Article

Natural Hydrogen System in Western Australia?

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Abstract: There is a marked momentum towards the use of clean hydrogen energy as an alternative for fossil fuels. Renewable energies such as solar and wind are being used to generate hydrogen from the water hydrolysis process. Indeed, this approach stores renewable energies in the form of combustible hydrogen for other energy uses. The other alternative that could be economically more cost-effective at the current technology stage is to explore the natural "Hydrogen System" where the natural hydrogen is generated and accumulated within the earth system, the same that stands for a "Petroleum System".

The Discovery of a large accumulation of relatively pure natural hydrogen (H₂) in Mali has triggered the opportunity of searching for natural hydrogen accumulations in other countries. The generation of hydrogen from a circular depression in Mali and some other countries is linked to the presence of geologically very old iron-rich basement rocks. Solid-liquid redox reactions between iron-rich minerals and groundwater that split water are a possible source of H₂ in deep basement rocks. It is believed that the hydrogen degassing may be detected by surface topographic circular to subcircular shallow depressions. Chemical processes such as dissolution by hydrogen are considered to play the main role in the formation of the circular depressions through preferential vertical hydrogen migration channel.

Archean iron-rich Yilgarn Craton that covers a vast area of Western Australia (WA) contains abundant iron-rich mafic-ultramafic rocks. The craton reveals many surficial circular depressions visible through satellite images. The area has abundant fault systems and is blanketed with Eocene sedimentary rocks containing high-quality reservoir rocks. All these characteristics seem to provide most of the required elements, such as hydrogen source, migration pathway, and reservoir rock for a "Hydrogen System" in this area.

Keywords: Natural Hydrogen System; circular depressions; Archean iron-rich Craton; Western Australia

1. Introduction

Hydrogen as the most abundant element in the Universe has the potential to become one of the sources of the future's clean energy. Natural hydrogen emanation could be of practical interest, as it serves as an environmentally friendly combustible fuel source. Hydrogen due to its high reactivity is rare in Earth's atmosphere but the presence of molecular hydrogen (H₂) is reported in many deep boreholes, with no clear explanation for the source (Ward, 1933; Tenenbaum, 2008). Although there are known sources of biotic production of hydrogen in the subsurface, hydrogen is also found in recent hot igneous rocks where no microbes can survive suggesting abiotic sources for hydrogen as well (Tenenbaum, 2008). There are some publications from several countries reporting the presence of naturally emanated H₂ (e.g., Larin et al., 2015; Zgonnik et al., 2015; Prinzhofer et al., 2018). The following paragraphs summaries some of the recent hydrogen discoveries:

Exploration of relatively pure hydrogen reservoirs in Mali (Prinzhofer et al., 2018) highlights the possibility of producing natural hydrogen from hydrogen reservoir rocks. In Mali, H₂ generation and accumulation are related to the presence of multi overlaid doleritic sills and aquifers to play as a seal. Soil analysis above a circular structure (Figure 1A) shows a hydrogen concentration profile in 1-m depth samplings suggesting regular upward hydrogen seeping. Hydrogen generation is believed to be sourced from the basement, as relatively large amounts of radiogenic Helium and Argon are

associated with hydrogen. The current estimate of Mali's hydrogen exploitation price seems cheaper than synthesizing from water hydrolysis.

Seeping of H₂ out from about 562 subcircular morphological depressions is also reported in Russia (Larin et al., 2015). The size of subcircular structures ranges from a hundred meters to several kilometers in diameter (Figure 1B). The perimeter of the structures generally shows a ring of soil-bleaching associated with growth anomalies of vegetation and the cores of the structures commonly covered by marshes or lakes. The subsoil gas composition of one of these structures was estimated as a daily hydrogen flow seeping out at the surface between 21,000 and 27,000 m3 (Larin et al., 2015).

A significant amount of H₂ is also reported in North Carolina (USA) emanating from circular depressions around the Carolina bays (Zgonnik et al., 2015) (Figure 1C). It is reported the surficial circular expressions are from hydrogen gas fluid flow pathways moving from depth to the surface. It is believed the alteration of rock along the deep pathways of H₂ migrating has resulted in local ovoid shape collapse surface patterns.

A circular depression in the Sao Francisco Basin (Brazil) (Figure 1D), emits H₂ that is believed to be generated from deeply seated basement rocks (Prinzhofer et al., 2019).

Yakymchuk and Korchagin (2020) reported the $\rm H_2$ degassing and accumulation in Azerbaijan, Tatarstan, and Latvia. They located hydrogen degassing in these areas in circular structures associated with channels of vertical migration of fluids and minerals matter filled above basaltic rocks (Figures 1E & 1F). They introduced the application of frequency-resonance methods of satellite images and photographs processing to identify the location and the depth of natural hydrogen accumulation sites.

Based on Guélard et al., (2017) several wells in Kansas have shown occurrences of H₂ rich gases. Near the H₂-bearing wells silicic igneous and metamorphic Precambrian basement rocks are found below the Paleozoic strata. They propose that the presence of iron-rich basement rocks has generated H₂ by coupled Fe⁺² oxidation and reduction of H₂O.

An abnormally high H₂ concentration in the eastern coastal area of China is reported by Hao et al., (2020) to be dominantly originated by the reduction of water and oxidation of Fe⁺²-rich pyroxene and olivine (serpentinization) in the basalt under near-surface conditions and migration to a sandstone reservoir. In this area, the basement is composed of Archaean and Paleoproterozoic metamorphic rocks.

Interestingly, most of the above H₂ discovery share two distinct similar conditions as follows:

- They are associated with circular to sub-circular depressions,
- They are located in areas with iron-rich, metamorphic, and igneous Precambrian basement rocks where Fe^{+2} oxidation and reduction of H_2O may occur.

In Australia, specifically in Western Australia (WA), the above two conditions exist. The satellite images of WA shows abundant magnificent circular to subcircular depressions formed over relatively thin Tertiary sediments that blanket Archean Yilgarn Craton. Yilgarn Craton is made of iron-rich metamorphosed granites, volcanic rocks, and abundant mafic-ultramafic dykes. Such a condition has promoted this question of whether some of the circular depressions in WA are related to the natural emission of H2. Although this study, at the current stage, does not provide a definitive answer to this question it provides some reasoning that supports the possibility of the presence of such a natural H2 system in WA.

The future step of this research study will be to measure the hydrogen content of the soils within and around circular depressions at different depths using small shallow holes and also to do geophysical imaging to understand the subsurface feature of one of the depressions.

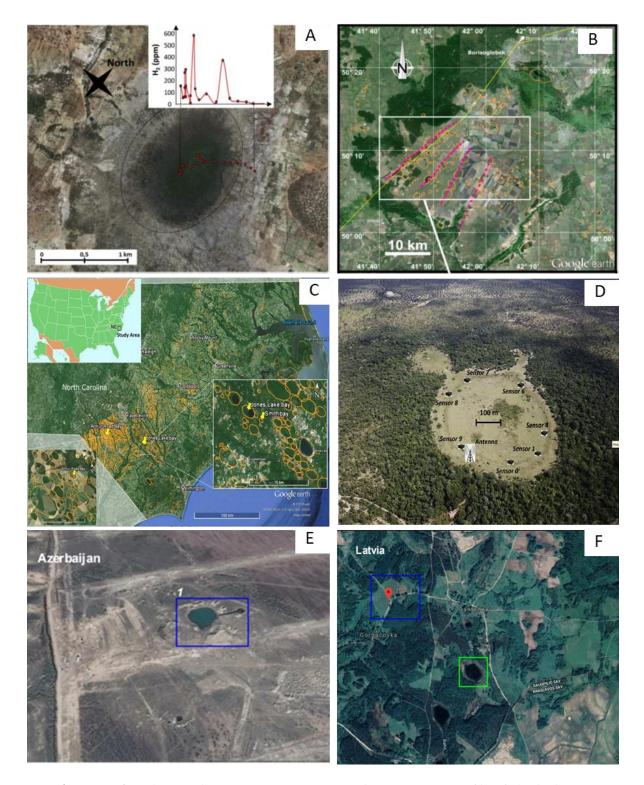


Figure 1. A) Mali's circular structure emanating nearly pure H₂. A profile of the hydrogen concentrations (in ppm) is also presented (From Prinzhofer et al., 2018); **B)** Distribution of depression sizes in Central Russia (Borisoglebsk–Novokhopersk) that are seeping H₂. Structures are outlined in orange polygons, and alignments of structures are shown as pink dashed lines (From Larin et al., 2015); **C)** Location of the Carolina bays seeping H₂ outlined by orange polygons (Zgonnik et al., 2015); **D)** A circular depression of the Sao Francisco Basin (Brazil), H₂ sensors positions can be seen in this photo (Prinzhofer et al., 2019); **E & F)** Satellite images of local sites in Azerbaijan and Latvia. Hydrogen degassing occurs from circular structures formed above basaltic rocks (Yakymchuk and Korchagin, 2020).

2. Hydrogen System in WA

Like a "Petroleum System" a "Hydrogen System" requires several elements including a hydrogen source, a migration pathway, a reservoir rock, and a suitable tarp and seal. Abiotic H₂ can be sourced through water reduction and oxidation of metals, especially iron, in the ultramafic-mafic rocks (Vacquand et al., 2018). In this process, Fe²⁺ is oxidized to Fe³⁺, and water reduction produces H₂. The proposed oxidation of ferrous iron-containing minerals and water, at high temperatures through the following reactions, can generate large quantities of H₂:

$$2FeO + H_2O \rightarrow Fe_2O_3 + H_2$$
 (Equ. 1)

$$2Fe_3O_4 + H_2O \rightarrow 3Fe_2O_3 + H_2$$
 (Equ. 2)

A study by Murray (1974) reveals the concentration of Fe³⁺ above a dolerite dyke in Darling Range, WA (Figure 2) that proves the oxidation of ferrous iron-containing minerals is happening in the area.

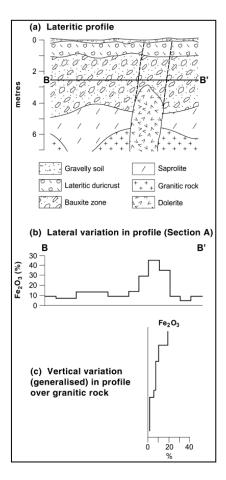


Figure 2. - Iron oxide concentration above granite bedrock intruded by a dolerite dyke (from Murray, 1974).

The serpentinization of the ultramafic rocks, rich in olivine and pyroxene, with water, is another source of hydrogen generation (Brazelto et al., 2012). It is also proposed by Freund et al., (2002) that the subsurface rocks could have comparatively large amounts of H₂, formed inside anhydrous minerals by the redox conversion of hydroxyl pairs. This molecular hydrogen expels out of the minerals when for any reason a fresh surface, such as natural fracture forms.

By cold crushing to liberate volatiles from granites and gneiss of Archean and Palaeoproterozoic (>1600 Ma) age, Parnell and Blamey (2017) demonstrated that they contain an order of magnitude greater hydrogen than in very young (<200 Ma) granites. Lollar et al., (2014) estimated that H₂

production rate via both radiolytic dissociations of water and hydration of mafic/ultramafic rocks from the Precambrian continental lithosphere is about 0.36–2.27×10¹¹ moles per year.

In WA although there are no direct pieces of evidence to report the presence of H₂ but some natural chemical processes may suggest that hydrogen must be involved with them. For example, diagenetic alunite is pervasive in many of Western Australia's salt lakes. It is believed that Yilgarn Craton has provided conditions for the generation of high acidity water and alunite formation. The formation of alunite requires acidic conditions so that Al³⁺ and Fe³⁺ may be mobilised (McArthur et al., 1991). Indeed, many salt lakes in south-western WA are acidic (McArthur et al., 1991) and their pH ranges from 2.8 to 6.7. Mann (1983) suggested that the widespread occurrence of acidic water in Western Australia could be explained by weathering of Fe²⁺ in bedrock minerals, followed by diffusion of the Fe²⁺ to the oxic zone of the water table where its oxidation and hydrolysis (ferrolysis) generated acidity. The Source of Aluminium in Alunite is suggested to be from kaolinite that has been disintegrated by H⁺ generated during ferrolysis (McArthur et al., 1991). Another example is the pervasive laterite formation in Yilgarn Craton reported by Mann (1983) that also forms as a result of ferrolysis. In general, the Western Australia ironstone pavements are linked to the transport and deposition of Si and Al generated by acidic weathering as a result of ferrolysis (McArthur et al., 1991). This has to be noted that both weathering of Fe²⁺ in bedrock minerals and ferrolysis produce hydrogen.

This has to be noted that one of the reasons that no hydrogen has been reported in the area is the standard analytical approach for gas chromatography often does not detect hydrogen and indeed nobody was searching for natural H₂ in the area to date. This is worth mentioning that a high concentration of hydrogen has been reported in a well drilled in Kangaroo Island, South Australia, in an area very similar to WA (Ward, 1933).

3. Yilgarn Craton

The Archaean Yilgarn Craton in Western Australia with the age of about 4000 Ma covers an area of about 650,000 km². The Craton is made of metamorphosed granites, volcanic, and sedimentary rocks and it has experienced strong metamorphism and hydrothermal activity at about 2700 Ma (Wilde et al., 1996). Cassidy et al. (2006) subdivided the tectonic evolution of the Yilgarn Craton into six terranes (Figure 3). There are series of dyke swarms of different orientations around the margins and into the craton interior (Hallberg, 1987; Wingate 2017). Most of the dyke swarms are dolerite or gabbro in composition. They are poorly-exposed but a high-resolution aeromagnetic study by Isles and Cooke (1990) indicated a high-density swarm of NE-trending dolerite dykes intruded on into Archean rocks of the southeastern Yilgarn Craton (Figure 3). The oldest mafic dykes belong to the east to northeast-trending Widgiemooltha dyke swarm (Pisarevsky et al., 2015). The Widgiemooltha dykes are up to 3.2 km wide, vertical to sub-vertical, and comprise predominantly massive olivine dolerite and gabbro (Myers, 1990). The next extensive dyke swarm is Marnda Moorn LIP (1210 Ma) intruding along the craton margins (Isles and Cooke, 1990). Other identified dyke swarms include the NW trending Boonadgin dyke swarm (1888 Ma) in the southwest (Stark et al., 2017).

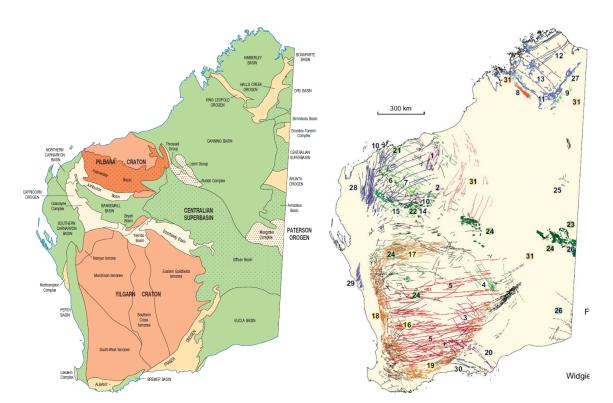


Figure 3. – Left) Yilgarn Craton of WA is subdivided, based on the tectonic evolution, into six terranes (Cassidy et al. (2006) in Myer and Hocking (1998). Right) Map of dyke and sill suites in Western Australia. Different sets of dykes are introduced in a different color (Modified from Wingate 2017).

4. Circular to subcircular depressions in WA

Satellite images from Google Earth reveal a large number of circular to subcircular depressions in WA (Figure 4). They are unique features that widely occur across south-western WA. In general, most of the depressions are along paleochannels. Some of the depressions are roughly oriented in the direction of NW-SE following the linear faults and dykes trends. There are some other relatively smaller depressions, specifically at the southwest of WA, that do not show a clear relation with any drainage systems and are scattered throughout the area. Indeed, some of the depressions are quite isolated with no connections to other depressions. The size of the depressions ranges from about less than 100 m to above 3.0 km in diameter. Based on the Satellite images from Google Earth Pro, the depth of depressions varies from about 1 to 10 m, for example, it is about 10m in Figure 4D. There are some signs of possible discoloration and variation in the type of vegetation due to gas emanation close to some of the depressions (Figure 5).

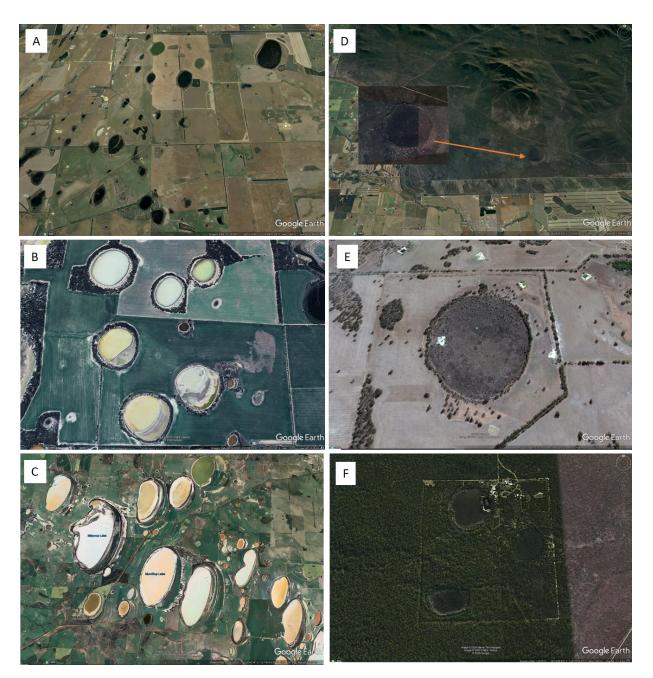


Figure 4. – Examples of circular/subcircular depressions in WA. Some of the depressions do not show a clear relation with drainage systems (e.g., A, and B) whereas some are aligned with paleochannels (e.g., C,). Some depressions are close to foothills (D, inset shows a close-up view), some are dry (e.g., E) and some are partially or completely covered by vegetation (e.g., F).

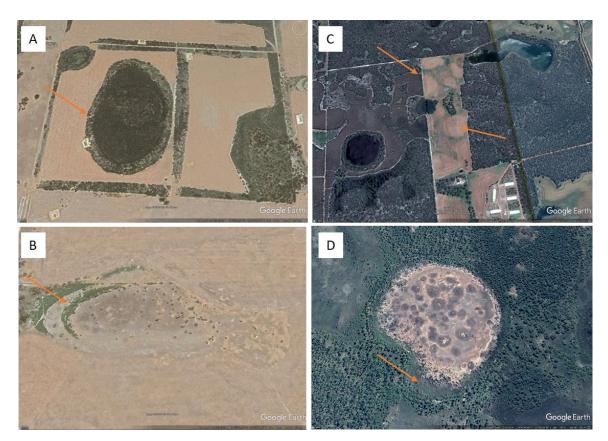


Figure 5. - Some of the depressions show clear discoloration and variation of vegetation (arrows) that could be due to gas emission.

Many publications relate the presence and the shape of the circular depressions to several natural processes such as tectonic rejuvenation, climate change, wind activity, paleomorphology, etc., and consider them as playas and salt lakes. Bettenay (1962), based on field studies and examination of aerial photographs, concluded that some of the salt lakes in WA are originated from river systems and wind modification in arid climatic phases during the Tertiary period. Van De Graaff (1977) related the oval depressions to playas or salt lakes formed along with paleodrainage systems which stopped flowing regularly when the Australian climate changed from humid to arid. English (2016) attributed some of the salt lakes that are disconnected from major drainages and that lie within basins containing ancient evaporite units, to the salt diapir process.

Although the formation of most of the large salt lakes of WA can be attributed to the above-mentioned processes the question is why these natural features are abundant in WA and do not exist in other places with nearly the same tectonic and climate conditions. This has to be noted even in WA the abundance of the circular depressions vary from one location to another. Besides, there are many other drainage channel systems in WA with no circular depressions associated with them.

This has to be noted that some of the circular depressions are very similar to playa lakes or salina and are formed within paleochannels. But there are so many other circular depressions in the area that cannot be easily related to any drainage systems and there is no wind-blown sediment around to shape them. Salama (1997) reported that the type of sediments in some of the oval lakes (e.g., Yenyening Lakes) indicate deposition in a closed system. This suggests that some of the circular depressions could be isolated features with no relation to any drainage system. Moreover, some of the depressions are dry and are close to the foothills with no indication of any drainage system (e.g., Figure 4D).

Zgonnik et al., (2015) and Larin et al., (2015) suggest that hydrogenation of rocks will generally produce acid that can be mobilized and dissolve the rocks during upward movement by gas creating a preferred vertical migration pathway or channel. The preferred migration pathway will generate excessive dissolution that can end up to a collapse structure and subsidence that may be evident as

circular to subcircular surface depressions. Besides, it is believed that the diffusive efficiency of hydrogen into other minerals such as calcite can make them prone to further change and make them brittle and prone to collapse.

The explanation of the formation of some of the circular depressions along paleochannels could be due to the tendency of paleochannels to align with paleotopography. Paleotopography lows, where surface water prefers to run, somehow can be controlled by faults vertical displacement. In such a situation running water acts as a source of oxidation agent penetrating deep into the iron-rich basement rock. Faults play the role of conduit to expose basement rock to oxidizing agents. Paleotopography lows may also coincide with low weathered exposed ultramafic rocks, such as olivine-rich doleritic dykes that are abundant in the area, where the serpentinization process and thus H₂ production may occur. This is supported by some publications reporting high concentrations of H₂ along some Japanese faults (Sugisaki et al., 1983), along the San Andreas and Calaveras faults in central California (Sato et al., 1986); and many other places (e.g., Wakita et al., 1980; Ware et al., 1985).

5. Discussion

There are many natural ways of H₂ generation such as ferrous minerals oxidation; water radiolysis; decomposition of methane; organic matter alteration; and H₂ emission from deep Earth. From published papers, it appears that the alteration of Fe⁺²-bearing minerals is the most commonly reported source of natural H₂ seepages on Earth (Zgonnik et al., 2015). A study by Lollar et al. (2014) reported that the H₂ generation from the Precambrian continental lithosphere has been underestimated and needs to be revisited.

The Archaean Yilgarn Craton in Western Australia has all the required elements for a Hydrogen System to generate and preserve a vast amount of H₂. The following pieces of evidence may support this opinion:

- The Archean Yilgarn Craton with abundant iron-rich rocks and ample mafic to ultramafic dyke swarms support the presence of the source for H₂ generation. The oxidation of ferrous iron-containing minerals and serpentinization of ultramafic minerals such as olivine and pyroxene that are reported to exist in WA doleritic dykes could be a vast sustainable source of H₂ generation.
- The presence of a complex set of fault systems that can play the migration pathway. Faults are believed to act as the fluid pathway from the deep subsurface to the surface or shallower part of the earth's crust. In some areas of the Earth degassing of the interior is often observed along with some deep faults (Sugisaki et al., 1983; Shangguan et al., 2000). The gas emission varies noticeably depending on the fault activity. Larin et al. (2014) reported hydrogen emission along some structural trends associated with basement faults in Russia.
- A blanket of Eocene sedimentary rocks, that covers the Yilgarn Craton, can provide suitable reservoirs and caprock to store and to prevent further upward migration of hydrogen.
- The presence of abundant circular depressions that are proved in many countries to be the sign of H₂ emanation from the subsurface.

There are many publications about the formation of the circular depressions in WA where most of them suggest they are associated with the tectonically uplifted paleodrainage systems in the arid and semi-arid environment. Based on the publications, the role of wind-blown dunes and the prevailing wind direction has also played an important role to shape these so-called playa or salt lakes. The above-mentioned elements that highlight the formation of the pervasive circular depressions in WA and other Australian states cannot be ignored since they might be correct for some of the lakes. But the following points urge us to reconsider other elements that may help to form some of the isolated peculiar circular depressions in the area:

 In WA there are so many other paleodrainage systems that do not show any circular depressions. Besides, there is no universal solid evidence to prove that paleodrainage develop circular lakes in arid condition,

- There are many circular depressions in WA that cannot be easily related to any paleodrainage system. Indeed, some of the circular depressions seem quite isolated with no connections to other depressions. Some of the circular depressions are on the topographically elevated area with no drainage systems present.
- 3. Not all of these structures are associated with any sand-blown dunes to shape them,
- 4. Many of the circular depressions are aligned with some linear deep faults and doleritic dykes where hydrogen generation and migration can occur. This is worth mentioning that some of the paleodrainage systems may have formed above paleovalleys of the Archean basement. The paleovalleys may have been aligned parallel with structural trends such as deep basement faults that are the conduit of hydrogen. Airborne electromagnetic conductivity depth image (AEM-CDI) is a method for geological mapping and mineral targeting by detecting variations in the conductivity of the ground to a depth of several hundred metres. The method perhaps enables us to locate the migration pathway of hydrogen where rock alteration and possibly concentration of conductive iron-rich mineral has occurred. Figure 6 shows a deeply rooted fluid migration pathway where conductive minerals (e.g., iron) are concentrated.
- 5. Most of the structures are associated with low pH acidic water,
- 6. The presence of high iron oxide concentration above doleritic intrusions,
- 7. Last but not least, based on Ward (1933) a high concentration of hydrogen (84%) was recorded at the depth of 1666 ft in a borehole drilled at Kangaroo Island, South Australia (Table 1). In Kangaroo Island, the same as Western Australia, the Archean basement is very close to the surface and many circular depressions can be seen in the area close to the drilled borehole (Figure 7).

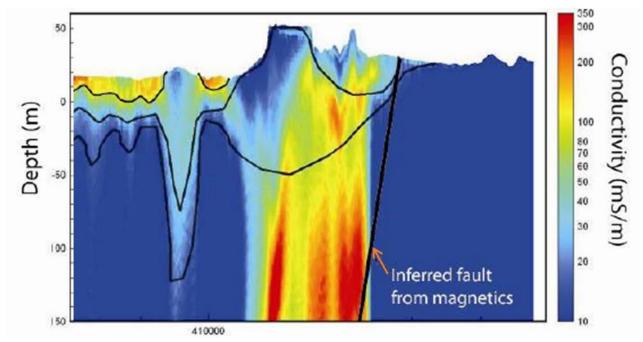


Figure 6. - Airborne electromagnetic conductivity depth image (AEM-CDI) at Fortescue River, WA. Although this profile does not cross any circular depression it shows that faults are the conduit of fluid migration and mineralization in the area (Modified from Department of Water Government of Western Australia, 2009).

Table 1. Gas sample analysis for a borehole in Kangaroo Island (Ward, 1933). Note the high concentration of H₂ collected at the depth of 1666 ft.

	I.	II.	III,	IV.	V.	VI.
Depth from surface	790 ft.	790 ft.	860 ft.	860 ft.	860 ft.	1,666 ft.
Carbon dioxide	0.8%	0.2%	0.8%	0.8%	0.6%	Nil
Oxygen	Nil	Nil	3.2%	2.4%	3.0%	1.2%
Ethylene, etc	Nil	Nil	Nil	Nil	Nil	Nil
Carbon monoxide .	Nil	Nil	Nil	Nil	Nil	Nil
Hydrogen	74.0%	76.0%	60.0%	64.4%	60.0%	84.0%
Methane	7.5%	7.5%	5.4%	7.0%	5.6%	Nil
Nitrogen						
(by difference)	17.7%	16.3%	30.6%	25.4%	30.8%	14.8%
	-					***************************************
	100%	100%	100%	100%	100%	100%

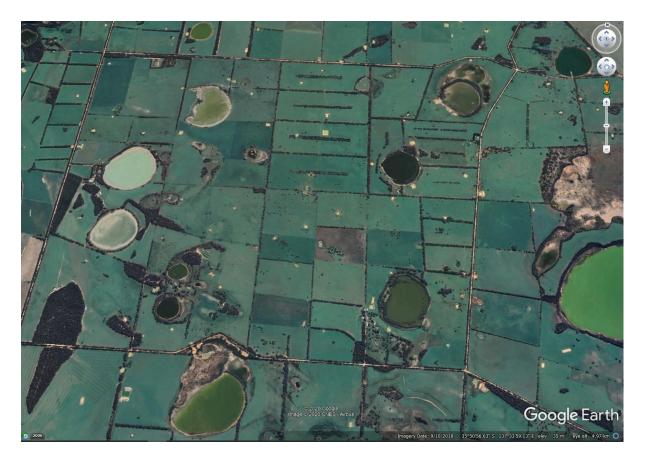


Figure 7. – Satellite image from part of Kangaroo Island, South Australia, where a high hydrogen concentration (84%) was reported by Ward (1933) while drilling a well in the area.

6. Conclusions

The knowledge of "Hydrogen System" is in its infancy stage and needs to be studied worldwide. There are many questions about the main global mechanism of natural H_2 generation, storage, fluid flow mechanism in porous media, and their trapping and sealing efficiency. The possibility of natural accumulation of the H_2 in the porous and permeable intervals and sealing capacity of caprock to efficiently prevent hydrogen to escape from reservoir rocks need to be evaluated.

Since the chemical and physical properties of hydrogen are different from other natural gases, such as methane, the effects of hydrogen on the reservoir rock, caprock would be different.

It is not clear yet in a high concentration of H_2 (e.g., Mali's H_2 natural accumulation) at reservoir condition hydrogen-bacteria, which use H_2 for reductive and energy-yielding purposes, would be active or not. Based on a modelling study by Hemme and van Berk (2018) at pressure and temperature around 2300psi and 80° C sulfate-reducing bacteria and methanogenic bacteria are not active.

Hydrogen has a higher diffusivity, lower viscosity, and lower density when compared to methane. This leads to high mobility and therefore the potential to escape through caprock (Ebigbo et al., 2013). The chemical reaction of hydrogen with caprock that may enhance or reduce the sealing efficiency is still unknown, although hydrogeochemical modelling by Hemme and van Berk (2018) suggest negligibly small effects of H₂ on the caprock mineralogy.

In conclusion, there is a pressing need for a detailed field study to prove the hypothesis of the presence of a complete "Hydrogen System" in WA. The first stage is to categorize different types of circular depressions using satellite images in the area and then choose the most suitable ones to test the presence and possible emanation of H₂ from them. The H₂ may be a continuous and sustainable generation that could be economically viable for collection or it may be commercially accumulated in some hydrogen traps in the area.

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