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1 Abstract

Maintaining the global warming well below 2°C, as stipulated in the Paris Agreement, will require a complete overhaul of the world energy system. Hydrogen is considered to be a key component of the decarbonization strategy for large parts of the transport system, as well as some heavy industries. Today, about 96% of current hydrogen production comes from the steam reforming of coal or natural gas (labeled Black and Grey hydrogen, respectively). If hydrogen is to become a solution, then Black and Grey hydrogen need to be replaced by a low-carbon option. One method that has received much attention is to produce so-called Green hydrogen by coupling water electrolysis with renewable energies. However, green hydrogen is expensive and energy intensive to produce. In this communication, we explore an alternative option and highlight the benefits of rock-based hydrogen (White and Orange) compared with classic electrolysis-based technologies. We show that the exploitation of native

hydrogen and its combination with carbon sequestration has the potential to fuel a large part of the energy transition without the substantial energy and raw material cost of Green hydrogen.

2 Main

To meet the Net Zero Emission Scenario (NZE) of the International Energy Agency [1], about 17,000 TWh of hydrogen-based energy will be consumed in the world in 2050. This hydrogen is usually seen as being provided by large facilities using renewable electricity to convert water to hydrogen through electrolysis (Green hydrogen). The energy cost for hydrogen production is related to the splitting of the water molecule into hydrogen and oxygen:

$$H_2O \xrightarrow{\text{energy}} H_2 + \frac{1}{2} O_2 \tag{1}$$

and is directly calculated from the Gibbs energy of formation of water (120 MJ/kg_{H_2} at 25°C – THERMODDEM database [2]). Considering an 80% efficient electrolyser, this corresponds to about 400 Mt of hydrogen, or about five times the amount consumed in 2020 [1]. These 17,000 TWh represent more than the electricity produced by both China and the USA in 2020, about 63% of the world's electricity production, and more than twice the current worldwide production of electricity from renewable energies. Meeting this figure will then require an unprecedented upscaling of both electrolysis and renewable energies.

Blue hydrogen does not fare better due to the extra cost related to CCS and is probably only a temporary solution [3]. Some authors are even more critical, arguing that the imperfect capture of CO_2 associated with methane losses along the whole chain leads to increased greenhouse gas emissions with only little benefits from pure Grey hydrogen [4]. Another critical shortcoming

of Blue hydrogen is the continued reliance on fossil fuel. This last issue also impacts an emerging production technique, sometimes referred to as Turquoise, and which consists in producing H_2 from methane but with graphite-C and not CO_2 as a resulting product. This technology is still in development but could also play a key role in the coming years.

3 A greener hydrogen

The Earth's subsurface is a giant and undervalued hydrogen factory. Long considered anecdotic, the current estimations give a rough estimate of 20 Mt of native hydrogen escaping from the surface towards the atmosphere each year [5]. Estimates vary widely but everything suggests that the subsurface is actually producing significantly more, and that this value is just the tip of the iceberg. Natural hydrogen is most commonly produced by the reduction of water to hydrogen following the anoxic and abiotic oxidation of ferrous iron to the orange-coloured ferric iron [6]:

$$2\,FeO + H_2O = Fe_2O_3 + H_2 \tag{2}$$

This oxidation occurs, for example, under the ocean floor [7, 8], where peridotites – magnesium silicate rocks which contain up to 10%wt of ferrous iron – become hydrated at temperatures between 200 and 400°C by the percolation of seawater. This process, known as serpentinization, also occurs at lower temperatures and pressures on land-based ophiolite-peridotite massifs [9]. Serpentinization is estimated to produce around 1-2 Mt of hydrogen per year [10] at oceanic ridges where it is the most active. However, serpentinization is not the only process producing hydrogen and anywhere water meets reduced iron, there is a possibility to produce hydrogen if the right conditions of temperature, fluid composition and pressure are met (e.g. archean

Banded-Iron Formations [11], peralkaline and biotite-rich granites [12, 13]). On land, numerous native hydrogen seeps (usually mixed with methane [14]) have been localized (e.g. Oman, Japan, New Caledonia, Greece, Spain, Italy, California) [15], France [16], Australia, Brazil [17], Kansas [18]). Some are even known since the antiquity such as the Mount Chimaera (Yanartaş, Turkey) which continuously produces a mixture of 87%*vol.* CH_4 and 10%*vol.* H_2 [14]. Exploitation of this hydrogen offers then an interesting alternative to Green and Blue hydrogen by completely skipping the electrolysis and simply collecting the hydrogen emitted by natural underground processes. This White hydrogen is already under production in Mali where a well drilled for water turned out to produce a 98%vol hydrogen stream currently used power the entire village of Bourakebougou for the last 10 years [19]. The actual source of the hydrogen remains unknown but chances are that the reduction of iron is involved deep underground.

4 Orange Hydrogen

It is possible to go even further and go Orange. This color, which refers to the color of oxidized iron, follows the same principles of White hydrogen and looks to make the Earth provide most of the work for hydrogen production. Yet, instead of using a passive approach of exploration/exploitation, Orange hydrogen follows a proactive way and searches to stimulate the reaction. This is done by injecting water *in situ* in identified reactive formations and collect the hydrogen-saturated water from recovery wells surrounding the injection point (Fig. 1). This approach can also be adopted for Fe-rich mine wastes and steel slags [20] in an *ex situ* surface reactor which has the advantage of being easier to control and set-up. Additionally, the magnetite particles (Fe_3O_4) resulting from the oxidation process have a commercial value, especially in

the pharmaceutical industry. However, the significantly lower volumes of raw materials available for *ex situ* Orange Hydrogen and the prohibitive financial and environmental cost of extracting and grinding new feedstock makes the *ex situ* solution less attractive than the *in situ* one.

In situ Orange hydrogen requires more energy than White hydrogen production, but the outputs will be significantly higher, while production costs will likely remain under Blue/Green ones. An average peridotite can provide around 2-4 kg_{H_2}/m^3 upon complete oxidation. With 10^{20} kg of peridotites in the upper crust (top 7km) [21], there are 100 trillions tons of hydrogen to be extracted from the subsurface, sufficient for 250,000 years at a rate of 400 Mt/year. This is not even considering that over these timescales, tectonic activity will refresh the peridotites at a rate of 10^{12} kg/year [21]. The exploitation of the whole volume is obviously unrealistic due to technical, economic and regulatory reasons, but even a small percentage would make Orange hydrogen a key player in the achievement of the NZE scenario.

The real game-changing impact of Orange hydrogen, is that the very same formations which naturally produce hydrogen are also the perfect location for carbon sequestration [22]. These formations are part of the deep carbon cycle, balancing the concentration of atmospheric carbon dioxide through its reaction with silicate rocks. This natural weathering has allowed the capture of 99.9% of the total carbon on Earth as solid, stable carbonates [23], but is now getting rapidly outpaced by the increase in anthropic emissions since the industrial era. Matter *et al.* [24] calculated for example that 30 trillions tons of CO_2 can be stored in the Oman Ophiolite (the largest on-land peridotite massif), and 100 trillions tons globally. For comparison, the CO_2 anthropic emissions in 2020 are estimated at 33 billions tons.

In Iceland, Carbfix, a company built on the results of several European Union's research projects, has been injecting 72,000 tons of carbon dioxide in the island basaltic formation for about 10 years with great success [25, 26]. A similar pilot injected 1000 tons of pure liquid CO_2 in the Columbia River Basalts near Wallula (USA). Cores retrieved 2 years after the injection presented unequivocal evidence of the mineralization of the injected CO_2 [27]. However, neither of these large-scale pilots considered the hydrogen production associated with the CO_2 mineralization process despite the iron-rich nature of the targeted basalts and thus the possibility of coupled production [28].

The list of Orange hydrogen advantages does not end here. In contrast to electrolysis, where only specifically tuned water compositions can be used, natural oxidation of iron as well as carbon mineralization works very well with seawater [29] or even wastewater, alleviating significantly the water cost of hydrogen production. Target formations can also contain elements of economic interest such as Li, Ni, Co... as is usually the case in serpentinite formations [30]. The dissolution of the primary minerals following the injection will release these elements in the percolating fluids, which can be extracted in parallel to hydrogen by fractionated precipitation. Similar processes are currently in use in Uranium recovery [31] and extraction of Li in parallel to heat is being considered in geothermal applications [32]. Orange hydrogen differs significantly from the alternatives as it does not rely on critical raw materials as electrolysis processes, and can even produce them as by-products of the main operation.

Laboratory experiments on the reactivity of magnesium silicates with carbon dioxide have shown that a mixture of $NaHCO_3$ and dissolved CO_2 can achieve complete carbonation over the course of a few hours to a few days [33]. Carbonation rate is a direct function of the CO_2 partial pressure but the rate of serpentinization seems either unaffected [34] or slightly accelerated [35] by

the presence of CO_2. Both carbonation and serpentinization reaction rates are also strongly temperature-dependent, and follow a bell-shaped curve with an maximum respectively at 185°C and between 250 and 280°C for serpentinization [36, 37]. Maintaining a high rate of H_2 production and CO_2 mineralization requires thus a careful control of the downhole temperature. Luckily, serpentinization and carbonation are very exothermic (respectively $\Delta H = 250$ kJ/kg and $\Delta H = 760$ kJ/kg). Once the process is initiated, the desired temperature can be obtained through self-heating with a heat production from the chemical reactions balancing the heat losses through convection and conduction, as well as the cooling induced by the injection of colder fluids [24, 38]. This self-heating behavior allows reducing production costs as no surface heating of the injected fluids is necessary, but requires a careful control of the injection rate. Interestingly, some of this heat can be extracted from the recovered fluids in a geothermal-like process and be reused to power the facility with recovered fluid temperatures up to 200-300°C (considering the combined action of exothermic reaction and formation temperatures e.g. 250°C for Carbfix2 [39]).

Kelemen *et al.* [38] calculate a carbonation rate of 4.10^9 t_{CO_2}/year for the stimulation of 1 km^3 of peridotite at 185°C (5.10^6 kg/s of carbonation, CO_2 partial pressure between 75 and 300 bars). Extrapolating their calculation with 10%wt FeO, this leads to a rate of 5 Mt_{H2}/year for 1 km^3 of peridotite (5.10^4 kg/s for serpentinization in the same conditions). In Figure 2 are represented hydrogen production rates from batch [40–42] and reactive percolation [43] experiments. In all cases, measured hydrogen production is significant and approaches the maximum production which can be expected from these rocks. A linear fit of the data gives rates from 0.1 to 3 Mt_{H2}/km^3/year, similar to the extrapolation from Kelemen *et al.* [38]. As a result, experimental

data from laboratory experiments support directly the preliminary estimations. Extra research is however necessary to assess the potential of other types of formations for hydrogen production combined with carbon sequestration.

This means that, while it is not expected that Orange hydrogen will solely meet the 400 Mt for the NZE scenario, it has the potential to provide a significant part of it, at a fraction of the energy and raw/critical mineral cost of electrolysers. In other words, Orange hydrogen promises to provide abundant, clean, rock-based, carbon-negative, and price-competitive energy to fuel the energy transition towards the NZE scenario.

5 Challenges for Orange hydrogen

Developing Orange hydrogen faces numerous scientific challenges, which can be sorted in 3 different categories:

1. Accurate description, quantification and mapping of the resource, taking into account the iron grade of the targeted rock formation and its technical and societal accessibility.
2. Identification of the ideal pressure, temperature, flow rate, fluid composition for the optimized production of hydrogen and carbon mineralization. These settings need to account for the precipitation of parasitic minerals competing with carbonate precipitation, the incorporation of Fe into precipitating minerals preventing its oxidation and H_2 production, as well as the careful control of porosity to prevent any clogging by the precipitating minerals. Parasitic reactions are for example at work in the experiment from Grozeva *et al.* [41] represented on Figure 2. The injection of a CO_2-rich fluid mid-experiment lead to a decrease in H_2 in the reactor. The process consuming the hydrogen was however, not clearly identified.

3. Field-scale optimization of the injection and recovery of the fluids to maximize the impacted zone and minimize hydrogen losses. This includes research on reservoir stimulation (hydraulic fracturing and chemical stimulation) and potential induced (micro)-seismicity, as well as the microbiology of low-temperature reservoirs which, if left unchecked, can consume the produced H_2 and impact significantly the yield.

These challenges cover a wide range of scales and disciplines and require a combined effort to offer a satisfactory solution. The first challenge falls within the scope of geologists and economists, the second is related to thermodynamics and geochemistry as well as the physics of coupled processes in porous media. The third one concerns reservoir engineering and simulations, geotechnics and microbiology. Given the scale of the projects, social acceptance will also be a key point in the development of the technology, in particular in the case of induced-seismicity.

One example of these multidisciplinary and multiscale challenges is the proper modeling of the porosity/permeability relationship during the process. Carbonation of target rocks leads to the precipitation of carbonate minerals within the porosity as well as the precipitation of serpentine, which is less dense than the minerals it replaces. As such, the porosity and thus the permeability of the rock will be a function of the dynamic balance between dissolution of the primary minerals and the precipitation of the secondary minerals. If the latter occurs faster, it is likely that some clogging will occur with dramatic effects on the long-term viability of the process. Yet, the only two large-scale pilots for geological mineralization of CO_2 have not evidenced any clogging or permeability decrease, even several years after the beginning of the injection in the case of Carbfix [26]. Moreover, natural settings are proofs that carbonation can be going on for tens to hundreds of thousands of years without any clogging [44].

Reaction-induced fracturing, which is often invoked to explain the maintained permeability during some natural processes despite adverse molar volume evolution is the process during which a growing mineral in a confined space is able to generate large stresses on the surrounding matrix, enough to fracture it and open new percolation paths for the fluid [45–47]. However, up to date, not only does the modeling of such fracturing still fail to reproduce the experimental observations but experiments of reactive percolation where such phenomenon is expected, usually do not present any evidence that reactive-induced fracturing occurred [43, 48, 49]. The control of reaction-induced fracturing and the overall control of the reactivity and injectivity during the production of Orange hydrogen (potentially through targeted hydraulic fracturing) is of paramount importance for the viability of the process and requires a deeper understanding of the microscopic behavior of dissolution/precipitation as well as a proper upscaling of these processes to reservoir and field scale.

6 Conclusion

We are at a tipping point for the climate, and the exploitation of Orange hydrogen is an additional technology which could prevent a dramatic evolution. Hydrogen is nowadays a controversial energy carrier as it is considered by some as an easy way to transport energy and decarbonize the transportation sector, while others see it as an unnecessary intermediate and advocate a full electric switch. This controversy becomes irrelevant if White and Orange hydrogen are included in the debate. Their production does not require as much electricity or raw materials as Green hydrogen and Orange hydrogen offers also the possibility to store enough CO_2 to curb global warming.

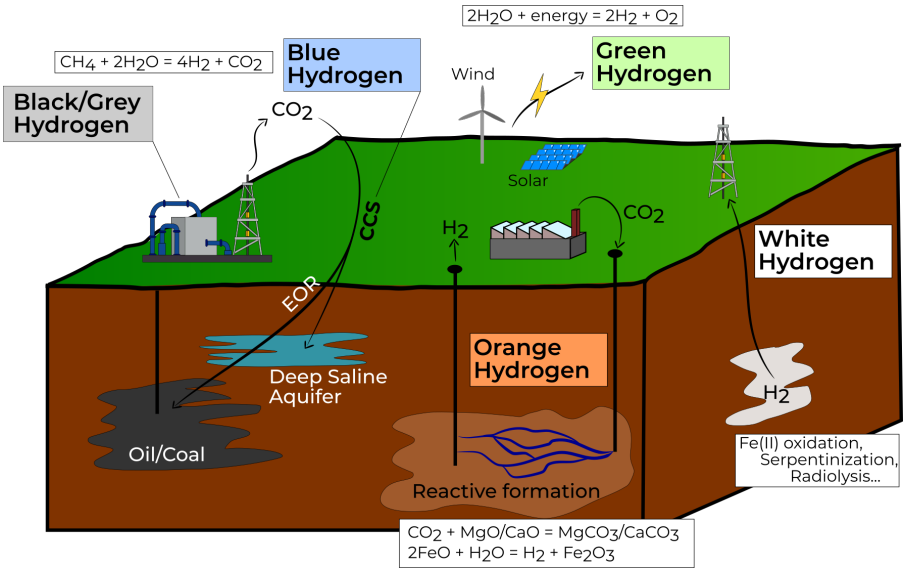

Fig. 1 The different colors of Hydrogen. Black/Grey H_2 is from steam reforming. It turns Blue through the addition of CCS, either through permanent storage (e.g. in deep saline aquifers) or through Enhanced Oil Recovery (EOR). On the other end of the spectrum, Green hydrogen is produced by electrolysis using renewable energies. White hydrogen corresponds to the exploitation of subsurface-sourced natural hydrogen. Orange hydrogen is a proactive take on White hydrogen and consists in injecting in a reactive formation a carbon-enriched solution. Geochemical reactions lead to the precipitation of solid carbonates while hydrogen is formed and recovered from the fluid.

In addition to energy considerations, the decarbonization of the world's energy mix will require an unprecedented upscaling in the production of renewable energies and the associated batteries and thus in ore mining, especially for Nickel-Cobalt ($\times 20-25$) and Lithium ($\times 40$) [50]. Every million tons of White and Orange hydrogen means at least 120 TJ (or 33GWh) of electricity saved. This saved electricity as well as the saved critical metals can be used for more batteries, electric car or wind turbines in order to accelerate the energy transition. If we add the carbon storage ability of Orange hydrogen, we might just have the solution for some of the energy problems of the coming decades.

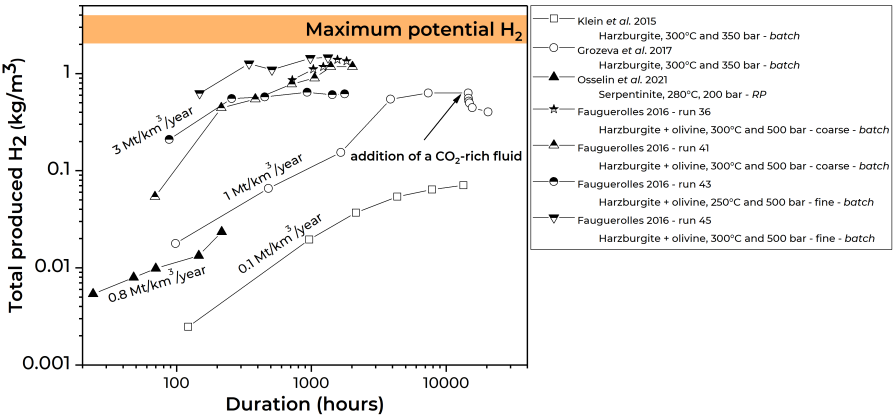

Fig. 2 Measured hydrogen in both batch and reactive percolation experiments. In controlled conditions, the production of H_2 is significant and reaches values close to the theoretical maximum. RP = Reactive Percolation

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9 Author contribution

FO - design of the study, drafting the article and data acquisition. CF - data acquisition. CS, EG and BS revisions. MP - design of the study.

10 Competing interests

The authors declare no competing interests.

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