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The effect of regenerated MEG on hydrate inhibition performance over multiple regeneration cycles

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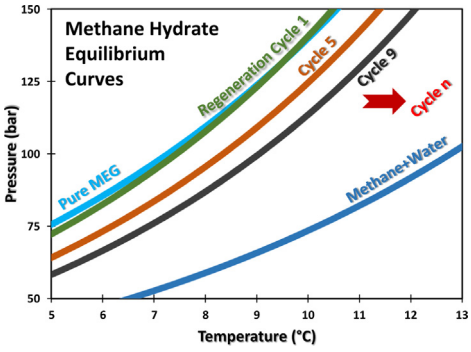
GRAPHICAL ABSTRACT



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ABSTRACT

Mono-ethylene glycol (MEG) is a favorable gas hydrate inhibitor mainly due to its recoverability through MEG regeneration facilities, and thus reducing costs. However, it is not clear how the hydrate inhibition performance of MEG is affected by multiple regeneration cycles. In this study, MEG samples that were regenerated and reclaimed over multiple cycles using an innovative bench-scale MEG pilot plant which can simulate field-like MEG operations, were assessed on their hydrate inhibition performance. The cycled MEG samples were carefully analyzed in the laboratory for their composition, and each sample was tested in a high-pressure sapphire cell for methane hydrate inhibition performance. The study found a directly proportional relationship between the number of cycles and the shift in hydrate equilibrium phase boundary. A maximum equilibrium shift of 2.21 °C was recorded for a 20 wt% MEG/deionized water sample that had experienced 9 regeneration cycles compared to pure MEG. The analysis suggests that the shift in hydrate equilibrium phase boundary was due to thermal degradation of MEG within the regeneration and reclamation units due to the presence of acetic acid. The study found that even though the operation was below MEG degradation temperature range, repeated heating of MEG may have caused its degradation. Additionally, the phase equilibria are empirically modeled as a function of the number of cycles to aid MEG end-users. Application of the model to experimental results provided accurate outcomes, and had an average relative difference of 1.24% when determining equilibrium temperatures.

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1. Introduction

An ongoing issue of concern in the field of flow assurance is the formation of gas hydrates in pipelines and process facilities. Gas hydrates can be the cause of serious damage to facilities, plugging in pipelines and even explosions near cornices [1–3]. Hydrates are classified as crystalline solids composed of host and guest molecules, or water and gas respectively. The water host forms a cage-like structure capturing gas molecules (such as carbon dioxide, methane, ethane and propane) within its cavities [4,5]. Hydrates, unlike ice, can form at a temperature higher than the ice formation temperature, and form when adequate water and gas molecules are present at high-pressure and low-temperature conditions which are typical sub-sea pipeline conditions [6]. Samimi (2012) has outlined various ways these conditions can be shifted to a hydrate-safe zone, either by depressurizing the pipeline, or through heating and thermal insulation, or to remove water through glycol dehydration [7]. Applying these techniques may not be suitable in all cases due to the lack of time, and economic constraints [8]. However, the conventional strategy the industry has adopted is to utilize chemical additives known as hydrate inhibitors to achieve hydrate inhibition. Methanol (MeOH) and mono-ethylene glycol (MEG) are common hydrate inhibitors, however, MEG is looked upon as more favorable due to its chemical stability, high regeneration efficiency, lesser environmental effect, and low solubility in final gas products [9].

An effective hydrate inhibition program requires a large quantity of MEG. This is troublesome if used-MEG is discarded into the environment, as well as costly to constantly replenish the MEG supply. The current best-practice is to recycle used-MEG and thus allowing the reuse of MEG. MEG recycling involves two critical processes, regeneration and reclamation. Regeneration, also known as re-concentration, utilizes distillation to re-concentrate MEG by removing the water present in the used-MEG/rich-MEG stream; the rich-MEG solution may be contaminated with chemical additives such as corrosion and scale inhibitors, drilling mud, and formation water. The deposition of these chemicals and precipitation build-up in process equipment can lead to equipment fouling, downtime in production, concern in safety, and economic losses due to maintenance [10]. The regenerated MEG is then pumped through to the reclamation unit, where the solution is thermally exposed under vacuum conditions to the vaporization temperature of MEG. This allows for the recovery of MEG and water whilst removing the contaminants as waste products [10].

Regeneration of MEG is a cost-effective strategy and has been the subject of numerous research with regards to design and process, but there is very little or no research into how the inhibition performance of MEG is affected by multiple regeneration/reclamation cycles. In this study, recycled MEG samples from an experiment simulating the switching between corrosion management strategies using a fully functional bench-scale MEG regeneration/reclamation plant was evaluated on their hydrate inhibition performance using a PVT sapphire cell (Fig. 1). The recycled MEG samples from a total of 9 consecutive cycles, were tested in the PVT cell to determine whether the number of cycles have an impact on the hydrate inhibition performance. The results of this study give rise to a whole new aspect of MEG recyling, and allowing users to take the necessary steps to ensure minimal loss by adequately adjusting MEG injection rates.

Furthermore, one of the purposes of this communication is to present a model that accurately depicts this new information of experimental hydrate equilibria data. Presenting this research's experimental data in the form of a model is not only more convenient but ensures it is more accessible to industry and research personnel. A balance between simplicity and ease of use was the desired outcome for this model and it is based on mathematically interpolating (linearly) experimental pressure (P)-temperature (T) hydrate equilibria for a specified MEG cycle number, n. Exponential functions are chosen as the data-fitting equations since hydrate pressure-temperature equilibria correlate very well when described exponentially and the exponential data-fitting equation just has one term with only one occurrence of P and T [11,12].

2. Methodology

2.1. Materials and equipment

As the MEG regeneration and reclamation process becomes increasingly complex, the complexity increases in terms of experimentation in the laboratory. An innovative approach is the bench-scale MEG pilot plant housed in the Curtin Corrosion Engineering Industry Centre (CCEIC). The bench-scale pilot plant is designed and built for thorough experimentation and study of the behavior of MEG in different field scenarios, in combination with production fluids, drilling mud and other chemical additives. The pilot plant has a real-time processing flow of up to 4 kg/h of lean-MEG. The plant comprises of four distinct yet related stages; a) preparation of brine, b) preparation of rich or contaminated MEG, c) reconcentration/regeneration unit, and d) reclamation unit. In this study, samples of reclaimed MEG from the reclamation unit were extracted to be evaluated on their hydrate inhibition performance.

A PVT sapphire cell (Fig. 1) located in the Clean Gas Technologies Australia (CGTA) laboratory was used as the experimental apparatus for testing the samples for hydrate inhibition performance. The essential process of the cell was to provide steady heating and cooling in a controlled environment. The cell is made from strong sapphire material that is able to sustain increased pressures allowing it to operate at a maximum pressure of 500 bar. To ensure there was no contamination, a ventilation and purging line was connected to the sapphire cell which allowed for gas to be released to a safe atmospheric zone above the building. The total volume contained within the system inclusive of the cell (60 cm^3) and tubing (26 cm^3) is 86 cm^3. Furthermore, the cell was insulated firmly from the outside surroundings by a tightly sealed accessible door with a window allowing for easy visual observations of the entire cell from the outside as well as through a camera system. The heating and cooling capability is within a temperature range of 60 °C to as low as -160 °C. A compressor was utilized for cooling, whilst for heating an integrated electrical heater within the PVT cell was utilized, and an external chiller was used to supply chilled water to enhance compressor performance. The air bath chamber wherein the cell was securely fixed has a fan mounted on the roof for enhanced circulation of cool or heated air. Specific to this study, the cell was operated at a pressure and temperature range of 50–200 bar and 0–30 °C respectively.

The cell was fitted with a magnetic stirrer to provide sufficient mixing between the gas and liquid. It also helps in the promotion of gas hydrate formation due to the disturbance it creates at the surface of the solution [13,14]. A lack of this disturbance leads to hydrate merely forming at the surface which prevents additional gas molecules from dissolving and hence severely delaying hydrate formation [15]. The magnetic stirrer (diameter of 2 cm) was operated at ~500 rpm during each run of the experiment. The gas was pressurized via a piston pump controlled by ABB Mint Workbench software (Build 5712). Pressure sensors (accuracy of ± 0.5 bar) linked to a proportional integral derivative (PID) controller which allows for pressure adjustment. The temperatures of the air bath chamber, vapor and liquid phases in the cell were monitored via multiple K-type thermocouples (accuracy of ± 0.03 °C). The Falcon application (version 4.30) was used to monitor and control temperature changes. Care was taken to ensure temperature changes were small (1 °C/h) in order to achieve steady state between the various phases within the cell. The heating and cooling system, piston pump and cameras were all controlled and maintained via the PVT computer system.

Pure mono-ethylene glycol (MEG) was sourced from Chem-Supply with a purity of 99.477 mol% (Table 1). Pure MDEA (purity ≥ 99 mol %) was sourced from Sigma-Aldrich, whilst FFCI is a proprietary film forming corrosion inhibitor (FFCI).

Analysis of the sample composition, ion concentrations and acids were determined using a ion chromatography and high performance liquid chromatography (HPLC) system (Thermo Scientific Dionex

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