Noble Gas Partitioning in Two-Phase Flow

Kiran Sathaye, Toti Larson, Marc A. Hesse, Esben Pederson University of Texas at Austin Department of Geological Sciences Email: kiransathaye@utexas.edu

Summary

- Noble gas isotope concentrations are used as tracers and indicators of subsurface fluid flow and origin
- Motivated by the case of the Bravo Dome magmatic O_2 field in New Mexico, we study the enrichment ∂^{σ} of ²⁰Ne and ³He in a natural CO₂ injection process
- We observe qualitative agreement between theory, experiments, and field observations of noble gas enrichment during gas injection processes
- Volumes of radiogenic noble gases can be used to inform thermal and tectonic history of regional crust

Gas Injection Theory: Two Phases, N Components	
Total Volume Fraction of $\mathbf{i} : C_i = c_{i,gas}S_{gas} + K_ic_{i,gas}(1 - S_{gas})$	(1)
Fractional flow of gas phase: $f_{gas} = \frac{k_{gas}/\mu_{gas}}{k_{gas}/\mu_{gas} + k_{liq}/\mu_{liq}}$	(2)
Total Fractional Flow of $\mathbf{i}: F_i = c_{i,gas} f_{gas} + K_i c_{i,gas} (1 - f_{gas})$	(3)
1D Equation for Gas Injection: $\frac{\partial C_i}{\partial t} + \frac{\partial F_i}{\partial x} = 0$ $i = 1 \dots N - 1$	(4)
(N-1) Independent variables: $C_N = 1 - \sum_{1}^{N-1} C_i$	(5)

• K_i : dimensionless Henry's law partitioning coefficient

• $c_{i,qas}$: volume fraction of component *i* in the gas phase

Theoretical Composition Profiles: (Equation 4)

- Solution to equation (4) with boundary conditions for magmatic gas injection into subsurface aquifer
- Injected gas: 1% ³He + 99% CO₂ (Mantle input)
- Initial Brine: $99.5\% H_2O + 0.5\% ^{20}Ne$ (Meteoric)
- Volatile noble gases enriched at the front
- ²⁰Ne completely swept from CO₂ plume
- Mathematically describes two-stage gas enrichment model proposed by Gilfillan et al., 2008





- Column initially filled with H₂O saturated with CH₄
- Injection gas: 80% CO₂ and 20% Ar
- Dissolved CH₄ swept and enriched into first arrival bank
- Argon is 20X less soluble in water than CO₂
- Enriched Ar bank reaches the end of the column before CO₂
- Experimental results match theory for both initial dissolved gas (CH₄) and injected noble gas (Argon)
- Gas composition measured away from source can be very different due to fractionation during migration
- After arrival of trace gas banks, gas returns to input mixture



Noble Gas Volumes: Air-Saturated Water Contribution

- Indicates volume of gas interaction with air saturated water (ASW): Gas cap currently contains roughly 9km³ residual H₂O
- All ASW derived gases indicate ASW equilibration volumes of <0.2 residual water volumes - evidence of noble gas sweep
- Noble gas volumes not indicative of ASW volume contacted

Isotope	Total Mols	Non-Mantle Mols	ASW Required	S_{rw} Volumes
²⁰ Ne	6500	3100	$0.4 \mathrm{~km^3}$	0.05
³⁶ Ar	$1.1 (10^5)$	$7.7(10^4)$	$1.7 \mathrm{km^3}$	0.2
⁸⁴ Kr	410	250	0.16km ³	0.02

Noble Gas Sourcing

Observations: Bravo Dome Natural CO₂ Field

- matic noble gas isotope signature
- source and interaction with initial brine
- gas emplacement age between **1.2Ma** and **1.5Ma**



- ⁴⁰Ar produced by ⁴⁰K decay in mica, K-spar
- ${}^{40}\text{Ar}/{}^{36}\text{Ar}$ Ratio $\approx 3400 = 11 R_{atm} \rightarrow \text{large crustal component}$
- Non-mantle ${}^{4}\text{He}/{}^{40}\text{Ar} = 0.9$, ${}^{4}\text{He}/{}^{40}\text{Ar}$ Production Ratio = 5
- Significant excess ⁴⁰Ar in Bravo Dome gas





- Hydrocarbon composition and isotopes are used to distinguish between bacterial and thermogenic gas
- Experiment shows composition change during gas injection
- Degassing of bacterial methane form groundwater into migrating gas plume can change C isotope value of methane gas

References

- 1. M. Cassidy, Ocurrence and Origin of Free Carbon Dioxide in the Earth's crust. PhD Thesis, University of Houston, 2005.
- 2. S.M.V. Gilfillan, et al. "The noble gas geochemistry of natural CO₂ gas reservoirs from the Colorado Plateau and Rocky Mountain provinces, USA." Geochimica et Cosmochimica Acta 72(2008): 1174-1198.
- 3. F.M. Orr. *Theory of gas injection processes*. Tie-Line Publications, 2007.
- 4. S.G. Osborn., et al. "Methane contamination of drinking water accompanying gas-well drilling and hydraulic fracturing." proceedings of the National Academy of Sciences 108.20 (2011): 8172-8176.
- 5. K.J. Sathaye et al. "Constraints on the magnitude and rate of CO₂ dissolution at Bravo Dome natural gas field." Proceedings of the National Academy of Sciences, 2014: 201406076.