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Review of common hydrogen storage tanks and current manufacturing methods for aluminium alloy tank liners

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A B S T R A C T

With the growing concern about climate issues and the urgent need to reduce carbon emissions, hydrogen has attracted increasing attention as a clean and renewable vehicle energy source. However, the storage of flammable hydrogen gas is a major challenge, and it restricts the commercialisation of fuel cell electric vehicles (FCEVs). This paper provides a comprehensive review of common on-board hydrogen storage tanks, possible failure mechanisms and typical manufacturing methods as well as their future development trends. There are generally five types of hydrogen tanks according to different materials used, with only Type III (metallic liner wrapped with composite) and Type IV (polymeric liner wrapped with composite) tanks being used for vehicles. The metallic liner of Type III tank is generally made from aluminium alloys and the associated common manufacturing methods such as roll forming, deep drawing and ironing, and backward extrusion are reviewed and compared. In particular, backward extrusion is a method that can produce near net-shape cylindrical liners without the requirement of welding, and its tool designs and the microstructural evolution of aluminium alloys during the process are analysed. With the improvement and innovation on extrusion tool designs, the extrusion force, which is one of the most demanding issues in the process, can be reduced significantly. As a result, larger liners can be produced using currently available equipment at a lower cost.

1. Introduction

According to the World Meteorological Organization (WMO) [1] and the Intergovernmental Panel on Climate Change (IPCC) [2], global warming is mainly due to human activities during which greenhouse gases such as CO_2 are emitted and accumulated in the atmosphere to trap heat on the Earth. An increase in temperature of more than 1.5 °C [3] above pre-industrial levels is believed to cause a loss in species, a rise in sea level, the spread of diseases [4], more frequent extreme weather conditions, and the migration of human beings [5]. To limit global warming to below 1.5 °C and maintain sustainable development, there is an urgent demand to reduce carbon emissions worldwide and a goal to achieve net-zero emission by 2050 was thus set by the Paris Agreement [6].

One of the major sources of carbon emissions is the burning of fossil fuels to provide the energy needed in daily life and it accounts for about 76% of the US emissions caused by human activities [7]. As a non-renewable energy source, the currently discovered fossil fuel reserves are rapidly running out [8]. Thus, many industries have taken a move to transfer from fossil fuels to other clean and renewable energy sources. For automotive and transport applications, the fuel cell vehicle (FCV) or fuel cell electric vehicle (FCEV) powered by hydrogen is a promising potential alternative to conventional combustion engine vehicles in the future.

Hydrogen is an abundant and clean energy source that produces no carbon emissions; the only products are water vapour and warm air [9]. It has an energy density of 120 MJ/kg, which is about three times that of diesel or petrol, and this makes hydrogen a desirable energy source [10]. Hydrogen has a wide range of applications across the chemical industry, electronics manufacturing, and power generation. In contrast to about 90% of the commercially available hydrogen used in the refining of petroleum and production of ammonia and methanol, only a very small percentage is used in the transportation sector [11]. The first FCEV prototype driven by

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hydrogen was introduced in the late 1990s and in the following years, many automakers such as General Motor and Toyota have developed more than 20 different types of FCEVs [12]. Over the recent decade, while battery electric vehicles (BEVs) have seen increasingly growing popularity and sales worldwide, the uptake of hydrogen vehicles is rather slow, and one of the main reasons for this is the challenges in on-board storage of the flammable hydrogen [13].

The common methods to store hydrogen on-board include the liquid form storage, the compressed gas storage, and the material-based storage, and the working principles and material used of each method have been reviewed by Zhang et al. [14] and Barthelemy et al. [15]. Due to the technical complexity of the liquid form storage and the material-based storage, the current FCEVs are dominated by the compressed hydrogen gas system, which stores compressed hydrogen gas in hydrogen tanks. However, little information on the manufacturing methods of these tanks was mentioned in the existing review papers. Some common manufacturing techniques used in the industry include roll forming, deep drawing and ironing (DDI), and backward extrusion. Halmos [16] has summarised the basics of roll forming, and some research has been done on the use of DDI and backward extrusion to produce cylinders [17–19], but there is a lack of systematic comparison of the different manufacturing methods.

The main objective of this paper is to review the common hydrogen storage tanks and the manufacturing methods for aluminium alloy liners of hydrogen tanks. First, different types of existing hydrogen tanks are analysed, and their respective advantages and disadvantages are compared. The key failure mechanisms of hydrogen tanks are also discussed, and common prevention measures are summarised. The main emphasis is on the different manufacturing methods of metallic tank liners, the working principle, and in particular, more recent development of the backward extrusion techniques.

2. Hydrogen tanks

2.1. Hydrogen storage methods

There are generally three hydrogen storage methods that can be applied to vehicles: the liquid form storage, the compressed gas storage, and the material-based (metal hydrides) storage. A detailed comparison of these three methods is listed in Table 1.

The liquid form storage gives a high hydrogen density of 70 kg/m^3 and this high density allows the storage of a large amount of hydrogen with relatively small tanks [20]. The ambient pressure required to store liquid hydrogen minimises the need for thick tank walls, and thus reduces the specific tank weight which is defined as the tank weight to store 1 kg of hydrogen, to about 11.5 kg. The liquid form storage also allows easy transportation and easy fuel refilling for cars. However, cryogenic temperatures as low as −253 °C are needed to keep hydrogen as liquid and thus, a refrigeration unit is required which causes extra cost and complexity on top of the storage tank itself [26]. Despite having the refrigeration unit, it is difficult to keep these cryogenic temperatures, which results in the evaporation and leakage of hydrogen gas under common environmental conditions [27]. Besides, due to the distinct contraction coefficient of different materials under low temperatures, high stress can be built up on tanks made of hybrid materials, causing structural failure of the tank which could lead to the massive leakage of flammable hydrogen fuel and thus pose severe safety concerns [25].

To ensure safety, the material-based storage method is currently in development. The common material used for this method is metal hydrides (e.g., $NaAlH_4$) which allow the formation of hydrogen bonds between the hydrogen and metal atoms (e.g., Al—H), and have good reversible hydrogen charge and discharge capabilities [28,29]. The ambient temperature and pressure adopted in this method simplify the technical complexity and the hydrogen density can vary from 40 kg/m^3 to as high as 100 kg/m^3 with different types of metal hydrides used [21]. However, one of the major disadvantages of this method is that both the weight and the cost of the whole storage system are relatively high. Taking $NaAlH_4$ as an example, the specific tank weight is about 85 kg and the specific cost is about $1,430, while the weight ratio of the stored hydrogen to the whole storage system is less than 2% [24]. Such high storage system weight will cause a rise in the overall vehicle weight and together with the low hydrogen release rate, reduce the fuel efficiency significantly. Besides, the refilling time of the material-based hydrogen is longer as compared to that of the liquid hydrogen, which makes long-distance travel inconvenient. Heat is also generated during the charging of hydrogen molecules, which poses a very high risk of explosion of hydrogen [24].

Table 1
Comparison of different hydrogen storage methods used for automotive applications.

Storage form	Liquid	Compressed gas	Metal hydrides
Temperature (°C)	−253	Ambient	Ambient
Pressure (bar)	1.013	350 or 700	1.013
Hydrogen density (kg/m^3)	70 [20]	23.3 (350 bar) 39.3 (700 bar) [20]	40–100 [21]
Specific internal tank size[a] (L/kg)	14 [22]	44 (350 bar) 26 (700 bar)	35
Specific system size[b] (L/kg)	22	60 (350 bar) 45 (700 bar)	/
Specific tank weight[a] (kg/kg)	11.5	12.5–25	85
Specific tank cost[a] ($/kg)	400 [23]	600–1600	500–1500 [24]
Advantages	– High hydrogen density – Easy transportation – Short refilling time	– Simple and well-developed technique – Short refilling time	– High hydrogen density – Good reversible hydrogen charge and discharge capability
Disadvantages	– High-cost refrigeration unit required – Evaporation and leakage of hydrogen gas – Structural failure of tank under high stress [25]	– High internal pressure/thick cylinder walls – Low density/large tank size – Risk of rupture/explosion	– High tank weight – Low hydrogen release rate – Heat generated during charging – Long refilling time – Weight ratio of hydrogen to storage system < 2%

[a] These parameters are defined as the size, weight, and cost of tank to store 1 kg of hydrogen.
[b] Specific system size is the size including both the tank size to store hydrogen and the size of the refrigeration unit for liquid form storage/the size of metal hydrides for material-based storage.

Due to the technical complexity and other disadvantages of the liquid form storage and the material-based storage, the current FCEVs are dominated by the compressed hydrogen gas systems. This storage technique is relatively simpler for using and better developed over the decades as compared to the other two storage methods as refrigeration unit and heavy material are not required and it allows a relatively short refilling time (about 3.7 min) of hydrogen fuel [30]. However, the density of hydrogen gas is very low (0.089 kg/m^3) and an extremely large volume (11,236 L) is needed to store 1 kg of hydrogen at ambient temperature and pressure [30]. Thus, to carry enough hydrogen fuel for a long-distance journey, the gas needs to be compressed to high pressures with higher densities. Current commercially available hydrogen vehicles normally adopt either 350 bar or 700 bar to achieve a hydrogen density of around 23.3 kg/m^3 or 39.3 kg/m^3 respectively [20]. If all-metal tank is used, wall thickness of greater than 22 mm is required because there is a high risk of rupture and explosion of the tanks under these high pressures. Also, as the density of hydrogen is still lower than that of the liquid form or material-based form, the specific tank sizes (60 L for 350 bar and 45 L for 700 bar) needed to store 1 kg of hydrogen are larger, and this restricts the amount of hydrogen gas that can be carried with the passenger cars due to limited space available on-board for the tanks. Thus, to improve the travel range of FCEVs as well as to prevent any failure of the tank and explosion of the flammable gas, the high-pressure tanks or vessels need to be carefully designed and manufactured to improve their mechanical properties and increase pressures tolerable.

2.2. Pressure vessels and applications

Hydrogen is normally stored in pressure vessels, which are containers used in the industry and daily life to store gases and liquids under high pressures. Common pressure vessels include cylindrical pressure vessels (Fig. 1(a)–(d)) and spherical pressure vessels (Fig. 1(e)); cylindrical pressure vessels can be further classified into the vertical pressure vessels (Fig. 1(a) and (b)) and the horizontal pressure vessels (Fig. 1(c) and (d)) according to their configurations. Cylindrical pressure vessels are cheaper and easier to manufacture and can be packed more efficiently during storage and transportation. However, weak areas are found at each end of the vessel as the joints between the body shell and the heads act as regions of high stress intensity. For instance, the stress at the joints can reach about 560 MPa, which is very close to the allowable limit (747 MPa) set by the American Society of Mechanical Engineers (ASME) code [31]. Differently, spherical pressure vessels have a larger volume to surface area ratio and the stress is evenly distributed across the whole area as there are no sharp edges. Thus, to withstand the same pressure, only half of the wall thickness is required for spherical pressure vessels as compared to cylindrical pressure vessels, and hence a smaller amount of material is needed [32]. As a result, spherical vessels are preferred for the storage of fluids under high pressures. However, it is very difficult and costly to manufacture the round-shape vessels and their packing efficiency is poor [33,34]. Thus, the dominant pressure vessels used in FCEVs currently are still the cylindrical type and the development mainly focuses on weight and cost reduction during the manufacturing process.

Pressure vessels have a wide range of applications across different industries besides the automotive sector. Table 2 summarises the products provided by some pressure vessel manufacturing companies and the corresponding intended applications. Common industrial applications include using pressure vessels for distillation and storage purposes in the oil and gas industry. In the chemical and power industry, pressure vessels are called reactors where the chemical reaction takes place. In private sectors, applications of pressure vessels can be found everywhere in our daily life such as cooking gas containers used in households, oxygen cylinders in the hospital, and nitrogen and argon gas containers in the laboratories. As the application of pressure vessels is so wide, cheaper and easier manufacturing methods will definitely tie in well with the market demand and have the potential to improve the overall industrial productivity and efficiency.

2.3. Different types of hydrogen tanks

Hydrogen tanks can be classified into five types according to the structure of the materials used [15]. The main features are summarised in Table 3.

Type I hydrogen tanks are made of metal only [48]. Common metals used include steel and aluminium alloys. Such hydrogen tanks are the easiest to manufacture among the five types and thus, the cheapest. They can be made to have a huge capacity and the working pressure is normally at around 200 bar [15]. Due to the high density of metal materials, Type I tanks are heavy and are mainly used for either stationary applications such as the large-scale storage of industrial hydrogen or large vessels such as submarines that have adequate space and can carry massive load [46]. Type II hydrogen tanks are also all-metal cylinders which are similar to Type I except that they have carbon fibre or glass fibre filament wrapped around their straight body part. These tanks are used for stationary applications as well and the working pressure is around 300 bar, higher than that of Type I due to the additional high-strength filament wrapping [15].

Type III and Type IV hydrogen tanks are solutions for automobile applications due to significant weight savings as compared to Type I and Type II [15]. Both types are fully wrapped with composite, normally carbon fibre Tory T700S or T800S. The only difference is that Type III uses metallic liner while Type IV uses polymeric liner [14]. The thin layer of liner functions to seal the gas in the tank and a thicker outer composite wrap is the main load-bearing component [14]. For Type III tanks, the metals used for the liner is commonly AA6061 or AA7000 as these alloy species have a good machineability and can be heat treated to gain better mechanical properties [48]. Due to the high strength (4900 MPa) and low density (1.80 g/cm^3) of T700S carbon fibre, both Type III and Type IV tanks allow higher internal pressures as compared to other types and the overall tank weight and wall thickness are significantly lower [48]. To withstand the same internal pressure, the thickness of Type III and Type IV tanks is less than half that of Type I tanks as indicated by Fig. 2 [49]. Thus, the hydrogen gas can be compressed to a higher density and more gas can be carried with smaller and lighter tanks which are favoured by lightweight vehicles.

Type V hydrogen tanks are fully composite. It is about 20% lighter than Type IV tanks and can withstand an even higher pressure of about 1000 bar [50]. However, this technology is still in development and the current high cost of composite materials restricts its applications commercially [47].

Currently both Type III and Type IV tanks are used commercially in FCEVs; some practical industrial information of the two types is collected and analysed as shown in Fig. 3.

For an internal pressure of 350 bar, Type III hydrogen tank is produced more favourably because its manufacturing technique is more mature as compared to that of Type IV. The specific cost of 350 bar Type III tank (600 \$/kg) is much lower than that of 700 bar Type III tank (1600 \$/kg) as seen in Fig. 3(a), but the specific tank size is significantly larger as indicated by Fig. 3(c). The 350 bar Type III tank has a specific internal tank size of 43 L/kg while the 700 bar Type III tank has a smaller size of only 25 L/kg, which represents a 42% reduction. Larger specific size means that a larger tank is

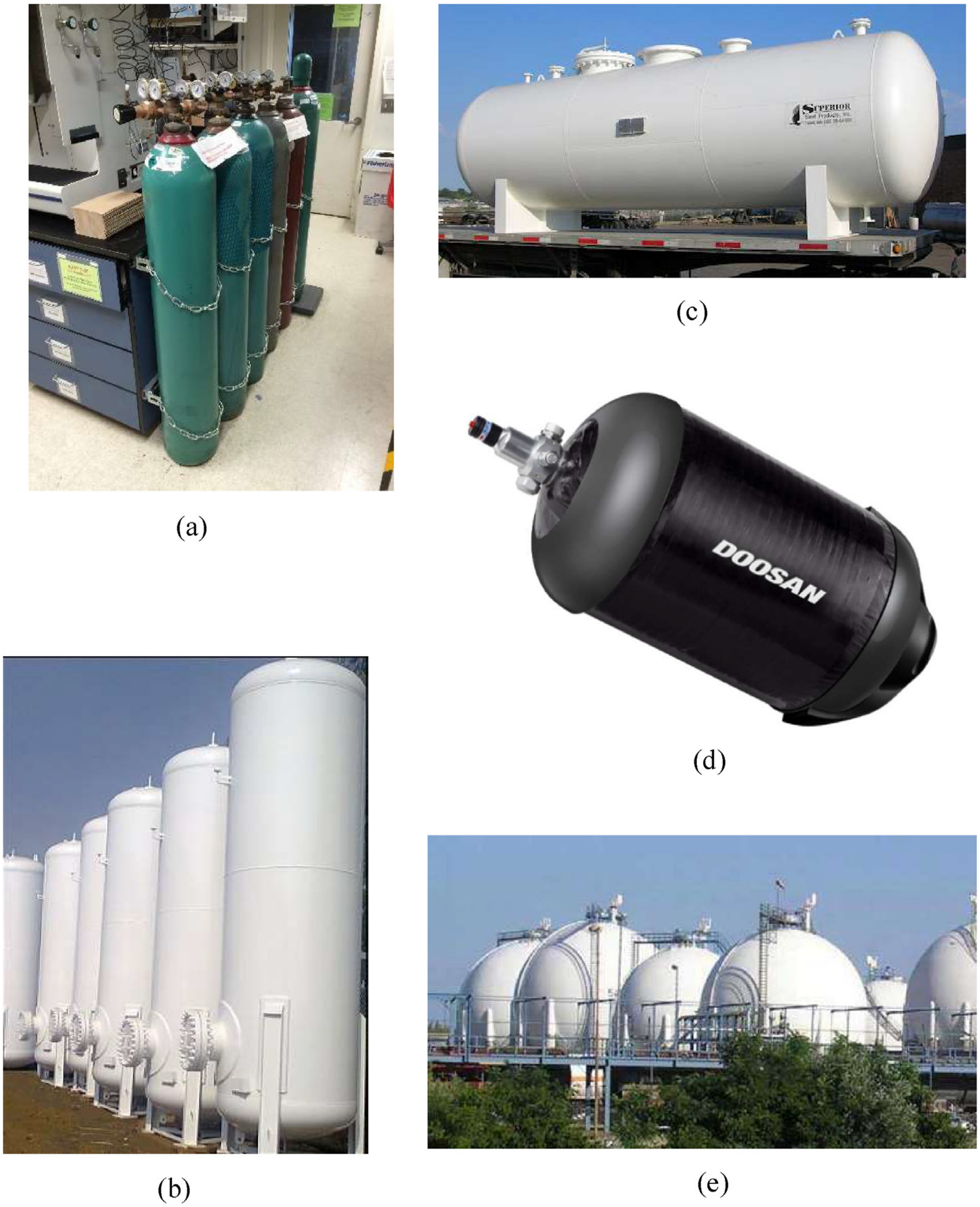
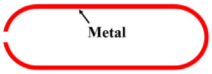
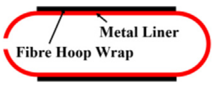
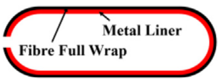
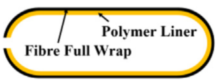
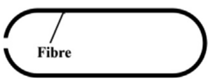


Fig. 1. Typical types and applications of pressure vessels: (a) vertical cylinders (gas cylinders used in the laboratory) [35], (b) vertical cylinders (stationary industrial storage cylinders) [38], (c) horizontal cylinders (cylinders for transportation of gas and liquid) [36], (d) horizontal cylinders (vehicle cylinders) [37], and (e) spherical cylinders (oil refinery cylinders) [39].

Table 2
Some examples of pressure vessel suppliers, their products, manufacturing methods, and potential applications.

Suppliers	Materials	Applications	Capacity (L)	Pressure (bar)	Manufacturing methods
Steelhead Composites [40]	Composite	Lightweight hydrogen storage	6–270	300, 500, 700	Metal spin forming, filament winding
Thielmann [41]	Stainless steel	Butane, propane, cooking, lighting, heating	27–36	30	/
MS Group [42]	Welded steel	Home cooking	/	/	Deep drawing
NEOS [43]	Steel and Aluminium	Air receivers, Large distillation columns	/	/	Rolled and welded tube
Luxfer [44]	AA6061	Oxygen cylinders	/	/	Backward extrusion
Ferromaxx [45]	Steel	Argon gas storage	30, 37, 50	/	/

Table 3
Classification and applications of different hydrogen tanks.

Type	Schematic	Materials			Maximum pressure (bar)	Applications
		Metal	Composite	Polymer		
I		Steel/Al	/	/	Al: 175 Steel: 200	Submarine applications [46]
II		Steel/Al liner	Filament windings around the cylinder part	/	Al/glass: 263 Steel/carbon fibre: 299	Stationary fuel cells and hydrogen (FCH) technologies
III		Al/Steel liner	Composite over-wrap (fibre glass/aramid or carbon fibre)	/	Al/glass: 305 Al/aramid: 438 Al/carbon: 700	Vehicles
IV		/	Composite over-wrap (carbon fibre)	Polymer liner	350 (buses) 700 [30]	Vehicles
V		/	Composite	/	1000	Aerospace applications [47]

needed to store the same mass of hydrogen. Such large tank is not suitable for passenger cars which usually have limited free spaces and thus, 350 bar tank is used more frequently in buses where multiple large tanks can be carried to store enough hydrogen for a fixed routine [51].

Hydrogen tanks of 700 bar are commonly used for passenger cars. Several largest car manufacturers are now transferring from Type III to Type IV due to the lower specific cost and weight of the latter as seen from Fig. 3(a) and (b), respectively. For both types, carbon fibre contributes to more than 90% of the total materials cost and as the development of carbon fibre is relatively mature currently, it is unlikely that the price of this material will drop significantly in the near future [27]. Type III uses metallic liners which can bear more load itself as compared to polymer liners. Thus, it is expected that thinner carbon fibre layer is needed to withstand the required internal pressures and the total cost of Type III can be potentially reduced. However, in industry, companies tend to make the composite layer thicker to ensure maximum safety. Also, as Type IV tanks are developed more recently and the main purpose is to reduce weight and cost, the design is smarter as compared to that of Type III and less carbon fibre is actually used as indicated by Fig. 3(a). Together with the higher cost and density of aluminium alloys as compared to polymer, the cost and weight of Type III tanks are thus higher than those of Type IV as a result. However, with more efficient tank design, it is theoretically that less carbon fibre can be used to make Type III a more economical choice instead, and the metallic liners can find a wider application as compared to the polymer liners. Besides the material cost, the processing cost also contributes to about half of the total product cost and there are higher possibilities for cost reduction by developing advanced manufacturing techniques as compared to materials. Thus, to promote the uptake of FCEVs in the automotive market, novel manufacturing methods of hydrogen tanks need to be developed.

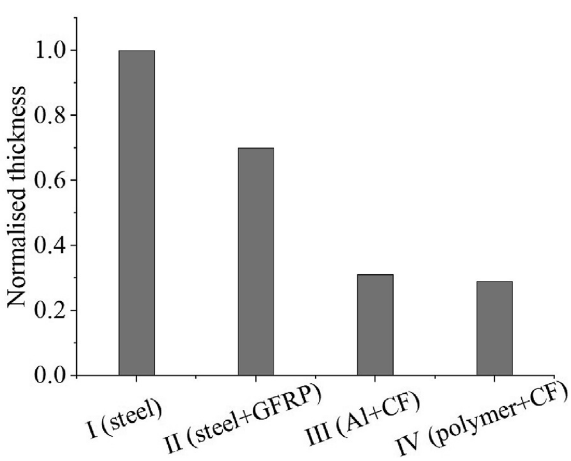


Fig. 2. Comparison of the overall thickness of different types of hydrogen tanks to withstand the same internal pressure of 350 bar, normalised with respect to the type I (steel) tank. Derived from Ref. [49].

2.4. Failure mechanisms of hydrogen tanks

As hydrogen is highly flammable, failure of the tanks and leakage of hydrogen gas need to be seriously prevented to ensure the safety of passengers. Hydrogen embrittlement (HE) is a main failure mode of hydrogen tanks; it is a phenomenon where crack is induced by hydrogen to cause fracture in metals [52]. This normally takes place in steels, but it was experimentally detected that aluminium alloy tanks used in vehicles can also be embrittled by gaseous hydrogen under high pressures [53]. The main HE mechanism under high hydrogen pressures is the hydrogen enhanced decohesion mechanism (HEDE) as illustrated in Fig. 4(a) [54]. During the manufacturing process of the aluminium alloy tanks, microcracks are formed at the corners as shown in Fig. 4(b). Under high pressures, hydrogen atoms move towards the tip of these microcracks through stress-induced diffusion. The accumulation of hydrogen atoms reduces the interatomic bonding strength at the crack tip and causes crack propagation along grain boundaries under stresses lower than the critical value [53]. As a result, intergranular fracture as shown in Fig. 4(c) occurs in the tanks. Such fracture allows hydrogen to diffuse out of the tank easily. Any leakage of hydrogen from pressure vessels will soon fulfil the closed environment with high concentration. Hydrogen is highly flammable and can be easily ignited to cause explosion. Thus, HE must be strictly prevented if aluminium alloy tanks are used.

Some prevention measures include improving the properties of the extruded aluminium alloy parts and applying a coating on the

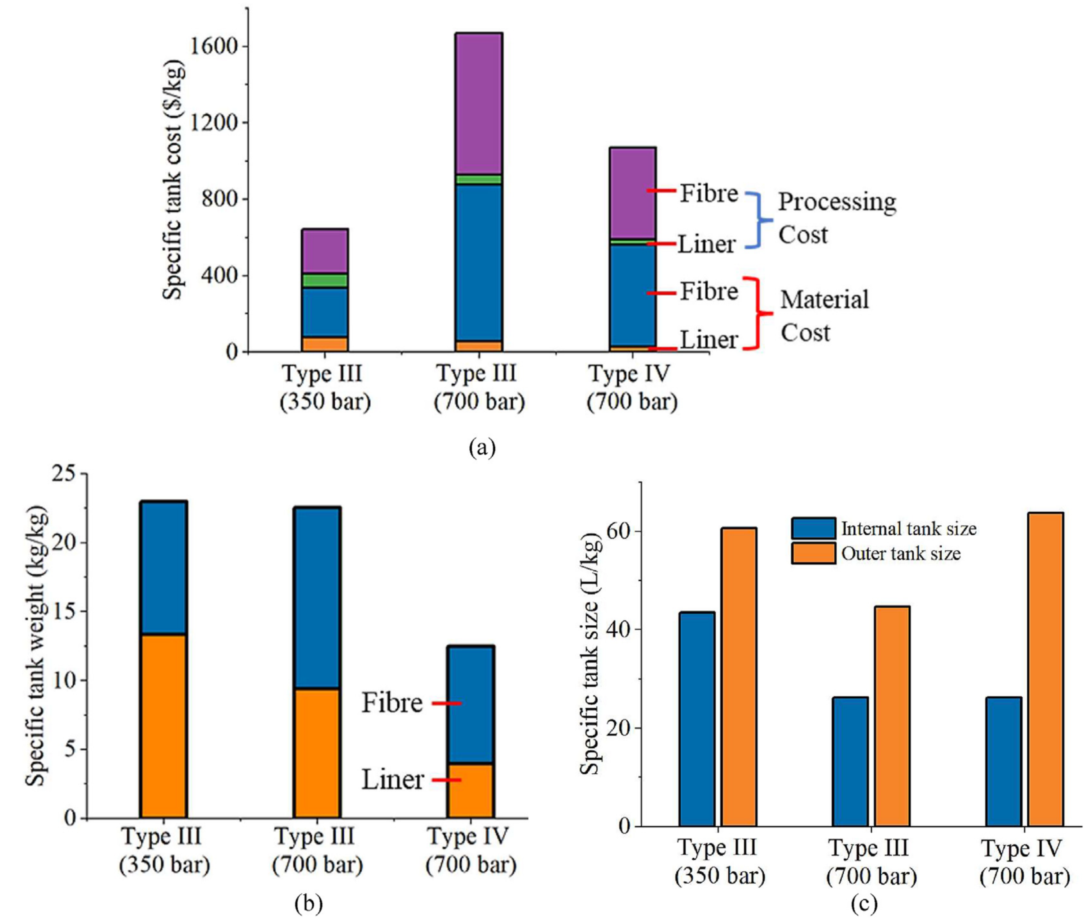


Fig. 3. Comparison of (a) specific costs, (b) specific weights, and (c) specific sizes of the three types of commonly used hydrogen tanks for storing 1 kg hydrogen.

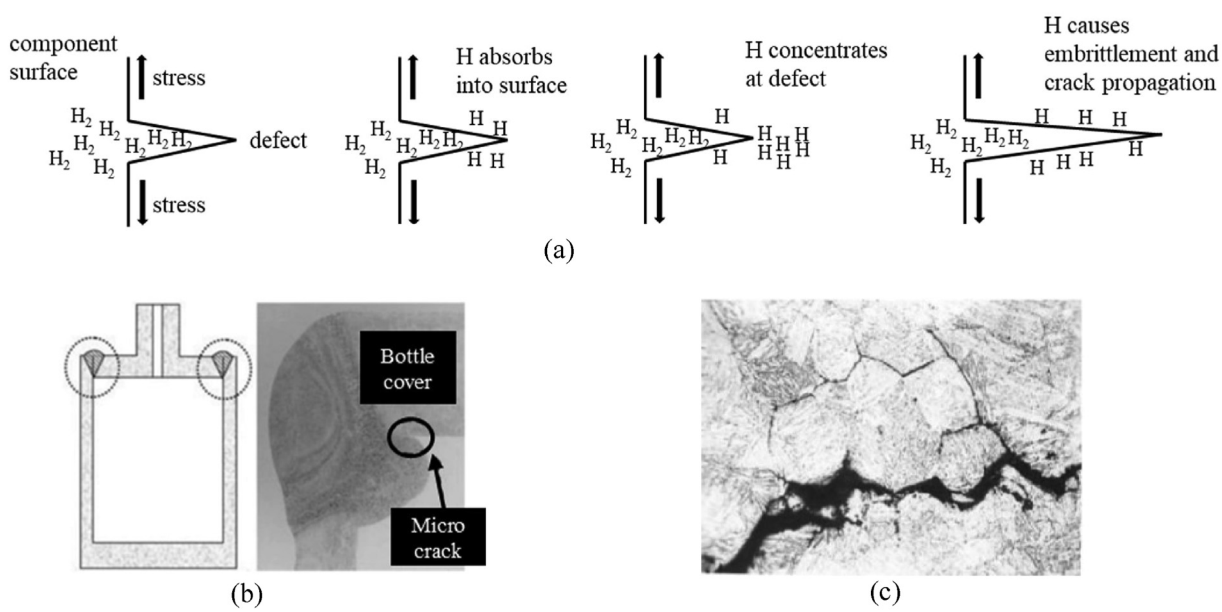


Fig. 4. Hydrogen embrittlement (HE) mechanism and its effect on hydrogen tanks: (a) hydrogen enhanced decohesion mechanism (HEDE), derived from Ref. [54], (b) microcrack at vessel corner [55], and (c) intergranular fracture [54].

alloy surface. The mechanical properties of aluminium alloys can be significantly improved through heat treatment. It was experimentally found that after artificially ageing within a temperature range of 140–170 °C, AA7175 achieved a higher strength and a better resistance to HE [56]. Another method is to refine the grain size. It was found that hydrogen atoms are more active in large grains [57].

Thus, by reducing the grain size, the movement of hydrogen atoms can be suppressed and the alloy's resistance to HE can be improved [58]. Coatings with low hydrogen permeability such as nitride or aluminium oxide can also be applied to the alloy surface to prevent the diffusion of hydrogen into the alloy, thus, increasing the resistance to HE [53]. With a better HE resistance, the application of aluminium alloy tanks can be expanded to more industrial sectors. However, the main promotion of HE is the presence of microcracks which are normally formed during the manufacturing process. The most important prevention measure is therefore to improve the manufacturing techniques to minimise the amount of microcracks formed during the tank manufacturing.

3. Manufacturing methods for gaseous tanks

There are three methods that are commonly used to manufacture cylindrical tanks: roll forming, deep drawing and ironing (DDI), and backward extrusion, as shown in Fig. 5, and a comparison of different cylinder-manufacturing processes is summarised in Table 4.

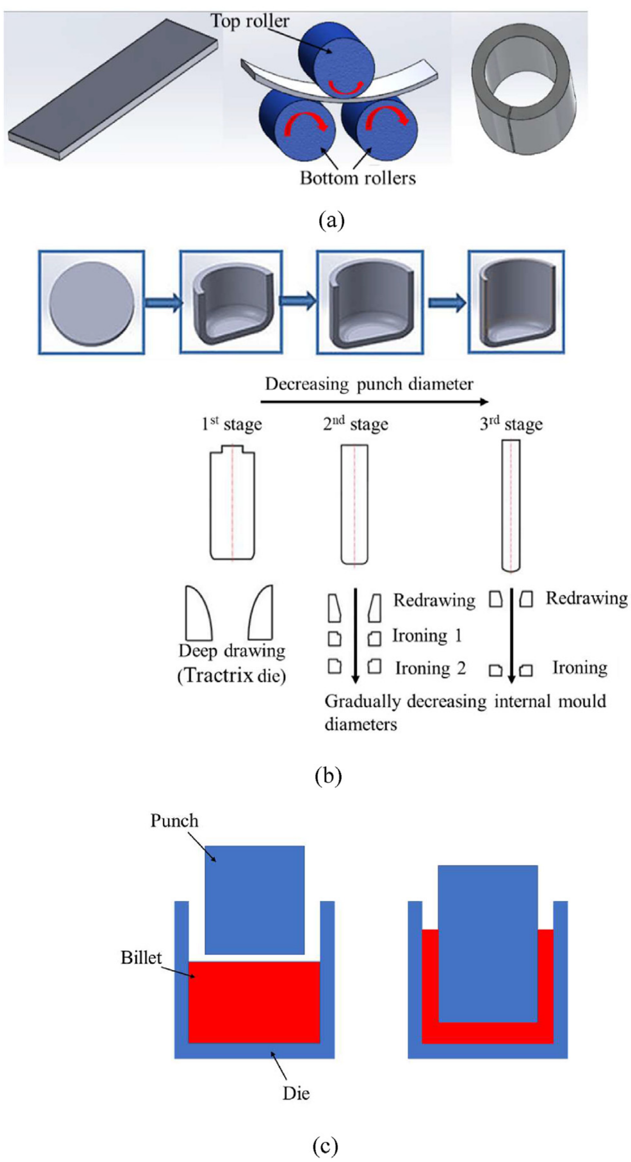


Fig. 5. Illustration of different manufacturing methods for producing cylinders: (a) roll forming, (b) deep drawing and ironing (DDI), replotted from Refs. [59,60], and (c) backward Extrusion.

Huge tanks used for transportation and stationary storage of hydrogen are commonly made by the roll forming technique shown in Fig. 5(a). During roll forming, large metal plates are cold rolled by a three-roll steel rolling mill at room temperature. The bottom rollers are driven by electric motors, and the top roller can move up and down vertically. The top and bottom rollers rotate in opposite directions and provide a (three-point) bending force which gives the plates a curved shape [61]. The curvature of the plate is controlled by adjusting the relative position between the top roller and the two bottom rollers [16]. The two ends of each formed profile and multiple segments of the cylinder are then joined together using arc welding, which is a process where metal is melted to bond discrete parts [63]. Such manufacturing method utilises universal equipment available in the industry with a low forming force, which leads to a low processing cost. Multiple cylinder configurations from small to very large size in diameter can be achieved using one machine setup by simply adjusting the roller position. However, during manufacturing, multiple passes are required to obtain the desired shape and thickness, which makes the production process time-consuming. Also, the welding quality may not be constantly good for all products, and the welded regions are prone to the formation of cracks and thus HE and fracture [64].

DDI, a multistage process as illustrated in Fig. 5(b) is one of the methods that can be used to produce cylinders without welding. During the first stage, a metal disk is put into a tractrix die, and a punch stamps the disk into a cylinder that is closed at one end and open at the other end. This forming process is known as deep drawing. Cold deep drawing at room temperature is commonly adopted in the industry to ensure a good surface finish and improved mechanical properties such as higher tensile strength as compared to hot drawing at elevated temperatures [65]. However, as the drawing ratio (diameter of metal disk/punch diameter) of most metals is limited between 1.6 and 2.2 [65], multiple redrawing and ironing stages need to be carried out to achieve the designed long cylinder shape. During the following stages, several punches with gradually decreasing diameters are used to form cylinders with smaller inner diameters. Going down the mould's setup in each stage, the diameter of the dies decreases gradually to push up the outer material to increase the cylinder length without changing the inner diameter. To form the final shape of a gas tank, the open end is then closed by hot spinning during which the metal is heated to increase its ductility and machined to have the designed features [66]. This method utilises a plate preform which gives a uniform wall thickness, but the billet preparation cost is high. The basic cylinder shape can be formed in a few seconds with the punches, which significantly saves the production time as compared to roll forming. Also, welding is not needed, and the product is a single piece of materials with good integrity. The probability of fracture and gas leakage is therefore reduced. However, wrinkles could form on the outer side of the cylinder during ironing, which reduces the mechanical properties of the product and thus increases the risk of HE. Numerous machineries need to be built in advance which requires high preliminary cost. Also, to manufacture tanks with different diameters, new designs of punches and moulds are necessary, which further increases the manufacturing cost [67].

Another method that is increasingly being used in recent decades is backward extrusion, a forging process where products are formed from metal billets [68]. As shown in Fig. 5(c), for conventional backward extrusion, a metal billet is firstly put into a container or die. A punch then strikes the billet and forces the material to flow in the opposite direction of the punch movement [69]. The inner diameter of the cylinder is the same as the diameter of the punch and the thickness is determined by the gap between

Table 4
Comparison of key characteristics of the different cylinder-manufacturing processes.

Process	Roll forming	Deep drawing and ironing (DDI)	Backward extrusion
Process operations	Plate roll bending, welding, and re-rolling	Multistage deep drawing, redrawing and ironing	Single-step backward extrusion
Preform	Rectangular plate	Circular disk	Cast billet
Preform cost	High	High	Low
Forming force	Low	Low	High [61]
Processing cost	Low	Low	High
Tank size	Small to very large	Limited size due to limiting drawing ratio (1.6–2.2) [62]	Limited size due to limited extrusion machine capacity
Manufacture equipment	Flexible. Change cylinder dimensions by adjusting the distance between rollers	Fixed punch and die to give one designed dimension only	Fixed punch and die to give one designed dimension only
Ease of operation	Medium	Multi-stage, complex	Simple
Other advantages	– Simple equipment – Flexible set-up & suitable for low & high-volume productions	– Uniform wall thickness – Good product integrity – Suitable for mass production	– High productivity – Good product integrity – Suitable for mass production
Other disadvantages	– Low productivity – Welding affects product integrity	– Wrinkles may occur – High machine cost – Complex process design – Low flexibility	– High machine cost – Low flexibility

the punch and the container wall. Common metals being extruded by steel punch are aluminium alloys such as the AA6XXX series due to their high ductility and strength-to-weight ratio [48]. Although these alloys are relatively soft with a yield stress of about 241 MPa [70], the force needed for extrusion is still high and can be a few hundred times that of the force needed for rolling and DDI. Small extrusion press normally has a power of around 10 MN. To form billets with a diameter larger than 250 mm, a force of over 50 MN of is required, which requires higher capital investment and running cost compared to smaller press. On the other hand, with high power, a few meters of metals can be extruded per second which ensures high productivity [61].

As compared to roll forming and DDI, backward extrusion uses metal billets instead of metal plates or disks as the starting material. Thus, extra process for material preparation is not required and the cost of the starting material preparation is reduced. Also, both productivity and efficiency in backward extrusion are reported to be high, which makes this technique suitable for mass production [67]. However, similar to the DDI process, the initial investment in machines is high and varying tank geometries and dimensions require redesign of dies and punches. Besides, the wall thickness is less uniform and less controllable, which could result in poor mechanical properties of the final components. Their mechanical properties are also affected by other processing factors such as temperature and extrusion rate [71]. Despite these disadvantages, the simple single-stage operation and the good product integrity as no welding is needed make backward extrusion favourable for manufacturing cylinders. The detailed working process of backward extrusion and its recent development are further discussed in Section 4.

4. Backward extrusion techniques for aluminium alloy containers

This section summarises the working process of backward extrusion and compares the different designs of the backward extrusion proposed recently in reducing the extrusion force.

4.1. Backward extrusion

Over the recent decades, backward extrusion has attracted an increasing attention in the cylinder making industry due to its simple operation and high productivity [61]. Backward extrusion can generally be classified into two categories depending on the operation temperature: cold backward extrusion and hot backward extrusion. Cold extrusion is a process where cold metal billets are extruded by a press at room temperature. During the process, the deformation work can be converted to heat and thus the temperature of the billet might increase by a few hundred degrees but not high enough to cause any microstructural changes [72,73]. Cold extrusion is favoured in the industry as no heating unit is required, which significantly reduces the manufacturing cost. Also, cold extrusion gives near-net shape products with better surface finish and higher production rates as compared to hot extrusion [73]. However, due to the low temperature in cold extrusion, the yield strength of the billet is high, resulting in more difficulty to make the material deform. Thus, a higher extrusion force is required which adds on the manufacturing cost and only limited extent of deformation can be achieved which restricts the product design [74]. To reduce the extrusion force and allow complex product designs, hot extrusion can be employed.

Hot extrusion is a process where the billet is heated to a temperature above the alloy's recrystallisation temperature [72]. With a higher temperature, the billet can be deformed to a larger extent because the ductility of the metal increases, and a lower deformation force is needed as compared to the cold extrusion due to a lower yield strength. Also, at temperatures above the recrystallisation temperature, finer new grains can be generated on the cost of the deformed old grains, and such finer grains could improve the mechanical properties such as the tensile strength of the final products [75].

In the industry, aluminium alloys have been commonly used to extrude cylinders and Fig. 6 shows the processing flow of aluminium alloy cylinders. There are a variety of aluminium alloys with different properties ranging from AA1XXX series to AA9XXX series according to different alloy compositions [76], while AA6XXX is most frequently used due to its superior extrudability, resistance to corrosion and ability to be heat treated to achieve better mechanical properties [77]. During the hot extrusion process, a billet is firstly heated to above the recrystallisation temperature (e.g., 300–600 °C) of the respective alloy type, before filling into the die [72]. Then, the heat-treated billet in the die is extruded by a hydrostatic press. With different alloys, as well as different billet sizes, the extrusion force varies from 10 MN to as high as 200 MN [61]. The extrusion rate can reach up to 1.5 m/s with sufficiently high extrusion force and for soft metals, the extrusion ratio (billet cross sectional area/product cross sectional area) can reach up to 100:1 [78]. After extrusion, the product is quenched to lock its

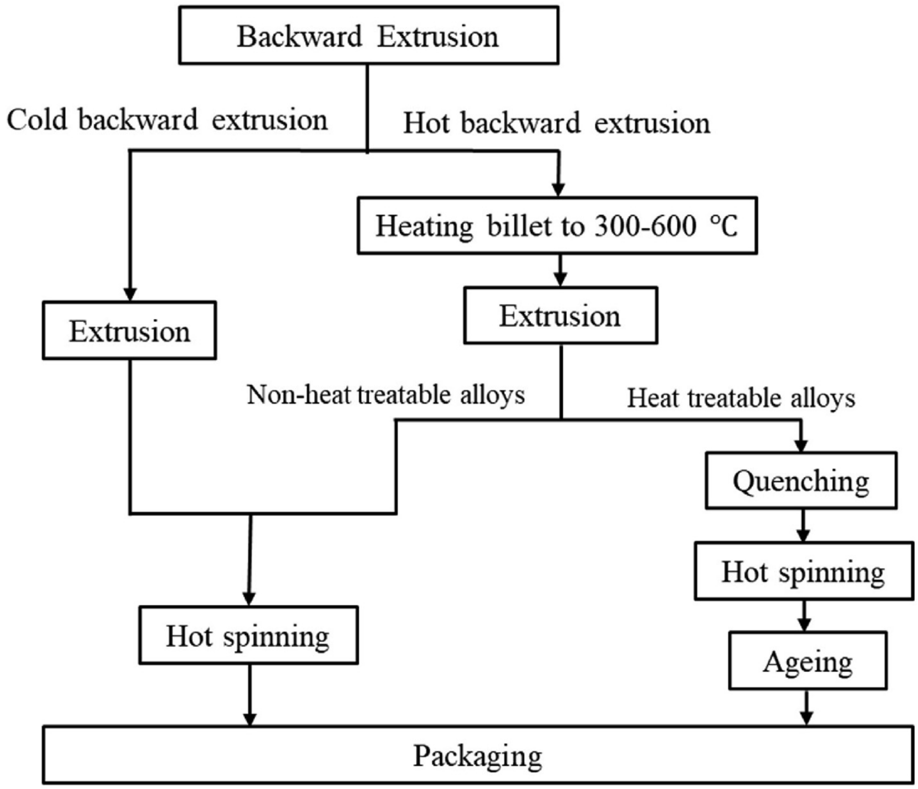


Fig. 6. Flowchart for processing aluminium alloy cylinders.

microstructure and followed by hot spinning to close the open end. Finally, before packaging, the heat treatable alloys are aged to further improve their mechanical properties [79].

4.2. Microstructural evolution during extrusion

Grain refinement is one of the biggest advantages of hot extrusion. As an example, Fig. 7 shows the microstructures of Al-0.2Sc-0.1Zr alloy before and after accumulative continuous extrusion forming, characterised by Guan and Tie [80]. The results illustrate that the average grain size is reduced significantly from about 100 μm before extrusion to about 0.8 μm after extrusion. Such grain refinement phenomenon normally occurs through a mechanism known as dynamic recrystallisation (DRX) [81]. During hot extrusion of aluminium alloys, one of the main mechanisms of grain refinement is continuous dynamics recrystallisation (CDRX) [82]. The continuous deformation of metal billets causes the generation of a large number of geometrically necessary dislocations (GNDs). Dynamic recovery (DRV) then takes place to annihilate these dislocations and subgrains with low angle boundaries (LAGBs) are formed due to the rearrangement of the dislocations by cross-slipping and climbing. Upon further deformation, misorientation of such subgrains increases continuously due to the occurrence of CDRX, and these subgrains then become new grains with high angle boundaries (HAGBs) and replace the old, deformed grains [83]. Due to the rapid extrusion rate, there is limited time for grain growth. Thus, the newly formed grains are much smaller in size as compared to the original grains.

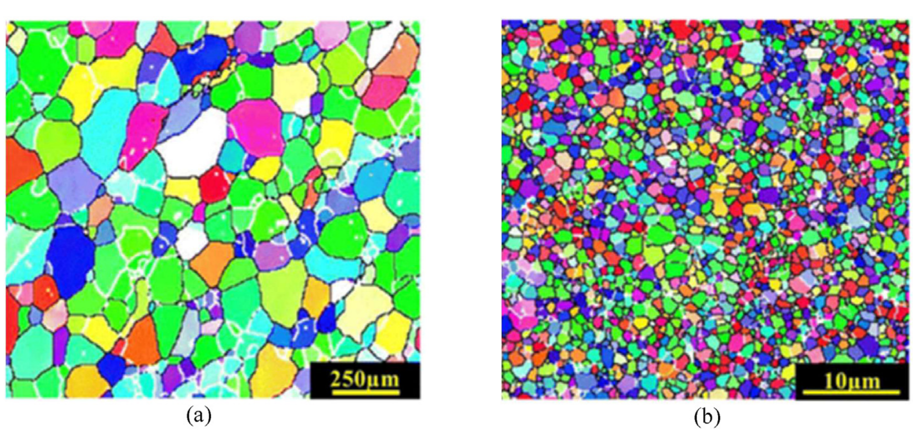


Fig. 7. EBSD maps of Al-0.2Sc-0.1Zr alloy showing the grain refinement (a) before and (b) after extrusion [80]. The average grain size is about 100 μm before extrusion and 0.8 μm after extrusion.

Another possible dynamic recrystallisation mechanism in hot extrusion of aluminium alloys is the geometric dynamic recrystallisation (GDRX). During hot extrusion, the aluminium alloys (e.g., AA6061) experience severe plastic deformation, and the grains are highly elongated and thinned with serrations. When the grain thickness is low enough namely below 1–2 times of subgrain size, the top and bottom grain boundaries can touch each other and break the elongated grains into smaller grains and such phenomenon is called pinch off [84]. As a result, the average grain size of the final product is reduced.

It is widely accepted that the grain size has a significant effect on the yield strength of materials according to the Hall-Petch equation $\sigma_y = \sigma_0 + k/\sqrt{d}$ where σ_y is the yield strength when average grain diameter becomes d, σ_0 is the yield strength for original materials, and k is a material constant [85]. According to the equation, with a smaller grain size, the yield strength of materials increases. Understanding what parameters affect the final grain size during extrusion is important for the achievement of better grain refinement and improved mechanical properties. Foydl et al. [84] examined the effects of temperature, strain rate and effective strain on the grain size refinement and the results are summarised in Fig. 8. From the experimental results, it is observed that with the same effective strain, changing extrusion temperature and extrusion rate does not have a clear effect on the grain size. In contrast, the grain size varies significantly with effective strain. Specifically, as the effective strain increases, the grain thickness decreases at a decreasing rate. At an effective strain value higher than 4, the reduction in grain thickness is no longer obvious. However, the evolution in grain length varies differently as compared to grain thickness. Initially, as the effective strain increases, the grain length increases as well due to the elongation of grains during extrusion. When the effective strain is above 3.5, the grain thickness is very low and the grain boundaries can touch each other to form smaller grains through GDRX, and the grain length starts to decrease [84]. Thus, an effective strain greater than 3.5 can considerably enhance the grain refinement effect. Lewandowska et al. [86–88] tested the potential of extrusion in reducing grain size and improving tensile strength of different materials with an effective strain of 3.8 and the results are summarised in Fig. 9. It is observed that with proper extrusion operation, nano-scale grain size could possibly be achieved in the laboratory, and such small grain size can greatly increase the yield strength of materials.

4.3. Modified backward extrusion designs

4.3.1. Reduction of extrusion force

The main disadvantage of backward extrusion is the high processing force. Many efforts have been dedicated for the reduction of the extrusion force. One of the effective methods is to improve the equipment design. Table 5 summarises several more recent backward extrusion designs proposed by different research groups to reduce the extrusion force.

The rotating backward extrusion (RBE) was proposed by Che et al. [89] and distinct from the conventional backward extrusion method; it uses a rotating open punch and a moveable die. During the RBE process, the punch rotates, and the die moves up to extrude the alloy. Due to the rotational movement of the punch, the billet experiences a complex stress state including both the shear stress and the compressive stress. According to the yield criterion, the primary stress of the billet is thus reduced, and lower force is required to deform the material. Besides, the rotating of the punch increases the temperature of the billet due to friction, and thus, the yield strength of the billet decreases. With lower yield strength, the material can flow more easily and thus, the extrusion force is reduced significantly. According to the experimental results reported by Che et al. [89], the extrusion force of AZ80 alloy was successfully reduced by 31.6% by using RBE. However, this extrusion machine and process design is much more complicated as compared to the conventional backward extrusion method. Also, high punch wear is encountered due to its rotation and frequent replacement of the punch causes an increase in the manufacturing cost. In addition, due to the slippage between the billet surface and the punch surface, not all rotational work is transferred into the billet for deformation, and thus the production efficiency is reduced as compared to the conventional backward extrusion method.

The hollow-cylinder backward extrusion was proposed by Wang et al. [90] to reduce extrusion force through reducing the contact area between the punch and the billet. The hollow-cylinder method has an additional mandrel in the hollow punch. As such, the extruded area of the billet is greatly reduced, and a lower extrusion force is required accordingly. The extrusion force can be further adjusted by changing the diameter of the mandrel. Also, with the internal mandrel, the diameter of the punch is larger and the mandrel act as a support for the punch. Thus, the problem of punch buckling is minimised. This method is suitable for producing

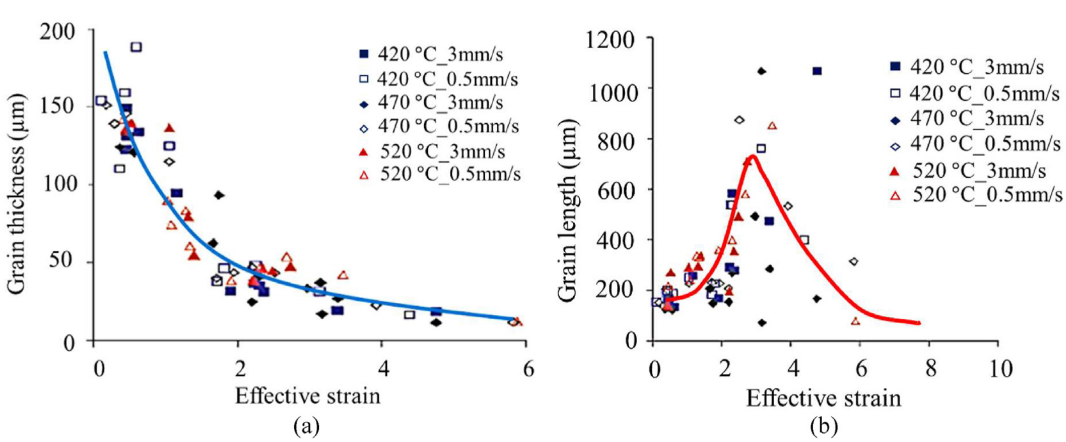


Fig. 8. The evolution of (a) grain thickness and (b) grain length with effective strain under different forward extrusion temperatures (420–520 °C) and speeds (0.5–3 mm/s) with an extrusion ratio of 11 for AA7020 in a cylinder profile of 6 mm diameter [84].

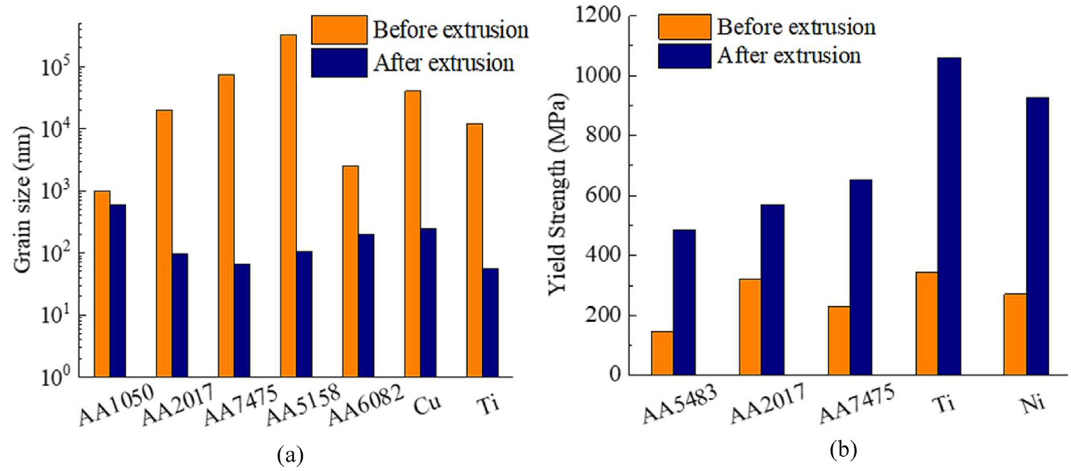
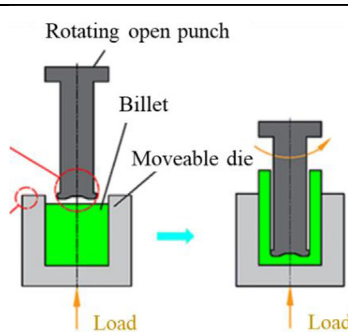
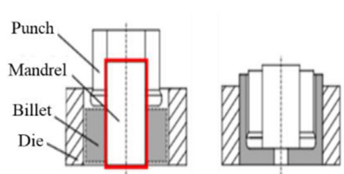
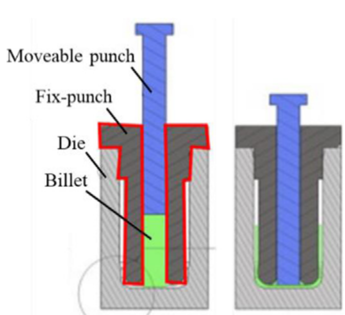


Fig. 9. Comparison of the (a) grain size and (b) yield strength of different materials before and after extrusion with the same effective strain of 3.8 [86–88].

Table 5
More recent backward extrusion designs for the reduction of extrusion force.

Designs	Schematic	Operation features	Merits and distinct features	Disadvantages
Rotating Backward Extrusion (RBE) [89]		The punch rotates to change the stress state of the billet	– High shear strain due to punch rotation – Low extrusion force due to low primary stress	– Complicated extrusion machine & process design – High punch wear due to rotation – Relatively low production efficiency
Hollow-cylinder Backward Extrusion [90]		Reduce extrusion force by reducing extrusion area using an internal mandrel	– Adjust extrusion force by changing mandrel diameter – Hollow punch with a mandrel to minimise buckling – Suitable for large diameter products	– V design
Small-diameter Backward Extrusion [91]		Reduce extrusion force by reducing billet diameter	– Low extrusion force	– Restricted product size – Buckling of the moveable punch – Failure of the fix-punch under high hoop stress

cylinders with large diameters. However, the starting billet preform required for this method is a hollow cylinder. Thus, the billet preparation is complex and costly. The tool design is also much more complicated as compared to the conventional backward extrusion method as both the punch and the mandrel need to be movable. In addition, further work is needed to close the hole at the bottom of the cylinder to be used for storage tank.

Besides the hollow-cylinder backward extrusion, Shatermashhadi et al. [91] proposed the small-diameter backward extrusion to reduce the extrusion area. This design utilises a fix-punch to reduce the diameter of the billet while keeping the inner diameter of the final product unchanged. By reducing the diameter to about one third of the original billet, the extrusion force is reduced to about a quarter of the conventional backward extrusion method [91]. However, the additional fix-punch makes the tool design more complicated as compared to the conventional backward extrusion method, but simpler than the rotating and the hollow-cylinder backward extrusions discussed above. Also, the closed fix-punch restricts the product size that can be extruded, and a longer and thinner punch is needed to strike the smaller-diameter billet, which makes the punch prone to buckling. Moreover, the hollow fix-punch will experience a high hoop stress during the process due to high extrusion force, and it thus needs to be thick enough to prevent burst of the hollow structure.

Other than changing tool designs, the extrusion force can also be reduced by controlling the extrusion parameters such as temperature and speed. As indicated in Fig. 10, both initial billet temperature and friction coefficient can affect the extrusion force. Specifically, with a higher billet temperature, the yield strength of the AA2024 alloy is lower, and the billet can be extruded more easily with a lower extrusion force. During extrusion, friction is found between the billet and the tool surface, and with a higher friction coefficient, a higher extrusion force is required. Such friction during extrusion is normally handled by applying a phosphate coating for lubrication [92]. However, this coating contains heavy metals and could cause environmental problems [93]. Over the years, liquid lubrication systems such as oil are continuously investigated and improved to ensure efficiency and enhance the tool life [94]. Uyyuru and Valberg [95] demonstrated that by eliminating the friction, the extrusion force can be reduced by 25%.

4.3.2. Increase of effective strain

Apart from the force reduction, some research groups have also worked to increase the effective strain of the backward extrusion for grain refinement. One popular design is the accumulative backward extrusion (ABE) as shown in Fig. 11 as an example. This novel setup has an additional hollow outer punch as compared to the conventional backward extrusion design. During the ABE extrusion, the inner punch firstly (Stage I) moves down, and the outer punch moves up to extrude a billet into a cup shape, and high plastic strains are induced due to the severe deformation. In Stage II, the billet is flattened and pressed into the original shape by the downward motion of the outer punch and upward motion of the inner punch. This process can be repeated several times and the plastic strain induced during each cycle can be accumulated to give a large overall effective strain. With a high effective strain, ultra-fine grain structure can be formed due to the occurrence of DRX to give the final product superior mechanical properties [97]. Faraji

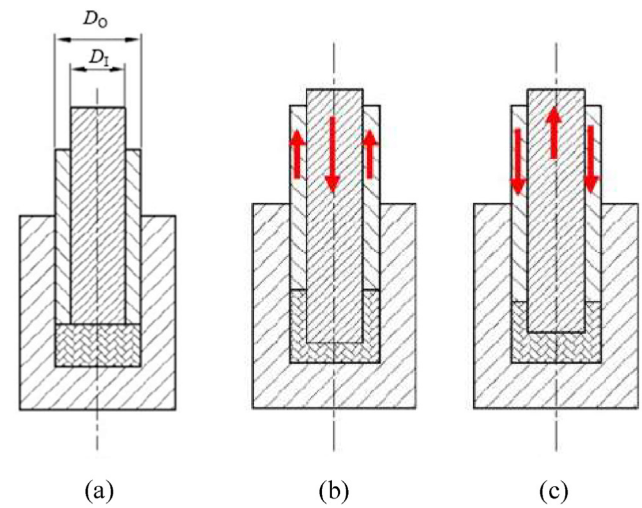


Fig. 11. Schematic illustration of the accumulative backward extrusion (ABE) process where D_O is the outer punch diameter and D_I is the inner punch diameter [97]: (a) Initial State, (b) Stage I, and (c) Stage II.

et al. [97] used this process and refined the average grain size of magnesium alloy AZ91 from 150 μm to 1.5 μm. Fatemi-Varzaneh and Zarei-Hanzaki [98] successfully extruded magnesium alloy AZ31 using ABE and they found that a single cycle operation can refine the grain size from 35 μm to 1 μm.

Besides the major changes on tool setup, other small features have been added to the conventional backward extrusion design to increase the effective strain or to minimise other technical problems. Some examples are shown in Fig. 12. Shatermashhadi et al. [91] changed the sharp corners of the punch and the die into rounded corners (Fig. 12(a)). Both the inner and outer radii of the punch and the matrix radius, as indicated in Fig. 12(a) help to promote the material flow. This makes the extrusion of the billet easier and reduces the extrusion force as well as increases the extrudability. Importantly, higher effective strain could be achieved, and grain size can be refined further to give better mechanical properties. In addition, voids can usually be formed during extrusion if there is a lack of material flowing to the right position. Cracks and fractures can then initiate, leading to failure of the product at a stress below the designed value. The better material flow shown in Fig. 12(a) prevents the formation of voids. Another example is that Labanova et al. [68] added a moveable punch sleeve to the conventional backward extrusion design, as shown in Fig. 12(b). The punch sleeve acts as a support for the thin punch and restricts the bending force which thus prevents the buckling of the punch. As such, both the replacement and maintenance cost are reduced.

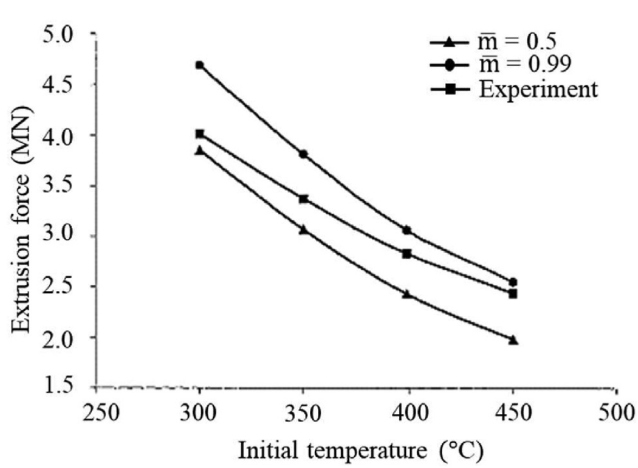


Fig. 10. Effect of initial billet temperature and friction coefficient $\overline{m}$ on the extrusion force of AA2024. The extrusion ratio is 30:1 and the punch speed is 3 mm/s [96].

4.3.3. Manufacture of products with complex shapes

The backward extrusion designs have also been modified to manufacture products with different shapes besides cylinders, and some examples are shown in Fig. 13. One simplest example is the production of metal rods using backward extrusion as illustrated in Fig. 13(a). The punch is designed to be hollow in the middle and there is no gap between the die surface and the container wall. When the punch moves into the container, the material flows through the hollow section in the opposite direction to form solid rods. The billet is stationary in relation to the container and thus, friction between the billet surface and the container wall is eliminated, unlike forward extrusion. As a result, the extrusion force can be reduced significantly [99]. Another example is the double-

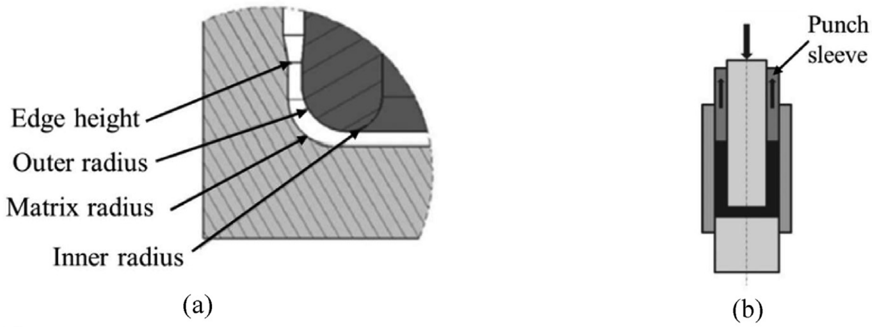


Fig. 12. More recent backward extrusion design features to promote material flow and prevent tool failure: (a) edge with radius [91] and (b) punch sleeve [68].

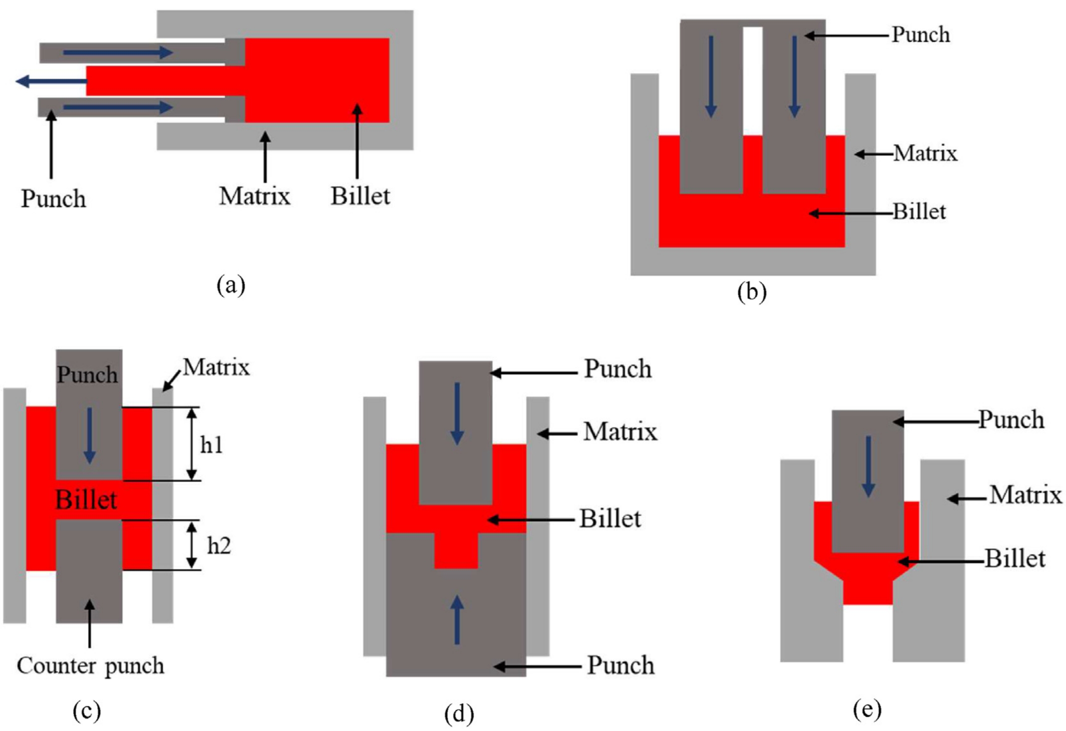


Fig. 13. Examples of modified backward extrusion designs to manufacture products with complex shapes: (a) backward bar extrusion [104], (b) double-backward extrusion [100], (c) double-cup extrusion [100], (d) double-punch extrusion [102], and (e) changing-radii extrusion [103].

backward extrusion which is the combination of both backward bar and cylinder extrusion as illustrated in Fig. 13(b). When the hollow punch moves into the die, the material flows through the central opening of the punch as well as through the gaps between the punch and the die. As such, cylinders with two cavities can be produced [100]. Other examples include the use of combined forward-backward extrusion to produce irregular cylinders as illustrated in Fig. 13(b)–(d). Design in Fig. 13(c) is a double-cup extrusion, and it contains a punch and a counter punch both with flat surfaces and the same diameter. The punches are positioned in the middle of the container, having gaps with the container wall on both sides of the punches. During extrusion, the upper punch moves down, and the counter punch is stationary. The material flows through the gaps and forms two cups with a common base. The ratio of h1/h2 (upper cup depth/lower cup depth) is determined by the friction between the billet surface and the container wall [101]. Design in Fig. 13(d) is similar to design 13(c) but with different counter punch designs. As the upper punch moves down, the material flows both backward through the gaps between the upper punch and container walls as well as forward into the cavity of the counter punch to form a cylinder with a small extrusion at the bottom [102]. Design in Fig. 13(e) produces cylinders with a similar shape to that of 13(d) but has one punch only. The internal radius of the die decreases from top to bottom. As the punch moves down, part of the material is extruded forward into the die cavity with smaller radii. The slope between the larger radius and the smaller radius block some of the material and force those material parts to flow backwards through the gaps between the punch and die walls. As such, cylinders with a smaller base are produced [103]. From these examples, it can be concluded that backward extrusion has a great potential to produce cylinders with complex shapes using modified tool designs.

5. Conclusions and perspectives

Hydrogen is a clean and renewable energy source that has great potential to replace fossil fuels. One of the promising applications of hydrogen is the fuel for fuel cell electric vehicles (FCEVs). In this review paper, different hydrogen storage tanks and the manufacturing methods of the associated aluminium alloy liners are discussed. Some key conclusions are summarised:

1. Hydrogen tanks can be classified into five types with Type III and IV being used for vehicles. Type IV tank with polymer liner is currently analysed to be more cost effective than Type III with metal liner due to similar carbon fibre thickness used for both types. However, considering that the metallic liners can withstand some load itself, there is a potential to reduce the cost of Type III tank with less amount of carbon fibre used, by improving the mechanical properties of the metallic liner through better processing designs which may also reduce the metal liner manufacturing cost.
2. Aluminium alloys are prone to hydrogen embrittlement (HE) through the hydrogen enhanced decohesion mechanism (HEDE) under high pressure. HE can lead to the formation of cracks and leakage of hydrogen gas which is dangerous. Thus, other than cost reduction, properties of the metallic liner need be improved to ensure safety at high pressure, and prevention measures need be taken to avoid HE.
3. Common manufacturing methods of metallic liners include roll forming, deep drawing and ironing (DDI), and backward extrusion. Roll forming is a simple process and can produce tanks with small to very large sizes. However, its productivity is low and poor welding quality could affect the product integrity. To avoid welding, DDI and backward extrusion can be used instead. Both techniques are suitable for mass production with high productivity. Although the tank size is restricted by the equipment size and the limiting drawing ratio of materials, it is still within the size range of on-board hydrogen tanks. Comparing these two methods, traditional backward extrusion requires higher processing force, but its operation is much easier and the simple billet preform helps save cost. Thus, backward extrusion might be the most promising manufacturing method for hydrogen tanks.
4. Improving tool designs can significantly reduce the extrusion force of backward extrusion. Several novel methods have been proposed but none is used commercially for mass production. One method is to employ a rotational punch, but this could cause severe punch wear and the tool design is very complicated. Other simple and straightforward designs include applying a fix-punch or using a hollow punch to reduce the contact area between the punch and the billet, but the product size is restricted.
5. Backward extrusion also has the advantage of grain refinement to improve the mechanical properties of the final products. With novel tool designs such as the accumulative backward extrusion process, the effective strain can be increased significantly, and the grain size can be refined to sub-micro range.

In conclusion, backward extrusion is a promising method for the mass production of metallic liners for hydrogen tanks. The future development of this technique lies in reducing extrusion force to minimise processing cost, increasing deformation strain to improve product mechanical properties, and enhancing the flexibility of tool designs to produce more complex geometries in one-single stage. With the commercialisation of advanced backward extrusion designs, the overall cost of FCEVs can be reduced, and the safety level can be improved. As such, the adoption of FCEVs in the market can be promoted to help with carbon-emission reduction.

Conflicts of interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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