

Reply to the “Comments on the Paper ‘Quantification of Density-Driven Natural Convection for Dissolution Mechanism in CO₂ Sequestration’ by R. Nazari Moghaddam et al. (2011)”

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In this note, we reply to the comments by [Meybodi \(2011\)](#) on our article “Quantification of density-driven natural convection for dissolution mechanism in CO₂ sequestration” (2011). Meybodi’s comments ([2011](#)) are discussed in details in the following:

We know that in the Zhang model ([2000](#)), the constant boundary condition is assumed. We used this model for interpretation of the experimental results. However, some notes must be considered: The main reason of using Zhang method in our paper is not determination of the exact value of CO₂ diffusion coefficient into water. We used this method just for comparison of two measured diffusion coefficients for initial and final stages of the experiments in order to show the activation of the “Convection”. In addition, this method is a simple and very easy for determination of hypothetical diffusion coefficient (Pseudo-diffusion coefficient) for using in diffusion equation and accounting the rate of mass transfer. We assumed the constant pressure boundary condition and compared the obtained results by this assumption with graphical method of [Sheikha et al. \(2006\)](#). It is shown that there is not considerable difference between two methods (see Tables 2, 4 in our article). Since we wanted to compare the order of magnitude of the measured coefficients, Zhang method could be reasonably applicable.

Meybodi in his comments stated that we claimed there are only two stages of mass transferring in real process of CCS. We believe that this point have been clearly enough explained in our paper and he did not read our article carefully enough. We reported only the processes which occurred in our experiments and we did not claim about the processes which are necessarily happen in the real media.

In the following, we try to calculate more precise value of the onset time in our experiments to show the occurrence of the natural convection **just at the start of the experiments**. The activation of this natural convection (due to high Rayleigh number of systems) happens so soon, and could not be detected even by precise measurement. Meybodi did not calculate the precise value of the onset time of our experiments and therefore, he concluded that (1) we used not accurate facilities in our experiments and (2) presented unjustifiable conclusion

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Table 1 Calculated onset time of convection based on linear stability analysis

Experiment number	Permeability (D)	Porosity (%)	Ra	t_c based on LSA (min)
1	2,546	34	14,866	0.018
2	555	33	2,552	0.48
3	121	32	798	6.86

of having only two stages in the mixing process! However, scientifically he could not reach to such a conclusion with his calculations.

Furthermore, it is very obvious and reported by many researchers that the first stage of diffusive mixing is omitted in high Rayleigh number systems. However, it always happens in the real porous media with smaller Rayleigh number. I think the author did not notice on the calculated Rayleigh number in his comments!

In addition, we measured the pressure and found two different trends in our experimental data and concluded that in the first part of the experiments, natural convection starts very soon and there is no pure diffusion mass transfer. Also we stated that in the later stage, there is a diffusion-dominated mass transfer (see the end of part 4.3 in our paper). It must be noted that, we did not claim about a real occurring process in reservoir during CCS. We just observed these periods in our experiments like pervious researchers (Farajzadeh 2009; Bahar and Liu 2008). On the other hand, we focused on the natural convection and quantification of this mechanism. We just used local Rayleigh number analysis to show the activation of the natural convection and it is not used for exact determination of onset of convection. However, we know that the local Rayleigh number analysis, which was given by Tan et al. long time ago, is just a ‘**quick and easy**’ approach to find the critical times. Obviously among widely different methods for determination of the critical times (Quasi-static analysis, local Rayleigh number analysis, energy stability analysis, amplitude theory) both the local Rayleigh number and the quasi-static theories are very definitely approximate and get a higher value (see Rees et al. 2008). Therefore, we reported the maximum critical time to insure the occurrence of the natural convection. However, it must happen sooner than reported critical time. In the following, the exact values of critical times are calculated based on linear stability analysis which was presented by Hassanzadeh et al. (2006):

$$t_c = 130 \left(\frac{\mu \phi}{Kg \Delta \rho} \right)^2 D.$$

The viscosity of water is calculated based on a correlation for aqueous NaCl solution presented by Kestin et al. (1981):

$$\mu^0(p, t, m) = \mu^0(t, m) + (1 + \beta(t, m)p)$$

where $\mu^0(t, m)$ and $\beta^0(t, m)$ are the hypothetical zero-pressure viscosity and pressure coefficient, respectively. The exact value of density difference is calculated using Garcia (2001), Spycher et al. (2003), Spycher and Pruess (2005), and Kestin et al. (1981).

Table 1 indicates that the natural convection can occur just at the start of the experiments and the first period of diffusive mixing omitted in our experiments.

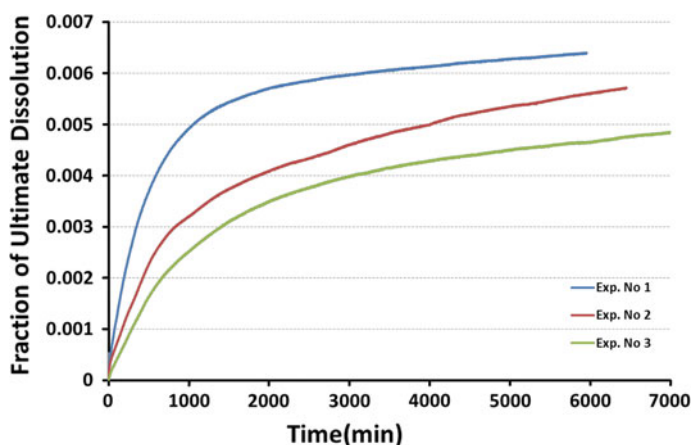
Emami in his comment claims that the behavior of the convective regimes in our experiments changed drastically with increasing Rayleigh numbers. We know that Kimura et al. (1986) used a pseudo-spectral numerical scheme to study time-dependent convection in a

Table 2 Rayleigh number for various transitions (Kimura et al. 1986)

Transition of regimes	Ra
Conduction to convection	$4\pi^2$
Steady convection to periodic	380–400
Periodic to Quasi periodic	500–520

Table 3 Duration time of experiments

Experiment number	Duration of the Experiment (min)
1	5,950
2	6,450
3	12,500

**Fig. 1** Fraction of ultimate dissolution versus time in the experiments in porous media

square box. They stated that with increasing Ra , the flow evolves from steady to non-periodic. The transitions presented by Kimura et al. (1986) are given in Table 2.

However, Kimura et al. (1989) in their earlier study had suggested that oscillations begin at $Ra = 575$. Based on this Rayleigh number, all our experiments are above the transition zone and therefore have a same condition from the convection regimes point of view.

In the last paragraph, the author expects to see the effect of the natural convection on the fraction of ultimate dissolution plots (Fig. 11 in our paper) and compared three graph of a, b and c with each other. But it must be noted that, this value for fraction of ultimate dissolution are reported in “Normalized Time” and it is obvious that, since each experiment has a different permeability and Rayleigh number, the final time of all experiments are not the same. Therefore, we cannot compare the fraction of ultimate dissolution in such a way. Table 3 shows the duration time of these three experiments. In addition, Figure 1 indicates the fraction of ultimate dissolution of the experiments in dimensional time. Now, we can compare the rate of mass transfer in all experiments. In original article, the main point of Figure 11, which is indicated in normalized time, was the comparison of the predicted result with the experimental data and it was not used for comparing all experimental data with each other.

References

- Bahar, M., Liu, K.: Measurement of the diffusion coefficient of CO₂ in formation water under reservoir conditions: implications for CO₂ storage. SPE 116513, Perth, 20–22 June 2008
- Farajzadeh, R.: Enhanced transport phenomena in CO₂ sequestration and CO₂ EOR. PhD Thesis, Delft University of Technology, Delft (2009)
- Garcia, J. E.: Density of aqueous solutions of CO₂. Berkeley, CA. Lawrence Berkeley National Laboratory, 10, LBNL-49023 (2001)
- Hassanzadeh, H., Pooladi-Darvish, M., Keith, D.W.: Stability of a fluid in a horizontal saturated porous layer: effect of non-linear concentration profile, initial, and boundary conditions. *Trans. Porous Media* **65**, 193–211 (2006)
- Hassanzadeh, H., Pooladi-Darvish, M., Elsharkawy, A.M., Keith, D.W., Leonenko, Y.: Predicting PVT data for CO₂–brine mixtures for black-oil simulation of CO₂ geological storage. *Int. J. Greenh. Gas Control* **2**(1), 65–77 (2008)
- Kestin, J., Khalifa, E., Correia, R.J.: Tables of the dynamic and kinematic viscosity of aqueous NaCl solutions in the temperature range 20–150°C and the pressure range 0.1–35 MPa. *J. Phys. Chem. Ref. Data* **10**(1), 71–87 (1981)
- Kimura, S., Schubert, G., Straus, J.M.: Route to chaos in porous-medium thermal convection. *J. Fluid Mech.* **166**, 305–324 (1986)
- Kimura, S., Schubert, G., Straus, J.M.: Time-dependent convection in a fluid saturated porous cube heated from below. *J. Fluid Mech.* **207**, 153–189 (1989)
- Meybodi, H.E.: Comments on the paper ‘quantification of density-driven natural convection for dissolution mechanism in CO₂ sequestration’ by R. Nazari Moghaddam et al. (2011). *Transp. Porous Med.* (2011, submitted)
- Rees, D.A.S., Selim A., Ennis-King J.P.: The instability of unsteady boundary layers in porous media. Emerging topics in heat and mass transfer in porous media, theory and applications of transport in porous media, 2008, 22, 85–110. doi:[10.1007/978-1-4020-8178-1-4](https://doi.org/10.1007/978-1-4020-8178-1-4) (2008)
- Sheikha, H., Mehrotra, A. K., Pooladi-Darvish, M.: An inverse solution methodology for estimating the diffusion coefficient of gases in Athabasca bitumen from pressure-decay data. *J. Pet. Sci. Eng.* **53**, 189–202 (2006)
- Spycher, N., Pruess, K.: CO₂–H₂O mixtures in the geological sequestration of CO₂. II. Partitioning in chloride brines at 12–100°C and up to 600 bar. *Geochim. Cosmochim. Acta* **69**(13), 3309–3320 (2005)
- Spycher, N., Pruess, K., Ennis-King, J.: CO₂–H₂O mixtures in the geological sequestration of CO₂. I. Assessment and calculation of mutual solubilities from 12 to 100°C and up to 600 bar. *Geochim. Cosmochim. Acta* **67**(16), 3015–3031 (2003)
- Zhang, Y. P., Hyndman, C. L., Maini, B. B.: Measurement of gas diffusivity in heavy oils. *J. Pet. Sci. Eng.* **25**, 37–47 (2000)