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Effectiveness of aluminium based coagulants for pre-treatment of coal seam water

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ABSTRACT

Brackish coal seam water produced from the coal seam gas (CSG) industry is often treated by reverse osmosis to make the water suitable for beneficial reuse. As such, pre-treatment technologies are required in order to minimise membrane fouling. It was our hypothesis that coagulants may be able to remove problematic dissolved ions such as alkaline earth ions and soluble silicate species which are responsible for scaling of membranes and equipment. Consequently, this study evaluated the performance of aluminium chlorohydrate and aluminium sulphate coagulants for both simulated coal seam water and water collected from operating CSG wells. For simulated coal seam water samples the degree of dissolved silica removal was generally high (>85%) and promoted by increasing water salinity. Aluminium sulphate was better at removing silica (*c.f.* 94–92%), barium (*c.f.* 87–20%), strontium (*c.f.* 66–15%) and magnesium (*c.f.* 16–7.5%) and worse at removing calcium (44–65%) compared to aluminium chlorohydrate from simulated coal seam water, respectively. Variation in coal seam water composition of relatively concentrated solutions detrimentally impacted alkaline earth ion removal and in some instances reduced the extent of dissolved silica removal. When coal seam water samples for operating wells were studied, the amount of aluminium chlorohydrate required was significantly increased and the silica removal process inhibited (50% compared to 92% for simulated CS water). It was proposed that the presence of organic species in the real coal seam water played an important role in the mode by which the coagulants operated. Future studies should focus on understanding in more detail the influence of organic matter and influence of dissolved cations and anions upon coagulant behaviour.

1. Introduction

Coal seam gas (CSG) exploration has accelerated in recent years and will continue to be an important part of the world's energy portfolio in the future [1]. Australia is a major gas CSG producer, with production mainly located in the Surat and Bowen basins of Queensland [2–4]. The gas extraction process from the coal seams involves the release of hydraulic pressure which allows the gas to flow. Consequently, the resultant coal seam (CS) water, which is typically saline in character, requires treatment to facilitate beneficial reuse [4–6]. Major components present in the coal seam water include sodium chloride and sodium bicarbonate with lesser quantities of alkaline earth, potassium, sulphate and fluoride ions also detected [6,7]. Reverse osmosis (RO) desalination plants have been installed as part of an overall strategy to manage the coal seam water [8]. Reverse osmosis systems require comprehensive pre-treatment of the incoming feedwater in order to minimise detrimental issues such as membrane fouling, which can reduce water recovery rates [9]. Generic approaches for water pre-treatment include both coarse and fine filtration stages [10–12], followed by a softening step such as that involving ion exchange resins [13,14].

With respect to the coarse filtration stage, both dissolved air flotation (DAF) and microsand ballasted flocculation are both prospective candidates for use [6,15]. The essential components of a dissolved air flotation system are: a rapid mixing stage where coagulant is added, a flocculation step; dissolved air flotation stage wherein air bubbles have been added *via* a saturator; and, a final filtration operation [16]. Dissolved air flotation has already found extensive application for wastewater treatment [16,17]. For instance, Hami et al. [18] evaluated a DAF unit wherein activated carbon was also added in order to promote removal of BOD and COD. Alternatively, Chen et al. [19] reported the efficient control

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of algae species in surface water by DAF. Veolia market microsand ballasted flocculation under the trade name ACTIFLO, and claim that it enhances the settling behaviour of suspended solids and allows creation of compact clarification units [20]. ACTIFLO has been employed for a wide variety of water treatment applications such as seawater desalination [21,22], drinking water purification [23] and refinery water treatment [24]. The basis of the technology is the combination of chemical coagulation, microsand addition and lamella settling [20].

Both the microsand ballasted flocculation and DAF processes rely on coagulant for destabilisation of colloidal matters and suspended particulates [25]. Commonly, two categories of coagulants are used in industry: inorganic and organic materials. Due to concerns with fouling of downstream membranes, the use of organic polymer coagulants is often not desired [26]. With respect to inorganic coagulants, the most common variants are either aluminium or iron based. Aluminium based examples include aluminium sulphate, aluminium chloride, sodium aluminate, aluminium chlorohydrate and polyaluminium chloride [27]. The iron based coagulants typically encompass ferric sulphate, ferrous sulphate, and ferric chloride.

Aluminium sulphate (alum) is one of the most studied coagulants due to its ready availability and relatively low cost. For example, Guida et al. [28] studied the impact of alum dose rate and solution pH upon the control of COD and suspended solids present in municipal wastewater. Similarly, Amuda and Alade [29] investigated the removal of COD, suspended solids and phosphate from abattoir wastewater and evaluated the performance of alum against a range of iron based coagulants. It was discovered that alum outperformed the other coagulants tested for reduction in the levels of total suspended solids and phosphate. In contrast, Yuheng et al. [30] compared the use of aluminium sulphate and polyaluminium chloride (PACl) to treat algae contaminated tap water and discovered that larger dose rates of alum were required compared to PAC. Ghafari et al. [31] confirmed the slight superiority of PACl compared to alum for leachate treatment, albeit alum was much better for COD elimination. These latter observations may relate to the study of Wu et al. [32] which identified different coagulation mechanisms for alum and polyaluminium chloride. It was concluded that charge neutralization and sweep flocculation were involved when alum was used and charge neutralization, polycation-patch coagulation for PACl. Aluminium chlorohydrate (ACH) is a modern coagulant which has a high aluminium content ($Al_2(OH)_5Cl$). ACH has been demonstrated by Ho et al. [33] to have some ability to remove silica species from RO brine. Yonge et al. [34] compared a range of inorganic coagulants for dissolved organic carbon (DOC) removal from wastewater and found that ACH had the highest performance of the aluminium based materials. Sadrnourmohamadi and Gorczyca [35] confirmed the superiority of ACH compared to alum or PACl for DOC removal from wastewater especially at higher pH values. The enhanced performance was correlated with the greater abundance of monomeric and medium polymeric aluminium species in solution.

Despite the proliferation of the coal seam gas industry, there appears to be minimal information published regarding the applicability of coagulants to coal seam water. It was our hypothesis that aluminium based coagulants may be useful in not only removing particulate matter present but also dissolved ions from coal seam water. The latter idea was based upon evidence that coagulants can remove substantial amounts of heavy metal ions from solution [36] and the observation that silica could be partially removed from brines [33]. The question arises if scale forming species such as silica, calcium, magnesium, strontium and barium can be removed from CS water by coagulation. In particular, this study was designed to determine the optimal dose of aluminium based coagulants such as aluminium sulphate and aluminium chlorohydrate (ACH). The impact of coal seam gas salinity and composition was also evaluated in relation to which species and to what extent were removed. Jar testing was the main approach used to determine the coagulant performance and both simulated and actual coal seam water samples were studied.

2. Materials and methods

2.1. Coagulants and chemicals

Two coagulants were used in this study, aluminium sulphate (alum) supplied by Omega Chemicals and aluminium chlorohydrate (ACH) supplied by Hardman Chemicals. The alum was of general formula $Al_2(SO4)_3\cdot 14H_2O$ and the ACH was $Al_2(OH)_5Cl$, and the content of each species per L of coagulant solution was 46.98 and 47.91%, respectively; which corresponded to 4.27% Al in alum solution and 14.8% al in ACH solution.

Simulated CS water solutions were made by addition of appropriate quantities of salt to purified water. The following salts were used: sodium chloride; sodium bicarbonate; potassium chloride; calcium chloride; magnesium chloride; barium chloride; strontium chloride; sodium fluoride; boric acid; potassium silicate and sodium sulphate. Chemicals were all of analytical grade or equivalent and supplied by both Sigma Aldrich and Rowe Scientific.

The CS water samples with different concentration but same ratio of elements tested had the compositions shown in Table 1. Similarly, the CS water samples with varying composition are illustrated in Table 2.

Simulated CS water solutions were used in order to allow precise control of the solution compositions in order to design experiments which explored the typical range of CS water concentrations found in the CSG industry [6]. The limited availability of real CS waters from operating gas fields necessitated the latter approach in order to elucidate the impact of variable CS water composition and salinity. In addition, as the main aim of this study was to investigate the ability of coagulants to remove dissolved ions from solution instead of the conventional approach to coagulation for removing suspended solids [37], a solution free of turbidity causing species was required in order to investigate the impact of these latter species upon coagulant performance.

2.2. Jar testing procedures

A bench top Platypus Jar Tester was used to carry out the coagulation and sedimentation experiments involving CS water. All jar testing experiments were performed using 2 L volume, square jars and large 35×80 mm impeller paddles. The standard procedure used for each experiment in the jar tests involved 10 s of premixing, followed by 1 min of rapid mixing at 190 rpm and then 10 min mixing at 50 rpm in order to encourage flocculation. The solution was then allowed to settle for 30 min before a small sample was taken for analysis. This latter approach was based upon accepted practices for performing coagulation wherein a rapid mixing, maturation and settling stage is present [38,39].

2.3. Water analysis

2.3.1. Inductively coupled plasma-optical emission spectroscopy (ICP-OES)

A PerkinElmer Optima 8300 DV Inducted Coupled Plasma - Optical Emission Spectrometer was used to analyse a range of ions in solution. Samples were acidified to pH 2 by adding the 100 μL of 70% nitric acid which was purified using a Savillex DST-1000 distillation unit. A multi-element standard supplied by Australian