I. Presentations: Energy Management and Flexibility Options

RECYCLING AND SECOND USE OF GREEN HYDROGEN FROM SEMICONDUCTOR INDUSTRY

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Abstract: Sector-coupling is a key component for decarbonization of the energy sector, industry and mobility. Hydrogen plays a vital role in these sector-coupling strategies. Amongst many other applications it is used in semiconductor industry as a carrier gas for e.g. epitaxy processes. After processing this highly valuable energy carrier is either burned or diluted and released to the atmosphere. This paper describes the technical and ecological evaluation of four alternative utilization paths for the hydrogen that re-use it for energy production, mobility purposes, and internal recirculation, respectively. The implementation of the utilization as fuel for zero-emission mobility in the public transport sector is described in more detail. After semiconductor production the hydrogen will be collected, purified, compressed and utilized to operate up to 40 fuel cell busses as well as other heavy-duty vehicles and cars. Thus, this system cuts emissions by avoiding hydrogen emissions to the atmosphere and by replacing diesel in the transport sector.

Keywords: Green hydrogen; sector-coupling; zero-emission mobility; hydrogen busses; semiconductor industry

1 INTRODUCTION

In order to reach the ambitious targets for the reduction of greenhouse gases (GHGs) in the EU major emission cuts in all sectors will be necessary. Sector-coupling plays a key role in these reduction efforts and (green) hydrogen serves as an important link between sectors. Produced with electrolysers using renewable electricity it can serve e.g. as carbon-free feed-stock for the chemical industry, as substitute for coke in steel-making, as carrier gas for industrial purposes, as substitute for fossil fuels for providing heat, for energy storage, or as fuel for mobility purposes. This paper gives an example for such sector-coupling using hydrogen as carrier gas in a semiconductor processing plant of Infineon Austria Technologies and, subsequently, as fuel for fuel cell (FC) vehicles in Villach, Austria.

In their production plant in Villach, Infineon Austria Technologies produces semiconductors for industrial power control, automotive, and power & sensor systems producing over 8.7 billion chips in 2020/21 [1]. One of the processes used in production is epitaxial growth of silicon layers on wafers (epi-wafers) through chemical vapor deposition (CVD) where silicon is deposited on wafers by reducing silanes like trichlorosilane (HCl_3Si) at high temperatures using hydrogen as carrier gas and reducing agent [2].

Until now, this carrier gas was supplied in form of highly pure liquid hydrogen made through steam methane reforming and transported over 700 km via lorries from a supplier in Germany. The hydrogen is then additionally purified using a cryogenic purification process (cryogenic temperature swing adsorption, TSA) to a quality of 8.0 (99.999999%). It is then mixed with 40-60% inert gas, silanes and traces of dopant precursors containing P, B, or As. The hydrogen itself is not consumed during the epitaxy process. The subsequent waste gas additionally contains HCl as reaction product and from cleaning steps in the reaction chamber. The waste gas is scrubbed from environmentally harmful substances, diluted with air below the lower explosive level (LEL) of hydrogen and finally released to the atmosphere.

In order to change this use of emission intensive hydrogen, to avoid wasting a highly valuable energy carrier, and to increase security of supply, Infineon decided to produce their own, green hydrogen with an on-site electrolyser using renewable energy. In the framework of the “H2Pioneer - Pave the way for green hydrogen for early adopters in the light industry” (H2Pioneer) project funded by the Klima- und Energiefonds Austria (see Figure 1-1) four potential alternative utilization paths for the waste gas have been investigated.

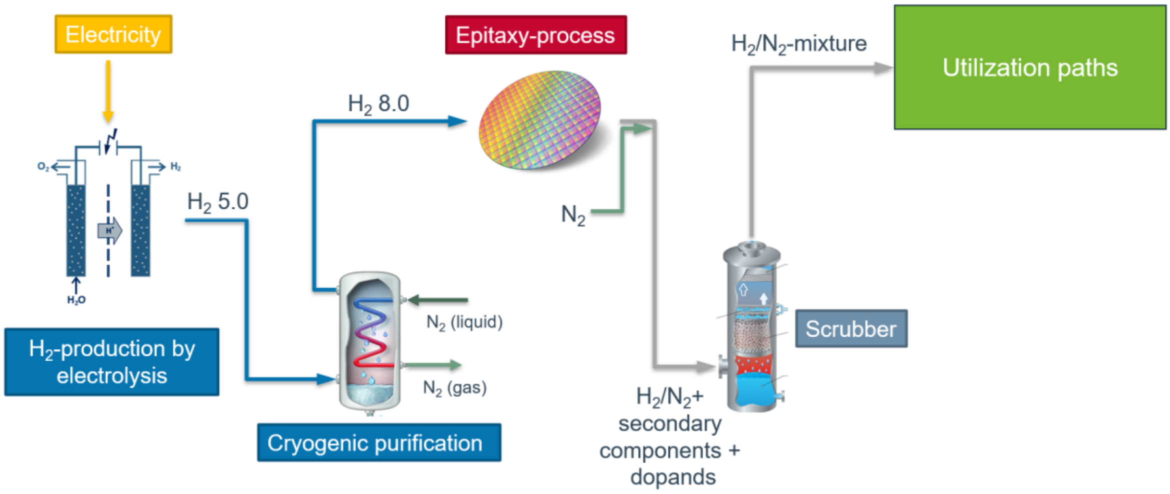


Figure 1-1: Schematic overview of the hydrogen production, purification, and utilization.

As a result of this project the decision was made to re-use the waste gas for zero-emission mobility purposes, especially in the public transport sector which is pursued in the framework of the “Reuse of Hydrogen for Bus Applications” (ReHyB) project funded by the Austrian Research Promotion Agency (FFG). Both projects are part of the H2Carinthia initiative and will be described in more detail in the following sections.

2 MATERIALS AND METHODS

One goal of the H2Pioneer project was to evaluate different paths for re-using the hydrogen from the epitaxy process in the semiconductor industry [3]. Four different paths were investigated, namely, using the hydrogen to drive a stationary internal combustion engine (ICE), purifying and using it in a stationary fuel cell for electricity generation, purifying and using it for H_2-moblitity at a hydrogen refuelling station (HRS), and internal recirculation of purified hydrogen back into the production process (see Figure 2-1).

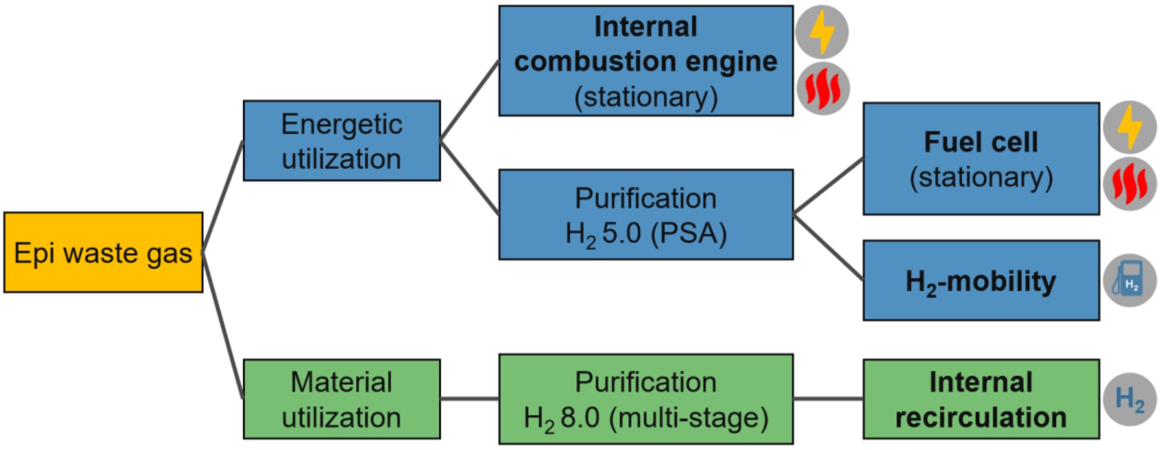


Figure 2-1: H_2 utilisation pathways for the epitaxy waste gas.

All four paths have been analysed on a technical and an ecological basis. The technical analysis was done on a conceptual level by means of mass and energy balances. Using characteristic curves of the major individual components zero-dimensional simulation models in MATLAB Simulink® have been implemented and analysed. Based on this analysis, an environmental analysis was accomplished by a calculation of greenhouse gas emissions following Greenhouse Gas Protocol guidelines [4].

Based on results from the H2Pioneer project re-use of the hydrogen for application in the mobility sector has been chosen as the path to be implemented through the ReHyB project. In this project coupling out and purification is simulated on 1D-/3D-computational fluid dynamics (CFD) simulations on component and system level. Data from real operation is then used for validation of these models. The HRS is simulated by model-based MATLAB Simulink tools. Instrumentation and control engineering will be done using Systems Modelling Language (SySML). For the operation of up to 40 busses in the public transport sector the hydrogen demand is simulated using longitudinal dynamic simulations with AVL CruiseM®.

3 RESULTS

3.1 Technical analysis of the utilization paths

For the utilization by means of a hydrogen ICE, the hydrogen is energetically converted into mechanical work and heat. The mechanical work is converted into electrical energy and can be used internally, for example, to operate the electrolysis. The heat can also be used internally for facility heating and reduces the heating demand that is currently provided by natural gas. This method requires the smallest number of components, namely a gas mixture system to secure a nitrogen content of at least 50% to avoid backfiring [5] and the ICE. Based on data from a hydrogen ICE *agenitor406* from the manufacturer *2G Energy AG* with a maximum power of 178 kW_{el} it was concluded that the electricity generated could lower the energy demand of the electrolyser by ca. 20%. Additionally, over 14 MWh of thermal energy per tonne of hydrogen could be utilized [3].

Utilization by a stationary PEM fuel cell could also generate electrical and thermal energy that can be used to cover part of the production plant needs. In contrast to the ICE, however, the PEM fuel cell can only be operated with high-quality hydrogen in accordance with the ISO 14687 standard. For this reason, the waste gas stream needs to be purified by e.g. a pressure swing adsorption (PSA). The system is more complex and consists of a compressor, a cooler for the compressor, a PSA and the fuel cell. The compression and purification use electric energy and the PSA can only recover a part of the hydrogen. Using data from a *PowerCell MS-100* system with a maximum power of 260 kW_{el} and a (simulated) hydrogen loss rate of 26% from the PSA (it has to be noted here that real PSA systems can have lower hydrogen loss rates in the range of 16%) a lower yield of electric power covering 11.5 % of the electricity consumption of the electrolyser can be achieved. The usable waste heat would amount to 10.9 MWh per tonne of hydrogen [3].

In case that the hydrogen is used for H_2-mobliity, the pre-treatment is very similar to the case of the stationary fuel cell. A first compression step including cooling of the compressor would be followed by purification using PSA. Subsequently, an additional further compression step to 350 bar and 700 bar for busses/heavy-duty vehicles and cars, respectively, is needed. The hydrogen could then be used to power e.g. fuel cell buses in public transportation. Assuming an average consumption of 30 l/100 km for a diesel bus and 7.5 kg_{H2}/100 km for a fuel cell bus one tonne of hydrogen can power latter bus for ca. 13300 km and replace 4000 liters of diesel.

Internal recirculation is the fourth alternative utilization path for the hydrogen. In this case it would in a first step be purified by a PSA yielding hydrogen with 5.0 quality. After this purification step it would have the same quality as the hydrogen from electrolysis and could be added to the hydrogen from the electrolyser. A second purification step using cryogenic TSA would then yield hydrogen of quality 8.0. If implemented, the hydrogen production would only need to replace the blow-off from the PSA.

3.2 Ecologic analysis

In accordance with the GHG Protocol [4], the environmental impact of the various epitaxy waste gas recovery concepts is evaluated by drawing up a CO_2 balance for the hydrogen path of the individual recovery methods. The total emissions (sum of direct and indirect emissions) of CO_2-equivalent emissions from the following areas are taken into account:

- Consumption of electrical energy during hydrogen production
- Hydrogen emissions during exhaust gas utilization
- Generation or consumption of electrical energy during waste gas utilization
- Generation of heat and replacement of natural gas during exhaust gas utilization
- Replacement of diesel by the use of hydrogen as a fuel

In order to achieve comparability of the greenhouse gas potential of the different forms of energy, all greenhouse gas emissions (GHG emissions) are converted to CO_2-equivalent emissions in the CO_2 balance. The factors for the CO_2-equivalents are summarized in Table 3-1.

Table 3-1: CO_2-equivalent emission factors of selected energy carriers [6] [7]

Emission factors (CO_2-equivalent of total emissions)		
Energy carrier	**kg_{CO2eq}/MWh**	**Source**
Electricity mix Austria	219	Umweltbundesamt [7]
Green electricity	14	Umweltbundesamt [7]
Natural gas	268	Umweltbundesamt [7]
Diesel (incl. 5.6% bio fuel)	321	Umweltbundesamt [7]
Hydrogen	174	Derwent et al. [6]

In order to assess the emissions for the different utilization paths in a first scenario the CO_{2eq} factor of the Austrian electricity mix (219 kg_{CO2eq}/MWH) is assumed. This represents the currently worst-case scenario in terms of emissions. Then the hydrogen production by means of electrolysis which accounts for high amounts of electrical energy consumption largely contributes to the CO_2 emissions (see Prod. in Figure 3-1). The provision of one tonne of hydrogen thus results in emissions of around 13.6 tonnes of CO_{2eq}

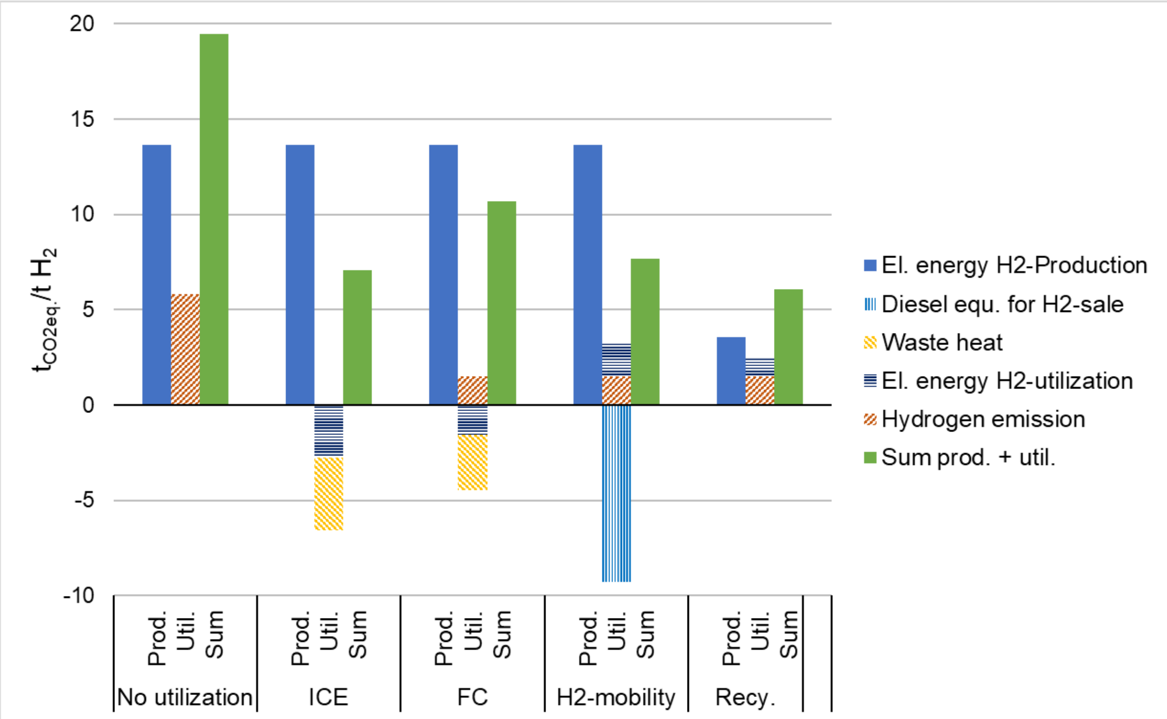


Figure 3-1: Emission balance of different utilization paths using the Austrian electricity mix.

Without a recycling method, all of the hydrogen is safely released into the environment after utilization (see Util. in Figure 3-1) and represents additional pollution, since hydrogen has an indirect greenhouse gas potential of 5.8 kg $CO_{2eq.}/kgH_2$ (equivalent to 174 $kgCO_{2eq.}$/MWh) [6] resulting in an additional emission of 5.8 tonnes of $CO_{2eq.}$ per tonne H_2. The indirect GHG potential results from a competing reaction to the decomposition of methane in the atmosphere. The case of hydrogen production via electrolysis and no further utilization (release to the atmosphere) is used as benchmark (see Sum in Figure 3-1).

In case of utilization through an ICE no hydrogen is released into the atmosphere and the obtained electrical and thermal energy reduces the emissions of hydrogen production and heating by natural gas, respectively, totaling in an emission reduction of ca. 64 % compared to the sum of the base scenario (assuming that accruing NOx emissions are avoided through aftertreatment). Due to the additional energy use for purification and compression as well as the hydrogen blow-off of the PSA, utilization through a fuel cell leads to an emission reduction of ca. 45 %. In the case of using the hydrogen for H_2-mobility, emissions from the additional energy use for purification and compression are more than offset by the replacement of diesel, resulting in a total emission reduction of ca. 60%. The internal recirculation of hydrogen yields the highest emission reduction in the range of 69%. Here the additional energy use for purification and compression is greatly offset by the recycling of the hydrogen and the accompanying energy savings at the hydrogen production via electrolysis.

If both the hydrogen production and all other electrical loads are considered to be powered by green electricity with an emission factor of only 14 kg_{CO2eq}/MWh (Umweltzeichen “Grüner Strom” in Austria [7]) the emissions of all utilization paths strongly decrease. The release of the hydrogen to the atmosphere is now the main contributor to emissions (except for the ICE). Through waste heat utilization and replacement of diesel, respectively, ICE, FC and H_2-mobility even abate more emissions than the production of the hydrogen emits in the first place (see Figure 3-2).

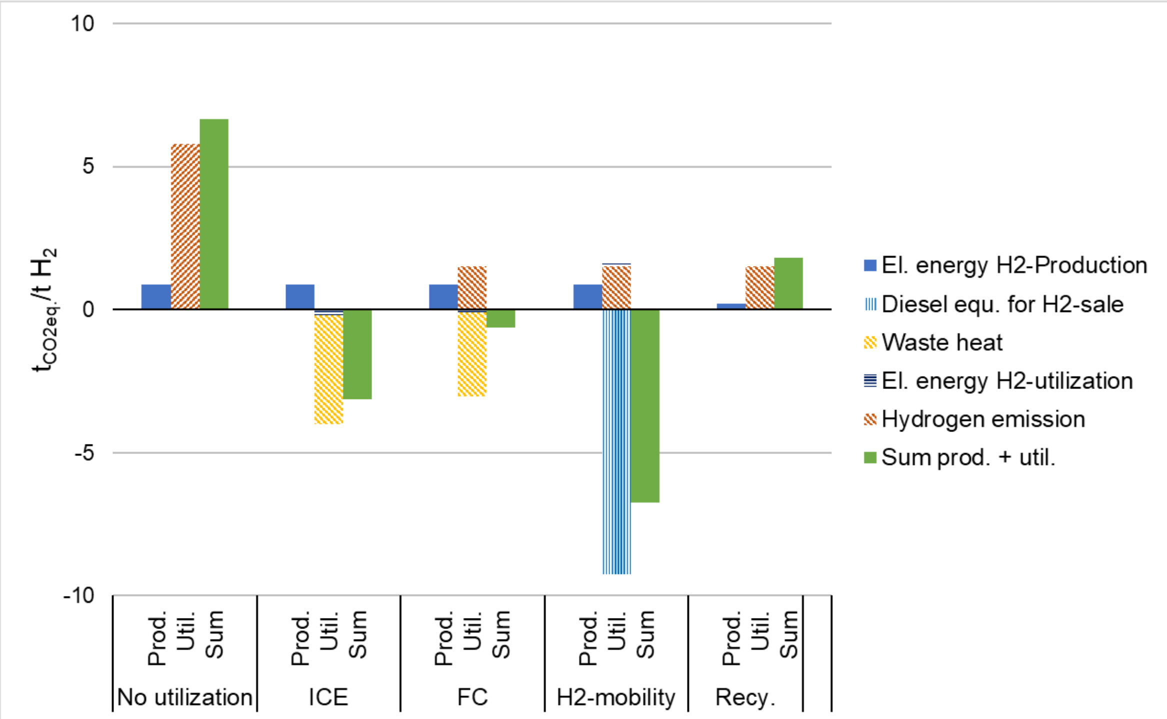


Figure 3-2: Emission balance of different utilization paths using green electricity.

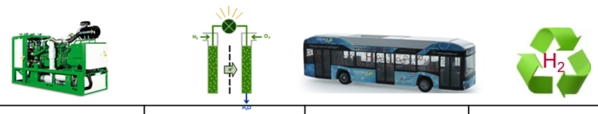
4 DISCUSSION

From a technical point of view all four utilization paths discussed above are realizable with existing technologies in principle although the technology readiness levels (TRLs) and degrees of innovation differ significantly. Hydrogen ICEs are already a well-established technology in this field thus their application poses a rather low degree of innovation. Reconversion of hydrogen to electricity via fuel-cells or using it in fuel-cell vehicles are also already established technologies, though the re-use of hydrogen from semiconductor industry would be a true innovation. Internal hydrogen recirculation also has not been implemented in semiconductor industry so far. One obstacle for this utilization path is the fact that some gas components from the epitaxy process like HCl or dopants cannot be measured online with an adequate resolution. Through multiple recycling steps they could accumulate in the hydrogen imposing a risk on the epitaxy production process. Since a negative influence on the sensitive process could prove to be very expensive such a risk will hardly be tolerated in semiconductor industry, prohibiting this path until adequate analytics for this purpose is available.

From an ecological point of view all four utilization paths lead to significant emission reductions. Depending on the assumptions for the emission intensity of the electricity mix the results for the emission reduction differ significantly, though. For the Austrian electricity mix (see Figure 3-1) the abatement is highest for internal recirculation due to the strong reduction of the need for energy intensive electrolysis in the hydrogen production. The paths using an ICE or using the hydrogen for zero-emission mobility both give similar, the use of a fuel-cell for reconversion into electricity gives the lowest emission reductions. If the use of green electricity is assumed (see Figure 3-2) emissions for all utilization paths as well as for no utilization are reduced dramatically and in three cases even leads to absolute emission savings if the replacement of natural gas for heating or diesel for mobility is considered. In this case the use for mobility purposes leads to the highest abatement followed by the ICE, the fuel cell and the recycling.

To fully evaluate the four presented hydrogen utilization paths an economic analysis would also be needed. This analysis strongly depends on assumptions for component and engineering prices and would go beyond the scope of this paper. Taking the results from the economic analysis of [3], though, some general remarks can be made. This work concluded that the fuel cell would under no scenario reach a break even while all other utilization paths would reach a break even in a range between 4 and 11 years with the ICE being the most profitable followed by recirculation and mobility.

Figure 4-1 gives a qualitative overview for the evaluation of the different utilization paths. Considering all three dimensions the fuel cell and the internal recirculation can both be excluded due to non-profitability and risks due to low TRL of available gas analysis systems, respectively. The ICE has many favorable features and the low degree of innovation is no show-stopper per-se. Nevertheless, use of the hydrogen from semiconductor production for zero-emission mobility has been chosen to be implemented due to its high potential for emission reduction and its degree of innovation to serve as a flagship project for the possibilities of sector-coupling.



Scale: 1 (bad) – 5 (good)	ICE	Fuel cell	H_2-Mobility	Recirculation
Criteria (technical, ecological, economic)	Evaluation	Evaluation	Evaluation	Evaluation
Period of amortization	5	1	3	4
CO_2 reduction (Austrian electricity mix)	4	3	4	5
CO_2 reduction (green electricity)	4	3	5	3
TRL	5	4	4	1
Innovation	1	4	4	5

Figure 4-1: Qualitative assessment of the four utilization paths proposed in this paper.

Presently, the implementation of this approach is being started in the framework of the ReHyB project. Figure 4-2 gives a schematic overview of the implementation concept. After pre-cleaning in a scrubber and drying of the waste gas the hydrogen/nitrogen mixture will be compressed in a first stage and pumped through a pipeline to a PSA situated at the HRS. After purification in the PSA the hydrogen with quality 5.0 is further compressed to be stored in low-pressure storage tanks.

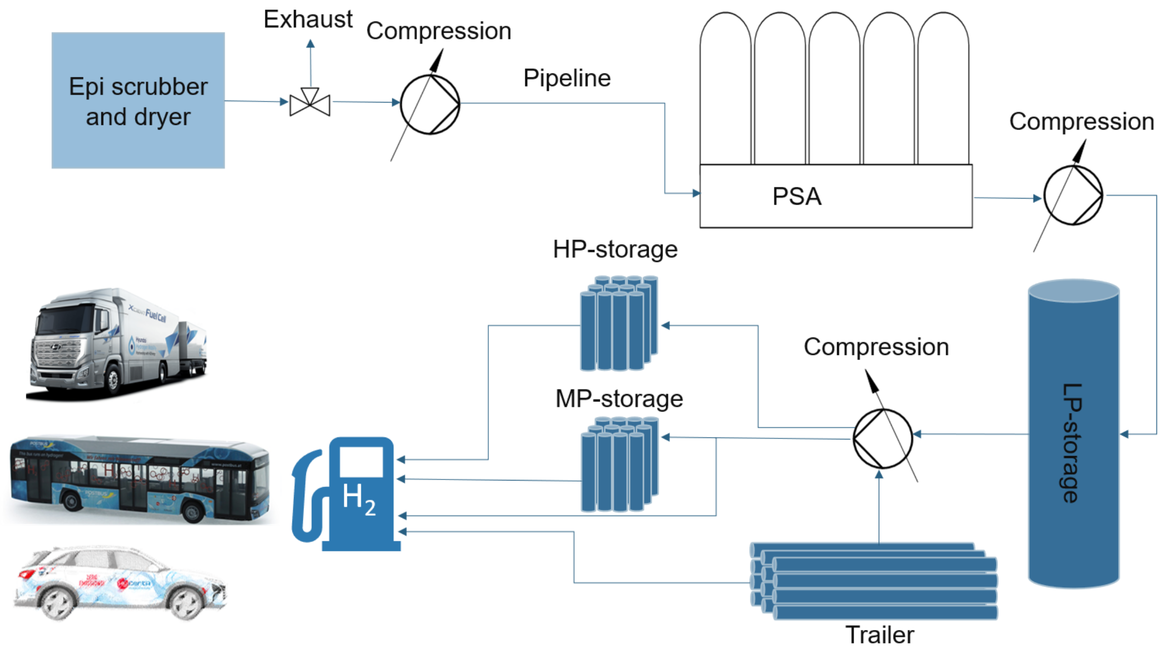


Figure 4-2: Schematic overview of the implementation of hydrogen collection, purification, compression and use in zero-emission mobility.

Another compression stage will raise the pressure to pressures apt to fill medium- and high-pressure storage tanks for refueling with a dual-use dispenser at 350 bar and 700 bar, respectively. A hydrogen trailer can also be connected to the HRS in order to increase security of supply e.g. in the case of maintenance works in the supply chain. By being close to three Austrian highways (A2, A10 and A11) and the "Baltic-Adriatic" TEN-T (Trans-European Transport Network) corridor the refueling station in Villach would also be placed in a strategically favorable place and filling a gap in the Austrian HRS network. The main use for the re-used hydrogen would be local public transport with a plan to fuel up to 40 FC-busses in and around Villach, decarbonizing most of the public transport in the region.

Another point to mention here is energy efficiency. One of the main arguments against hydrogen in the discussion fuel cell vs. battery vehicles is the lower round-trip efficiency of the former with a well-to-wheel efficiency in the range of 30%. The energy demand for re-using the hydrogen (drying, purification, compression) is less than 10 kWh per kg of hydrogen which has an energy density of 33 kWh/kg. With an efficiency of FC vehicles in the range of 50-60% the well-to-wheel efficiency would be up to 200% if the production of the hydrogen is fully accounted for in the semiconductor production already. This example underlines how wasteful it is to not re-use hydrogen whenever possible and the advantages of sector-coupling.

5 CONCLUSIONS

This paper showed that hydrogen can play a vital role in the decarbonization of our energy system through sector-coupling of energy systems, industry and mobility. All four presented paths for re-utilization of hydrogen from semiconductor industry show great potential to lower energy demand as well as GHG emissions. A technical, ecological and economic analysis of the four utilization paths conducted in the H2 Pioneer project and presented in the paper led to the decision to implement the re-use of hydrogen from industry for zero-emission mobility in the framework of the ReHyB project. Besides global GHG emission reduction, less emission of pollutants and noise will also benefit the health of the local population and increase the quality of living in the region.

6 ACKNOWLEDGEMENTS

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7 REFERENCES

[1] Infineon Technologies Austria AG, „www.infineon.com,“ [Online]. Available: https://www.infineon.com/dgdl/IFAT+Unternehmenspr%C3%A4sentation_DE.pdf?fileId=8ac78c8b7e7122d1017f26ee5cfa00f4. [Zugriff am 23 August 2022].

[2] Wikipedia, „Epitaxy,“ [Online]. Available: https://en.wikipedia.org/wiki/Epitaxy. [Zugriff am 23 August 2022].

[3] J. Trattner, „Energieeinsparungsmöglichkeiten durch Verwertung oder Aufreinigung von Wasserstoff in Abgasen von Industrieprozessen,“ Graz, 2021.

[4] „Greenhouse Gas Protocol,“ [Online]. Available: ghgprotocol.org. [Zugriff am 10 March 2021].

[5] J. Gausling, *H2-Motor - 2G Energy AG,* E-Mail Aug. 2019.

[6] R. Derwent, P. Simmonds, S. O'Doherty, A. Manning, W. Collins und D. Stevenson, „Global environmental impacts of the hydrogen economy,“ *IJNHPA,* Bd. 1, Nr. 1, p. 57, 2006.

[7] Umweltbundesamt, „Berechnung von Treibhausgas-Emissionen verschiedener Energieträger,“ [Online]. Available: https://secure.umweltbundesamt.at/co2mon/co2mon.html. [Zugriff am 25 August 2022].

[8] M. Klell, H. Eichsleder und A. Trattner, Wasserstoff in der Fahrzeugtechnik: Erzeugung, Speicherung, Anwendung, Wiesbaden: Springer Vieweg, 2018.