Optimization of Heat Transfer in Metal Hydride Reactor

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Heat management is the design bottleneck in creation of efficient metal hydride systems. Inefficient heat transfer in metal hydride beds is connected with relatively low effective thermal conductivity (0.1-1 W/m K) and high reaction heat (25-70 kJ/mole H_2) of activated hydrogen storage materials, which are the fine powders with particle sizes down to 1-10 micrometers. As the result even for water cooled metal hydride reactors heat transfer coefficient from hydride bed to coolant is only about 100 - 120 W/m^{2}K (Borzenko, 2008; Kaplan, 2009) and determined mostly by poor heat transfer in the metal hydride bed.

The goal of current investigation is to find relationship between operational parameters of metal hydride accumulator (hydrogen capacity V_H, inlet/outlet pressure P_{in}, P_{out} and hydrogen flow rate g), reactor design (heat transfer area A) and properties of hydrogen storage material (*PCT*-diagram, heat of reaction ΔH, thermal conductivity k, heat transfer coefficient α).

Heat and mass transfer was experimentally investigated at sorption of pure hydrogen and desorption in metal hydride reactor filled with m_{MH} =4.69 kg of $Mm_{0.8}La_{0.2}Ni_{4.1}Fe_{0.8}Al_{0.1}$ alloy, ΔH_{in} = 13,1 MJ/kg H_2, ΔH_{out} = 13,5 MJ/kg H_2. The reactor consists of O-ring shaped capsules filled with MH bed, the capsules are assembled in cylindrical casing with water heat exchanger at outer wall (see Figure 1).
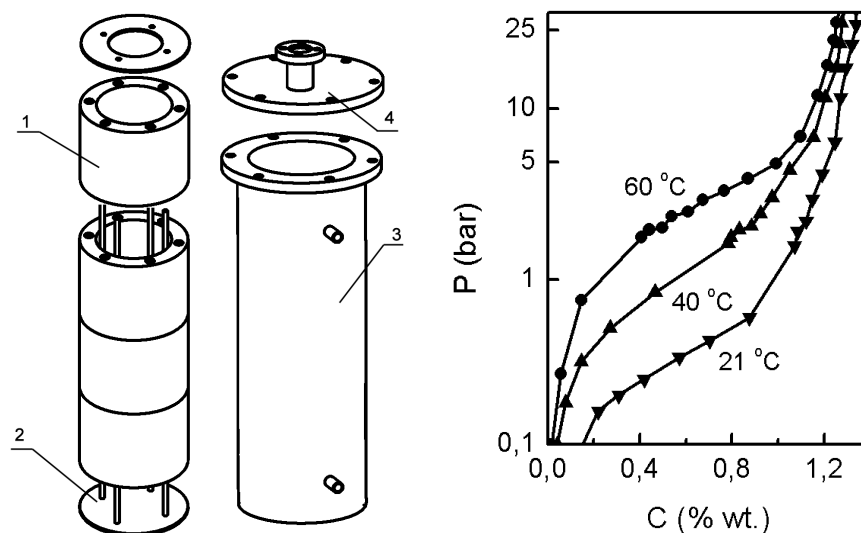

Figure 1: Reactor design (left) and desorption isotherms of alloy (right):
1 – capsule with storage material, 2 – hydride cartridge,
3 – casing with water heat exchanger, 4 – cover.

During charging and discharging of the reactor initial hydrogen flow at inlet and outlet was limited by flow regulator at different values between g_{max} = 24 (10% of maximal flow) and g_{max} = 240 st. l/min (100%) in order to measure parameters of heat and mass transfer for different operational regimes, hydrogen pressure in supply system was set to P_{in} = 9 bar for charging and discharged hydrogen was thrown into the atmosphere P_{out} = 1 bar. Hydrogen capacity of the reactor is found to be V_H = 690±5 st. l (1.33 %wt.) and charging time was t_H =18±1 min for g_{max} > 60 st. l/min.

It is found that the reactor for most of the regimes of charging and discharging cannot maintain the initial hydrogen flow rate g_{max}, and with increasing of g_{max} amount of hydrogen charged at initial flow rate decreasing significantly (Figure 2). Results for discharging are similar.
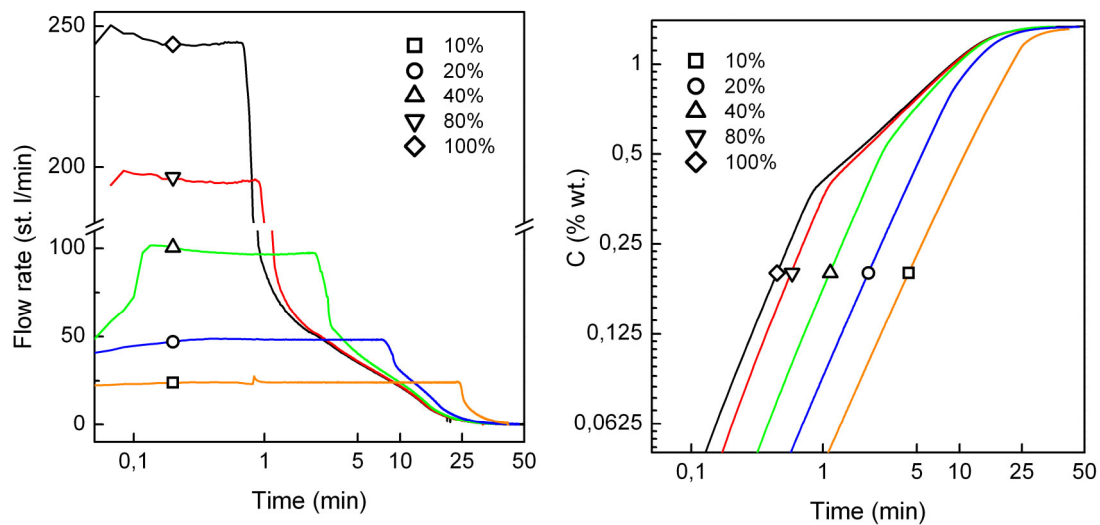

Figure 2:   **Hydrogen flow rate at inlet (left) and hydrogen content in MH bed (right) for different charging regimes.**

We can divide the process of charging and discharging of MH reactor into three stages (Figure 3). Transition from the first to the second stage is connected with dramatic decrease of hydrogen flow rate at reactor inlet – and we can call this crisis of heat and mass transfer in MH reactor.
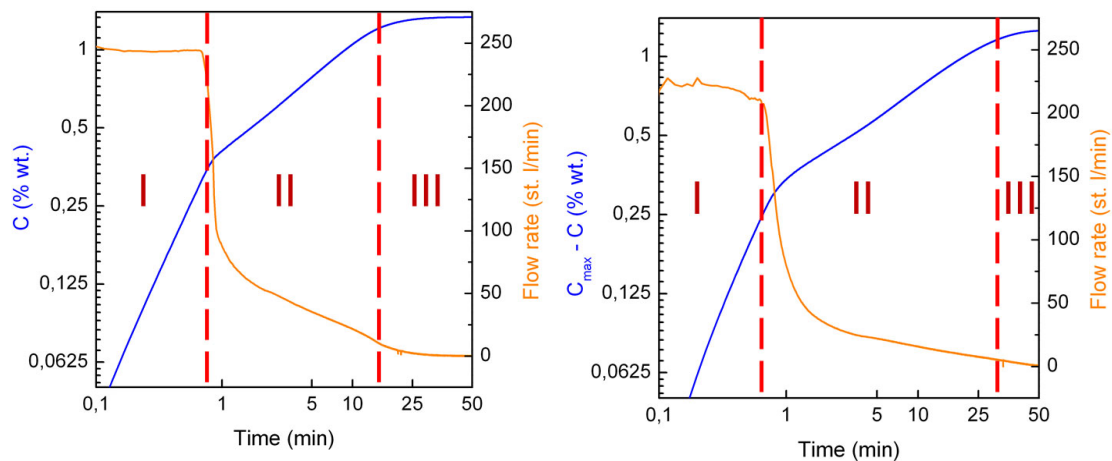
Figure 3:   **Hydrogen flow rate and hydrogen content in hydride bed for charging at 100% (left) and discharging at 90% (right) and three stages of the process.**

These three stages can be explained from the analysis of temperature and pressure evolution inside the metal hydride bed during the process (Figure 4).
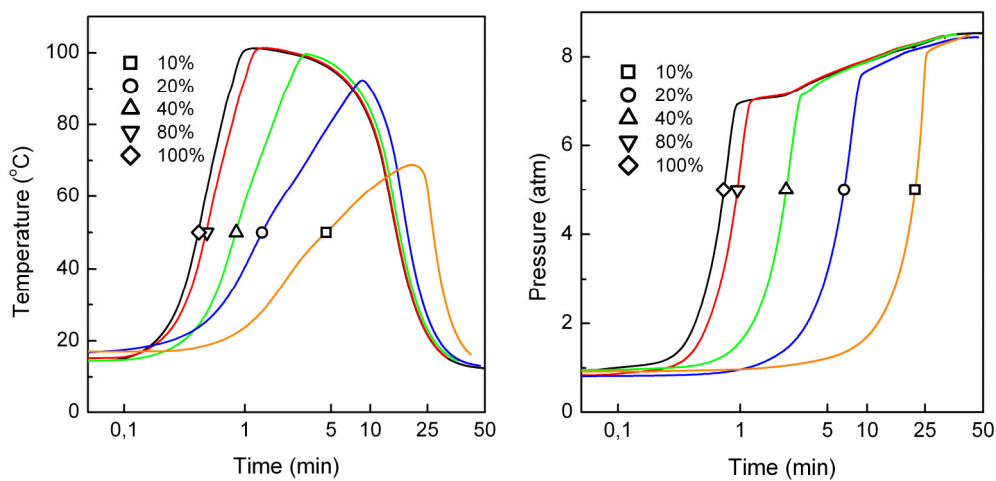

Figure 4:   **Temperature in the center of MH bed (left) and pressure in the reactor (right) for different charging regimes.**

Stage I is characterized by rapid heating of the MH bed connected with pressure increasing. Transition I – II is connected with heat and mass transfer crisis: when maximum temperature is reached, hydrogen flow rate dramatically decreases. Stage II is characterized by steady temperature and pressure changing, hydrogen flow decreases, and at stage III pressure inside the reactor reaches inlet pressure and temperature goes down to coolant temperature. Discharging of the reactor shows similar picture: minimum temperature is reached at stage I, crisis, steady temperature and pressure changing at stage II and finishing at stage III. We can connect these stages with heat and mass transfer processes in the MH bed (see Figs. 5 and 6):

I stage – heating/cooling to reaction temperature. Hydrogen sorption in hydride bed is limited only by regulator at reactor inlet. Heat of reaction cannot be efficiently removed from the bed.

II stage – equilibrium sorption/desorption. Most of hydrogen (>50%) is charged at this stage, and it corresponds to plateau of *PCT*-diagram.

III stage – finishing. Reaction slows down and MH bed is cooled down or heated up to coolant temperature.
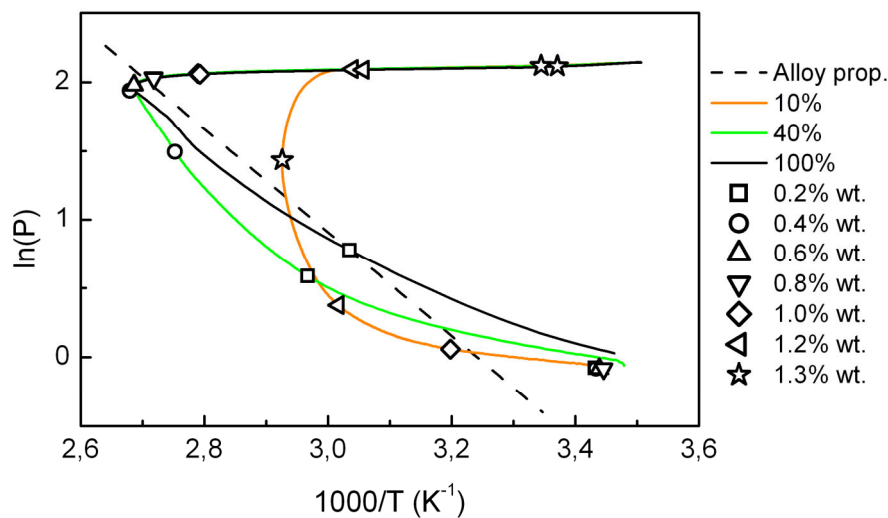
Figure 5: **Charging of the reactor (solid lines) in comparison with alloy properties (dashed line, corresponds to the middle of plateau of *PCT*-diagram), symbols on the solid lines show hydrogen content in the MH bed during the process.**
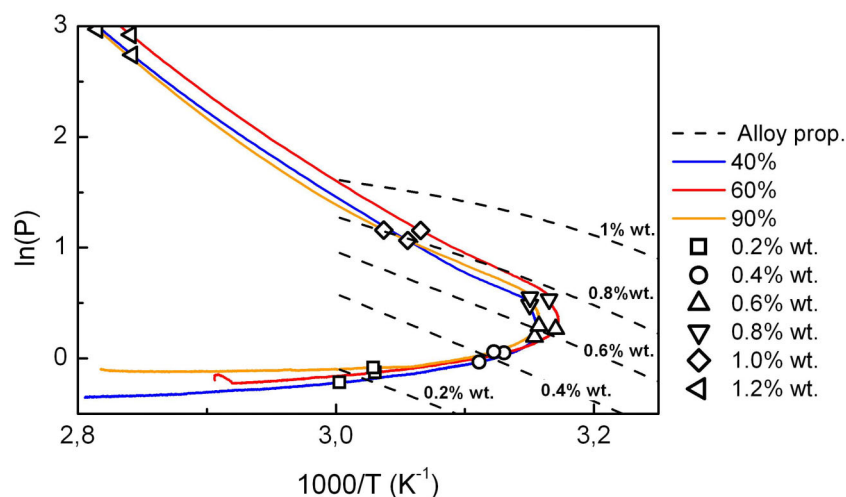
Figure 6: **Discharging of the reactor (solid lines) in comparison with alloy properties (dashed lines, corresponds to hydrogen content on the plateau of *PCT*-diagram), symbols on the solid lines show hydrogen content in the MH bed during the process.**

Two important things can be noticed after the analysis:

Unbalanced regimes with extreme reaction temperature – due to inefficient heat transfer for regimes with high hydrogen flow the hydrogen sorption and desorption reaction temperature is determined by inlet pressure not by coolant temperature, in this case the process is accompanied with crisis.

Heat-balanced regimes – in some regimes with low hydrogen flow rate the heat of reaction is efficiently removed from (moved to) the MH bed, these regimes are not accompanied with the crisis and constant hydrogen flow rate is preserved during the whole process (for example see regime 10% at Figure 5). These regimes are useful for practical applications.

Let determine a criteria for hydrogen flow rate at heat-balanced regime (Figure 7). Reaction heat is equal:

$$Q_{reaction} = m_{MH}\, g\, \Delta H,$$

Heat flow at heat exchanges is equal:

$$Q_{heat\ exchanger} = \alpha\, A\, (T_{MH\ bed} - T_\infty)$$

Heat-balanced regime necessary condition:

$$Q_{heat\ exchanger} > Q_{reaction}$$

Thus for sorption:

$$g_{balanced} < \alpha\, A\, (T_{eq}(P_{in}) - T_\infty)/\, m_{MH}\, \Delta H$$

And for desorption:

$$g_{balanced} < \alpha\, A\, (T_\infty - T_{eq}(P_{out}))/\, m_{MH}\, \Delta H$$
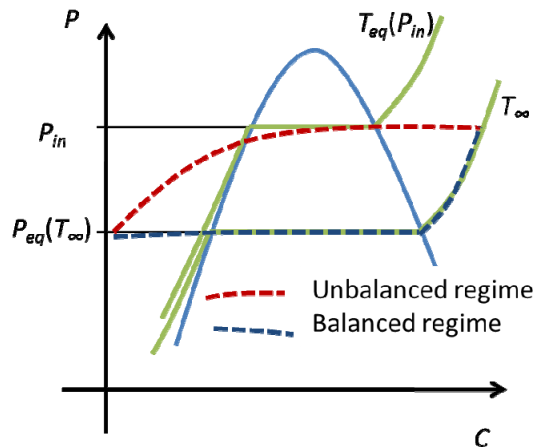

Figure 7:   **Scheme of heat-balanced and unbalanced regimes for charging of MH reactor.**

Conclusions

Charging and discharging of MH reactor is the three stage processes:

I – Heating/cooling to reaction temperature $T_{eq}(P_{in})$ or $T_{eq}(P_{out})$

II – Equilibrium sorption/desorption at reaction temperature

III – Finishing (cooling/heating to T_∞)

I – II transition is connected with heat and mass transfer crisis due to insufficient cooling or heating of MH bed.

There exist heat-balanced regimes with sufficient heat transfer and constant flow rate at reactor inlet, which are useful for practical applications. Criteria for maximum flow rate in heat balanced regime:

for sorption: $\quad\quad\quad\quad g_{balanced} < \alpha\,A\,(T_{eq}(P_{in}) - T_\infty)/\,m_{MH}\,\Delta H$

for desorption: $\quad\quad\quad g_{balanced} < \alpha\,A\,(T_\infty - T_{eq}(P_{out}))/\,m_{MH}\,\Delta H$

Acknowledgments

References

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[2]   Kaplan Y. Effect of design parameters on enhancement of hydrogen charging in metal hydride reactors, International Journal of Hydrogen Energy. - 2009. V. 34. p. 2288.