



International Conference on Ecology and new Building materials and products, ICEBMP 2016

Inhibition of hydrogen evolution in aluminium-phosphate refractory binders

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Abstract

The study deals with the possibilities of inhibition of the reaction of aluminium-phosphate binders with iron scale, which is contained in bauxite. This material is mixed with phosphate binders to form refractories. The hydrogen is produced by this reaction causing the mixture foaming. The amount of evolved hydrogen was observed by the measuring by the Jank's calcimeter. Also the shape of oxidation-reduction potential of the binders was measured before and after mixing with aggregate using potentiometer. The influence of the addition of various cations on the hydrogen evolution and on the oxidation-reduction potential was observed. The effectiveness of the inhibitors was studied also visually by forming test elements.

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Peer-review under responsibility of the organizing committee of ICEBMP 2016

Keywords: Phosphate binder; bauxite; refractories; wear iron; inhibition; calcimeter; potentiometer

1. Introduction

High alumina ceramics are preferred materials for number of reasons. Their strength is valuable for high load bearing applications, and they are resistant to corrosion in high temperature environments such as steam and CO atmospheres. Alumina ceramics are also well known for their low electrical and thermal conductivity. Therefore they are most useful materials in refractory bricks and electrical insulating components. Because of their technological importance, their low-temperature processing by chemical bonding has considerable technological significance. Phosphate bonded alumina ceramics consist of particles, whose surfaces are coated with berlinite

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(AlPO₄), a crystalline orthophosphate. The bonding phase AlPO₄ is formed by the reaction between the phosphoric acid and alumina. This phase is a solid formed by covalent network of oxygen-bridged alternating PO₄ and AlO₄ tetrahedra. This structure is isomorphous with that of various forms of silica. Consequently, aluminium phosphate shares many chemical and physical properties of silica, yet AlPO₄ is formed at a much lower temperature. This lower temperature formation of AlPO₄, possibly with less internal stresses in it that sintered counterparts, may translate into an economic advantage in producing ceramics suitable for high temperature service at low processing costs. [1]

Aluminium phosphate ceramics is often prepared by using an excess of acid because the solubility of alumina in phosphoric acid is highest in ultra-acidic mediums (pH<2) [1]. The abundant acid reacts with wear iron, which is contained in refractory filler (e.g. bauxite) which was milled in steel ball mills. Hydrogen is produced by this reaction, which negatively affects the shape of final products.

Most well-known acid inhibitors are organic compounds containing nitrogen, sulfur, and oxygen atoms and multiple bonds or aromatic rings, which allow an adsorption on the metal surface through lone pairs of electrons and/or pi electrons present in these molecules [2,3]. The inhibition efficiency increases in the order O < N < S < P [3]. The mechanism of the adsorption mainly follows Langmuir adsorption isotherm [2,3,4,5,6,7] or Temkin adsorption isotherm [8,9]. Some nitrogen containing heterocyclic compounds adsorb by the mechanism of simple chemisorption [10,11]. Extracts of some plants, e.g. Zenthoxylum alatum [12] or Coriandrum sativum [13], also showed themselves as good inhibitors.

Another possible method of inhibition consists in an addition of inorganic ions with positive standard electrode potential. The relation between standard electrode potential (E₀) and reduction potential of a half-cell (E) is described by Nernst equation (Eq. 1):

$$E = E^0 - \frac{RT}{|z|F} \ln \frac{a_C^c \cdot a_D^d}{a_A^a \cdot a_B^b} \quad (1)$$

where R is the universal gas constant (R = 8,314 J·K⁻¹·mol⁻¹); T is absolute temperature; z is the number of moles of electron transferred in the cell reaction of half-reaction; F is the Faraday constant, the number of Coulombs per mole of electrons (F = 9,649·10⁴ C·mol⁻¹) and a are activities of products (C, D) and reactants (A, B) raised to a higher power by their stoichiometric coefficients (a, b, c, d,) [14].

This paper is focused on a comparison of inhibiting activity of Cu²⁺, Ag⁺, Bi³⁺ and Sb³⁺ cations which were added in a form of suitable compounds (chlorides, nitrates, and citrates).

2. Materials and methods

2.1 Materials

First of all the phosphate binder was prepared. The solution of phosphoric acid was prepared by mixing 300 cm³ of H₃PO₄ with 150 cm³ of distilled water. Then 114.1 g of Al(OH)₃ was dissolved in this solution. Finally, additional 5 cm³ of an acid was added. The product was diluted with distilled water to the final volume of 800 cm³ and filtered through Büchner funnel. The filtrate was poured into a volumetric flask and diluted with distilled water to 1 dm³.

It was necessary to choose inhibitors with both cation and anion having positive standard electrode potential. For this study CuCl₂, Cu(NO₃)₂, Cu₃(C₆H₅O₇)₂ (copper(II) citrate), AgCl, AgNO₃, Ag₃C₆H₅O₇, BiCl₃, Bi(NO₃)₃, BiC₆H₅O₇ and SbCl₃ were chosen as inhibitors. Some inhibitors (AgCl, BiCl₃ and SbCl₃) needed to be prepared in laboratory (Eq. 2, 3, 4):

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