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Chemical Stabilization of Coal Fly Ash for Simultaneous Suppressing of As, B, and Se Leaching

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Additional information is available at the end of the chapter

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Abstract

The discard of coal fly ash produced from the combustion of pulverized coal in a coal-fired boiler of thermal power plants has led to environmental concerns. Due to the interaction of fly ash particles with weathering and hydrological processes, the rainfall leaches out toxic elements in coal fly ash from the ash heaps. This situation has been pointed out as a potential contamination of soil, surface, and groundwater. In this chapter, the available fly ash treatment techniques to minimize future release of toxic trace elements (arsenic, boron, and selenium) have been documented, and the recent investigations dealing with leaching suppression effect of arsenic, boron, and selenium from coal fly ash have been reviewed. The leaching characteristics of arsenic, boron, and selenium are discussed, and a simple and low-cost leaching control method is presented in the context of treating the fly ash through chemical stabilization technique using additives containing high levels of calcium. Experimental results described in this chapter show the chemical stabilization technique utilizing Ca-containing additives is an effective technique for simultaneous suppressing of As, B, and Se leaching from coal fly ash.

Keywords: chemical stabilization technique, coal fly ash, leaching, calcium, arsenic, boron, selenium

1. Introduction

Coal fly ash from coal burning power generation is one of the major sources of environmental pollution due to the discharge of large amounts of fly ash into the environment. Coal fly ash has been utilized in different ways such as a substitute material for Portland cement, structural fills (usually for road construction), soil stabilization, and mineral filler in asphaltic concrete because of its physical (selfhardening) properties [1, 2], and in recent years, coal fly ash has

been utilized as a potential material to treat acid mine drainage (AMD) because of its chemical (high degree of alkalinity) properties to neutralize AMD [3–8]. However, most of the fly ash is disposed in landfills [1], this disposal involves the interaction of fly ash particles with weathering and hydrological processes where rainfall leaches out toxic elements, anions, and cations from the ash heaps, which pose an environmental hazard through contamination of soil, surface, and groundwater [9–11]. During combustion, the organic matter in coal is destroyed, and as a result, the concentrations of trace elements are increased relative to the source coal. Among the elements that can be leached from fly ash, Ag, As, B, Ba, Cd, Co, Cr, Cu, Hg, Ni, Pb, Se, Sb, Sn, and Zn are of the greatest concern [12, 13] as environmental hazard. The leaching of trace elements such as As, B, and Se from coal fly ash (CFA) is likely to occur as these elements tend to form hydrophilic oxides that are dissolved as oxyanion forms [10]. The beneficial reuse of fly ash as a potential material to treat acid mine drainage (AMD) has great potential in minimizing the amount of disposed fly ash [3–8]; however, since the coal fly ash itself contain leachable trace elements and the usage of fly ash in treating AMD could lead to trace element accumulation with negative consequences to the environment [14], it is particularly important to be able to assess the leachability of coal fly ash (since the results can determine not only whether the ash is environmentally acceptable for use as the soil supplements, construction material, or neutralization material to treat AMD but to extend of isolation that might be required for disposal in landfill) and to treat the coal fly ash before the utilization to avoid the trace elements leaching into the environment.

To predict the possible effect of coal fly ash on the environment, it is particularly important to understand the factors that control the leaching behavior of trace elements in coal fly ash. Research studies on the leaching behavior of As, B, and Se in coal fly ash have been carried out with the promising results. The results demonstrated that the leaching behavior of arsenic, boron, and selenium from fly ash was affected by pH, solid-to-liquid ratio, leaching time, and ash type [15–19]. The leaching of As increased with increasing pH values in acidic fly ashes [19] and increased with decreasing pH values in alkaline fly ashes [20], while the leaching of B decreased with increasing pH values [18, 21] and the leaching of Se tends to decrease as pH was raised for an alkaline ash with high Ca composition [18]. The leaching of As, B, and Se from CFA generally increased with increases in the S/L ratio and leaching time, and adsorption/desorption played a major role in As and Se leaching from the CFA [22]. The leaching of As and Se from acidic ashes could be described by sorption of iron oxide, while the leaching from the alkaline ashes appeared to be controlled by sorption in the alkaline calcium phase [20, 23]. The presence of Ca in fly ash plays an important role in the leaching behavior of As, B, and Se, in which the leaching of As, B, and Se may involve the trapping of As, B, and Se species by the ettringite phase ($3\text{CaO}\cdot\text{Al}_2\text{O}_3\cdot3\text{CaSO}_4\cdot32\text{H}_2\text{O}$), leading to a decrease in leaching under alkaline conditions [18, 19, 24–28]. Therefore, Ca content and the sorption process are known to play important roles in the release of As, B, and Se from CFA.

Different treatments and stabilization processes of fly ash have been proposed by the scientific community. The most common used techniques for removing toxic elements from fly ash and APC (air pollution control) residue are (1) extraction and separation, (2) chemical stabilization, (3) solidification, and (4) thermal treatment [29]. Several studies have chemically treated fly ash produced in power plants to immobilize the toxic trace elements before

disposal. These studies used chemicals such as calcium aluminates [30], phosphoric acid [31], NaOH [32], and chelating surfactants such as ED3A [33]. Arsenic trioxide contaminated soil was successfully treated with a ferrous sulfate solution to promote the formation of insoluble metal-bearing phases [34] and the coal fly ash treatment with ferrous sulfate shown to be an effective method for the sequestration of As, B, Cr, Mo, Se, and V associated with coal fly ash, where the mobility of As, B, Cr, Mo, Se, and V were substantially reduced [35]. Recently, the coal fly ash chemical treatment using other waste sources containing high level of calcium has been applied [27]. The advantage of this technique is that it can be used for reducing As, B, and Se leaching at low cost with the abundant chemical available, since it utilizes wastes from other industry. The aims of this chapter are to review what is known about the factors which control the leaching of As, B, and Se in fly ashes and the method to reduce As, B, and Se leaching from fly ash. Experimental results of simultaneously leaching suppression of As, B, and Se were described in this chapter.

2. Coal fly ash and overview of leaching characteristics of arsenic, boron, and selenium from coal fly ash

2.1. Coal fly ash

Coal fly ash, a by-product of coal-fired power plants, produced from the combustion of pulverized coal in a coal-fired boiler of thermal power plants. The fly ash particles are removed from the flue gases using electrostatic precipitators, FGD systems, or bag houses and are collected and stored dry for recycling. Fly ash consists of fine particles, predominantly spherical in shape, either solid or hollow, ranging in diameter from $<1\ \mu\text{m}$ up to $150\ \mu\text{m}$ formed from the mineral matter in coal, consisting of the noncombustible matter in coal plus a small amount of carbon that remains from incomplete combustion. Properties of fly ash vary significantly with coal composition and plant-operating conditions. Fly ash contains the primary inorganic components of SiO_2 , Al_2O_3 , Fe_2O_3 , and CaO , less amount of MgO , Na_2O , K_2O , SO_3 , MnO , TiO_2 , and C and varying levels of trace elements [36–38]. Based on its chemical composition (ASTM C618), fly ash can be classified into two classes, C and F. Class C ash (high-calcium, $\geq 10\%$ CaO) is normally produced from lignite or subbituminous coals and contains less $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$ ($>50\%$) but more calcium hydroxide or lime and higher amount of alkalis (combined sodium and potassium). Class F ash (low-calcium, $<10\%$ CaO) is generally produced from burning anthracite or bituminous coal and contains at least 70% of $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3$. [38–43]. The mineralogy of fly ash is greatly influenced by the parent coal from which it was derived. Owing to the rapid cooling of burned coal in the power plant, fly ashes consist of amorphous glass ($\leq 90\%$) and a small amount of crystalline material [44–49]. This predominant portion of the glass gives fly ash its pozzolanic properties (harden with water after activation with an alkaline substance such as lime) [50]. The major crystalline phases in fly ashes are quartz (SiO_2), mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$), magnetic spinel includes; magnetite (Fe_3O_4), and hematite (Fe_2O_3). Although the total percentage of magnetic matrix of the ash is small, particular attention should be given because of its reactivity and potential for carrying and releasing toxic elements. [47].

Fly ash particle surfaces are often enriched in highly environmentally reactive trace elements (such as As, Se, Cd, Cr, Ni, Sb, Pb, Sn, Zn, and Mo [10, 51]) due to the largely emission of various trace elements in coal during coal combustion [52–55]. It has been reported that trace element concentrations in fly ash are sometimes 4–10 times higher than their original concentrations in coal samples due to the condensation of elements, which are volatilized during combustion, onto solid particles at different rates and in varying amounts as the combustion gases cool down [56–60]. As a result, these elements readily react when the fly ash particle is exposed to water and release into the environment [35, 59]. However, the release of the elements from fly ashes is controlled by precipitation or dissolution mainly and possibly by desorption. Understanding the factors that control the leaching behavior of trace elements is critical in predicting the potential impacts of fly ash on the environment. Since As, B, and Se have recently become a major problem in soil contamination in Japan, the leaching behavior of As, B, and Se has been growing in the recent investigations and described in the following sections.

2.2. Overview of leaching characteristic of arsenic, selenium, and boron from coal fly ash

Several mechanisms have been observed to elucidate arsenic, boron, and selenium leaching behavior from fly ash. Generally, the leaching behavior of As, B, and Se was affected by pH, solid-to-liquid ratio, leaching time, and ash properties, in which these ash properties including chemical composition, chemical/mineralogical speciation, the particle morphology, and the fraction of a species are available for leaching [16–19, 26, 28, 61].

Arsenic and Selenium releases from acidic fly ash increase with pH, whereas in alkaline fly ash, this trend is reversed and the leaching of As and Se from acidic fly ash could be described by sorption on iron oxide, while leaching from alkaline fly ash seems to be controlled by sorption on an alkaline Ca phase [20]. This finding is relevant with that from Zielinski et al., where in a highly acidic fly ash, the mode of occurrence of arsenic is associated with iron oxide, oxyhydroxide, or sulfate, while in a highly alkaline ash, arsenic is associated with a phase similar to calcium arsenate (detected using XAFS (X-ray absorption fine structure) spectroscopy) [62]. It is well known that Ca-rich ash tends to make the formation of insoluble Ca-arsenate [63], whereas low-lime fly ash provides less chance for this phase to precipitate. At pH > 11.5, the precipitation of ettringite ($3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{CaSO}_4 \cdot 32\text{H}_2\text{O}$) contributes to the dramatic reduction of As in leaching solution, along with other oxyanionic species [10, 64]. It is also reported that the formation of ettringite ($3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{CaSO}_4 \cdot 32\text{H}_2\text{O}$) contributes to the stabilization of selenium in subbituminous coal ash [65, 66]. The adsorption of selenite in fly ash was also likely controlled by aluminum oxide [23, 51, 67].

Boron is the most mobile trace element in coal ash [21], since it is associated frequently with the smallest particles in the ash, where it can accumulate on the surfaces of particles and in water-soluble fraction which promote high leachability rates [51]. Although the leachability of B does not depend significantly upon pH at nearly neutral values (pH 6–8), the initial rate of leaching is increased by an increase in acidity and it decreases with further increase in pH, and the trend shows no substantial difference between acidic and alkaline ash [21]. On the

contrary, the boron leachability depends on its element concentration in coal fly ash [18], and the ligand exchange mechanisms are considered as a reason for the lower concentrations of B under mildly acidic conditions [68, 69], although much greater leaching rates are achieved when under strongly acidic conditions [69, 70]. Previous studies indicate the tendency for ettringite to incorporate B within its mineral lattice and lead to the decreasing of solubility/dissolved concentrations of B at pH $\approx$ 11.5 [18, 71]. It has been also indicated that under alkaline conditions, the co-precipitation of B with CaCO_3 may significantly captured B in alkaline fly ash [18], to some extent, in acidic-natured fly ash, this process is unlikely to take place [19].

3. Arsenic, boron, and selenium: properties, contamination pathways, and their harmful effects

3.1. Arsenic

Arsenic is a semi metallic element with atomic number 33, atomic mass 74.92 g mol^{-1} and density 5.72 g cm^{-3} at 14°C [72]. It is an odorless, tasteless, and notoriously poisonous metalloid with various allotropic forms: black, yellow, and gray, in which the gray form is the most common, and it is used for industrial purposes. It is distributed throughout our environment, mainly in earth crust, air, water, soil, sediments, etc. Arsenic is emitted into the atmosphere by high-temperature processes such as coal-fired power generation plants, burning vegetation, and volcanism [73]. Some forms of arsenic are inorganic which do not contain carbon, and others are organic, which always contain carbon. The examples for organic arsenic compounds are arsanilic acid (4-aminophenylarsonic acid, $\text{C}_6\text{H}_8\text{AsNO}_3$), arsenobetaine (2-trimethylarsoniumylacetate, $\text{C}_5\text{H}_{11}\text{AsO}_2$), methylarsonic acid (monomethylarsonate, CH_3AsO_3), etc. [74]. Inorganic arsenic compound exists in four oxidation states: -3 (arsenide), 0 (metallic arsenic), $+3$ (arsenite) and $+5$ (arsenate). The examples for arsenide compounds are alkali and alkaline earth metal arsenides (e.g., sodium arsenide Na_3As), arsenides of group III elements (e.g., gallium arsenide GaAs) [75], etc. The most common forms of arsenite compounds are arsenic sulfide (As_2S_3), arsenic trichloride (AsCl_3), potassium arsenite ($\text{AsO}_2\cdot\text{K}$), sodium arsenite ($\text{AsO}_2\cdot\text{Na}$), etc. [76]. The general form of arsenate is AsO_4^{3-} and exists in different forms such as arsenic acid (H_3AsO_4) at strong acidic condition, dihydrogen arsenate (H_2AsO_4^-) at weak acidic condition, hydrogen arsenate (HAsO_4^{2-}) at weak basic condition, and arsenate ion (AsO_4^{3-}) at strong basic condition. Examples of arsenate compounds are arsenic pentoxide (As_2O_5), calcium arsenate ($(\text{AsO}_4)_2\cdot 3\text{Ca}$), lead arsenate ($\text{HAsO}_4\cdot\text{Pb}$), potassium arsenate ($\text{H}_2\text{AsO}_4\cdot\text{K}$), sodium arsenate ($\text{H}_2\text{AsO}_4\cdot\text{Na}$), etc. [76]. Arsenate is thermodynamically stable at aerobic condition, while arsenite is stable at anaerobic conditions [77]. Generally, inorganic forms of arsenic are more toxic to the environment than organic forms, and among inorganic forms, arsenite is more toxic than arsenate due to higher cytotoxic, genotoxic, mobile, and soluble nature of arsenite [78].

Humans may be exposed to arsenic through the usage of arsenic containing water for drinking purposes, food, cosmetics, cigarettes, etc. [79]. The sources of arsenic contamination in water medium can be classified as natural and anthropogenic sources. The natural sources of

arsenic contamination in water medium are due to the mobilization of arsenic in natural condition which depends on its oxidation states, mobilization mechanism, and the parent mineral form [80]. The anthropogenic sources of arsenic contamination in water medium include the wastes generated from industries manufacturing wool and cotton, glass, ceramics, semiconductor, pesticide, etc. and from industries like rare earth industry, chemical industries, petroleum refining, etc. [81, 82].

The health effects on arsenic may come both from different forms of arsenic (inorganic and organic forms) and categorized based on the level of contamination. Among the effect of inorganic arsenic exposure, disturbance of the stomach and digestive organs, diminished generation of red and white blood cells, skin changes, and lung irritation have been reported in many health cases. The take-up of significant amounts of inorganic arsenic has been proposed to increase the possibility of cancer development, particularly the possibility of skin cancer development, lung, liver, and lymphatic cancer. Barrenness and miscarriages with women, skin irritation, declined protection from diseases, heart disruptions, brain damage with both men and women, and DNA damage have been reported as the effects of a very high exposure to inorganic arsenic. The effects of organic arsenic are almost the same with inorganic arsenic, which can cause neither cancer nor DNA damage, but high doses exposure may cause nerve injury.

3.2. Boron

Boron is a nonmetallic element with atomic number 5, atomic mass 10.81 g mol^{-1} , density 2.3 g cm^{-3} at 20°C , and a poor electrical conductor at room temperature. It has several allotropes in the form of amorphous boron, a dark powder, unreactive to oxygen, water, acids, and alkalis; crystalline boron, silvery to black, and extremely hard. It is found in nature in a low concentration in oceans, earth crust, rock, soil, and water [83] and mostly in the form of over 200 minerals with different amounts of calcium, sodium, or magnesium and available as calcium, sodium, and magnesium borates. Among them, the most popular are borax, tincal, colemanite, ulexite, and kernite [84]. Boron has various oxidation states in compounds, but the most significant and common is +3. It appears in lower oxidation states +1, 0, or less than 0, but these states are found in compounds such as higher borates only [85, 86]. Boron enters the environment mainly through the weathering of rocks, boric acid volatilization from seawater, and volcanic and geothermal activity. Boron is released to the environment from anthropogenic sources e.g., via industrial air emissions, fertilizer and herbicide applications, and municipal and industrial wastes from industries manufacturing borosilicate glass, detergents, semiconductor, cosmetics, flame retardants, fertilizers, and dyestuff production [87]. Two anthropogenic boron compounds, boron trichloride and boron trifluoride, are listed as toxics release inventory (TRI) chemicals.

Human causes of boron contamination include releases to air from power plants, chemical plants, and manufacturing facilities. Contamination of water can come directly from industrial wastewater and municipal sewage, as well as indirectly from air deposition and soil runoff. Borates in detergents, soaps, and personal care products can also contribute to the presence of boron in water. Boron can be found in surface water in the form of undissociated

orthoboric acid, partially dissociated borate anions in the form of polyborates, complexes of transition metals, and fluoroborate complexes, and it is also found in ground water, brackish water, or hot springs, especially at geothermal or tectonic areas [88].

Among the effect of long-term consuming of food and water with increased boron content, cardiovascular, coronary, nervous, reproductive systems, changes in blood composition, neurological effects, physical disorders, and intellectual development of children have been reported in many health problems. For pregnant women, abundance of boron can be notably harmful as it raises the risk of birth pathology. Testicular decay and degeneration have been detected as the effects of high daily doses of boron. Queasiness, retching, diarrhea, and lack of appropriate weight gain or weigh loss have been detected as the effects of consuming of food and water with doses greater than 500 mg/day [89].

3.3. Selenium

Selenium is a nonmetallic element with atomic number 34, atomic mass 78.96 g mol^{-1} , and density 4.79 g cm^{-3} at 20°C . Selenium has a number of allotropes including a gray crystalline hexagonal selenium, a red crystalline form, an amorphous (which has a disordered atomic structure) red powder, and a black vitreous (glass-like) form. Amorphous selenium is a photoconductor (a light-dependent semiconductor), and it has had a long history of use in light-based applications—it was used to make the first solar cell in 1883. Selenium is a rare component of the Earth's crust that is mostly found as selenide associated with heavy metal sulfide ores, such as copper and lead sulfides [90]. Selenium species can be grouped into the four major categories: (1) inorganic selenium, (2) volatile and methylated selenium, (3) protein and amino acid selenium, and (4) nonprotein amino acids and biochemical intermediates. Selenium compounds commonly exist in the oxidation states -2 , $+2$, $+4$, and $+6$. It is usually found as the oxyanions selenate (SO_4^{2-}) and selenite (SO_3^{2-}) in oxidized systems and as elemental selenium ($\text{Se}[0]$) and selenides (HSe-) in aerobic zones and unweathered mineral formations [91]. Though complexed selenium is of low toxicity, selenate (SeVI) and selenites (SeIV) are very toxic. These two forms of selenium are generally found in water and display bioaccumulation and bioavailability. Under acidic conditions, the extremely toxic and corrosive hydrogen selenide gas can be generated from selenium containing species.

Selenium contamination typically occurs in the aqueous stream and bioaccumulation of selenates and selenites in waste water can threat all aquatic life downstream. Natural processes that redistribute selenium include volcanic activity, terrestrial weathering of rocks and soils, wildfires, and volatilization from plants and water bodies. The anthropogenic sources of selenium to aquatic systems are including mining, fossil fuel combustion, oil refining, and discharge of seleniferous drainage water from irrigated agriculture [92]; these sources will end up in groundwater or surface water through irrigation.

Humans may be exposed to selenium through food or water or contact with soil or air that contains high concentrations of selenium. The health effects of various forms of selenium can vary from brittle hair and deformed nails, to rashes, heat, swelling of the skin, and severe

pains. Exposure to selenium through air can cause dizziness, fatigue, and irritations of the mucous membranes. Among the effect of selenium overexposure, red staining of the nails, teeth, and hair have been reported in many health cases. Due to the fact that selenium is an eye and upper respiratory irritant and a sensitizer, selenious acid produced from the reaction of selenium dioxide with moisture may cause corrosive to the skin and eyes. Accumulation of fluid in the lungs, pneumonitis, bronchitis, bronchial asthma, shortness of breath, sore throat, chills, fever, headache, conjunctivitis, queasiness, retching, abdominal pain, diarrhea, and enlarged liver has been reported as the effects of overexposure of selenium fumes.

4. Fly ash treatment techniques

Treatment of fly ash before utilization is important to minimize leaching of contaminants and/or utilize in the best possible manner in which methodologies for detoxifying fly ash including reducing the concentrations of the contaminants (e.g., through washing), astringing the leaching of the contaminants by forming steady blocks or inertial compounds with additives or binders (e.g., by stabilization methods), or reducing the mobility of the contaminants (e.g., through S/S methods) [93]. Several researchers have produced numerous treatments and disposal solutions for fly ash over the recent decades, some of these are only tested in the laboratory, while others are available commercially. Based on the main principle of operation, the treatment techniques may be classified in four categories [29, 94]:

1. extraction and separation,
2. chemical stabilization,
3. solidification, and
4. thermal treatment.

4.1. Extraction and separation

Extraction and separation techniques have the main purpose to remove or recover specific components or fractions from fly ash which focus on removing heavy metals and to some extent salts from fly ash, mainly using water or acidic solutions. The processes of this treatment technique are typically relatively simple, but the main disadvantage is the generation of process water with high content of metals and salts.

The main extracting agent used in this technique including water [95], acid [96], and micro-organisms [97]. The electrodialysis process which involve ion exchange technique [98] and particle size fractionation process based on the settling velocities of fly ash particles in a water filled reactor [99] are also including in this extraction and separation techniques. For the comprehensive overview of the principles of these techniques, see Refs. [94, 100].

4.2. Chemical stabilization

Chemical stabilization technique has the main purpose to bind and immobilize pollutants in the fly ash matrix which focus on binding heavy metals by alterations to the fly ashes geochemical properties [101, 102]. Generally, the processes of the chemical stabilization technique are simple and low cost, which significantly improve the leaching properties of the fly ash, such as water extraction, chemical reactions, and then, de-watering. The main disadvantage is the generation of metal and salt containing process water.

Various chemical stabilization processes have been developed, and most of them involve the use of FeSO_4 [35, 95, 103], $\text{FeSO}_4 + \text{CaCO}_3$ [35], CO_2 [104, 105], CO_2 , and H_3PO_4 [106] Phosphate (PO_4^{3-}) [107], Sulfide (S^{2-}) [108], $\text{Ca}(\text{OH})_2$, and paper sludge ashes [27], $\text{Ca}(\text{OH})_2$ and cement [109, 110], colloidal silica [111], and silica fume [112].

4.3. Solidification

Solidification technique is often discussed as stabilization processes (S/S: solidification and stabilization) in which solidification involves the transformation of a liquid or sludge into solids and may not lead to a chemical interaction of the constituent of concern with the solidifying agent. This technique has the main purpose to physically and hydraulically encapsulate the fly ashes and reduces the mobility of the contaminants in fly ashes because of the reduced surface area and low permeability. On the other hand, the main goal of stabilization is to convert the contaminants into less soluble or less toxic forms, with or without solidification [113]. The main advantages of this technique come from the fact that this technology is simple, well established, and low cost, which significantly decrease the leaching of contaminants and improve the mechanical properties. The main disadvantages of this technique are related to significant increase in the mass disposed of, and the physical integrity of the product may deteriorate over time resulting in increased metals leaching.

The main solidifying agent used in this technique including water [114], cement [93], and cement [115]. For the comprehensive overview of the principles of these techniques, see Refs. [94, 100, 116].

4.4. Thermal treatment

Thermal treatment technique involves a heating of fly ashes and thereby changing the physical and chemical properties of fly ashes in which the stable and very dense product can be produced with sufficient leaching properties [117]. This method very efficient at destroying dioxins, furans, and other toxic organic compounds due to the high temperatures used [118]. In some cases, encapsulation of fly ash is also occurred [101]. The main disadvantages are the high cost due to the high-energy demands for the process and generation of flue gas containing volatile metals.

Three major types of thermal treatment technique are sintering, vitrification, and melting.

1. Sintering processes involves heating to a level at which individual particles are bound together and the chemical species of interest can achieve a reconfiguration [119]. Temperatures are around 900–1300°C, and a denser and less porous material is produced.
2. Vitrification processes involves melting of a mixture of fly ash and additives (glass precursors (i.e., Si)) to fix the contaminants in the final matrix (alumina silicates). This mixture is typically heated to around 1300–2000°C and then cooled to form a single solid phase (an amorphous and homogenous glassy material). In this process, fly ash components are bound in the glassy materials thereby also encapsulating the fly ash.
3. Melting (or fusion) processes are very similar to vitrification processes; however, in this case, no additives (glass forming materials) are added. The final product consists of multiple metal phases [118] which can possibly be separated utilizing different melting temperature of individual metal phases. Therefore, the product obtained can be used as a construction material, for example, as a sub-base in road construction or for pavements [120]. The temperatures involve in this melting processes are like vitrification processes.

5. Experimental results of simultaneous leaching suppression of As, B and Se from coal fly ash

Considering the leaching of arsenic, boron, and selenium from fly ash has recently become a major problem in soil contamination in Japan [27, 121], the current research focuses on the development of effective technologies for the improving leaching properties of fly ash. As described in section 4, several pre-treatment options have been proposed for minimizing the leaching of toxic trace elements, including addition of Ca(OH)_2 to fly ash [109, 110]. Based on this method, the Laboratory of Environmental Engineering Systems of Gifu University-Japan, one of the founding members of the Next Generation Energy Research Center, developed its scientific research on “Experimental study on simultaneous leaching suppression of trace elements including As, B, and Se from coal fly ash” which analyzes the leaching properties and promotes the methods for control of the trace elements leachates from coal fly ash, utilizing the chemical stabilization technique by using Ca(OH)_2 , paper sludge ash, and filter cake, which are waste generated in the papermaking and lime industry processes [27].

In this study, a low calcium content fly ash sample named fly ash H (FAH) (2.05% of CaO, detected using X-ray fluorescence) with a high concentration of trace elements leaching (As 48.66 $\mu\text{g/L}$, B 5.39 mg/L , Se 86.9 $\mu\text{g/L}$, detected using ICP-AES) that was collected from coal-fired power plant in Japan was treated through chemical stabilization technique with the addition of additives (Ca(OH)_2 , three kinds of paper sludge ashes (PS Ash 3, 4, 5) and one kind of filter cake (FC)) under 5 and 10% Ca content in additives and subjected in a detailed characterization and leaching test regarding its chemical properties and its leaching behavior. **Table 1** lists the composition of inorganic elements in FAH and additives. **Figure 1** depicts experimental process of chemical stabilization in this study. Fly ash sample and additives characterization

Sample			FAH (1s) ¹	PS Ash 3	PS Ash 4	PS Ash 5	FC
Ash composition	SiO ₂	(%)	59.25	31.47	44.21	42.36	23.31
	Al ₂ O ₃		25.63	12.40	22.23	19.80	13.87
	TiO ₂		1.99	0.38	2.56	2.11	0.06
	Fe ₂ O ₃		7.49	5.13	2.63	5.56	2.33
	CaO		2.05	46.31	18.77	19.51	59.18
	MgO		0.79	3.28	3.42	3.30	0.96
	Na ₂ O		0.60	0.24	0.95	0.41	0.03
	K ₂ O		1.56	0.20	2.09	1.99	0.25
	P ₂ O ₅		0.18	0.18	1.75	1.54	0.00
	MnO		—	0.03	0.05	0.06	0.04
	V ₂ O ₅		0.03	0.01	0.02	0.02	0.02
	SO ₃		0.42	0.36	1.31	3.33	0.00
pH of the leachate			10.38	13.72	12.71	12.26	7.28
Leached Ca ion		(mg/L)	121	1405	246.5	597	15.45
Leaching concentration	As	(μg/L)	48.66				
	B	(mg/L)	5.39				
	Se	(μg/L)	86.9				

¹Sample fly ash H, from the chamber 1 of electrostatic precipitator.

Table 1. Composition of inorganic elements in fly ash and additives.

include the following measurements: pH, elemental composition, trace elements (As, B and Se) leaching and Ca ion concentrations (**Table 1**), thermogravimetric (TG) analysis, X-ray diffraction (XRD) analysis, and leachate analysis (pH, trace elements (As, B and Se) leaching and ions concentrations). This treatment technique, using additives addition, was examined in order to evaluate their effectiveness in the improvement of fly ash leaching properties. Additives may reduce the leachability of As, B and Se contained in ash by promoting the formation of ettringite or precipitation with calcium in ash, which are resistant to leaching. For the classification of untreated and treated ash samples, the procedure of standard leaching tests for fly ash (Notification No. 13 by the Environmental Agency of Japan) was employed as the protocol for leaching tests in this study.

Figure 2 depicts the leaching concentration of As, B, and Se for fly ash H alone and fly ash H under five kinds of additives addition for 5% and 10% Ca content samples, and pH values of leachate. As indicated in **Figure 2**, the results showed that the leaching concentrations of As, B, and Se in FAH tremendously decreased below the soil environmental standard in Japan for both 5% and 10% Ca content in additives and further decreased with the increase of Ca content. Especially, the leaching concentration of As, B, and Se reduced to 91–100% with Ca(OH)₂ addition. Among three kinds of paper sludge ashes (PS Ash

Figure 1. The experimental scheme of chemical stabilization process.

3, 4, 5) addition, As and B leaching concentrations reduced to 89–96% and 83–92%, respectively, with PS Ash 3 addition; and Se leaching concentration reduced to 87–96% with PS Ash 5 addition. While As and B leaching concentration did not influenced by filter cake (FC) addition (owing to the composition of FC is calcium carbonate based, which is not a water-soluble calcium compound (**Figure 3**)), but Se leaching concentration reduced to 58–78% (owing to the presence of Al_2O_3 and Fe_2O_3 in FC contributing for selenium to being hard to leach). **Figure 3** depicts XRD patterns of several calcium compounds and additives. According to the leaching mechanism of selenium, Al_2O_3 and Fe_2O_3 may provide additional surface area for positively charged ions to be attached through the sorption reaction process, which made contributions to lower concentration of Se in aqueous solutions (FAH-FC mixture leachates) [122–124]. It is also indicated in **Figure 2** that pH values of chemical treated products range from 9.23 to 12.98. It could be seen that the pH of mixture leachates increased with the addition of all additives excepting FC. Especially, pH values of FAH- $\text{Ca}(\text{OH})_2$ and FAH-Ps Ash 3 mixture leachates increased to approximately 12.98 and 11.94, respectively. This exhibited that relatively high CaO content included in PS Ash 3 and $\text{Ca}(\text{OH})_2$ itself being a water-soluble calcium compound could promote the pH values of leachates. However, FC which contained the highest CaO content did not have any influence on the pH values of mixture leachates, this was likely to be caused as calcium compound included in FC is composed of CaCO_3 being an insoluble substance in water (**Figure 3**). **Figure 2** demonstrated that the minimum solubility of As, B, and Se was obtained when the pH of mixture leachates became 11.5 or higher. Obviously, higher pH

Figure 2. Trace element leaching concentration for fly ash H alone, and fly ash H under five kinds of additives addition for 5% and 10% Ca content samples: (a) arsenic; (b) boron; and (c) selenium.

Figure 3. XRD Patterns of several calcium compounds and additives.

of mixture leachates in this study was favorable to lowering leaching concentrations of As, B, and Se and the mobility of As, B, and Se indeed decreased in **Figure 2**, implying that the efficiency of method in this study was susceptible to pH value of leachate. The results revealed that all additives (other than filter cake) showed a simultaneous leaching suppression effect of As, B, and Se from coal fly ash.

6. Conclusion

Treatment of trace elements (arsenic, boron, and selenium) contained in coal fly ash is a necessity to minimize leaching of trace elements and utilized in the best possible manner. Chemical stabilization of trace elements (arsenic, boron, and selenium) contained in coal fly ash offers a simple and low-cost environmentally friendly technique that if properly and thoroughly carried can bring our environment into a better place for both human and animal well-being due to its enormous advantages over other treatment methods. According to fly ash treatment through the addition of Ca-containing-additives experimental results, the leaching concentration of As, B, and Se can greatly be reduced simultaneously until 89–96%, 83–92%, and 87–96%, respectively. Paper sludge ash 3 as a waste generated in the papermaking process has been found as an effective and best additive (suppressing material) to reduce the leaching concentration of As, B, and Se from coal fly ash. Therefore, the chemical stabilization technique utilizing Ca-containing-additives is an effective technique for simultaneous suppressing As, B, and Se leaching from coal fly ash.

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