MRI Measurements of CO\textsubscript{2} Hydrate Formation and Dissociation in Porous Medium

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ABSTRACT

A series of experiments were carried out to study CO\textsubscript{2} hydrate formation and dissociation in porous medium using Magnetic Resonance Imaging (MRI). A new vessel was used to satisfy the demands of high pressure and no influence on the magnetic signal in the experiments. Images were obtained by using a fast spin-echo sequence, and field of view was set to 40×40×40 mm. The vessel pressure was recorded during CO\textsubscript{2} hydrate formation and dissociation, which was used to compare to MRI data. The results indicated that the MRI data could visualize hydrate formation and dissociation, and the MRI data was good agreement with the pressure change. The CO\textsubscript{2} hydrate formation rate was also quantified using MRI data.

KEY WORDS: CO\textsubscript{2}; hydrate; porous medium; MRI;

INTRODUCTION

The explosive growth of fossil fuels consumption has caused global problems, including air toxics and greenhouse gases (GHG). Carbon dioxide (CO\textsubscript{2}) is the largest contributor in regard of its amount present in the atmosphere contributing to 60 percent of global warming effects (Yang, Xu, Fan, Gupta, Slimane, Bland, and Wright, 2008). So sequestration of CO\textsubscript{2} has become an important issue, and scientists are exploring the option of sequestering CO\textsubscript{2} in the deep ocean (Servio and Englezos, 2001). Since the pressure and temperature conditions in ocean and in marine sediment may well be within the CO\textsubscript{2} hydrate stability zone (Kvamme, Graue, Buanes, Kuznetsova and Ersland, 2007), the CO\textsubscript{2} can be sequestrated in hydrate form. In order to obtain high quality data, the formation and dissociation of CO\textsubscript{2} hydrate in these instances have been widely studied.

MRI is an effective tool for investigations in physical, chemical, life, and clinical sciences as it non-invasively maps the water proton with high-space resolution in three-dimensions. Since its perfect performance for proton, MRI has been used for hydrate investigation. Baldwin et al. (2003) measured the formation and dissociation of tetrahydrofuran/water hydrate at ambient pressure using MRI. The formation and dissociation was monitored both in a THF/water-saturated Berea sandstone plug and in the bulk. The dissociation temperature of hydrate formed in the sandstone plug was not significantly different from the dissociation temperature in bulk. Having designed and constructed a high-pressure vessel to safely withstand 40MPa, Hirai et al. (2000) observed CO\textsubscript{2} hydrate growth in a water droplet injected into liquid CO\textsubscript{2} at 20MPa with the high pressure apparatus. Their experiments demonstrated not only the effective performance of the apparatus but also the perfect performance of MRI for hydrate investigations. Then they measured hydrate thickness growth with MRI (Hirai, Tabe, Kuwano, Ogawa and Okazaki, 2000). They concluded that CO\textsubscript{2} droplets dissolve during the process of sinking from their release point into Deep Ocean, by forming fine hydrate particles inside CO\textsubscript{2} droplets. The distribution of supercritical CO\textsubscript{2} injected into a packed bed of glass beads containing water was also directly visualized by them using MRI (Suekane, Soukawa, Iwatani, Tsushima and Hirai, 2005).

Kvamme et al. (2007) applied MRI to visualize the conversion of CH\textsubscript{4} hydrate within Bentheim sandstone matrix into the CO\textsubscript{2} hydrate. And then they carried a number of experiments to investigate the rates and mechanisms of hydrate formation in coarse-grain porous media. Many of these experiments were conducted in a sample holder fitted within a MRI instrument that allowed for a unique method of monitoring hydrate formation by the loss of signal intensity as water and free gas are converted into a solid phase. The rates and efficiency of the exchange process were reproducible over a series of initial conditions, with the notable observation that no free water was observed during the exchange process. Permeability measurements on hydrate saturated core indicate a finite level of permeability to gas, even in cores where all of the free water was converted to hydrate. Husebo et al. (2009); Ersland et al. (2009a, 2009b); Bernard et al. (2009) were also focused on the exchange of CO\textsubscript{2} for CH\textsubscript{4} in the hydrate using MRI.

Although lots of studies have been carried to investigate hydrate using MRI, there is little investigation on CO\textsubscript{2} hydrate in porous medium. In this study, CO\textsubscript{2} hydrate was formed and dissociated in porous medium using high pressure vessel. The experimental processes were recorded by MRI which can visually detect the processes. The pressure was also recorded to analysis the formation and dissociation of CO\textsubscript{2} hydrate.

EXPERIMENTAL INVESTIGATION

Experimental apparatus