Metal hydride hydrogen compressors for energy storage systems: layout features and results of long-term tests

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Abstract
This work summarises the results of development and long-term testing of two prototype models of industrial-scale metal-hydride thermal sorption hydrogen compressors, TSC1-3.5/150 (up to 11 Nm\textsuperscript{3} h\textsuperscript{−1}; water cooling/steam heating) and TSC2-3.5/150 (up to 15 Nm\textsuperscript{3} h\textsuperscript{−1}; heating and cooling by circulating oil). Both compressors have a two-stage layout utilising LaNi\textsubscript{5} for the stage 1 and La\textsubscript{0.5}Ce\textsubscript{0.5}Ni\textsubscript{5} for stage 2, thus providing hydrogen compression from 0.35 to 15 MPa at a cooling temperature of about 20 °C and a heating temperature between 150 °C and 160 °C. The compressors were operating for one year each and exhibited a satisfactory performance. The results of this work can be used in various medium-to-large scale hydrogen compression applications, including renewable energy systems.

1. Introduction

Energy systems on the basis of renewable energy sources (RES), such as solar and wind, do not provide an uninterrupted power supply because of non-uniform and irregular energy generation. To solve this problem, it is favourable to develop hybrid energy complexes comprising of hydrogen energy storage systems. In such complexes, the surplus energy supplied by the RES is used for hydrogen generation by water electrolysis followed by hydrogen compression and storage. When the primary energy is in deficit, electricity needed for an end-user is generated by hydrogen fuel cells [1, 2].

Hydrogen energy storage systems are also important for the infrastructure supporting hydrogen fuelled vehicles, particularly at refuelling stations which require the compression and storage of hydrogen at pressures of several hundred atmospheres [3, 4]. Backup power systems require efficient hydrogen storage and compression solutions as well [5, 6].

Hydrogen compression is one of the key components of the hydrogen energy storage method, to densify hydrogen gas and, as a result, to increase energy storage density. Conventional methods used in industry, mechanical hydrogen compressors (piston, diaphragm, etc) have a number of disadvantages including a complicated design, insufficient reliability, high operating costs, a probability of hydrogen leakage and hydrogen contamination, noise and vibration during operation [3, 7].

An alternative to mechanical hydrogen compressors is a metal hydride thermal sorption compressor (MH TSC) whose operation is based on the reversible interaction of metals of alloys with hydrogen gas. MH TSC’s have a number of advantages including practically unlimited (up to 500 MPa) discharge pressure, good scalability (from several litres to several normal cubic metres of hydrogen per an hour), modular design, simplicity in service and operation, as well as the possibility to use low-grade heat and high purity of the delivered...
hydrogen. In addition, the MH TSC does not contain moving parts that simplifies its design, increases reliability and eliminates noise and vibration [6, 8, 9].

Presently, there exist several practical applications of MH TSC’s including the handling of hydrogen isotopes in the nuclear industry [8, 10], cryo-coolers in space engineering [8, 11], thermally driven actuators [8, 12], and laboratory practice [6, 9]. These compressors used in quite narrow, niche areas have low productivity and efficiency.

It is promising to integrate MH TSC’s in the infrastructure of hydrogen production which includes filling of composite (up to 700 atm) and steel (up to 150–200 atm) gas cylinders with pure hydrogen, at the productivity up to 20 Nm³ h⁻¹. Though some recent publications [13–15] present information about development of industrial-scale metal hydride hydrogen compressors (discharge pressure up to 200 atm at 5–13 Nm³ h⁻¹ productivity), the published information is still scarce.

For the development and industrial operation of the MH TSC’s, the solutions of a number of research and engineering problems are necessary including: (i) selection of existent or development of new metal hydride materials, (ii) design optimisation taking into account providing of effective heat-and-mass-exchange, (iii) optimisation of the operating parameters to provide the required productivity, together with long service lifetime and (iv) integration of the compressor in energy storage systems and its operation.

This work summarises the results of joint development, by SKTBE and IPCP, and long-term operation of two models of two-stage metal hydride hydrogen compressors, TSC1-3.5/150 and TSC2-3.5/150. Further details have been published in [16–22].

2. Results and discussion

2.1. Metal hydride materials

Analysis of the literature data and results of our preliminary experiments showed that for hydrogen compression to 150 atm over temperature range 15 °C–150 °C two-stage compression is suitable using LaNi₅ as the first- and La₀.₅Ce₀.₅Ni₅ as the second-stage MH material. These alloys forming only one hydride phase, can be easily activated, have high rates of reversible hydrogen absorption/desorption, narrow hysteresis, satisfactory cycle stability, and are tolerant to impurities in hydrogen produced by electrolysis. As it can be seen in figure 1, the cycle productivity of two-stage H₂ compression utilising these AB₅-type alloys (~80 NL H₂/kg) is limited by hydrogen transfer from stage 1 to stage 2 at PM ≈ 35 atm. This drawback was, however, mitigated by the increase of the amount of the MH material on stage 1 of the hydrogen compressor by 20%–25% in the weight as compared to stage 2 [19, 20, 22].

2.2. Metal hydride bed and hydrogen compression module

Despite of quite good hydrogenation/dehydrogenation kinetics, the powdered beds of the selected MH materials have a low effective thermal conductivity (ETC) originated from micron-range particle size. This limits the rates of hydrogen absorption/desorption. Thus heat-and-mass transfer in the MH bed significantly influencing on its H₂ charge/discharge performance should be optimised, taking into account design features
Modelling of heat transfer performance of MH beds on the basis of the AB5-type alloys showed that externally heated and cooled cylindrical beds with a high length-to-diameter ratio is an optimal solution due to (i) high heat exchange area between the MH and the heating/cooling fluid, (ii) simplicity and lower cost and (iii) less pronounced reduction of hydrogen storage capacity at the same dimensions as compared to the solutions with internal heat exchangers [21]. According to the modelling results verified by experiments [22], the externally heated and cooled cylindrical stainless steel containers, about 20 mm in the internal diameter and about 800 mm in the length, filled by the selected AB5-type materials, are characterised by satisfactory H2 absorption/desorption (H2 charge/discharge times shorter than 20 min), if averaged heat transfer coefficient between the MH container walls and the heating/cooling fluid is about 500 W m\(^{-2}\) K and higher. In this case, hydrogen absorption/desorption dynamics is almost completely determined by the heat transfer in the MH bed (effective thermal conductivity about 1.5 W m\(^{-1}\) K without heat transfer augmentation). Further increase of the effective thermal conductivity (up to 12 W m\(^{-1}\) K) was achieved by a placing the MH powder into porous high thermally conductive matrix (copper foam) [22]. The latter solution was used in the TSC2-3.5/150 model. The optimal two-stage hydrogen compression (3.5–150 atm) performance of such layout of the MH container was achieved at the duration of the heating (140 °C–160 °C) and cooling (20 °C–40 °C) between 10 and 24 min (full compression cycle duration 20–48 min) [19, 22].

A module for H2 compression (see figures 2 and 3; left) comprises a set of the tubular MH containers (1) as described above connected to one or two gas manifolds (2 in figure 2, left) whose common ends are connected to hydrogen input/output pipelines (4) equipped with replaceable in-line gas filters (3 in figure 3, left). The MH containers are placed in the cylindrical case of heat exchanger (3 in figures 2, 2 in figure 3) thermally isolated from the environment and equipped with pipelines for the input (5) and output (6) of the heating/cooling fluid.

(geometry, dimensions, materials) and operating conditions (H2 input pressure, temperature and flow rate of the heating/cooling fluid, etc).

Figure 2. Drawing of metal hydride module (left) and general view (right) of TSC1-3.5/150 compressor. 1—MH container, 2—gas manifold, 3—case of heat exchanger, 4—H₂ input/output, 5, 6—input and output of the heating/cooling fluid.

Figure 3. Drawing of metal hydride module (left) and general view (right) of TSC2-3.5/150 compressor. 1—MH container, 2—case of heat exchanger, 3—replaceable gas filter, 4—H₂ input/output, 5, 6—input and output of the heating/cooling fluid.
2.3. Compressor assemblies

Two prototype models of two-stage metal hydride thermal sorption compressors, TSC1-3.5/150 and TSC2-3.5/150, utilising MH materials and containers described in the previous section have been manufactured and tested. The drawings of compression modules (identical for stages 1 and 2) used in the compressors and images of the developed models are presented in figures 2 and 3. Table 1 presents overview of characteristics of the developed prototypes.

Both models use the same AB5-type materials loaded in similar MH containers and differ by the inner layout of the MH containers (presence of copper foam for TSC2-3.5/150), layout, size and arrangement of the H2 compression modules, as well as the kind of heating and cooling fluids.

The TSC1-3.5/150 compressor comprises of three sections each of which includes two modules for stage 1 (16 kg LaNi5 per a module) and two modules for stage 2 (12 kg La0.5Ce0.5Ni5 per a module). The compressor uses steam (150 °C/5 atm) for the heating and water (15 °C) for the cooling.

Figure 4 presents a piping diagram of one section of the TSC1-3.5/150 compressor. For simplicity, each of the pairs of compression modules for Stages 1 and 2 are shown as one module (MHM1.1, MHM1.2). The gas piping includes four lines:

- Low pressure hydrogen line comprising hydrogen input pipeline (H2 in), auxiliary N2 purge pipeline, manual shut-off valves (V1, V2), pressure sensor (M1) and buffer gas cylinder pack (BG1) with manual shut-off valve (V5);
- STAGE I line includes metal hydride module of the first stage (MHM1.1) with gas filter (F1) and manual shut-off valve (V1.1), as well as pressure sensor (M1.1);
- STAGE II line includes metal hydride module of the second stage (MHM1.2) with gas filter (F2) and manual shut-off valve (V1.2), as well as pressure sensor (M1.2);
- High pressure hydrogen line includes output gas filter (F3), pressure sensor (M3), hydrogen output pipeline (H2 out) as well as manual shut-off valve (V4) and safety relief valve (RV5) connecting high-pressure gas

Table 1. Characteristics of the developed MH hydrogen compressors.

| Characteristic [units]                      | TSC1-3.5/150 | TSC2-3.5/150 |
|--------------------------------------------|--------------|--------------|
| Suction pressure [MPa]                     | 0.35         | 0.35         |
| Discharge pressure [MPa]                   | 15.0         | 15.0         |
| Number of stages/compression modules in a section | 2            | 2            |
| MAX productivity [Nm³ h⁻¹]                 | 11           | 15           |
| Number of sections                         | 3            | 4            |
| Duration of the heating/cooling cycle [min]| 20           | 28–48        |
| Heat transfer augmentation                  | —            | Copper foam  |
| Heating T [°C]                             | 140–150      | 150          |
| Fluid                                       | Steam        | Oil          |
| Cooling T [°C]                             | 15–20        | 15           |
| Fluid                                       | Water        | Oil          |
| Stage 1 modulesa                           | LaNi₅        | LaNi₅        |
| Total amount [kg]                          | 16           | 41           |
| Number of MH containers                    | 16           | 41           |
| Stage 2 modulesa                           | La₀.₅Ce₀.₅Ni₅ | La₀.₅Ce₀.₅Ni₅ |
| Total amount [kg]                          | 12           | 32           |
| Number of MH containers                    | 16           | 41           |

a for TSC1-3.5/150, each section includes two modules connected in parallel.
manifold with venting pipeline (Release) which is also connected to the STAGE I line via auxiliary solenoid valve (PV1) for the purging of the first stage when necessary.

The operation is provided by the periodic heating and cooling of compression modules controlled by opening/closing solenoid valves PV1.1–PV1.5 (module MHM1.1) and PV1.6–PV1.10 (module MHM1.2) thus providing alternate heating and cooling of the compression modules while hydrogen flows are switched automatically with the help of check valve arrangement CV1.1–CV1.3. The beginning of the heating/cooling cycle (20 min in total), for different sections 1–3, is shifted by 1/3 of the cycle duration as shown in [20], to provide the required cycle productivity during permanent operation.

The gas piping of the TSC2-3.5/150 compressor (figure 5) is similar to the one for the TSC1-3.5/150 model described above. In the TSC2-3.5/150 model, the alternate heating and cooling of the compression modules is provided by oil (Marlotherm N; Sasol). The heating/cooling system includes cooling and heating circuits shown in figure 5 in blue and red, respectively; the parts of the circuits periodically connected to the heating/cooling jackets of the compression modules are shown in figure 5 in brown. The cooling circuit includes a heat exchanger (HE1) also connected to CHILLER, cold oil tank (OT1) with manometer (M4), main (P1) and
auxiliary (P3) pumps. The heating circuit comprises of a hot oil tank (OT2) with manometer (M5), induction heater (IH1), main (P2) and auxiliary (P4) pumps. The control of oil flows is provided by pneumatically actuated valves: PV2–PV4 for the cold oil and PV5–PV7 for the hot one. The compressor assembly includes four sections as shown in figure 5 when the beginning of the heating/cooling cycle (38 min in total) for different sections is shifted by 1/4 of the cycle duration (9.5 min).

2.4. Test results

Figure 6 shows the changes of temperatures of metal hydride modules for the first and the second stages in one section of the TSC1-3.5/150 compressor during its operation. The modules are periodically heated and cooled in an opposite phase; the duration of both heating and cooling was set to 10 min. Low-pressure hydrogen (0.35 MPa) is absorbed with LaNi5 in the cooled module of the first stage followed by its desorption at 3–4 MPa when the module is heated up. The medium-pressure H2 is then absorbed with La0.5Ce0.5Ni5 in the cooled module of the second stage in another section (second part of the curve for MHM1.2 in figure 6) followed by its desorption at P = 15 MPa when the module is heated up (beginning of the curve for MHM1.2 in figure 6). The presence of three sections provides continuous H2 compression from 0.35 to 15 MPa at productivity up to 11 Nm3 h–1.

The operation of the TSC2-3.5/150 compressor is illustrated by figures 7, 8. The cyclic heating and cooling of the MH modules was provided by the periodic switching of hot and cold oil flows, using pneumatically actuated valves PV2–PV7 (figure 5); the duration of both heating and cooling varied between 14 and 24 min (20 min in figure 7), so the complete cycle duration was from 28 to 48 min.

Figure 8(A) illustrates the operation of one section of the TSC2-3.5/150 compressor (figure 5) during filling a receiver (cylinder pack, total water volume 480 l) with hydrogen (starting pressure 4 MPa, final pressure 15 MPa).

During the cooling of the first stage module (MHM1.1), low-pressure hydrogen (in; P = 0.35 MPa) is absorbed therein while the simultaneous heating of the second stage module (MHM1.2) results in the desorption of hydrogen into a high pressure discharge line (out; gradual increase of pressure in the receiver from 4 to 15 MPa). Further heating of the module MHM1.1 results in H2 desorption there from at P ~ 3.5 MPa to be absorbed in the simultaneously cooled module MHM1.2. In doing so, the increase of pressure in the discharge line (out) and absorption of the low-pressure H2 (in) are provided by the heating of the second-stage modules and the cooling of the first-stage modules, respectively, in the other sections of the compressor.

As can be seen from figure 8(B), the increase of cycle time from 28 to 48 min results in a slight increase of the compressor’s productivity, particularly, at high output H2 pressure. Further increase of the cycle time results in a gradual drop of the productivity. The cycle time of 38 min was selected as a setpoint for further operations.

Long-term tests of both compressors were carried out. The total operation time of the compressors within one year was about 6000 h that corresponds to 18 100 hydrogen absorption/desorption cycles for each module in the TSC1-3.5/150 compressor and 10 000 cycles for each module in the TSC2-3.5/150 compressor. The test

Figure 6. Changes of temperatures of metal hydride modules of the first (MHM1.1) and the second (MHM1.2) stages in one section of the TSC1-3.5/150 compressor during its operation.
results are summarised in figure 9 as dependencies of the average compressor’s productivity (in % of the starting value) on the operation time (A) and number of hydrogen absorption/desorption cycles (B).

The main problem identified during the tests of the TSC1-3.5/150 compressor was in the decrease of average compressor’s productivity, from 10–11 to 7–8 Nm³ h⁻¹, or down to <80% of the initial productivity (figures 9(A), (B); curve 1). Comparative analysis of MH materials sampled from the MH containers of the first and the second stage after the 10th and 18th H₂ absorption/desorption cycle showed decrease of reversible hydrogen storage capacity for the 1st stage alloy (LaNi₅) by 23% mainly caused by the disproportionation of the intermetallic in H₂ to yield LaH₃–ₓ and Ni. At the same time, the 2nd stage alloy (La₀.₅Ce₀.₅Ni₅) showed only minor changes in its hydrogen absorption/desorption performance and did not exhibit disproportionation, due to higher thermodynamic stability of Ce-substituted intermetallic. More details including detailed characterisation of the samples are presented in [20].

Comparison of the decrease of the productivities of the compressors TSC1-3.5/150 (figure 9; curve 1) and TSC2-3.5/150 (figure 9; curve 2) during their operation showed that the performance degradation depending on the number of cycles was similar for both prototypes (figure 9(B); deviations below 5%). At the same time, the rate of productivity drop after ~1000 h of the operation was significantly higher for TSC1-3.5/150 than for TSC2-3.5/150 (figure 9(A)). As a result, at the end of the one-year operation the productivity of the former (cycle time 20 min) was only ~80% of the initial one, against ~95% for the latter (cycle time 38 min). This allows us to
conclude that performance degradation of the MH compressors is mainly determined by the number of hydrogen absorption/desorption cycles rather than the time of exposing the MH materials at high temperatures and H₂ pressures (same for both compressors). Accordingly, to prolong service life of the MH TSC’s, it is recommended to set an acceptably long cycle time.

Apart from the thermodynamic origin of the degradation (also called as ‘hydogenolysis’) of RTₓₙ intermetallic compounds to yield binary hydride of the rare earth metal RH₃ₓ₋ₓ and individual transition metal T, or RTₓₙ₊ₘ(Hₓ) intermetallic (hydride) [20, 23, 24], the degradation process requires movement of the metal atoms from their initial positions in the metallic matrix to form the new phases. At low temperatures the degradation is frequently impeded kinetically, and exposing the intermetallic under high H₂ pressures and temperatures (‘thermal aging’) is necessary to observe this process in a reasonable time [23, 24]. At the same time, there are several known systems (e.g. H–CeCo₂) where the degradation of the intermetallic matrix was observed even at near-ambient temperatures and low (down to 1 atm) partial H₂ pressures; in most cases amorphous products were formed at the beginning of this process [23]. Taking this into account, we assume that main contribution in the observed effect is made by the movement of metal atoms accompanying ‘intermetallic ↔ hydride’ phase transitions during H₂ absorption/desorption cycling rather than the thermal movement/oscillations of the atoms at the higher temperatures.

The TSC2-3.5/150 compressor was integrated in a pilot hydrogen production line developed at SKTBE. The line is intended for the production of up to 15 Nm³ h⁻¹ of hydrogen (purity of 99.9999%; dew point –60 °C) supplied to metal hydride hydrogen storage units at P = 0.4 MPa and to hydrogen cylinders at P = 15 MPa generated by the compressor.

The line (figure 10) comprises of the following main components:

- Hydrogen generator on the basis of 20 Nm³ h⁻¹ alkaline electrolyser (EL), 0.4 MPa output pressure;
- Low pressure buffer cylinder (BC);
Compressor, the total amount of the compressed H2 was of 40 000 Nm3. In doing so, the compressor worked for 10 000 H2 absorption/desorption cycles at average productivity between 12 and 13 Nm3 h⁻¹.

3. Conclusions

This work showed the feasibility of industrial-scale applications (up to 15 Nm³ h⁻¹ in productivity) of metal hydride technology for hydrogen compression from 0.35 to 15 MPa in the operation temperature range from 15 °C–20 °C to 150 °C–160 °C, by the use of La₅Ce₁₋ₓNiₓ intermetallics as hydrogen compression alloys.

Two models of the MH compressors, TSC1-3.5/150 and TSC2-3.5/150, have been developed within this work. The compressors have two-stage layouts, using LaNi₁₀ (0.35–3.5 MPa) as a first-stage material and La₅.₅Ce₃.₃Ni₅ (3.5–15 MPa) as a second-stage one. Each compressor was in operation for one year and exhibited a satisfactory performance. The TSC2-3.5/150 compressor was successfully integrated in a pilot hydrogen production line developed by SKTBE, for the filling of standard gas cylinders with ultra-pure hydrogen.

Comparative analysis of long-term performance tests of the TSC1-3.5/150 and TSC2-3.5/150 compressors operated at the same conditions but with different cycle times has shown that the drop in productivity associated with the degradation of the hydride forming materials is mainly contributed by the number of hydrogen absorption/desorption cycles rather than the time of exposing the MH materials at high temperatures and H₂ pressures. Accordingly, it is recommended to set longer cycle times to slow down the drop of compressor productivity during the operation.

Results of this work can be used in various medium-to-large scale applications including renewable energy systems utilizing hydrogen for the energy storage.

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