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Transformation of planar Mögel Al_{13} coagulant during the dilution and aging process

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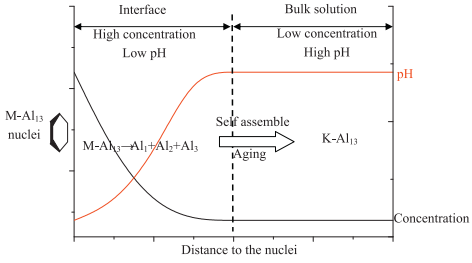
HIGHLIGHTS

- The Mögel Al_{13} would decompose into oligomeric species at high concentration.
- Mögel Al_{13} would transform to Keggin Al_{13} at lower concentration.
- Transformation of Mögel Al_{13} to Keggin Al_{13} is a time dependent reaction.
- More Mögel Al_{13} transformed to Keggin Al_{13} under longer aging period.
- The concentration profile is used to interpret transformation process.

GRAPHICAL ABSTRACT



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ABSTRACT

Transformation of a novel planar Mögel Al_{13} ($Al_{13}(OH)_{24}(H_2O)_{24}^{15+}$, simplified as M-$Al_{13}$) coagulant in the dilution and aging process was investigated with Ferron colorimetric method, electrospray ionization mass spectrometry (ESI-MS) and ^{27}Al NMR spectroscopy. Special attention has been paid to the transformation process of M-Al_{13} to stereo Keggin Al_{13} (ε-$AlO_4Al_{12}(OH)_{24}(H_2O)_{12}^{7+}$, simplified as K-$Al_{13}$). M-$Al_{13}$ was found to be an unstable aluminum species and could be decomposed into oligomers as Al_1, Al_2 and Al_3 when dissolved in solution. These newly formed oligomers were also found to polymerize into K-Al_{13} by self-assembly during the dilution process. More K-Al_{13} species could be continuously formed at the initial aging period of the diluted solution. On the whole, the hydrolysis and aging behavior of planar M-Al_{13} in aqueous solution is quite different from those of the K-Al_{13} species, which may provide insight into the structure re-organization mechanism of polymeric Al species in solution.

1. Introduction

Polyaluminum chloride (PACl) is wildly used as an effective coagulant in water purification. The performance of coagulation was proved to be closely related to the distribution of aqueous aluminum (Al) species in water [1–3]. Commercial PACl generally are composed of Al species as monomers, K-Al_{13} ($AlO_4Al_{12}(OH)_{24}(H_2O)_{12}^{7+}$) and uncharacterized oligomers or colloidal species. Among these components, K-Al_{13} has often been claimed to be the critical species with higher charges, higher molecular weight, and thus contributed to the higher performance of PACl than traditional monomeric Al coagulants [4–6]. Consequently, the stability and hydrolysis behavior of K-Al_{13} has been the focus of researches in the past several decades.

More recently, another Al_{13} species with planar structure (i.e., M-Al_{13}, $Al_{13}(OH)_{24}(H_2O)_{24}^{+15}$) has also been proved to be an efficient coagulant [7]. With the same polymerize degree with K-Al_{13}, M-Al_{13} with 15 positive charges might also show higher destabilization ability. Although M-Al_{13} has attracted increasing attention as a novel class of inorganic coagulant, the stability and hydrolysis of M-Al_{13} in aqueous solution has not been studied yet. While the overwhelming majority of studies on hydrolysis of Al species were based on K-Al_{13} [8–10], the hydrolysis behavior of M-Al_{13} is a problem to be clarified.

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