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## A NOVEL HYDROGEN COMPRESSOR

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### INTRODUCTION

The absorption of hydrogen by intermetallic compounds has, during the past few years, been receiving increasing attention. For instance, at Philips Research Laboratory in Eindhoven, The Netherlands, Lanthanum Pentanickel ( $\text{LaNi}_5$ ) has been studied since 1969 (reference [1]). These compounds offer the potential to separate, compress and store hydrogen, and to operate between room and boiling water temperature. Philips Laboratories built a hydrogen compressor using  $\text{LaNi}_5$  as the working medium. The compressor uses no pistons or motors (references [2], [3]) and can be powered by low grade heat.

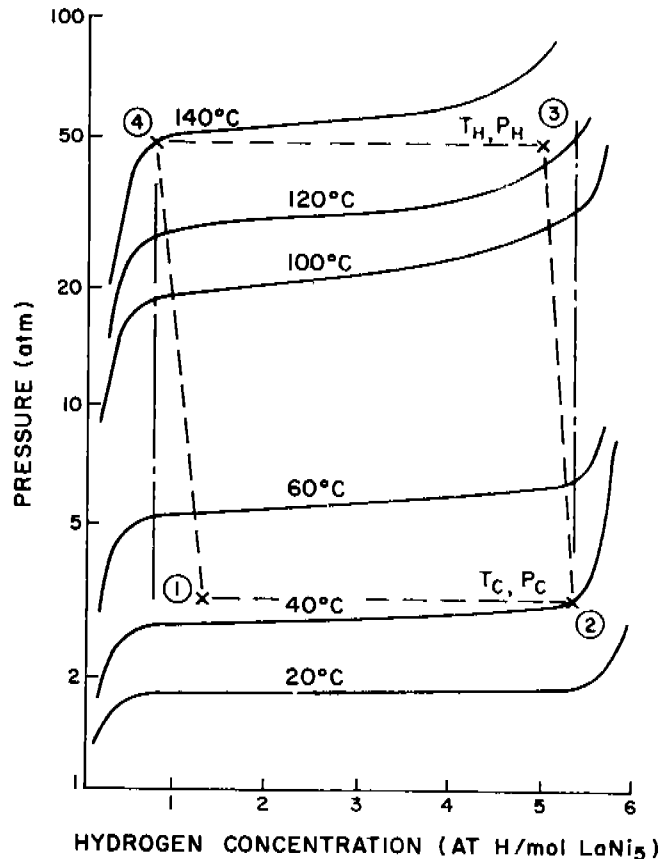
### THE HYDROGEN COMPRESSOR

The compression of hydrogen gas in the subject compressor is based on the unique properties of  $\text{LaNi}_5$ . The phase diagram of  $\text{LaNi}_5$  and H is shown in Figure 1. The most significant features are:

- An extended two phase region characterized by an almost constant equilibrium pressure (plateau pressure) at 2.5 atm at room temperature.
- A rapid increase of the plateau pressure with rising temperature. For example, a 100°C temperature increase over room temperature yields a ten-fold pressure increase.

In practice, a continuously operating compressor is created by combining at least three separate compressor modules and sequentially cycling them between a high and a low temperature. The phasing of the three modules is shown in Figure 2. Note that a constant delivery is assured provided that the time for heat-up or cool-down is one-half that for sorption.

A basic concern in designing the module that contains the  $\text{LaNi}_5$  is effective heat transfer. This is important since during absorption the formation energy of the hydride ( $7.2 \frac{\text{kcal}}{\text{mole H}_2}$ ) has to be removed, and conversely, the formation energy has to be supplied during desorption of hydrogen. The size of the  $\text{LaNi}_5$  bed required in any application decreases with faster cycling times which is a se-



#### PROCESSES:

- ① → ② ABSORPTION - HEAT REMOVAL
- ② → ③ HEAT ADDITION - NO GAS FLOW
- ③ → ④ DESORPTION - HEAT ADDITION
- ④ → ① HEAT REMOVAL - NO GAS FLOW

Figure 1.  $\text{LaNi}_5$  - A Phase Diagram and Compression Cycle.

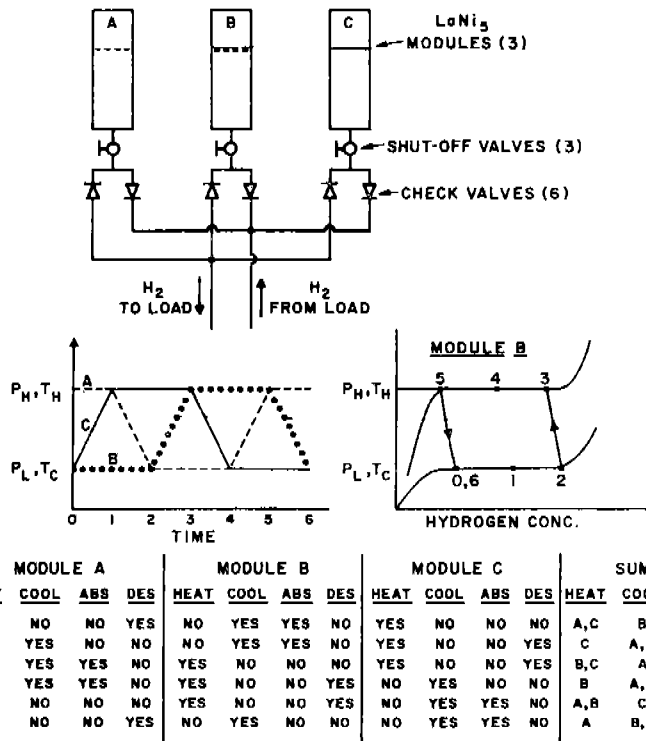


Figure 2. Phasing Three Modules to Form a Sorption Compressor.

rious consideration for non-stationary applications. These cycle times are limited by transient heat transfer considerations. The design of a module with efficient heat transfer is made difficult by the fact that the  $\text{LaNi}_5$  decays to a powder of less than  $5 \mu\text{m}$  particle size after repeated cycling. This activated powder cannot be exposed to air because it is pyrophoric and would therefore degrade. Layers of powder beds have a very low overall thermal conductivity. In designing a module with good heat transfer, one should use beds with a layer thickness of about 1 mm. A compromise between compactness and good heat transfer was found in the triple thickness design as shown in Figure 3 and Table I. The length of the module is plotted versus the average diameter at constant volume, with sorption time and bed thickness as parameters. The electric heating makes it convenient to vary the input power. To fill the above described module, a second chamber with a transparent window was charged with the required amount of  $\text{LaNi}_5$  in large chunks and attached to the module. In this second chamber the  $\text{LaNi}_5$  was activated until a fine powder was obtained (after 20 cycles). This powder was then transferred into the module by shaking. Finally, the connecting tube was pinched off.

Tests were performed on the prototype  $\text{LaNi}_5$  module, Figure 4, to determine its performance characteristics. Specifically, the heat-up and desorption input heat requirements were investigated. During operation, the sorption processes of an actual three module compressor would occur at constant pressure. To accomplish this with a single  $\text{LaNi}_5$  module the test setup shown in Figure 5 was devised. A large hydrogen reservoir was charged to a pressure between

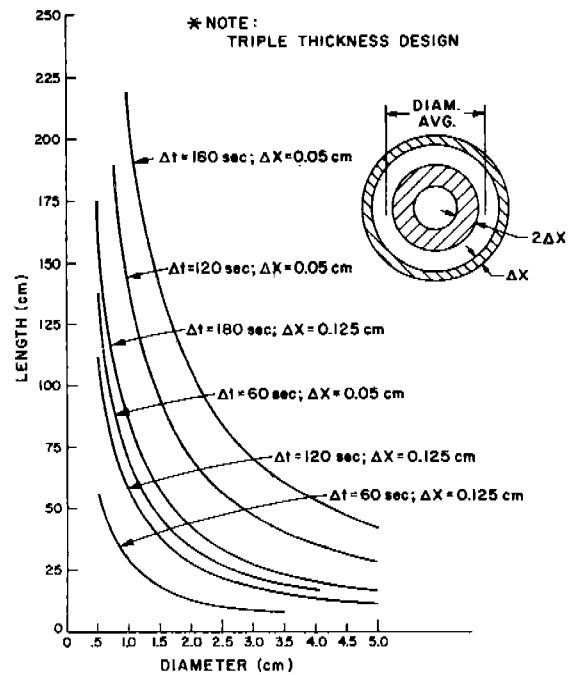


Figure 3.  $\text{LaNi}_5$  Module Length vs. Diameter, Sorption Time, and Bed Thickness (Triple Thickness Design).

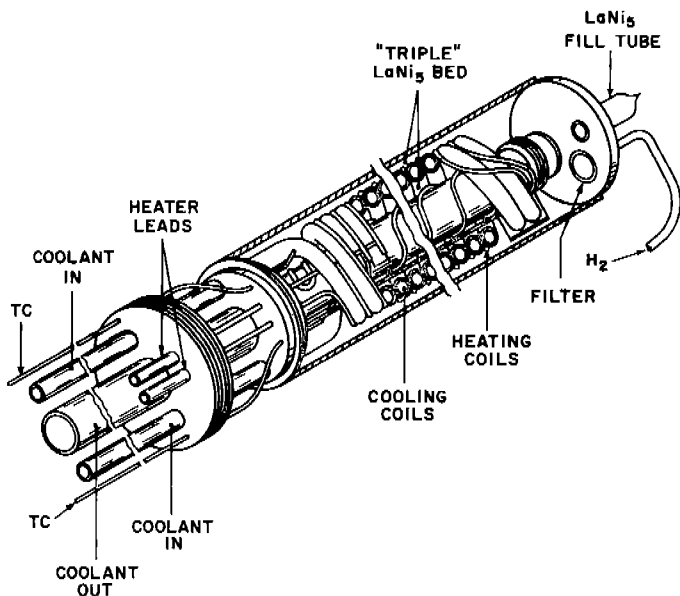


Figure 4. Compressor Module Design.

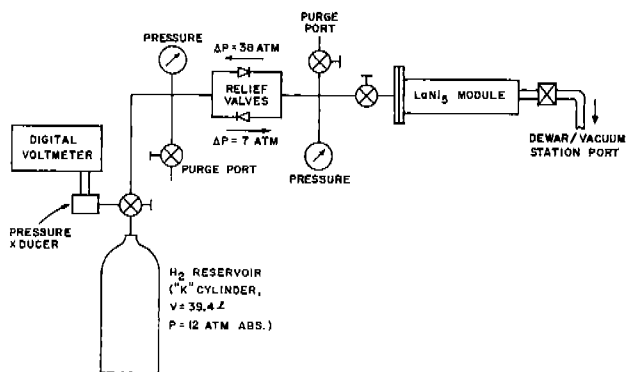


Figure 5. Schematic of H<sub>2</sub> System

the desired absorption and desorption levels. The reservoir was sized to be sufficiently large so that its pressure variation during sorption would be acceptably small. Figure 6 shows the heating and cooling system used for the tests.

Each test cycle was conducted as follows:

(a) Starting at state point (2) (concentration 5.35, temperature 20°C, pressure 5 atm), a fixed input heat rate was applied to the module and the time to reach state point (3) (concentration 5.0, temperature 105°C, pressure 50 atm) was recorded. At point (3) the automatic desorption relief valve shown in Figure 5 opened, and desorption began.

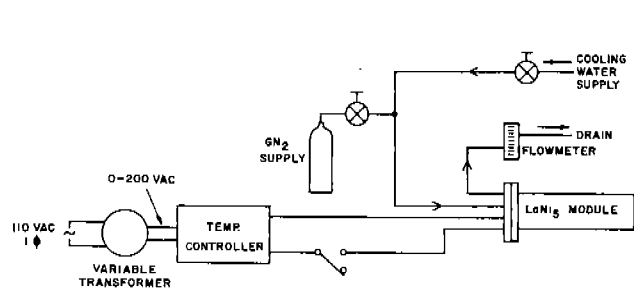


Figure 6. Schematic of Heating/Cooling System

(b) While still maintaining the input heat rate constant, the reservoir pressure versus time data was recorded for later conversion to hydrogen flow rate until state point (4) (concentration 1.0, temperature 185°C, pressure 50 atm) was reached.

(c) At state point (4) the heater power was switched off, and a coolant flow rate of approximately 0.03 l/s was initiated (all test runs were accomplished with the same coolant flow rate). The time to reach state point (1) (concentration 1.35, temperature 40°C, pressure 5 atm) was recorded.

(d) At state point (1), the automatic relief valve opened and the reservoir pressure versus time was recorded for subsequent reduction to hydrogen absorption flow rates.

These test cycles were performed with input powers ranging from 500 to 900 W. The average heat input requirement,  $Q$ , during heat-up was found to be 60,000 Ws. During desorption the average heat input,  $Q_s$ , was found to be 85,000 Ws. Since the quantity of input heat required for process 2 → 3 ( $Q$ ) and 3 → 4 ( $Q_s$ ) has been determined, this information can be used to relate the way in which the input heat rate would have to vary with the time available to complete the process.

- Process 2 → 3 Heat-up

From a measured energy input of  $Q = 60,000$  Ws, one obtains with a heat-up time of 75 s, 800 W input power.

- Process 3 → 4 Desorption

From a measured energy input of  $Q_s = 85,200$  Ws, one obtains with a desorption time of 150 s, 570 W input power.

In an actual compressor cycling automatically, an electric control circuit could be used to limit the maximum module input power to 800 W during heat-up. During sorption, the average input power

could automatically be limited to 570 W by using a heater control circuit which senses module pressure.

### DISCUSSION

The construction and testing of the compressor module, as described above, is only a first step in the development of a commercial hydrogen compressor. Certain problem areas still remain, such as the relatively low ratio of active to inactive material and the involved filling procedure. The latter can be overcome by casting or pressing--from powder metallurgical materials--LaNi<sub>5</sub> in thin cylinders. These cylinders are inserted into the proper beds and activated in situ. The low ratio of active to inactive material can be improved by a new, light-weight design.

Another problem area is the cycle life of the LaNi<sub>5</sub> compound. No systematic investigation of this problem is known to the authors. However, Brookhaven National Laboratories has determined that FeTi shows no deterioration in the hydrogen absorption after 10,000 cycles in a closed cycle high purity environment. Furthermore, the exposure of activated LaNi<sub>5</sub> powder to air produced a temporary deterioration of the hydrogen absorption kinetics, which disappeared after a number of cycles [4]. It has also been shown that CO concentrations of about 3% spoil the hydrogen absorption very rapidly. It will therefore be necessary to investigate the life time problem for every specific application.

It is obvious that faster cycling reduces the amount of LaNi<sub>5</sub> necessary to pump a given amount of hydrogen. Such considerations are very important for all automotive applications. Therefore, the rate limiting process under isothermal conditions has been measured for LaNi<sub>5</sub>. A minimum absorption time of approximately 10 s was found at room temperature. The phase transition between metal and hydride was determined as the rate limiting process. (Reference [5]). In practice, heat transfer conditions might limit the absorption time even further.

The pressure range of the compressor can be varied over a wide range by the use of different hydrogen absorbing materials. One example is shown in Fig-

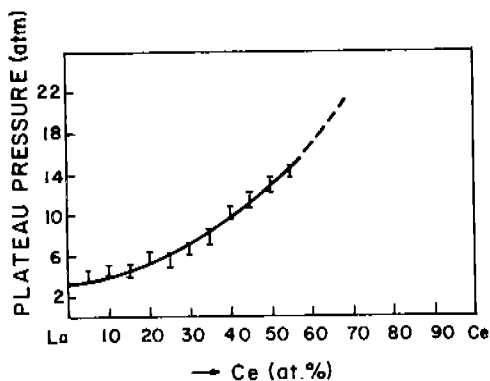


Figure 7. Plateau Pressure at Room Temperature as a Function of  $\chi$  In the  $La_{1-\chi}Ce_{\chi}Ni_5$  System.

ure 7, where the equilibrium pressure at room temperature is shown as a function of Ce concentration (reference [1]). Replacing La by Ce increases the room temperature equilibrium pressure five-fold from 3 atm to 15 atm. Correspondingly, the pressure at 81°C increases from 20 atm to 100 atm. Other compounds exist that allow the achievement of even higher pressures.

Most important, in addition to its long life potential, the hydrogen compressor can be driven with waste heat at less than 100°C and produce a tenfold pressure increase. The electric heating supply in the test module was chosen for the convenience of regulating the power input. The use of waste heat should make this compressor ideally suited in many industrial situations where high pressure hydrogen is needed and waste heat is available.

Table 1. Summary Of Triple Bed Design

Bed	Dimensions	LaNi <sub>5</sub> %	Module Mass %	Heater Power %	Cooling Surface %
*3 Concentric Beds; $\Delta X = 0.053"$					
Inner	.394"ID x .500"OD	23	15	15	15
Middle	.500"ID x .606"OD	29	85	85	85
Outer	.889"ID x .995"OD	48			
*D effective = 0.65"					
*L effective = 17.0"					
*M <sub>LaNi<sub>5</sub></sub> = 270 g (total), 256 g (effective)					
*Filling Factor: 42% (absorbed state)					
*Bed Density: 2.84 g/cc					
*Prototype Weight: 1400 (excluding dewar)					

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