MODELING LABORATORY EXPERIMENTS OF FLUID FLOW AND HEAT TRANSFER IN
SUPERCritical-CO2-SATURATED CORES WITH ECO2N V2.0
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ABSTRACT
We have investigated heat transfer in a set of laboratory experiments where we flowed dry CO2 through a bed of heated sand. Our laboratory apparatus is capable of operating at temperatures up to 200°C, pressures up to 340 bar, and flow rates up to 400 ml/min. In our system, we measure temperature throughout the sample. We designed the experimental system such that measurements and controls at the boundaries could be readily modeled with TOUGH2 and the ECO2N fluid property module. Significant cooling occurs over the course of an experiment resulting in large CO2 property changes. Past modeling attempts failed to achieve a good match between simulation results and experimental data, likely due to the lack of effective thermal conductivity updating of CO2-saturated grid blocks during the course of the simulation.

We found that temperature changes caused the effective thermal conductivity in our sample to vary from 1 to 0.2 W/m-K during a single experimental run. In order to take this behavior into account, optional code was included in the improved ECO2N V2.0 distribution package that estimates the thermal conductivity of the saturated rock as a function of rock properties, NaCl concentration, CO2 saturation, and the varying thermal conductivity of CO2. We performed five CO2 flood experiments at various flow rates and system pressures to collect temperature, pressure, and mass flow rate measurements for use in model validation. A detailed model of our system was implemented in TOUGH2 and ECO2N using the optional thermal conductivity code. We found that the new code capabilities provided a significantly better fit to the experimental data than when a single effective thermal conductivity value was used.

INTRODUCTION
Numerical modeling tools are necessary for studying, planning, and operating geologic-based CO2 sequestration and CO2-based geothermal energy projects. Project viability and safety is dependent on the results of computer simulations of heat and mass flow in porous media, therefore model validation is an important concern. Models can be tested against closed form solutions, compared with other models (Pruess, 2004), or ideally they can be compared with measurements of actual physical systems. The data sources for validation either come from field data, that is usually sparse in space and time and very expensive, or laboratory experiments, which are often denser in space and time but usually lack the proper scaling.

Previous experiments collected data from a specially constructed apparatus that injected cold CO2 into a heated porous sample and compared the results to a numerical model of the system implemented in the TOUGH2 family of codes using the ECO2N equation of state module (Magliocco et al., 2015). TOUGH2 is a general-purpose non-isothermal, multiphase, multicomponent fluid flow simulator for porous and fractured media developed at Lawrence Berkeley National Laboratory (LBNL) (Pruess, 2004). In previous work it was impossible to achieve a reasonable match between experimental and model results by trial and error. Our analysis found that the constant effective thermal conductivity of the saturated medium assumption used in TOUGH2/ECO2N was a likely source of error in the model results.

Since the initial experiments were performed, the ECO2N module has been updated and optional code (TCSUB) created that allows for
more accurate effective thermal conductivity modeling (Pan et al., 2015). Using an updated version of our apparatus a new set of data was captured that includes the mass flow at the outlet of the vessel and was used to perform the first comparison of the results of the new thermal conductivity modeling code to measured data. This paper presents the results of the experimental and numerical studies and examines what effects the updated thermal conductivity model has on the simulation accuracy.

CORE FLOOD EXPERIMENTS

Experimental Apparatus

Temperature, pressure, and flow rate measurements were taken with the experimental apparatus shown in Figure 1, which was an updated version of the system used for previous work (Magliocco et al., 2015). The experiment was designed to generate data for numerical model validation purposes by careful implementation of boundary conditions, the layout and density of temperature measurements, and the selection of experimental parameters such as flow rate and porous medium grain size. The inlet to the test vessel was controlled as a constant temperature and constant mass flux boundary by means of the computer controlled pumps and a laboratory chiller. The outlet was controlled as a constant pressure boundary condition by means of a back pressure regulator. Constant pressure and constant mass flux boundary conditions were easily implemented in a TOUGH2 model. The vessel was insulated by a custom fabricated aerogel insulation blanket in order to impose a relatively low heat flux at the exterior surfaces of the vessel. The pressure vessel was a hollow type 304 stainless steel cylinder with an inside diameter of 9.1 cm, outside diameter of 12.7 cm, and a 50.8 cm height between the type 316 stainless steel end caps secured by 4430 alloy steel caps.

Instrumentation and flow access to the interior of the vessel was through three axial passages through the bottom end cap, and one passage through the top. The vessel was oriented vertically such that the flow path was in the same orientation as the gravity-induced pressure gradient in order to minimize instabilities and maintain radial symmetry in the system.

Figure 1. Diagram of experimental apparatus. Fluid was supplied by a siphon style CO₂ tank. Fluid was driven by a pair of pumps and fed through air-actuated valves. The fluid was chilled before it passed into the bottom inlet of the vertically oriented pressure vessel. A differential pressure sensor was connected hydraulically to the inlet and outlet of the vessel. Pressure and mass flow sensors were located at the outlet (top) of the vessel.
Temperature measurements within the sample were made with 23 small diameter (0.79mm) stainless-steel clad type-T thermocouples. The thermocouples were arranged at several elevations and radii in the sample (Table 1). At one elevation in the porous media, two thermocouples were mirrored so that they were both at the same radial distance from the central axis of the vessel to test our assumption of a radial symmetry in the heat transfer process.

<table>
<thead>
<tr>
<th>TC#</th>
<th>Z (cm)</th>
<th>R (cm)</th>
<th>TC#</th>
<th>Z (cm)</th>
<th>R (cm)</th>
</tr>
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<tr>
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<td>0</td>
<td>12</td>
<td>25.4</td>
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</tr>
<tr>
<td>2</td>
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<td>30.5</td>
<td>0</td>
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<td>30.5</td>
<td>1.5</td>
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<tr>
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<td>0</td>
<td>15</td>
<td>30.5</td>
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<tr>
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<td>3.0</td>
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<td>0</td>
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<td>0</td>
</tr>
<tr>
<td>11</td>
<td>20.3</td>
<td>4.6</td>
<td>22</td>
<td>50.8</td>
<td>2.3</td>
</tr>
</tbody>
</table>

Table 1. Thermocouple location and numbering. This table excludes the thermocouple that duplicates the radial position of TC15.

Porous Core Sample

The core sample consisted of dry-packed, well-sorted, spherical shaped quartz silica sand. The shape and sorting of the media were chosen to further simplify the system (Magliocco et al, 2015). Many conceptual models of porous media flow make use of a packed bed of spheres to represent the solid matrix. 4.9489 kg of prepared sand was packed into the mounted vessel in 15 separate lifts with manual tamping with a rounded aluminum rod between lifts. The porous sample properties are listed in Table 2.

<table>
<thead>
<tr>
<th>Porous Core Properties</th>
</tr>
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<tbody>
<tr>
<td>Total Core Length</td>
</tr>
<tr>
<td>Cross Sectional Area</td>
</tr>
<tr>
<td>Crystalline Quartz Density</td>
</tr>
<tr>
<td>Crystalline Quartz Specific Heat</td>
</tr>
<tr>
<td>Crystalline Quartz Thermal Conductivity</td>
</tr>
</tbody>
</table>

Experimental Results

Five single-phase CO₂ experiments were performed under the conditions listed in Table 3. All experiments were operated above the critical pressure of CO₂ to ensure no gas phase was present in the system.

Table 3. Experimental conditions.

<table>
<thead>
<tr>
<th>Experiment #</th>
<th>Injection Flow Rate</th>
<th>Outlet Pressure</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>200 ml/min</td>
<td>108 bar</td>
</tr>
<tr>
<td>2</td>
<td>50 ml/min</td>
<td>147 bar</td>
</tr>
<tr>
<td>3</td>
<td>200 ml/min</td>
<td>108 bar</td>
</tr>
<tr>
<td>4</td>
<td>100 ml/min</td>
<td>108 bar</td>
</tr>
<tr>
<td>5</td>
<td>50 ml/min</td>
<td>108 bar</td>
</tr>
</tbody>
</table>

The CO₂ was injected in the liquid phase at a temperature of 11°C. The temperature data from the twenty-two thermocouples (ignoring the redundant thermocouple) from a typical experimental flow run are shown in Figure 1. The sample was saturated with CO₂ at the desired experimental pressure and heated to the desired initial temperature and allowed to equilibrate. Before injection initiation a vertically oriented thermal gradient was present in the vessel with the highest temperature at the top of the vessel and the lowest temperatures at the bottom. Injection initiation can be seen as the temperature at the sample inlet (solid green line) drops shortly after flow initiation at time zero. The temperature at the vessel wall (dotted lines) decreases at a slower rate due to the heat energy stored in the stainless steel. After the initial temperature front has passed a strong thermal gradient develops in the inward radial direction as the heat is drawn out of the vessel wall by the passing CO₂.
Figure 2. Temperature vs time data from twenty-two thermocouples from experimental flow run #3 operated at 200ml/min flow rate, 108 bar pressure, and an initial vessel temperature of 100°C. Line color indicates elevation of the sensor, green being at the vessel bottom and yellow at the top. Line style indicates radial location with solid lines at the vessel axis and dotted lines at the exterior. Thermocouples are numbered in order of radial location and then by elevation starting at the bottom of the sample axis.

MODELING

A 2D axisymmetric model of the system was developed for evaluation in TOUGH2, a numerical simulator for non-isothermal flows of multi-component, multiphase fluids in one, two, and three-dimensional porous and fractured media, and the ECO2N property module which is capable of modeling mixtures of water, NaCl, and CO₂. Version 2.1 of TOUGH2 and version 2.0 of ECO2N were used with the included optional TCSUB code which implements effective thermal conductivity updating as a function of CO₂ thermal conductivity, rock thermal conductivity, and a pore shape parameter (Zimmerman, 1989; Pan et al., 2015).

During simulations, the standard TOUGH2/ECO2N code will vary the thermal conductivity of grid blocks based only on the degree of saturation by calculating a value based on the thermal conductivity of the dry and fully saturated block. Without TCSUB enabled, ECO2N does not update the effective thermal conductivity of CO₂ saturated grid blocks despite the fact that the thermal conductivity of CO₂ can vary greatly as a function of pressure and temperature (Magliocco et al., 2015). The functionality of TCSUB was enabled and disabled by changing the value of the IE(10) parameter in the SELEC block of the input file. An IE(10) value equal to zero will use the default TOUGH2 thermal conductivity handling, and an IE(10) value equal to 1 makes use of the new updating scheme based on effective medium theory (Zimmerman, 1989).

TOUGH2 with the ECO2N module was compiled on 64-bit intel Core i3 and Core 2 Duo processors running Ubuntu 14.04 and Mac OS X 10.10 respectively using the GNU Fortran compiler GFortran (4.8.2 on Ubuntu and 4.9.2
on the Macintosh). The Mac system was used primarily for model development with coarse meshes while the more computationally robust Linux system was used for finer resolution simulations.

A suite of custom Matlab scripts was created that would sequentially generate the model mesh, initialize the model input files based upon the experimental data, initiate the TOUGH2 simulation, import the results into Matlab and analyze and plot the results compared to experimental data. The Matlab mesh generation function was capable of producing a mesh of the appropriate dimensions with user selectable resolution. Besides the pressure and mass flow rate, the TOUGH2 simulation was initialized with an initial temperature distribution that was derived from the experimental temperature data.

**Mesh Design**

The model mesh geometry was based upon measurements of the experimental vessel with the exterior surface of the vessel taken as the system boundary (Figure 3). The mesh is 2D axisymmetric described on the R-Z plane and revolved around the Z axis creating a series of stacked and nested annuli with the appropriate 3D volumes and surface areas. The mesh blocks were assigned to one of seven different domains: the inlet block, outlet block, passage through the end caps, stainless steel vessel body and end caps, carbon steel vessel nuts, packed sand sample, and sand in contact with the vessel walls.

Interpolation was employed in order to approximate the modeled temperature values at thermocouple locations when a larger than thermocouple diameter grid-block size is used, the results of which vary depending on the temperature gradient and the mesh resolution especially near boundaries. To explore the sensitivity of the results to grid-block size, simulations were run with various mesh resolutions. Ultimately a mesh resolution was chosen of approximately 4 mm in the z direction and 8 mm in the r direction.

**Domain Properties**

The majority of the domain properties such as density, specific heat, and thermal conductivity were based on standard reference values (Avallone, Baumeister, and Sadegh 2006). Other values such as the sample porosity and permeability were based on laboratory measurements. To impose a constant temperature on the injection fluid, the injection cell domain was given a very large density and specific heat, and was initialized with the desired injection temperature.

![Figure 3. Partially revolved model mesh, with colors indicating material type. Not to scale and not representative of final mesh resolution.](image)

**Boundary Conditions**

Three boundary conditions were included in the model, a constant pressure condition at the outlet, a mass flux at the inlet, and heat loss to the lab environment at the vessel exterior. The inlet cell was set as a generation cell with the type set to single phase CO$_2$ at either a constant mass flow, or a flow rate that varied over time, based on the measured experimental conditions. The outlet cell behavior takes advantage of a TOUGH2 computational shortcut in which the cell is marked as “inactive” and is not included in any of the mass or energy balance equations to ensure that the state does not change from the initial values. It was found that including heat loss at the boundary did not significantly improve the fit of the model to measured data.
Model Calibration

The choice of the thermal conductivity input parameters for the model was dependent on the thermal conductivity handling method chosen in the model input file using the IE(10) parameter which enables the TCSUB option. The standard TOUGH2/ECO2N code only changes the thermal conductivity of the grid block based on fluid saturation. The experiments we conducted were under fully saturated conditions at all times so the chosen saturated thermal conductivity value was used throughout the simulation. The modeler must choose a single pressure and temperature at which to make their estimate for thermal conductivity out of the range of pressures and temperatures that occur over time and space during a single experiment. This choice was problematic due to the fact that any estimate will only be valid for limited locations and times within the sample. Furthermore, the value would have to be estimated and calibrated separately for each experimental run based on the unique operating parameters, making the model less deterministic.

Due to the characteristics of the prepared core sample, estimates of effective thermal conductivity made use of a well-tested (Woodside and Messmer, 1961) model based on an unconsolidated packed bed of uniformly sized spheres (Kunii and Smith, 1960). Estimated effective thermal conductivity is a function of the sample porosity and reference values for the thermal conductivities of CO₂ and quartz. The calculated values ranged from approximately 0.2 to 1 W/(m K) with the lower value associated with cold injection CO₂ and the higher value with the hot CO₂ present at the initial conditions.

When the new TCSUB option was enabled, the model inputs are the thermal conductivity of the dry rock and a parameter, α, that relates to the shape of the pore space. By basing the thermal conductivity on physical parameters, the model becomes more deterministic and should produce accurate results for all operating conditions. Three limiting shapes have been identified that describe the pore spaces as flat discs (α = 0), spherical (α = 1), and needle-like pores (α > 1) (Zimmerman 1989). For the initial choice we used an α value equal to one, and a thermal conductivity reference value for quartz grains situated in random orientations (Woodside and Messmer, 1961). These values generated simulation results with a good initial fit to experimental data and allowed us to study the sensitivity of our model to other parameters such as heat loss, the effect of higher porosity at the vessel wall, and mesh resolution.

During calibration it was found that the modeled temperature consistently mismatched the experimental data near the end caps of the vessel. This was most likely due to the radial flow that occurs near the end caps, which results in very high pore velocities near the injection and outlet ports. The high pore velocities around the inlet and outlet are well outside of the Darcy flow regime and the theoretical capabilities of TOUGH2.

The model also consistently under-predicted the temperature front arrival time at the lower elevations in the sample. This was more apparent in the higher flow rate experiments, indicating this may be due to the upstream prediction errors at the injection end cap propagating up the sample column. When comparing the misfit between simulations, the temperature data at the bottom two thermocouple elevations (numbers 1 through 7) and the highest thermocouples located near the outlet end cap (numbers 21 and 22) were disregarded.

In order to differentiate results, a quantitative approach was applied using the weighted mean square error summed over all experiments as a measure of model misfit

\[
\Phi = \frac{1}{5} \sum_{j=1}^{5} \frac{1}{k_j} \sum_{t=1}^{k_j} \frac{1}{13} \sum_{i=8}^{20} \frac{(d_{ij}(t) - s_{ij}(t))^2}{\sigma^2},
\]

where \( s_{ij}(t) \) is the simulation result at thermocouple number \( i \) at time \( t \) from experiment number \( j \), \( k \) is the number of simulation time steps, \( d_{ij}(t) \) is the recorded experimental data, and \( \sigma \) is the standard deviation of the measurements (estimated from mirrored thermocouple data). The misfit values for some different thermal conductivity choices are shown in Table 4.

Table 4. Misfit values for various thermal conductivity parameter choices.
The lowest misfit value was achieved with an $\alpha$ value of 0.08, while the lowest misfit for a single, constant effective thermal conductivity was 3 W/(m K) which was out of the range of expected values. Without the TCSUB code enabled, it would have been difficult to calibrate the model using realistic effective thermal conductivity values using this calibration method.

**MODELING RESULTS**

Using the TCSUB option ($\alpha = 0.08$), the calibrated model simulation results for the central thermocouples (radial location = 0) are shown in Figure 4 with diamond markers, along with the experimental results shown without markers. The general temperature trends and front arrival time predictions produced by the simulation are relatively good at locations which are not in contact with the end caps.

The use of the TCSUB option allowed for relatively good fits with a wide range of $\alpha$.
choices (Table 4), and allowed for much easier model calibration. This indicates that the theoretical basis of the TCSUB code was more accurate than the assumption of constant thermal conductivity of the CO$_2$ saturated rock. In the original attempts at modeling experiments without TCSUB (Magliocco et al 2015), it was difficult to choose a reasonable $\lambda_{\text{eff}}$ that could be used to identify and correct other deficiencies in the model. A good overall fit could be achieved for individual experiments using a carefully chosen and often unrealistic effective thermal conductivity value that was outside of the range supported by theory and research findings. Using reasonable values for $\lambda_{\text{eff}}$ resulted in an overall poor match for all experiments.

CONCLUSION

We measured experimental data using an experimental apparatus capable of producing temperature, pressure, and mass flow measurements of cold CO$_2$ flow through a heated porous sample. Five experiments were conducted under well-controlled conditions, and the resulting data was subsequently used for model validation. The results of the experiments and the modeling show that TOUGH2 with the TCSUB option enabled in the ECO2N module is capable of simulating heat transfer in CO$_2$ saturated porous media with reasonable accuracy with minimal calibration using reference values for material properties and direct measurements of our system. This was an improvement over the previous version of ECO2N that required the modeler to choose a possibly unreasonable value for the effective thermal conductivity of the CO$_2$ saturated media for each separate experiment in order to get a reasonable model fit. The new method enabled by the use of TCSUB was more deterministic in nature and more conceptually sound. While the TCSUB code does require the modeler to choose a pore shape parameter, the sensitivity of the system to the choice was not as great as the sensitivity to the choice of effective thermal conductivity when TCSUB was disabled, allowing more rapid and reasonable model calibration.

REFERENCES


