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石炭フライアッシュからの微量元素溶出におけるカルシウム化合物の特性

Calcium compound characteristics in trace element leaching mechanisms from coal fly ash

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Calcium has been shown good decreasing in leaching concentration of several trace elements. In the term of suppressing material, based on the previous research, paper sludge ash (PS 3) conducted good influences in controlling As, Se and B leaching concentration from coal fly ash (FA H). Paper sludge ash is waste generated by the paper recycling industry which is consisting high calcium compound concentration and it is used as an additive in trace element leaching experiment. In order to make an improvement in decreasing of trace elements leaching concentrations, coal fly ash and paper sludge ash conducted in variant ratio and waiting time. One coal fly ash sample (FA C) has been tested with variant of paper sludge ash in order to ensure the effect of suppressing material. The result initiated that PS 3 and PS 8 had the good effect in decreasing the leaching of As, Se and B. PS 8 also tested into FAC in variant addition ratio and compared with the effect of $Ca(OH)_2$, the result showed that PS 8 showed very close effect to $Ca(OH)_2$, therefore it could be used as the single suppressing material or be combined with $Ca(OH)_2$ with the appropriate ratio. FA C also been tested with PS 8 in variant days of waiting time before the leaching process is carried out. The results shows that sample which directly carried out to leaching process without waiting time did not show the good decreasing in Se and B leaching concentration and samples which carried out after one day of waiting shows rapidly decreasing then almost constant for the next seven days.

Key Words : Coal fly ash, Trace elements, Calcium, Suppressing material, Leaching methods,

1. Introduction

Globally, approximately 100 million tons of coal ashes are produced annually from coal combustion, but less than 50% is recycled [1]. Coal fly ash finds reuse primarily in cementitious products, construction areas such as highway road bases, grout mixes, stabilizing clay based building [2]. Because the rate of coal fly ash production is greater than it`s consumption, the unused fly ash will be disposed into ponds, lagoons, landfills and slag heaps. Coal contains harmful elements such as arsenic, selenium and boron. So that, the development and improvements method on leaching of trace elements is still an interesting topic to discuss.

When coal ash comes into contact with water, its toxic constituents can “leach” or dissolve out of the ash and percolate through water [3]. Leaching experiment with the addition of suppressing material already discussed in the previous research. The addition of suppressing material evidenced the linier result with the decreasing of trace elements leaching concentration. Suppressing material is by-products from other industries which contains high calcium contents, which is added to coal fly ash sample before leaching process to control the trace elements leaching concentration [4].

In order to produce the good decreasing of trace elements leaching concentrations, this study is discussed the method improvements through the variation of calcium addition ratio and variation of waiting time after the mixing process.

2. Methodology

Calcium has been known to play an important role in the release of arsenic (As) from coal fly ash. Previous research also found that the calcium also affect the selenium (Se) and boron (B) leaching concentrations through the addition of suppressing material. In this study, several paper sludge ash have been tested into coal fly ash sample C (FAC) in order to ensure the effect of paper sludge ash and also to find the appropriate addition ratio and waiting time in leaching process. Calcium hydroxide ($Ca(OH)_2$) is used as the standard for the suppressing materials tested (paper sludge ashes).

Leaching test No. 13 which is notified by the Japanese Environmental agency was basically employed as the leaching test in this study. Amount of sample added with distilled water with the ratio 1:10. Then, the mixture will be shaken in with shaking speed 200 r.p.m in room temperature. After six hours, the mixture will be filtered by using

vacuum filtration and 0.45 µm membrane filter to separate the solid and liquid (leachate). Then, the leachate could be analysis with ICP-AES, ion chromatography, and pH meter.

The different with leaching experiment with suppressing material is coal fly ash sample need to be treated before the leaching process. Figure 1 explained the sample preparation process. Coal fly ash mixed with suppressing materials in the mixed bag. The mixture added with distilled water with the ratio 25% of sample weight, then the sample is kneaded for 1 minute, scraped then continue kneaded the sample for two minutes. Before carried out to the leaching process, sample need to be waiting for seven days in room temperature, then sieved using 2 mm sieve.

This study is intended to improve the leaching process result through studied the effect of variation addition ratio of suppressing materials and also the variation of waiting time.

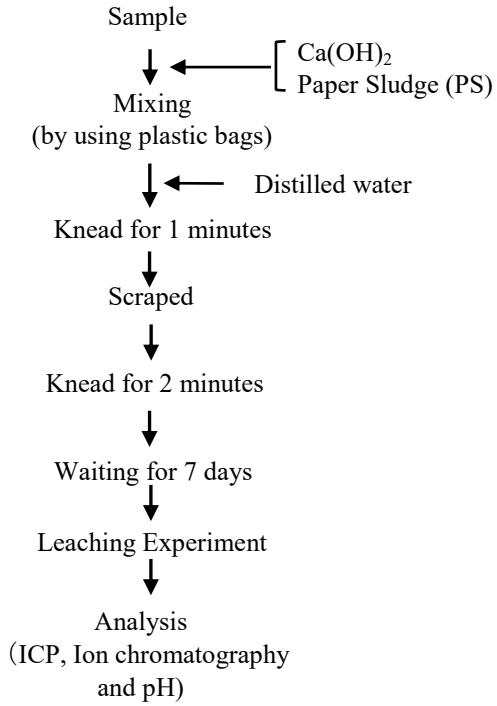
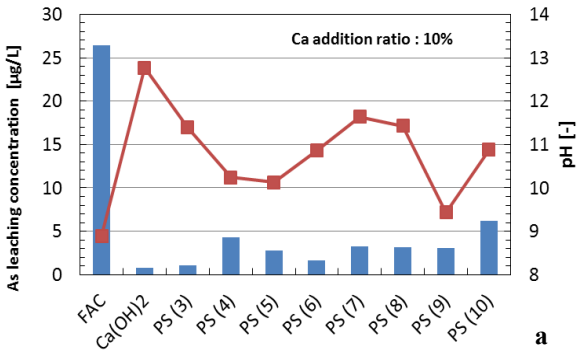


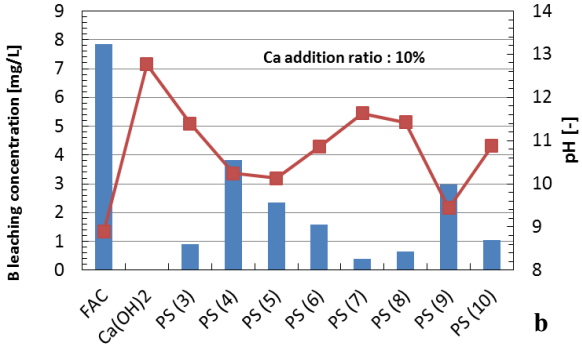
Fig 1. The procedure of leaching process with the addition of suppressing material

3. Result and Discussion

3.1. Effect of suppressing material into trace elements leaching concentrations

In the previous study, paper sludge ash and filter cake have been tested into coal fly ash H (FA H) and the result report that PS 3 give the best decreasing for As leaching concentration. Then, PS 3 and other paper sludge ashes have been tested into coal fly ash sample C (FA C) to ensure the suppressing material effect into trace elements leaching concentration. Fig 2 a-c showed the effect of several paper sludge ash into arsenic, selenium and boron leaching concentrations. Calcium hydroxide has been used as the standard to compare the calcium effect of other paper sludge ashes tested. Could be known that PS 3 and PS 8 showed the closest effect into $Ca(OH)_2$ in the decreasing of As, Se, and B leaching concentrations than others paper sludge ashes. Calcium affected the As, Se and B leaching concentration by the reacting with oxides of As, Se and B and then form stable compound which is slightly soluble in water.





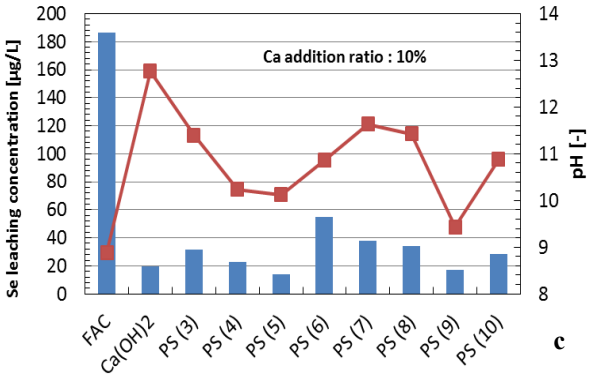


Fig 2. The effect of suppressing material (paper sludge ash) into trace element leaching concentration in FAC, (a) Arsenic, (b) Boron, (c) Selenium

The addition of suppressing material also affected the leachate pH. In the reaction, alkalinity also has an important role. Bora Cetin et al wrote that selenium start to have effect in high alkalinity solution because at that pH, selenium starts producing anionic form $HSeO_3^-$ released from fly ashes and boron is very sensitive to the pH of the aqueous solution and shows a cationic leaching pattern indicating that its solubility is very high at low pH. Then, Tian Wang et al also said that As and Se have high environmental mobility at alkaline pH.

3.2. Suppressing material addition ratio effect into trace elements leaching concentration

The addition amount of suppressing material into coal fly ashes is one important thing to discuss since it will affect the trace elements leaching concentration and also will affect pH of the leachate. The aim of this discussion is to find the appropriate combination and ratio of suppressing material which could decrease the trace elements leaching concentrations. As the initial step, PS 8 and $Ca(OH)_2$ has been chosen to learn the different in arsenic leaching concentration between the addition ratio base on the amount of sample and base of calcium amount on sample. Fig 3 explained that there is no specific different between the both of them in arsenic leaching concentration. The next step will be discussed later.

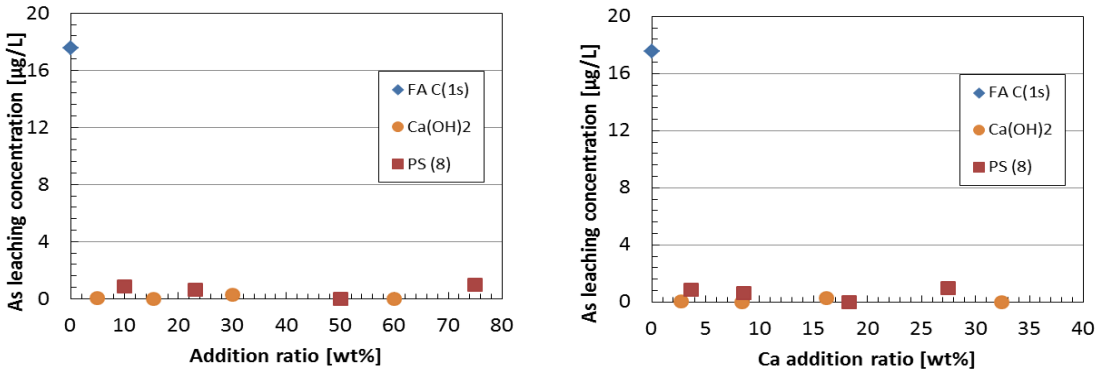


Fig 3. The effect of suppressing material addition ratio in arsenic leaching concentration in FAC

3.3. The effect of waiting time in sample preparation into trace element leaching concentration

In the very first experiment before the sample ready to carry out in a leaching process, it should wait for seven days in room temperatures. The purpose is to make sure the pre-reaction between trace elements in coal fly ash and water from distilled runs good, but at the same time the waiting times need to be tested in order to find the efficient time in leaching process. Therefore, this discussion is intended to find the appropriate waiting time in leaching of trace elements.

Fig 4 showed the effect of waiting time on coal fly ash sample C with the addition of 10 % of $Ca(OH)_2$ as the suppressing material. The results explained that while FA C directly carried out to leaching process after the preparation sample, it did not give any good effect on the leaching of arsenic, selenium and boron. The trace element leaching decreased on the one day waiting time and then kept stable on the next day until day 7. Therefore, in the next experiment, before the leaching process is carried out, the sample mixtures only need one day to complete the reaction.

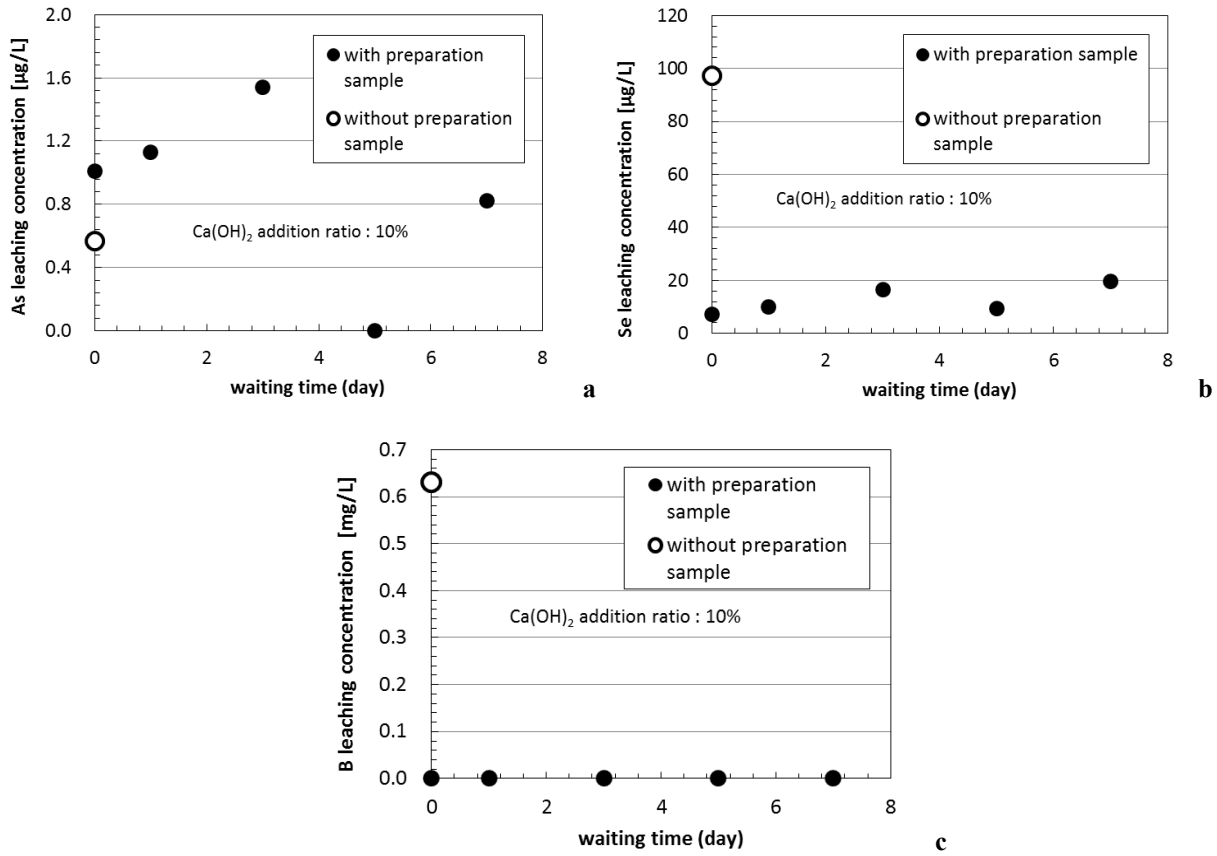


Fig 4. The effect of waiting time in trace elements leaching concentration (a) Arsenic, (b) Selenium, (c) Boron

4. Conclusion

In leaching of trace elements with suppressing material, coal fly ash should be prepared first before carried out into a leaching process. The amount of suppressing material added and waiting time discussed in this study. The addition amount of suppressing material could be decided by the total amount of sample and sample only need to be curing for one day before the leaching process is carried out.

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