Weathering mechanisms of municipal solid waste incineration air pollution control residues: A state of the art review

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1. Introduction

Municipal solid waste incineration fly ash and air pollution control (APC) residues, hereafter referred as APC residues, contain high level of soluble salts (e.g. chlorides), heavy metals (e.g. Hg, Cd, Pb, etc.) and persistent organic pollutants (PHAs, dioxins, etc.) and, thus, they are considered as hazardous waste. Recent research on weathering or aging of fly ash has demonstrated that this treatment can possibly decrease the leaching of heavy metals, making it suitable for either landfilling or reusing. However, there are still several issues regarding this technology that need to be clarified, such as the optimum conditions for heavy metal immobilization and the mechanisms involved. Thus, this paper aims to report on the state of the art research on the weathering of APC residues that are currently available in the literature and discuss the topics that need to be addressed in the future.

2. Weathering of air pollution control (APC) residues

Fly ash is a highly unstable and reactive material. As such, it may be subjected to a natural or induced mineralogical alteration called weathering or aging. Weathering occurs as a consequence of several factors such as pH, redox potential, temperature, humidity and the concentration of certain components, such as CO₂ (Sabbas et al., 2003). This process leads to mineralogical, physical and chemical changes over time which may impact the leaching behavior of trace elements present in APC residues. Due to the presence of lime, APC residues are highly alkaline with pH values around 12. Carbonation, which is one of the mechanisms involved in the weathering process, is the responsible for a pH decrease to 7 – 10 (Li et al., 2007). Together with this decrease in pH, immobilization or release mechanisms of elements present in APC residues takes place. A series of the following equations were proposed by different authors in order to understand this phenomenon:

Hydrolysis of species containing Ca, Al, etc. starts immediately due to water input (Sabbas et al., 2003):

\[ \text{CaO + H}_2\text{O} \rightarrow \text{Ca(OH)}_2, \text{or Al}_2\text{O}_3 + 3\text{H}_2\text{O} \rightarrow 2\text{Al(OH)}_3 \]

As a consequence of hydration, Ca and Al containing phases can also dissolve and other secondary minerals such as ettringite can be formed (Meima and Comans, 1997):

\[ 6\text{Ca}^{2+} + 2\text{Al}^{3+} + 3\text{SO}_4^{2-} + 38\text{H}_2\text{O} \rightarrow 12\text{H}^{+} + \text{Ca}_6\text{Al}_2(\text{SO}_4)_3(\text{OH})_{12} \cdot 26\text{H}_2\text{O} \]

The reaction can go further when the initially alkaline APC residues take up CO₂ from the atmosphere. During this process, the pH decreases due to the formation of minerals such as calcite:

\[ \text{Ca(OH)}_2 + \text{CO}_2 \rightarrow \text{CaCO}_3 + \text{H}_2\text{O} \]

Supporting this hypothesis are the results of accelerated carbonation experiments and subsequent mineralogical analysis using XRD reported by several research groups (Li et al., 2007), which showed the disappearance of portlandite (calcium
hydroxide) and the formation of calcite. Studies regarding the factors affecting carbonation were also reported in the literature. Ecke (2003) studied carbonation factors such as partial pressure of CO$_2$, addition of water, temperature and reaction time and concluded that the concentration of CO$_2$ in the gas phase has the greatest impact followed by the reaction time, temperature and addition of water. Furthermore, Li et al. (2007) reported that water to solid (W/S) ratio and temperature had major influence on carbonation. This is because water is the reaction medium for dissolution, ionization and CO$_2$ transportation, with high temperatures increasing the reaction velocity. However, the authors mentioned that high temperatures do not imply high CO$_2$ sequestration and, thus, they concluded that optimum conditions for carbonation reactions take place at room temperature with W/S ratio of 0.3.

Research on the immobilization of critical heavy metals in response to carbonation was also reported by several authors. Wang et al. (2010), for instance, found that the release of Pb, Zn and Cu decreased significantly, while the release of As decreased slightly and the release of Cd and Sb increased as carbonation process evolved. They concluded that pH and liquid to solid ratio (L/S) were the main factor that impact metal mobility or leaching, the optimum final pH being between 9.5 to 10.5 and the optimum L/S ratio of the fly ash being 0.25. Furthermore, they found that carbonation does not decrease the leaching of soluble salts such as chlorides, sulfates or fluorides. Thus, they suggested the use of carbonation as a pre-treatment method for heavy metals and mentioned that washing or other treatments should be considered to remove soluble salts.

A relationship between the formation of new mineral phases due to weathering and the immobilization of certain species was reported by Bayuseno and Schmahl (2011) and Cornelis et al. (2008). They found that Pb, Zn and Mn may be absorbed by hydroxides and carbonates. For instance, the sorption of Cd and Pb was controlled by the neo-formed iron-(hydr)oxides such as lepidocrocite. Besides, the formation of ettringite-type structures, in which SO$_4^{2-}$ can be replaced by AsO$_4^{3-}$, CrO$_4^{2-}$, MoO$_4^{2-}$, Sb(OH)$_6$$^-$, and VO$_4^{3-}$, has been reported for alkaline environments, which explains the immobilization of oxyanions. Moreover, they found that hydrocalumite, which is a mineral more stable than ettringite at high pH values, has an important role in the immobilization of oxyanions.

3. Further research

As part of further research, the impact of redox potential on the mobilization of Cd, Sb and Cr need to be studied because their leaching are likely controlled by redox reactions. Furthermore, long term stability of heavy metals immobilized in the neo-formed minerals due to weathering need to be assessed.

4. References


