 19 September 2024).
77. The Hydrogen Europe Quarterly Magazine, Q1 of 2024. Available online: https://hydrogeneurope.eu/wp-content/uploads/2024/03/The-Hydrogen-Europe-Quarterly_6_24DIGITAL_FINAL.pdf (accessed on 19 September 2024).
78. Wang, D.; Liao, B.; Zheng, J.; Huang, G.; Hua, Z.; Gu, C.; Xu, P. Development of regulations, codes and standards on composite tanks for on-board gaseous hydrogen storage. *Int. J. Hydrogen Energy* **2019**, *44*, 22643–22653. [CrossRef]
79. Cui, Y.; Kumar, S.; Konac, B.R.; van Houcke, D. Gas barrier properties of polymer/clay nanocomposites. *RSC Adv.* **2015**, *5*, 63669. [CrossRef]
80. Lin, J.; Zheng, C.; Dai, Y.; Wang, Z.; Lu, J. Prediction of composite pressure vessel dome contour and strength analysis based on a new fiber thickness calculation method. *Compos. Struct.* **2023**, *306*, 116590. [CrossRef]
81. Genovese, M.; Cigolotti, V.; Fragiacomo, E.J.P. Current standards and configurations for the permitting and operation of hydrogen refueling stations. *Int. J. Hydrogen Energy* **2023**, *48*, 19357–19371. [CrossRef]
82. *ISO 11439:2013*; Gas Cylinders-High Pressure Cylinders for the Onboard Storage of Natural Gas as a Fuel for Automotive Vehicles. ISO: Geneva, Switzerland, 2013. Available online: https://www.iso.org/standard/44755.html (accessed on 19 September 2024).
83. *ISO 16111:2018*; Transportable Gas Storage Devices–Hydrogen Absorbed in Reversible Metal Hydride. ISO: Geneva, Switzerland, 2018. Available online: https://www.iso.org/standard/67952.html (accessed on 19 September 2024).
84. Kangal, S.; Kartav, O.; Tanoğlu, M.; Aktaş, E.; Artem, H.S. Investigation of interlayer hybridization effect on burst pressure performance of composite overwrapped pressure vessels with load-sharing metallic liner. *J. Compos. Mater.* **2020**, *54*, 961–980. [CrossRef]
85. Nebe, M.; Soriano, A.; Braun, C.; Middendorf, P.; Walther, F. Analysis on the mechanical response of composite pressure vessels during internal pressure loading: FE modeling and experimental correlation. *Compos. Part B* **2021**, *212*, 108550. [CrossRef]
86. Magneville, B.; Gentilleau, B.; Villalonga, S.; Nony, F.; Galiano, H. Modeling, parameters identification and experimental validation of composite materials behavior law used in 700 bar type IV hydrogen high pressure storage vessel. *Int. J. Hydrogen Energy* **2015**, *40*, 13193–13205. [CrossRef]
87. Mair, G.W.; Scherer, F.; Hoffman, M. Type approval of composite gas cylinders e Probabilistic analysis of standards' requirements concerning minimum burst pressure. *Int. J. Hydrogen Energy* **2015**, *40*, 5359–5366. [CrossRef]
88. Patra, S.; Mallisetty, P.K.; Murmu, N.C.; Hirani, H.; Samanta, P. Study on fracture evaluation in hydrogen environment in 316L stainless steel used in high pressure hydrogen tank. *Mater. Today Proc.* **2022**, *66*, 3723–3728. [CrossRef]
89. Tretsiakova-McNally, S. Safety of hydrogen storage. HyResponse, Grant Agreement No. 325348. Available online: https://cordis.europa.eu/project/id/325348/reporting (accessed on 19 September 2024).
90. US Department of Energy. Office of energy efficiency & Renewable Energy, Hydrogen and Fuel Cell Technologies Office. Energy Earth Shots. In *The 2nd DOE Energy Earthshot*; US Department of Energy: Washington, DC, USA, 2021.
91. Katsivalis, I.; Signorini, V.; Ohlsson, F.; Langhammer, C.; Minelli, M.; Asp, L.E. Hydrogen permeability of thin-ply composites after mechanical loading. *Compos. Part A Appl. Sci. Manuf.* **2024**, *176*, 107867. [CrossRef]
92. Jiang, P.; Liu, C.; Yang, C.; Zhang, L.Y.; Liang, J.W.; Yang, Z.H. Study on acoustic emission parameter fusion-based NOL ring damage assessment method for modeling hydrogen storage cylinder shells. *Int. J. Hydrogen Energy* **2024**. [CrossRef]
93. Okolie, O.; Latto, J.; Faisal, N.; Jamieson, H.; Mukherji, A.; Njuguna, J. Manufacturing Defects in Thermoplastic Composite Pipes and Their Effect on the in-situ Performance of Thermoplastic Composite Pipes in Oil and Gas Applications. *Appl. Compos. Mater.* **2023**, *30*, 231–306. [CrossRef]
94. Mohamadiana, N.; Ghorbanib, H.; Bazrkarc, H.; Wood, D.A. Carbon-nanotube-polymer nanocomposites enable wellbore cements to better inhibit gas migration and enhance sustainability of natural gas reservoirs. In *Sustainable Natural Gas Reservoir and Production Engineering*; Elsevier: Amsterdam, The Netherlands, 2022. [CrossRef]
95. Wei, R.; Wang, X.; Zhang, X.; Chen, C.; Du, S. Fabrication of High Gas Barrier Epoxy Nanocomposites: An Approach Based on Layered Silicate Functionalized by a Compatible and Reactive Modifier of Epoxy-Diamine Adduct. *Molecules* **2018**, *23*, 1075. [CrossRef] [PubMed]
96. López-González, M.; Flores, A.; Marra, F.; Ellis, G.; Gómez-Fatou, M.; J. Salavagione, H. Graphene and Polyethylene: A Strong Combination Towards Multifunctional Nanocomposites. *Polymers* **2020**, *12*, 2094. [CrossRef] [PubMed]
97. Al Sheheri, S.Z.; Al-Amshany, Z.M.; Al Sulami, Q.A.; Tashkandi, N.Y.; Hussein, M.A. The preparation of carbon nanofillers and their role on the performance of variable polymer nanocomposites. *Des. Monomers Polym.* **2019**, *22*, 8–53. [CrossRef]

98. Lizundia, E.; Sipponen, M.H.; Greca, L.G.G.; Balakshin, M.; Tardy, B.L.; Rojas, O.J.; Puglia, D. Multifunctional lignin-based nanocomposites and nanohybrids. *Green Chem.* **2021**, *23*, 6698–6760. [CrossRef]
99. DNV. *Energy Transition Outlook 2023—A Global and Regional Forecast to 2050*; DNV: Baerum, Norway, 2023. Available online: https://www.dnv.com/energy-transition-outlook/ (accessed on 19 September 2024).
100. IEA. *The Future of Hydrogen Seizing Today's Opportunities*; IEA: Paris, France, 2019.
101. Rahman, M.A.; Haque, S.; Athikesavan, M.M.; Kamaludeen, M.B. A review of environmental friendly green composites: Production methods, current progresses, and challenges. *Environ. Sci. Pollut. Res.* **2023**, *30*, 16905–16929. [CrossRef]
102. Crupi, V.; Epasto, G.; Napolitano, F.; Palomba, G.; Papa, I.; Russo, P. Green Composites for Maritime Engineering: A Review. *J. Mar. Sci. Eng.* **2023**, *11*, 599. [CrossRef]
103. Shrigandhi, G.D.; Kothavale, B.S. Biodegradable composites for filament winding process. *Mater. Today Proc.* **2021**, *42*, 2762–2768. [CrossRef]
104. Major, Z. Applicability and Limitations of Thermoplastic Composites for Hydrogen Pressure Vessels. *Acad. J. Polym. Sci.* **2023**, *5*, 555675. [CrossRef]

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.