Superconductivity for hydrogen economy

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Abstract. The emerging hydrogen economy is expected to deal with a large amount of liquid hydrogen produced from the renewable energy resources. The main advantage of liquid hydrogen in comparison with other forms of its storage and transportation is in allowing wide use of superconductivity, which would optimise energy efficiency of the economy. The basic element of the infrastructure for hydrogen economy is a network of superconducting pipelines carrying simultaneously liquid hydrogen and loss-free electricity. The most likely material for such infrastructure is MgB$_2$, the only superconductor efficiently working at boiling temperature of liquid hydrogen and not showing strong critical current reduction on grain boundaries. The cheap techniques for the preparation of MgB$_2$ are hot isostatic pressing, resistive sintering and paint coating. These and other advanced techniques are able to provide MgB$_2$ with suitable for the infrastructure structural and superconducting properties. The preparation of a large-area superconducting joint between two pieces of MgB$_2$ as a technique enabling this infrastructure is reported. A potential of synergy between liquid hydrogen and superconductivity is revealed in a range of possible new energy applications.

1. Introduction
In a few tens of years, the age of fossil fuels with their wide use in transport and energy generation will mainly be over. Still, the demand for energy is expected to grow, and only a short time is available to reshape the energy economy making it fossil fuels-free. Moreover, a significant reduction in the use of fossil fuels is needed to decrease CO$_2$ emissions leading to global warming. The only reasonable response to growing energy demand and necessity to remove fossil fuels from transport and energy generation is development of hydrogen economy.

Hydrogen is a convenient clean fuel giving no CO$_2$ emissions. It could be produced in abundance from renewable energy resources such as solar, wind, marine, hydro, geothermal, biomass and also from controllable nuclear and thermonuclear reactions. The abundance of liquid hydrogen would allow wide use of superconductivity, making energy use more efficient and eliminating its unnecessary losses. This work describes materials basis for the development of hydrogen economy starting with its infrastructure in the form of a network of superconducting pipelines carrying simultaneously liquid hydrogen and loss-free electricity. The MgB$_2$ pipes can be produced by hot-pressing, hydro-extrusion or by coating MgB$_2$-compatible materials with a superconducting paint. The superconducting properties of hot-pressed bulks, paint-coated surfaces and superconducting MgB$_2$
joint are reported. A potential of the synergy between liquid hydrogen and superconductivity is described in a range of applications.

2. Experimental

Three experimental techniques for the preparation of MgB$_2$ are used in this work; they are hot isostatic pressing (HIP), resistive sintering (RS) and paint coating (PAC). HIP is a standard technique that was very successful in application to MgB$_2$ [1-6]. RS [6,7] is an original technique. Its closest analogue is spark plasma sintering (SPS) [8,9]. PAC is an original innovation of the author.

HIP and RS techniques are schematically shown in figures 1 and 2. In HIP (figure 1), a pre-compacted powder is enclosed in a steel container, evacuated and pressed in a high-pressure (typically at 1 kbar) argon chamber at temperature of 940–1000 °C for 4–12 hours. A medium size chamber allows to process thousands of cubic centimetres of MgB$_2$. Much bigger volume could be processed in industrial chambers.

The RS (figure 2) is performed in a vacuum chamber. The MgB$_2$ powder is uniaxially pressed in a graphite die with tungsten rods simultaneously acting as electrodes carrying high electrical current up to 1000 A. The primary role of the electrical current is to heat the powder when, on the initial stage, MgB$_2$ particles are in weak contact and the heat mainly comes from the current flowing in the carbon die. At a temperature of about 700 °C the powder becomes dense and current flows mainly through MgB$_2$. Moreover, high current ignites plasma between the isolated MgB$_2$ particles. The term ‘spark plasma sintering’, which is frequently used to describe the process [8,9], comes from this phenomenon. The plasma welds particles into dense sintered pellet with strong superconducting properties. RS is an express-method allowing to produce MgB$_2$ samples quickly. It is a method of choice for testing nanoparticle additions to MgB$_2$. Typically porosity in RS samples is about 15%.

![Figure 1. Schematics of hot isostatic pressing. The MgB$_2$ in the centre of the evacuated stainless steel can is subjected to high pressure and high temperatures. The arrows show the direction of isostatic pressure applied through the pressurised argon.](image)

![Figure 2. Schematics of resistive sintering. The MgB$_2$ is in the centre of the carbon die between the tungsten rods. It is subjected to the high pressure, high temperatures and high current. The arrows show the direction of uniaxial pressure applied by the tungsten rods that simultaneously carry high electrical current.](image)

HIP allows producing dense samples with lower porosity (5-7%), bigger volume and better superconducting properties than in RS samples. However, it is a slow process involving long preparation procedures.
MgB$_2$ samples prepared by HIP or RS are cut on a wire spark erosion unit into small, about $0.2 \times 0.2 \times 3-4$ mm$^3$, bars and their magnetic moment ($m$) is measured on a Quantum Design Magnetic Properties Measurement System MPMS-XL in DC field $\mu_0H$ up to 7 T. The critical current density is derived from $m$ using equation:

\[ J_c = \frac{4m}{a^2bc(1-\frac{a}{3b})}, \]

where $a$, $b$ and $c$ are dimensions of the bar ($c>b>a$) and magnetic field is applied along the longest $c$-axis. The small size of the samples is necessary because a very high $J_c$ in MgB$_2$ generates large magnetic moment exceeding the limit of $m$ measurable in MPMS.

The equation (1) represents a critical state model, which assumes that the current is constant and equal to $J_c$ in every point of the cross-section of the sample. The $J_c$ is considered to be field ($H$) dependent and the gradients of $H$ are ignored. This equation can be obtained by integration from the definition of $m$. In specific case of the MgB$_2$ superconducting joint, a more general formula has been derived treating the case of a weak link between the two pieces of a superconductor.

The superconducting joint was produced using the RS set-up. First, two 5-mm diameter and 3-mm thick sintered pellets of MgB$_2$ were produced by RS. The jointing surfaces were polished and the pellets were placed back into the carbon die with a thin layer of MgB$_2$ powder between them. After that, routine resistive sintering has been performed on the stack. The jointed pellets were sliced by the spark wire cutting and the bars were cut perpendicularly to the joint surface as shown in figure 3.

![Figure 3. Procedure of jointing two pellets of MgB$_2$. The position of the joint is shown by thin black line. The red arrow shows the direction of pressing and the green arrow shows the order of the jointing and cutting the sample.](image)

The paint in the PAC method was prepared by mixing fine MgB$_2$ powder with a liquid hydrocarbon. The paint is of a brown colour, has good adhesion to the surfaces and can be spread evenly using a brush. When treated at a high temperature in oxygen-free atmosphere, it produces a thick superconducting MgB$_2$ cover.

The magnetisation measurements have been performed on all samples and a variety of visualisation techniques was applied to investigate their microstructure, namely polarised optical microscopy, scanning electron microscopy, transmission electron microscopy (TEM), electron backscatter diffraction (EBSD), atomic force microscopy (AFM) and scanning conductivity probe microscopy (SCPM).
The described methods, together with other techniques developed for MgB$_2$ and high temperature superconductors (HTS), form a critical mass for wide penetration of superconductors into developing hydrogen economy. The examples of other successful MgB$_2$ techniques include magnesium infiltration [10], reactive evaporation [11] and shock consolidation [12]. Together with high demand for energy, scarcity of fossil fuels and dangers of CO$_2$ emissions, superconductivity is posed to be a major driving factor of hydrogen economy.

3. Results and discussion
The main parameters in applications of superconductivity are critical temperature ($T_c$) and critical current density ($J_c$), or, more precisely, total critical current ($I_c$) and total critical current per centimetre of width: $I_{c-w} = J_c d$, when a flat superconductor of the thickness $d$ is considered. The value of $J_c$ in high $H$ is of primary importance due to the dominant use of superconductors for high magnetic field generation. Because $J_c$ rapidly decreases when temperature approaches $T_c$, liquid hydrogen applications require critical temperature well above boiling temperature of liquid hydrogen. Both MgB$_2$ and HTS satisfy this requirement. However, in HTS $J_c$ exponentially decreases with the increase of misorientation angle between the grains. This restricts the use of HTS to their low-$I_{c-w}$ applications with samples prepared on textured substrates or high-$I_c$ applications with small-volume textured bulks. In addition to MgB$_2$ and HTS there are several other superconducting materials, including recently discovered iron-based superconductors [13], but all of them are sensitive to grain misorientations. The only exception is MgB$_2$ and this makes it most suitable superconducting material for hydrogen applications. The following sections describe structural and superconducting properties of MgB$_2$ obtained by techniques described in section 2 in comparison with other materials and techniques.

3.1. Structural properties of MgB$_2$ samples
Figure 4 shows a dense MgB$_2$ sample produced by hot isostatic pressing. The material is of gold-like appearance in the center of bakelite disk surrounded by the white stainless steel ring. Several bars are cut from the HIP MgB$_2$ and placed on the disk. MgB$_2$ is a hard light material three times lighter than stainless steel. The density of the samples is close to that of single crystalline MgB$_2$ with the pores accounting for about 7% of the volume.

Figure 5 shows an AFM image of the surface of HIP MgB$_2$ on a sub-micron scale. Few pores and inclusions are seen. Nano-inclusions of secondary or external phases play an important role in grain boundaries generation. One of such nano-inclusions is shown in the center of the image marked by the red arrow. The nano-inclusion forms two straight grain boundaries with an angle of about 50° between them. Such grain boundaries contain large number of dislocations important for increasing $J_c$ in the sample. The AFM imaging allows to visualize few grain boundaries etched by the water in the process of sample polishing, while an ultimate method to image grain boundaries that does not require etching is EBSD. The AFM and EBSD images of the same area of MgB$_2$ are shown in figures 6 and 7. The EBSD allows to determine accurately the orientation of all grains on the surface of the sample. The application of EBSD to MgB$_2$ was very successful and clarified the role of grain boundaries in pinning providing large $J_c$ in high magnetic fields [6,14].

More detailed information about grain boundaries could be obtained by the application of scanning conductivity probe microscopy (SCPM) that measures local conductivity of the surface with the nanometer resolution. The comparison of SCPM and AFM is given in figures 8 and 9. In SCPM, contrast represents local conductivity of the surface. This method allows to identify grain boundaries that are not seen in AFM, like the boundary in figure 9, which crosses the center of SCPM image at the angle of about 15°. The combination of AFM, CSPM and EBSD gives complete information that allows determining relation between the local resistance of the grain boundaries and the angle of their misorientation, as shown in figure 10.
Figure 4. MgB$_2$ sample produced by hot isostatic pressing. It is in the centre of bakelite disk surrounded by the stainless steel ring and several bars cut from pressed HIP MgB$_2$. These are placed around the MgB$_2$ disk in a form of pentagon.

Figure 5. A sub-micron scale AFM image of the surface of HIP MgB$_2$. The red arrow shows a nano-inclusion that forms two straight grain boundaries of the angle of about 50°. Such grain boundaries are important for increasing $J_c$ in the material.

Figure 6. AFM image of a polished surface of HIP MgB$_2$. The areas of different orientation have different rate of polishing, those with $c$-axis perpendicular to the surface having the highest rate.

Figure 7. EBSD image of the surface of HIP MgB$_2$ shown in figure 6. The areas of different orientation are clearly distinguished and shown in the different colours.
Figure 8. AFM image of a surface of HIP MgB$_2$. Several grain boundaries are exposed due to water etching during polishing.

Figure 9. SPCM image of the surface of HIP MgB$_2$ shown in figure 8. The contrast of the lines reflects local conductivity of the boundaries.

Figure 10. Surface resistance as a function of misorientation angle for grain boundaries shown in figures 8 and 9.
Figure 10 shows that a strong change in local resistance of grain boundaries takes place at an angle of about 40°. It is likely that at this angle the density of dislocations in the grain boundaries is high enough to overlap their cores, which, in its turn, decreases $J_c$. This identifies a subset of highly misoriented grain boundaries that have negative effect on $J_c$. Engineering samples without these boundaries would improve effectiveness of MgB$_2$ in practical applications.

3.2. Superconducting properties of MgB$_2$ and YBa$_2$Cu$_3$O$_x$ samples

Figure 11 compares $J_c$ at temperature of 20 K and different magnetic fields for a range of superconducting samples that are important for the development of hydrogen economy. The black filled squares show $J_c(H)$ of conventionally sintered (CS) MgB$_2$ sample. It has the lowest critical current density in a wide range of magnetic fields. The HIP procedure (black open squares) significantly increases $J_c$. The addition of SiC [3] (blue open squares) makes $J_c$ even higher in high magnetic fields. The RS sample (filled green squares) has $J_c$ comparable with that of HIP samples in low magnetic fields, but shows a lower $J_c$ in high fields.

Figure 11. Magnetic field dependence of critical current density in MgB$_2$ and YBa$_2$Cu$_3$O$_x$ samples at temperature of 20 K.

The MgB$_2$ powder-in-tube tape with addition of nano-carbon [15] (filled magenta squares) has lower $J_c$ than in bulk HIP in low field, but high $J_c$ in high fields. When HIP procedure is applied to the power-in-tube tapes [1], it increases $J_c$ in high field (filled olive squares) above that in CS MgB$_2$. The red open circles show $J_c(H)$ for an epitaxial MgB$_2$ film [16]. Such film does not have high-angle grain
boundaries. It is likely that this is the reason why $J_c$ of the film is much higher than in all other MgB$_2$ samples in low magnetic fields. However, due to small amount of dislocations residing on grain boundaries, $J_c$ in high fields is low. The specially designed HTS films, for example YBa$_2$Cu$_3$O$_x$ film composed of nano-columns [17] (filled red squares) have much higher $J_c$ than in any other sample in high fields. Although the thickness of HTS films is limited by few microns, they can be used in many liquid hydrogen applications. Overall, critical current density in the samples shown in figure 11 is sufficient for most liquid hydrogen applications and the relevant techniques only need a scale-up to the industrial level.

3.3. Superconducting MgB$_2$ joint

The superconducting properties of the samples shown in figure 11 were measured on small pieces of material, typically few millimetres in length. The infrastructure for hydrogen economy would, however, require much longer superconductors. The pipelines, for example, may need to be thousands kilometres long. Although it is easy to maintain high $J_c$ on long distances in continuous pipes, they are likely to be constructed from sections, and the joints between these sections may strongly affect $J_c$. HTS are unlikely candidates for the pipelines because of difficulties in making HTS superconducting joints. Fortunately, low MgB$_2$ sensitivity to grain misorientations makes its jointing much easier than in HTS. As it is described in section 2, the procedure could be based on the combination of moderately high pressure of few hundred bars, high temperature of few hundred centigrade and a buffer layer of MgB$_2$ powder. This is now a general approach to MgB$_2$ jointing, see for example [18].

Figure 12 shows the joint area of the sample produced by the RS technique shown in figure 2. The buffer layer, shown by arrows in the lower part of the plot, is about 200 µm thick. It is hardly distinguishable from the rest of the sample.

\[ J_c (10^6 \text{ A/cm}^2) \]

\[ T (\text{K}) \]

\[ H = 0 \]

**Figure 12.** Optical image of the MgB$_2$ joint. The buffer layer is about 200 µm thick. It is marked by the arrows in the lower part of the plot.

**Figure 13.** Temperature dependence of the critical current density of the joint shown in figure 12 (black squares) and bulk MgB$_2$ adjacent to the joint (red squares).

The joint in figure 12 is prepared from the material containing nano-inclusions of Dy$_2$O$_3$ [19]. In spite of high porosity partially coming from the non-evenly distributed Dy$_2$O$_3$, the critical current density in this sample is high, as shown by the temperature dependence of $J_c$ in figure 13. Most importantly, $J_c$ of the current through the joint (black squares) is close to $J_c$ in the bulk (red squares).
3.4. *MgB$_2$ paint coating*

The critical current density of HIP MgB$_2$ is so large that a pipeline of inner diameter of 18 centimetres and outer diameter of 20 centimetres would be able to carry a power of up to 10 GW without losses. This is sufficient to connect a large city and a big power station. However, only a small amount of hydrogen could be pumped through such a pipeline. A long distance pipeline supplying large amount of liquid hydrogen would require MgB$_2$ pipes of a bigger diameter. Such pipes would be difficult to produce by HIP, even if this method is extended to the continuous process of hot hydro-extrusion. The ideal solution for such a pipeline would be paint technology. A simple painting of a stainless-steel pipe by MgB$_2$-based paint followed by thermal treatment may produce a thick superconducting layer suitable for the transport of supercurrent. Such a technology would be intrinsically free from the superconducting joint problem.

An optimal solution would be a pipe of inner diameter of 1.3 meters delivering 10 GW of electrical energy and 20 GW of energy in the form of liquid hydrogen. Such a pipeline will be suitable for transportation of electricity and hydrogen on distances of several thousand kilometres. Similarly to the small-diameter pipeline described above, this pipeline would feature double layer of MgB$_2$ for the two-way transport of current necessary to close delivery loop and to cancel strong magnetic field induced by the current. An important feature of the pipeline would be its magnetic suspension based on the property of superconductor to levitate above the magnet. Such a suspension would reduce stresses and vibrations that may affect its integrity.

To clarify the viability of large-diameter pipes, the paint technology was tested with MgB$_2$ paint prepared as described in the section 2. Figure 14 shows temperature dependence of the real (lower part of the plot) and imaginary (upper part of the plot) AC magnetic moment of a thick paint cover on Ta (black squares) and stainless steel (blue and red squares). Although $T_c$ of the cover is lower than that of pure MgB$_2$, the diamagnetic response at 20 K is strong. The first layer of MgB$_2$ on Ta is superconducting (black squares), whereas first layer on stainless steel shows only traces of superconductivity (blue symbols) due to chemical interaction of the paint with iron. However, second layer above the first one shows strong superconductivity (red symbols). There is no restriction on the number of layers or the thickness of the paint cover.

![Figure 14. Temperature dependence of real (lower part of the plot) and imaginary (upper part of the plot) AC magnetic moment of thick paint cover on Ta (black squares) and stainless steel (blue and red squares).](image1)

![Figure 15. Total critical current per centimetre of width for the layers shown in figure 14 with the matching colours in magnetic field of 0.1 T. The $I_{c,w}$ of the first MgB$_2$ layer on stainless steel is not shown.](image2)
Figure 15 shows total critical current per centimetre of width in 0.1 T magnetic field for the layers shown in figure 14. It is the first test of the paint and it is believed that its optimisation together with proper treatment of the substrate would significantly increase $I_{c-w}$.

The hydrogen pipeline infrastructure is designed to serve carbon-free economy. Unexpectedly, production of MgB$_2$ offers another possibility to tackle CO$_2$ emissions. A disadvantage of pure MgB$_2$ in comparison with HTS is in its relatively low critical magnetic field. This could restrict its use in high-field applications. A way to increase critical field is incorporation of a small amount of carbon into the crystal lattice of MgB$_2$ [20]. In coated paint technique, it is achieved by mixing hydrocarbons with MgB$_2$ powder. During the thermal treatment hydrocarbons decompose, carbon enters crystal lattice of MgB$_2$ and is trapped there permanently (no CO$_2$ emission, no necessity for CO$_2$ storage) while hydrogen is released and can be collected and used as a fuel. This process could be one of the most effective ways of decomposing hydrocarbons. With well-developed MgB$_2$ powder metallurgy producing thousands (possibly millions) tons of MgB$_2$, this process would significantly reduce emissions of CO$_2$ from otherwise burned hydrocarbons.

4. Synergy of hydrogen economy and superconductivity

The pipeline infrastructure is one of the applications of superconductivity for hydrogen economy. When supply of liquid hydrogen from renewable resources is secured, superconductivity would find wider applications. It could be implemented in car manufacture. There are already several models of cars using liquid hydrogen. The tank of a car could contain up to 100 litres of liquid hydrogen and, utilising its low temperature, car could be fitted with superconducting devices.

The car can feature compact and efficient superconducting motor; superconducting generator to cover electrical needs of the car, superconducting energy storage unit; loss-free superconducting wiring; superconducting current limiter, protecting electrical system of the car; superconducting (possibly quantum) computer etc. Some of the superconducting components, for example, flexible flat MgB$_2$ superconducting cable [21], are already developed. It would also be possible to design superconducting train, plane, submarine, spaceship, and built superconducting home energy unit. The home unit would contain some 200-300 litres of liquid hydrogen and have superconducting devices as in the superconducting car adapted for the home use. The house would be self-sufficient in energy for some time and immune to blackouts. This would be of special importance for hospitals and defence installations.

Superconductors could also find wide use in the screening of magnetic field. In particular, MgB$_2$ is attractive for Space applications, since it is a light superconductor and due to low temperatures in Space, it would work there with limited amount or even without liquid hydrogen. The MgB$_2$ paint could be a method of choice for these applications, especially on the Moon’s surface or Earth’s orbit, where it can be treated in oxygen-free environment. Small MgB$_2$ cylinders showing complete screening of magnetic field have already been demonstrated [22].

When liquid hydrogen is produced on a large-scale, magnetic screening could be extended to larger objects. From materials point of view, it would even be possible to magnetically protect the whole planet by a single superconducting pipeline. For Earth, for example, necessary amount of boron for MgB$_2$ pipeline could be mined just in 5 years. Other global projects like generating energy from Solar wind or extracting it from below the crust in magnetically active volcanic regions would also be possible.

5. Conclusions

The disappearance of fossil fuels, global warming and transition to renewable energy resources lead to synergy of superconductivity and hydrogen economy. The superconductivity offers compactness, efficiency, energy savings and wide range of applications that are not possible without using this macroscopic-scale quantum phenomenon. It gives hope to cope with threatening energy crisis and may addresses global problems on the planetary scale.
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