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Storage system of renewable energy generated hydrogen for chemical industry

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Abstract

Chemical industry is the base of the value chains, and has strong influence on the competitiveness of almost all branches in economics. To develop the technologies for sustainability and climate protection and at the same time to guarantee the supply of raw material is a big challenge for chemical industry. In the project CO2RRECT (CO2 - Reaction using Regenerative Energies and Catalytic Technologies), funded by the German federal ministry of Education and Research, carbon dioxide is used as the source of carbon for chemical products with certain chemical processes. Hydrogen that is needed in these processes is produced by electrolyzing water with renewable energy. To store a large amount of hydrogen, different storage systems are studied in this project, including liquid hydrogen tanks/cryo tanks, high pressure tanks, pipelines and salt cavities. These systems are analyzed and compared considering their storage capacity, system costs, advantages and disadvantages. To analyze capital and operational expenditure of the hydrogen storage systems a calculation methodology is also developed in this work.

Keywords: hydrogen storage; storage capacity; cost analysis; system comparison

1. Introduction

Fossil fuel-based power plants produces large amount of carbon dioxide and this aggravates the greenhouse effect and speeds up global climate change. The usage of the exhausted carbon dioxide is one solution to diminish the negative effect to environment. On the other hand, renewable energy is going to be the main energy resource because of the limited petroleum reservoir on the earth. But many renewable energy sources, such as wind energy and solar energy, are highly fluctuant and difficult to forecast. To store the excess renewable energy is also a challenge. In the study under the framework of the project

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CO2RRECT (CO2 - Reaction using Regenerative Energies and Catalytic Technologies) the excess renewable energy is used to electrolyze water to produce hydrogen. The hydrogen reacts with carbon dioxide to produce raw materials for the chemical industry, such as carbon monoxide and formic acid. Because of the variability of renewable energy a hydrogen storage system is necessary for the surplus hydrogen. There are in general three categories for hydrogen storage: physical storage, adsorption storage and chemical storage. In physical storage system hydrogen is either cooled down through heat exchanger to be stored in liquid hydrogen tanks (LH2) [1][2], or compressed by high pressure to be stored in compressed gaseous hydrogen tanks (CGH2) [2][3][4] or in salt cavities [5][6][7]. Cryo-compressed hydrogen (CcH2) storage system is a combination of liquid hydrogen and compressed gaseous hydrogen storage systems [8].Taking full advantage of low temperature and high pressure CcH2 can store hydrogen under a lower pressure than compressed gaseous hydrogen storage system at a higher temperature than liquid hydrogen storage system that lowers the technical difficulties for liquefication and compression. In adsorption storage system hydrogen is bound on the surface of the adsorbent by high pressure at certain temperature. Carbon nanofiber (CNF) [1], metal organic frameworks (MOFs) [9][10] and zeolite [11] belong to this category. In the last storage type chemical storage hydrogen is bound with materials through chemical bonding. The technologies are for example metal hydride storage (MH2) [2][12], chemical hydride storage [13][14][15], liquid organic hydride storage (e.g. methanol) [16][17], and iron sponge storage [16]. Cryo-compressed hydrogen storage in category physical storage, adsorption storage and chemical storage are still in development and unsuitable for large scale hydrogen storage in range of tons. Therefore, in this work only liquid hydrogen storage, compressed gaseous hydrogen storage and salt cavity are studied. Besides the hydrogen storage peripherals are also taken into account for the cost analysis including purification plant, liquefier, compressor, buffer storage and so on.

In this work, boundary conditions for hydrogen storage system are set up firstly. Then the storage systems between the electrolysis chain and the hydrogen consumption chain in the chemical process with different hydrogen storage technologies are analyzed with the given boundary conditions. Finally, a calculation methodology is developed for the comparison of the capital expenditure and operational expenditure for the different hydrogen storage systems.

2. Boundary conditions for hydrogen storage system

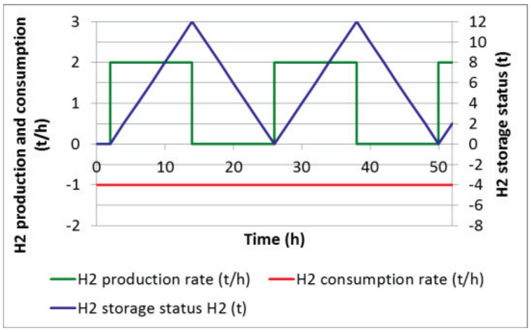


Fig. 1 Boundary conditions for hydrogen storage system

In order to analyze hydrogen storage system, an ideal hydrogen production/consumption profile is set up in Fig. 1. The graphic illustrates the capacity of hydrogen production (green line), hydrogen consumption (red line) and hydrogen storage status (blue line). Hydrogen is produced through the electrolyser 2 t/h continually in the first 12 hours and the electrolyser is shut down in the second half day. Meanwhile the hydrogen consumption is 1 t/h continually for one day. Therefore, the hydrogen storage system must be at least 12 t and the flow rate is 1 t/h both for charging and the discharging. System fluctuations such as electrolysis disruption and consumption interruption are not taken into account in this work. If the storage tolerance should be considered, it is convenient to increase the hydrogen storage volume by specific requirement.

3. Hydrogen purification

In order to avoid the possible disruption and corrosion in hydrogen storage system led by the contamination in hydrogen (in this case water and oxygen), the electrolyser produced hydrogen is purified before it is transported to hydrogen storage system or consumer. In the purification plant the input gas is heated at first to a reaction needed temperature before the reactor converts oxygen and hydrogen into water. Because the reaction is highly exothermic, the temperature of the gas increases further during the reaction. Through the cooler the purified gas is cooled down and the reaction produced water is condensed from the purified gas. The content of the water is reduced further through the drying unit. This kind of purification plant costs 3 to 4 million euros [18]. In this work, the purification plant is used for all the hydrogen storage systems.

4. Liquid hydrogen storage system

In order to store hydrogen in liquid form, hydrogen must be cooled down to -252.8 °C (20.4 K). After liquefication the volume of the gas reduces down to 0.1% [1]. In order to store the liquid hydrogen as long as possible, the cryo tank must be thermally insulated that in general consists of two vacuum separated steel containers. In the vacuum area heat reflecting coating is used to avoid heat conduction and heat radiation. Even with this tank structure the temperature in cryo tank can increase. After-cooling process or hydrogen evaporation process is used to keep the tank temperature at an allowable level. After-cooling is relatively expensive especially for small system. Regarding the second method the evaporated hydrogen gas causes high pressure in the tank, then the hydrogen gas must be discharged to keep the tank pressure at an allowable level, this is so called boil off. The boil off rate is in general between 0.1% and 3% according to the system size. One serious problem of liquid hydrogen storage is the large energy requirement for liquefication that is around 30% of the lower heating value of hydrogen (33.33 kWh/kg) [19]. In addition, for liquefication the purity of hydrogen gas must be higher than 99.999%. Many liquefication processes are available, such as Joule-Thompson-Process, Claude-Process, and Linde-Hampson-Process [20].

4.1. Liquefier

Liquefier is a core component for hydrogen liquefication. The largest liquefier in the world has a liquefication capacity 2.25 t/h (Union Carbide, Linde Div. in USA) [21]. According to the study from LBST a liquefier with possible capacity up to 6.5 t/h costs ca. 140 million euros and requires a floor space of 10,000 m^2 [21].

4.2. Cryo tank and system

The storage volume of a cryo tank is 300 m^3 and it costs about one million euros (3,330 €/$m^3_{geom.}$) [22]. According to the volume specific storage density 64 kg/m^3 [23] the cryo tank can store more than 20 tons hydrogen. To store 12 tons hydrogen with one tank costs 7.5 €/nm^3 (hydrogen density: 0.0899 kg/nm^3) that is more expensive than to store 20 tons hydrogen because of the lower volume usage rate 60%. 100 m^3 are needed to install the cryo tank if it is spheriform, and the life time of the tank is between 20 and 30 years. But the input flow rate is limited to ca. 3 t/h [22]. The liquefier and cryo tank described in [21] and [22] can satisfy the system set up listed in section 2: the flow rate of the liquefier and the cryo tank are 1 t/h. But what should be mentioned is, if the required flow rate of liquefication and cryo tank are higher than the allowable values, a buffer storage is needed to insure the system stability, and the volume of the buffer storage can be calculated after demand. In Fig. 2 buffer storage is introduced into the system between the purification plant and the liquefier. Since hydrogen pressure from electrolysis is at 50 bars, a pressure tank storing hydrogen under 45 bars is one of the optimal buffer storages, because compressor is unnecessary. If the buffer storage is needed, its cost should be taken into account as peripheral cost. The amount of this buffer storage can be calculated based on the cost per nm^3 according to the needed buffer storage volume (cost for pressure tank see section 6.1). In this work, considering the low system set up buffer storage cost is not considered for system analysis. Part of the purified hydrogen is transported directly to the consumer, and the decreased temperature caused by expansion from 45 bars to 20 bars can be compensated with heater (like ambient air or water). The excess hydrogen is liquefied and stored in cryo tank. When needed, the liquid hydrogen can be discharged through a thin cable and heated up by ambient air (heater) to room temperature (see Fig. 2). The heated hydrogen gas must be compressed from 2.5 bars to 20 bars, which is customized to the specified needs in CO2RRECT project. By compression the hydrogen is heated up further, and then the compressed hydrogen must be cooled down by cooler back to room temperature before it is transported to the consumer. In general, a compression plant has already been equipped with a cooler (for the compressor refer to section 5.1). All the main components are counted in the system cost. The advantage of the liquid hydrogen storage system is the low cost and small floor space of the cryo tank. But the flow rate of liquefier and cryo tank are limited; the additional cost for liquefier and energy loss for liquefication are very high, and the floor space for liquefier is pretty large. In addition to this, a compressor is also needed in the system that also increases the total investment. To sum up, liquid hydrogen storage is very expensive.

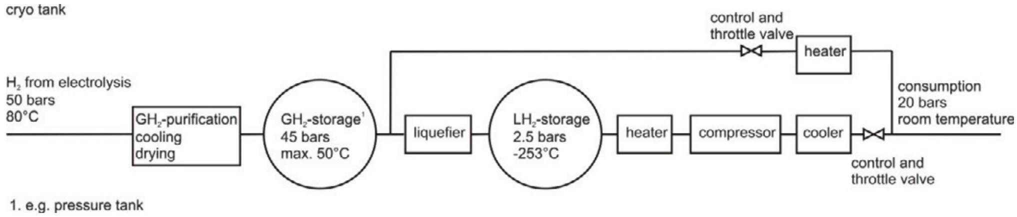


Fig. 2 Liquid hydrogen storage system with buffer storage and liquefier

5. Compressed gaseous hydrogen storage system with buffer storage and compressor

The compressed gas hydrogen storage system is the most common technique used to store hydrogen. Hydrogen is stored in pressure tank under the pressure up to 700 bars. This technique is relatively mature and simple compared to other techniques, but the weight and the cost of the system must be further decreased in the future. In the system with buffer field and salt cavity, hydrogen must be compressed before storage, since the storage pressure is higher than the input hydrogen pressure of 50 bars. If the flow rate of compressor can't meet the requirement of the electrolyser, a buffer storage is needed between the purification plant and the compressor (see Fig. 3). In this work, based on the set up in section 2 the buffer storage is not in consideration for the cost analysis. Because of the temperature raise caused by compression, the hydrogen must be cooled down before charging into the storage. During discharge expansion leads to a temperature decreasement, and then hydrogen must be heated up back to room temperature by a heater (like ambient air or water) when needed before it is transported to customer (see Fig. 3). All the main components are also counted in the system cost.

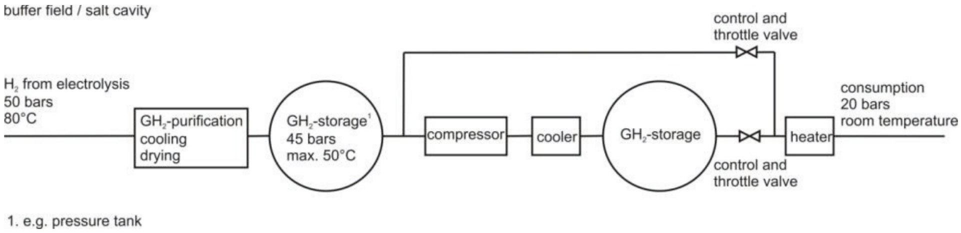


Fig. 3 Compressed gaseous hydrogen storage system with buffer storage and compressor

5.1. Compressor

One core component for high pressure hydrogen storage system is compressor. A reciprocating compressor with a maximal flow rate of 4 t/h and working pressure between 50 bars and 300 bars costs around 3 million euros and the price is going to be doubled according to the experience if it also includes costs of planning, foundation, control system installation, etc., besides the costs for oil plant, pulsation damper, drive system, cooler and instrumentation [24]. 600 m^2 are required for the installation.

5.2. Buffer field

A buffer field system stores hydrogen under the pressure of 300 bars and consists of many bundles, and one bundle consists of 16 compressed gas bottles with 80 liters volume each. To store 12 tons hydrogen 580 m^3 (volumetric density refer to [25]) storage volume i.e. 460 bundles are needed, they cost ca. 4.8 million euros (36 €/nm^3, 8,300 €/$m^3_{geom.}$) [26], and cover an area of 640 m^2. The compressed gas bottles can achieve up to 35,000 cycles. Note that the storage volume 580 m^3 is realizable only when the storage output pressure can reach 0 bars. But in this case the output pressure is 20 bars, that means the tanks can't be discharged down to a pressure below 20 bars. On one hand, it causes an additional investment for the hydrogen to keep the tank pressure at 20 bars that is called cushion gas, and on the other hand, the installation volume must be increased to insure a daily output volume of 12 tons hydrogen. So the total installation volume of the storage should be calculated according to the DOD (depth of discharge) of the storage (calculation details refer to section 7.1). The DOD of buffer field is 90%. The cushion gas is the remaining hydrogen in the tanks under pressure of 20 bars: 1.2 tons. The total thermal energy content of cushion gas is calculated according to the lower heating value of hydrogen 33.33 kWh/kg firstly, and the efficiency of electrolyser 60% for hydrogen production must be also taken into account, then the 1.2 tons cushion gas costs 6,000 € based on an estimated price of wind energy generated electricity 9 €ct/kWh. For the cost analysis the cushion gas cost is converted to €ct/nm^3, that means the cost of cushion gas per normal cubic meter stored hydrogen. This calculated price will not be affected by the changing of installation volume. Buffer field is quite flexible for rebuilding and repositioning. But additional investment and floor space for compressor and additional energy requirement for compression are needed.

5.3. Salt cavity

In general a salt cavity has a volume of about 500,000 m^3, and it is economically inefficient if the volume is smaller than 150,000 m^3 [27]. In this section a salt cavity with volume 500,000 m^3 is considered. Since a compressor is already included in the investment cost of the salt cavity as above-ground facility, the cost of an additional compressor is not considered for salt cavity hydrogen storage system. A salt cavity (500,000 m^3) that works under pressure between 60 bars and 180 bars costs about 25 million euros [27]. In general, the usable volume of the cavity is 70% and the other 30% volume is for cushion gas. If this 70% volume of salt cavity is complete used, it costs 0.5 € per nm^3 or 71 € per m$^3_{\text{geom.}}$. To put the salt cavity into operation, 2,000 tons cushion gas (ca. 10 million euros, for the calculation refer to section 5.2) is needed to keep the pressure in the cavity at least at 60 bars. In this work, salt cavity is used only to store 12 tons hydrogen, the cost is 187 €/nm^3 (volumetric density refer to [25]) that is much more expensive than the price when the cavity volume is completely used. Salt cavity is the cheapest storage per standard cubic meter of hydrogen because of the large storage volume, only if the volume usage rate is high enough. It is worthy to use salt cavity for this project only if the cavity is shared with other projects. At the same time, the salt cavity is geology dependent and requires a long construction period. It takes about 10 years from planning to implementation. Besides this the extra costs for cushion gas and pipeline system that connect the salt cavity and hydrogen consumer are very high. Energy losses exist for compression process. Salt cavity shows outstanding life time despite its relatively high capital cost. The first salt cavity in Germany has worked for more than 40 years.

6. Compressed gaseous hydrogen storage system without buffer storage and compressor

In the system with hydrogen storage under 45 bars compressor is unnecessary, since the storage pressure is lower than the pressure of hydrogen gas from electrolyser. The system shown in Fig. 4 is much simpler than the systems introduced in the above sections. After purification process hydrogen is charged into storage and transported to consumers. A heater is needed to compensate the temperature drop caused by hydrogen expansion.

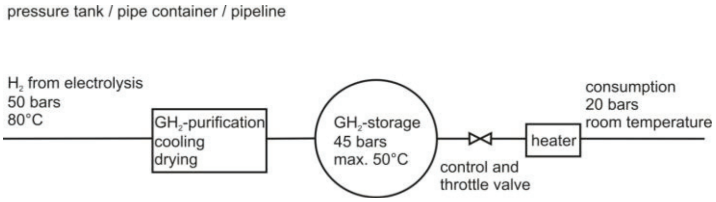


Fig. 4 Compressed gaseous hydrogen storage system without buffer storage and compressor

6.1. Pressure tank

Pressure tanks store hydrogen under a pressure of 45 bars. 3,330 m^3 storage volume is needed for 12 tons hydrogen (volumetric density refer to [25]) that is 29 pressure tanks with 115 m^3 volume each, and the cost is 4.5 million euros in total (33.7 €/nm^3, 1,350 €/m$^3_{\text{geom.}}$) [28]. Horizontal tank system covers an area of 1,600 m^2 and vertical tank system 260 m^2. Up to 155.000 cycles the pressure tanks can be used when the loading condition is between 42 bars and 22 bars [28]. Similar to the buffer field in section 5.2, the total storage volume must be calculated with the DOD of the pressure tank, here is 45%. The cushion gas costs 33,000 euros (for the calculation refers to section 5.2). Besides there is no compressor necessary, the cycle life is long, and the floor space of vertical tank system is small.

6.2. Pipe container

There are already some existing pipe container systems for natural gas storage. This kind of system can also be used for hydrogen storage. Pipe container is similar to pipeline, but built underground in a serpentine shape with larger diameter than pipeline. To store 12 tons hydrogen for example under the pressure of 45 bars, 3,330 m^3 storage volumes is needed, and the length of the pipe container is 2.1 km with the diameter 1.42 m and wall thickness 23.5 mm. It costs 1,400 €/m for material [29], 1,000 €/m for pipework (calculated based on information in [30]) and 500 €/m for underground mining (calculated based on information in [30]). In total, a pipe container system costs above 6 million euros (45.6 €/nm^3, 1,830 €/$m^3_{geom.}$). As for the pressure tank, because of the same storage pressure, the total storage volume must also be calculated according to DOD 45%, and the cushion gas costs 33,000 euros (for the calculation refers to section 5.2). In this system, no compressor is needed, and the pipe container is installed underground, the above-ground surface is free for original agricultural use. But the costs for pipework and underground mining are very high.

6.3. Pipeline

Hydrogen pipeline is also a possibility to store hydrogen. There are many existent hydrogen pipelines until 2001 [21]. In Germany the Rhein-Ruhr-Pipeline is 225 km long and in operation since 1938. The allowable maximal pressure is up to 30 bars. The second hydrogen pipeline in Germany is the Leuna Pipeline (Linde) that is 100 km long and the allowable maximal pressure is 25 bars. The cost for pipelines consists of material cost, working related cost, and costs for rights of way and damage, and other costs like inspection, engineering, building supervision, etc. [21]. Based on the cost for a natural gas pipeline system 400 €/m with a diameter of 400 mm [31], the cost of a hydrogen pipeline system can be estimated. In order to minimize hydrogen losses, the working related cost for the connection sealing for hydrogen pipeline is higher than for natural gas pipeline. And also because of the usage of special steel against the embrittlement effect of hydrogen, the material of hydrogen pipeline is also more expensive than for natural gas pipeline. If the working related cost is 25% more expensive and material cost 50% more expensive than for natural gas pipeline, the hydrogen pipeline costs up to 480 €/m [31]. In order to store 12 tons hydrogen, the storage volume is 3,330 m^3, and the length is 26.5 km with a diameter of 0.4 m, if the maximal working pressure is 45 bars. The total cost is around 12.72 million euros (95.3 €/nm^3, 3,820 €/$m^3_{geom.}$). The same as pressure tank and pipe container, the total storage volume must be calculated also according to DOD 45%, and the cushion gas costs 33,000 euros (for the calculation refers to section 5.2). The land scope of the system is about 14,000 m^2. Many experiences for natural gas pipeline systems are good references for hydrogen pipeline systems, and it is also possible to share the existent hydrogen pipeline systems. But the intermediate compression stations are needed for every 100 km pipeline to compensate the pressure losses.

7. Hydrogen system costs analysis, estimation and comparison

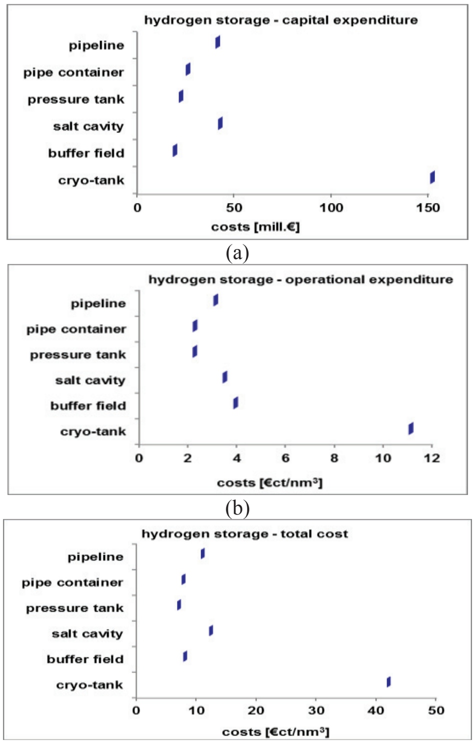
In this work, a calculation methodology has been developed to analyze different hydrogen storage systems. All the hydrogen amounts are related to volume of hydrogen (nm^3), and in order to calculate the energy losses for liquefication, compression consistently the energy losses are also related to volume of hydrogen (nm^3). The application time is set up to 30 years. The cost of hydrogen is calculated according to the lower heating value of hydrogen and the efficiency of electrolyser 60% for hydrogen production based on an estimated price of wind energy generated electricity 9 €ct/kWh.

7.1. Hydrogen system costs analysis and estimation

Input data varies in different hydrogen storage systems. For every parameter a best case and a worst case is defined. Efficiency of storage is used together with efficiency of discharging and DOD to calculate the installed capacity of storage system. That means, caused by hydrogen losses and DOD of the storage, the installed capacity of the storage must be increased, in order to achieve the customer required amount of hydrogen getting out of the storage system. The efficiency of the charging is not considered for the calculation of installed capacity of storage, because it only affects the amount of hydrogen for full charging the storage, but not the stored hydrogen in storage. Total round trip efficiency is calculated with external energy demand for hydrogen treatment, e.g. liquefication or compression, efficiency of charging, storage system and discharging. Capital cost and costs for peripherals depend on the data in the sections for different storages. The cycle life and lifetime of the storage system are also defined in case the application lifetime is longer than storage lifetime, if so, storage is required to be replaced. The costs for maintenance and repair is also been considered. With this calculation methodology, the capital cost including the costs for storage and the possible peripherals can be calculated. And the operational cost does not only include the energy requirement for hydrogen compression or liquefication but also the hydrogen losses during charging, storage and discharging and the cost for system maintenance. What must be mentioned is the flow rate of hydrogen is also inclusive in the cost calculation, since it affects the amount of hydrogen going through the storage system that affects the cost per nm^3.

7.2. Hydrogen system comparison

From all the hydrogen storage systems the cryo tank system requires the most expensive capital investment because of the high cost of liquefier, and the buffer field system has the cheapest capital investment (see Fig. 5 (a)). However, including the operation cost the buffer field is not the cheapest storage system any more, since in buffer field system hydrogen must be compressed before charging into tanks, the energy requirement, that is not needed in the systems without compressor, increases the total cost of buffer field system. Because of the high maintenance cost, the operational cost of pipeline system is much higher than pressure tank system and pipe container system (see Fig. 5 (b)). In salt cavity system, since the pressure difference between input hydrogen 50 bars and hydrogen storage 180 bars is less than the difference in buffer field system between 50 bars and 300 bars, the operation cost of salt cavity is lower than buffer field (see Fig. 5 (b)). The cryo tank system has the most expensive operational cost because of the highest energy losses in liquefication process. Fig. 5 (c) shows the total cost per nm^3 that includes capital expenditures and operational expenditures for all hydrogen storage systems. With the operational expenditure pressure tank system is the cheapest system. All the cost analysis results have interval that is caused by the best and worst case setting for specific data of different storage systems. But because of the small different of best and worst case assumption and large value of cryo tank cost, it isn’t shown so clearly in Fig. 5.



(a)

(b)

(c)

Fig. 5 Comparison of hydrogen storage systems (a) capital expenditure (b) operation expenditure (c) total expenditure

8. Conclusions

In this work, many hydrogen storage methods and systems have been investigated for the application of large scale hydrogen storage in range of tons. The physical hydrogen storage is more suitable than other technologies for this kind of applications. Cryo tank, buffer field, salt cavity, pressure tank, pipe container and pipeline and their systems have been studied in detail referring to system efficiency, system capital expenditure and operational expenditure. To analyze capital and operational expenditure of the hydrogen storage systems a calculation methodology is also developed in this work. Because of the high investment for liquefier and the low efficiency of liquefication process, cryo tank hydrogen storage system is out of consideration. Buffer field system has also unacceptable because of the inefficiency of compression. Pressure tank system and pipe container system are relatively cheap and can be considered for the application. But if the system fluctuation must be taken into account, like electrolyser disruption or consumption black out, the storage system capacity must be increased accordingly. Salt cavity has a large storage volume and is capable for system fluctuation. The possibility of sharing the existent salt cavity highlights its advantages. Similar to this, to share the existent pipeline system can help to decrease the cost of pipeline storage system. As long as the location of the electrolyser and chemical plant is determined, the conditions of existent salt cavity or pipeline nearby can be investigated. Considering all the factors, optimal hydrogen storage can be determined through the cost estimation. Based on this study the whole chemical production system can be analyzed. Furthermore, this cost estimation mechanism is not only suitable for the project CO2RRECT but also for other applications.

Acknowledgements

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