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System Level Permeability Modeling of Porous Hydrogen Storage Materials

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Abstract

A permeability model for hydrogen transport in a porous material is successfully applied to both laboratory-scale and vehicle-scale sodium alanate hydrogen storage systems. The use of a Knudsen number dependent relationship for permeability of the material in conjunction with a constant area fraction channeling model is shown to accurately predict hydrogen flow through the reactors. Generally applicable model parameters were obtained by numerically fitting experimental measurements from reactors of different sizes and aspect ratios. The degree of channeling was experimentally determined from the measurements and found to be 2.08% of total cross-sectional area. Use of this constant area channeling model and the Knudsen dependent Young & Todd permeability model allows for accurate prediction of the hydrogen uptake performance of full-scale sodium alanate and similar metal hydride systems.

ACKNOWLEDGMENTS

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NOMENCLATURE

κ	Permeability	cm^2
d_p	Pore Diameter	cm
g	Gravitational constant	cm/s^2
ϕ	Porosity	–
τ	Tortuosity	–
ψ	Channel area fraction	–
μ	Viscosity	Poise
ΔP	Pressure drop	barye and psi
L	Bed length	cm
ρ	Density	g/cm^3
v	Velocity	cm/s
Kn	Knudsen number	–
Re	Reynolds number	–
Ga	Galileo number	–
Ψ	Pressure drop parameter	–

1. INTRODUCTION

Hydrogen-based power systems have been developed for a variety of applications including transportation applications and materials handling solutions [1, 2]. These systems provide high-efficiency energy storage and utilization solutions that exceed the performance of battery systems [3, 4]. In particular, the development of a practical hydrogen-powered vehicle system has been the focus of numerous recent projects and initiatives [5]. The primary technical challenge in the realization of such systems is storage of hydrogen efficiently in terms of volume and mass. The primary methods of storing hydrogen are as a gas, as a liquid, and in solid-state storage materials such as metal hydrides, high surface area cryosorbents, or chemical hydrides [6]. The latter, solid-state methods, hold promise to exceed the energy densities of both gaseous and liquid hydrogen storage.

Many solid-state hydrogen storage materials consist of fine powders or porous structures packed into a vessel where hydrogen is introduced from an inlet tube when refilling the tank. Many of these materials provide significant resistance to hydrogen flow during this process, which can limit hydrogen mass transport within the storage system. Additionally, the small pore diameters in these packed systems can introduce non-continuum flow effects, which complicate analysis of mass transport phenomena.

In this paper, we consider the mass transport properties of a sodium aluminum tetra-hydride ($NaAlH_4$) storage system. As a result, a model of hydrogen transport in the hydride bed was developed which can be used for subsequent design and optimization of systems of different sizes, geometries, or storage materials.

2. BACKGROUND

2.1 Sodium Alanates

The discovery in 1997 that sodium alanates (Na_3AlH_6 and $NaAlH_4$) could be destabilized to decompose at lower temperature by the addition of a titanium catalyst introduced a new complex hydride to the field of possible hydrogen storage materials [7, 8]. While these materials do not meet the current DOE hydrogen storage targets [9], they present an opportunity to study coupled hydrogen heat and mass transport and chemical kinetics. Investigation of chemical kinetics and thermal properties has been done extensively in previous work, and will be used for the analysis herein [10, 11].

2.2 Flow in Porous Media

There are many models which have been presented in the literature for flow through porous media. Some of the most common models are the Ergun model, the Darcy-Brinkman model, and the Young & Todd permeability model.

The Ergun model was originally introduced by Ergun in 1952 [12]. The Ergun model treats the porous media as a series of capillary flow channels which induce viscous and inertial pressure losses similar to those resulting from porous media.

$$\frac{\Delta P}{L} = A\mu v + B\rho v^2 \qquad (1)$$

Ergun also went on to derive the values of the *A* and *B* coefficients for beds of packed spheres as follows.

$$A = 150\frac{(1-\phi)^2}{\phi^3 d_p{}^2} \qquad B = 1.75\frac{(1-\phi)}{\phi^3 d_p} \qquad (2)$$

Equation 1 can be normalized as in Equation 3, using the modified Reynolds and Galileo numbers. Here *A* and *B* are set to 150 and 1.75, respectively, using the original Ergun relationship [13, 14].

$$\Psi Ga^* = A\,\mathrm{Re}^* + B\,\mathrm{Re}^{*2} \qquad (3)$$

$$\Psi = \frac{\Delta P / L}{\rho g} \qquad (4)$$

$$\mathrm{Re}^* = \frac{\rho v d_p}{\mu(1-\phi)} \qquad (5)$$

$$Ga^* = \frac{\rho^2 g d_p{}^3 \phi^3}{\mu^2 (1-\phi)^3} \qquad (6)$$

Several variations on this equation have also been proposed, including the Carman-Kozeny equation, which uses A = 180 and B = 0 [15]. Other authors have adjusted A and B experimentally to match their measurements for flow through packed beds of non-spherical particles [16]. The Ergun equation, however, is most easily applied to 1-D continuum flow systems, so while it can be applied to laboratory-scale permeability experiments, it is not easily applied to more complex 3-D systems where cross-flow and non-continuum effects are often present.

The Darcy-Brinkman equation combines Darcy's law with the Stokes equation to produce the following relationship [17, 18].

$$\mu_e \nabla^2 v - \nabla p - \frac{\mu}{\kappa} v = 0 \qquad (7)$$

This relationship allows for modeling of gas flow through multi-dimensional porous media using finite element analysis. It does, however, require knowledge of the permeability of the porous media (κ) and the effective viscosity (μ_e). It has been shown experimentally that the effective viscosity term is required to reconcile experimental data with the model [19, 18, 20], however, here it is assumed that the effective viscosity is equal to the "real" viscosity ($\mu_e = \mu$).

The permeability of the bed, for use in the Darcy-Brinkman equation, is calculated by the model presented by Young & Todd [21], which accounts for slip flow by including a linear Knudsen number dependence. Other recent work has discussed a second order Knudsen dependence, and also incorporated a Knudsen dependence into the traditional Ergun equation [22]. Tang and colleagues present the permeability of a single microtube as shown in Equation 8, where the constants C_1 (typically 1 to 1.15) and C_2 (typically -0.5 to 1.3) have been given different values by different authors [22].

$$\kappa = d_p^{\,2}\left(\frac{1}{32} + \frac{C_1}{4} Kn + \frac{C_2}{2} Kn^2\right) \qquad (8)$$

The Young & Todd relationship for permeability (given in Equation 9) neglects the second order Knudsen term (C_2 = 0), uses C_1 = 5/3, and introduces the ϕ/τ^2 term to average the permeability over all the "microtubes" in a porous media and adjust for their actual tortuous length [21].

$$\kappa = \frac{\phi d_p^{\,2}}{\tau^2}\left(\frac{1}{32} + \frac{5}{12} Kn\right) \qquad (9)$$

For continuum flow ($Kn \rightarrow 0$) in a single capillary tube of diameter d_p, this relationship simplifies to the well known Darcy friction factor relationship for laminar flow in a circular tube ($f = 64/Re$), with the term ϕ/τ^2 acting as a pore number and length adjustment factor such that when they equal unity, the relationship simplifies to the well-known equation for laminar tube flow, which is similar to the derivation done by Endo [23], who also adjusted flow based on the tortuosity.

2.3 Channeling

The process of channeling, whereby the gas creates preferential flow channels, usually near the walls or edges of the porous media, is another significant physical process which the permeability models noted above neglect. It has been shown by Schlunder that a system of capillary tubes with one tube much larger than the others is not well represented by a model with the average capillary diameter [24]. This indicates that a wall-channeling model cannot simply rely on pore size averaging. At application-scales, this phenomena can affect overall mass transport significantly. In systems where the ratio of overall diameter to particle diameter is small (< 20), this effect can be explained by the increased porosity in the region of the wall, which is several particle diameters thick [25, 26]. However, in a large system with small particles this does not adequately explain wall channeling. One proposed method to deal with wall channeling is to create a region of higher permeability near the wall which is a constant fraction of the overall area (Figure 1) [27, 28]. This method is still based on the different porosity near the wall for large particles, but is applied in this model to describe the formation of "wall channels" which form naturally in the presence of high gas flows.

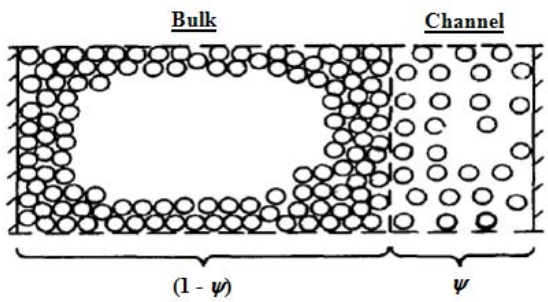

Figure 1 - Constant area fraction (ψ) channeling model proposed by Martin [27]

This approach requires additional parameters for the channel permeability, as well as the channel area fraction, but can be applied more successfully to different geometries. The presence of a wall channel in the large-scale sodium alanate system has been observed (Figure 2).

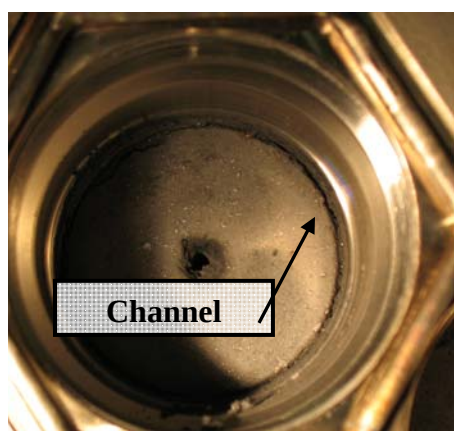

Figure 2 - Large scale sodium alanate system showing visible wall channel

3. METHOD

3.1 Laboratory-Scale Permeability Measurements

In order to study the permeability characteristics of the sodium alanate system, three small-scale permeability measurement setups were developed (Figure 3). In this system, different gasses of different temperature are flowed through a packed bed of sodium alanate (including Ti and ENG additives) held in place by a pair of porous stainless steel frits. These sample holders have different aspect ratios, so that the effect of wall channeling will be variable across the measurements and can be analyzed

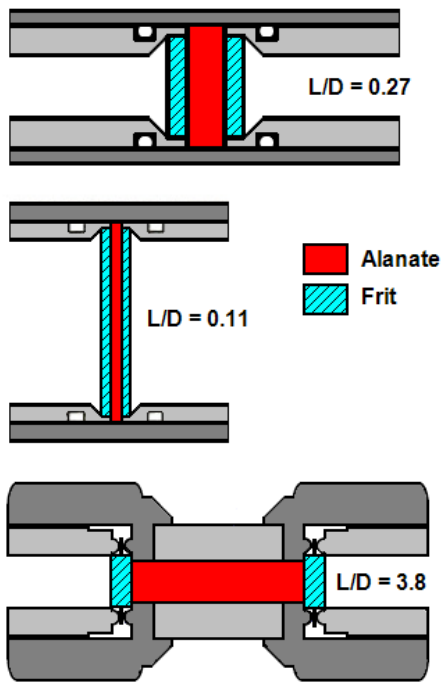

Figure 3 - Permeability measurement setups: standard aspect ratio (STD), short aspect ratio (SAR), and long aspect ratio (LAR), shown from top to bottom.

For an initial analysis of these experiments, the Ergun relationship is compared to the experimental data in Figure 4 through Figure 6. The effect of the frits was determined first (Figure 4), then subtracted from the alanate data so the pressure drop over the alanate only could be examined (Figure 5). It can be seen that the sample holders with low aspect ratios (large diameter, small thickness) fall very close to the Ergun relationship, while the long aspect ratio sample measurements show lower pressure drop and higher flow rates than expected. The difference between the data and the Ergun relationship corresponds to an “effective” porosity which is 1.2 times greater for the standard sample holder, and 1.5 times greater for the long aspect ratio sample holder (Figure 6). This indicates that there are flow paths with less resistance than a homogeneously packed bed in these geometries which are less prominent in the short aspect ratio geometry.

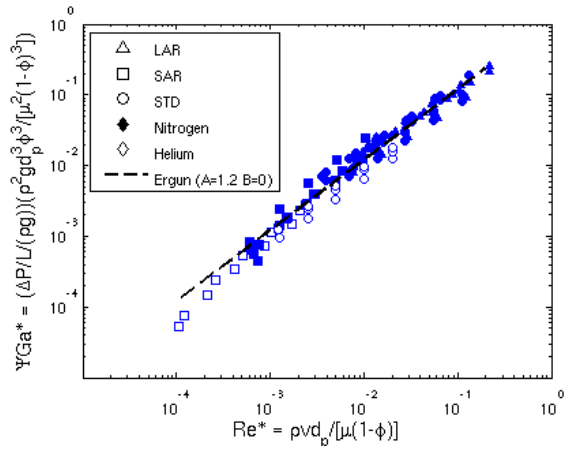

Figure 4 - Frit pressure drop compared with modified Ergun relationship

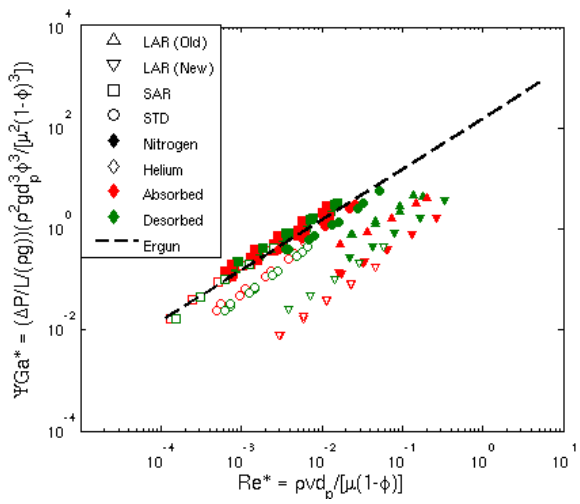

Figure 5 - Nondimensionalized flow data compared with Ergun relationship with no channeling effects and frit effect removed

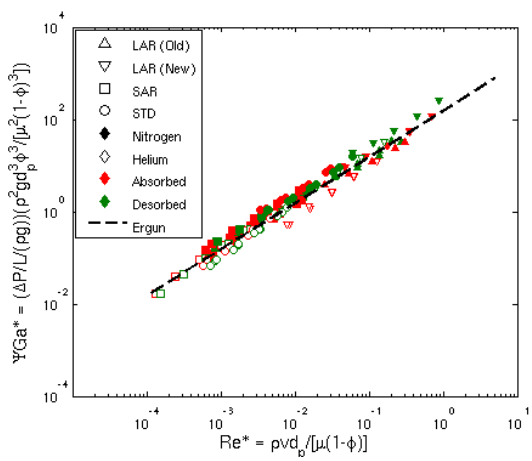

Figure 6 - Nondimensionalized flow data with a "channeling factor" on the porosity for the STD, SAR, and LAR sample holders

To analyze the permeability results in a manner which accounted for the geometry's deviation from 1-D, separate axially symmetric models using the finite element method (FEM, implemented in Comsol™) of the three permeability setups were constructed (Figure 7). These models solve the Darcy-Brinkman equation for flow through porous media over the experimental domain.

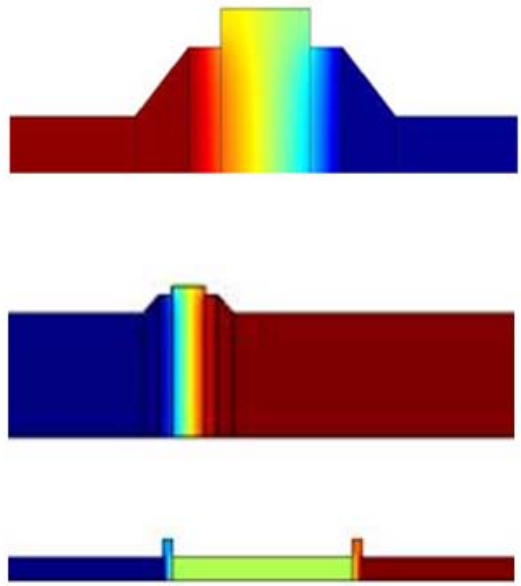

Figure 7 – Axially symmetric FE models of small-scale permeability setups without a wall channeling layer shown: standard aspect ratio, short aspect ratio, and long aspect ratio (top to bottom)

The first permeability measurements performed in each test campaign were performed with only the frits present. These results were used to determine the permeability parameters of the frits. Using these parameters, experiments conducted with alanate were analyzed to determine the appropriate permeability parameters for the alanate.

3.2 Application-Scale Permeability Measurements

In the system studied herein, a 90 cm long, 5 cm diameter stainless steel tube immersed in oil is filled with 1.6 kg of sodium alanate at a packing density of 1 g/ml (corresponding to a porosity of 51%), 3 mol% Ti catalyst ($TiCl_3$ precursor) and 10 wt% expanded natural graphite (ENG) for thermal conductivity enhancement. This tube is used to conduct full-scale hydrogen fueling experiments by introducing hydrogen at high pressure at one end, and controlling temperature so the reaction proceeds at a reasonable rate.

A FE model of the full tube was developed which couples previously determined kinetics for sodium alanates and thermal properties [10], as well as flow through porous media using the Darcy-Brinkman equation and the Young & Todd permeability model. To determine the validity

of the model, the transient amount of hydrogen absorbed in the model is compared with experimental data of actual hydrogen inflow measured with a coriolis flow meter.

3.3 Results

Using the permeability parameters shown in Table 1 and a channel area fraction of 2.08%, the channeling model was applied to all permeability measurement geometries. In all these geometries, the channel was placed along the perimeter of the alanate sample (Note that another possibility not considered here is bulk channeling, where channels form through the bulk of the material).

Table 1 - Channeling model parameters

Parameter	Bulk	Channel
ϕ	Measured	0.8
τ	1.8	1.2
d_p	1.3 μm	15 μm

3.4 Small-Scale Results

The comparison between the modeled results and measurements (Figure 8 through Figure 10) indicates that the model matches the measurements best when using the porosity calculated from the solid density of the desorbed material (ρ_{solid} = 2.04 g/cc). This is likely due to changing morphology, indicated in SEM images of absorbed and desorbed alanate, which indicate that the absorption process fills the material micropores, but does not significantly affect the prominent flow paths through the materials.

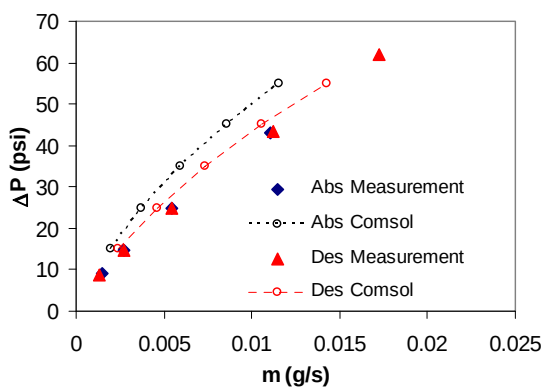

Figure 8 – Standard sample holder data compared with 107 μm channel FE model

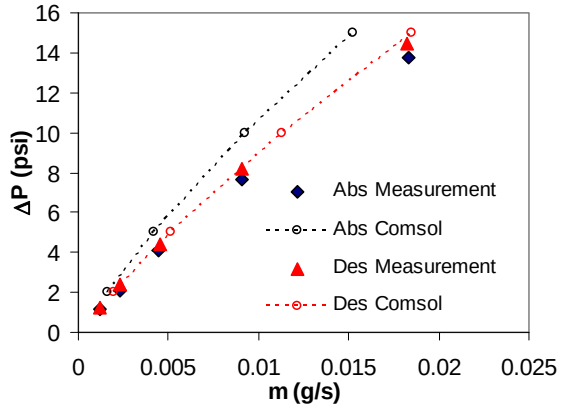

Figure 9 – Short aspect ratio sample holder compared with 230 μm channel FE model

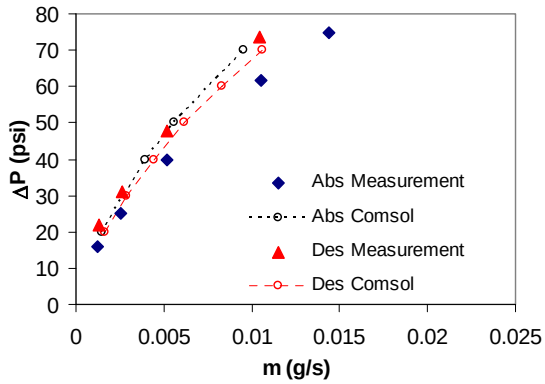

Figure 10 - Long aspect ratio sample holder (old geometry) compared with 50 μm channel FE model

3.5 Application-Scale Results

Upon the successful development of a permeability and wall channeling model that is geometry-independent, the characteristics of application-scale hydrogen transport were investigated in the sodium alanate system. The need for the channeling model was illustrated by the long aspect ratio tests and the application-scale tests, where measured flow rates were significantly higher than those predicted by a permeability model without wall channeling.

A comparison between the initial, no channeling model results and the results with the current channeling model shows much better adherence between measured data and calculations (Figure 11). Note that the modeled reaction continues beyond the point where the actual reaction ends. This is likely due to scarcity of NaH/Al reaction sites caused by depletion of the NaH concentration, which is not explicitly captured in the kinetic model used in the simulation.

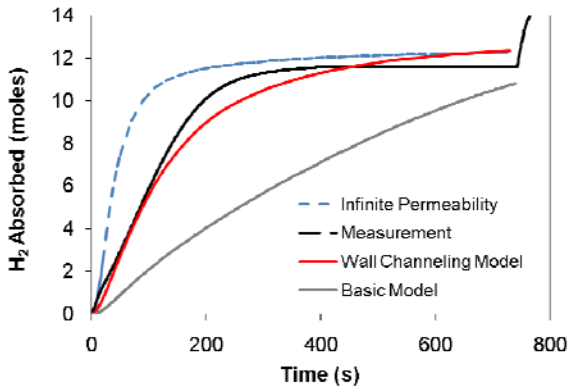

Figure 11 - Comparison between FE simulation with 520 µm channel and measurement of hydrogen absorbed in the module tube, showing good agreement over the basic (no channeling) model

4. CONCLUSIONS

The use of a strict permeability model, such as the Ergun relationship [12] or the Young & Todd model [21] does not accurately predict system-level hydrogen transport performance in large beds if wall channeling is not included. A wall channel model consisting of 2.08% of the total cross-sectional area was found to most closely match the flow characteristics of both high and low aspect ratio bed geometries. A subsequent publication will quantify the physical geometry of the wall channel in the application scale system [29].

Use of this constant area channeling model and the Young & Todd permeability model allows for accurate prediction of the hydrogen uptake performance of full-scale sodium alanate systems. It is likely that a variety of solid hydrogen storage materials will exhibit similar behavior; the application of this model to these alternative systems will enable the design and optimization of hydrogen uptake and release within the integrated system.

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