Phosphorus Transfer in the Ash through Acid or Alkali Extraction Processes

Masaaki Takahashi¹, Yukimasa Takemoto¹ and Eiji Yuki²
¹. Yokkaichi University, Research Laboratory on Environmental Technology, Kayo-cho 1200, Yokkaichi, Mie, Japan
². Mie Chuo Kaihatsu Co., Ltd, Hachiya 4713, Yono, Iga, Mie 518-1152, Japan

Abstract: In order to find the best method of the phosphorus recovery, some kinds of the phosphorus recovery methods were applied using the same raw material. The acid treatment shows the highest recovery performance, on the contrary, the alkali treatment indicates the lowest recovery rate. The recovery rate was improved by the application of the heat treatment or hydrothermal method. In the acid treatment, the recovered phosphorus is mainly composed of aluminum phosphate, conversely, in alkali treatment, recovered phosphorus was mainly composed of sodium phosphate containing a small amount of aluminum. But the aluminum content in the phosphorus decreased by introduction of the heat treatment or hydrothermal method.

Key words: Phosphorus, recovery rate, sewage sludge, extraction method.

1. Introduction

Sewage sludge contains significant amounts of phosphorus, and in order to recover the phosphorus in them, some kinds of recovery methods using acid [1] or alkali [2] are investigated. In the phosphorus recovery by the acid treatments, the recovery rate is high but recovered phosphorus is mainly composed of aluminum phosphate [3], which has little utilization. On the contrary, alkali treatment can recover the phosphorus as a form of sodium phosphate which has many usages. However, the recovery rate is low compared to the acid treatment [4], and to solve the matter, incineration treatment [5] or hydrothermal method [6] are considered. Like this, phosphorus extraction performance differs depending on the recovery method or extraction condition and also composition of the raw materials.

In order to find the best method of the phosphorus recovery, we investigated the phosphorus transfer in these treatment (acid treatment, alkali treatment, heat treatment or hydrothermal method) using the same raw material [7]. In this experiment, as a raw material, charcoal of the sewage sludge and incinerated ash of same charcoal were used.

2. Methods

2.1 Raw Material

As a test material, charcoal of the sewage sludge and ash which was made by incineration of the same charcoal, were used. The charcoal is made from sewage sludge treated in the oven at 750 °C. The chemical composition of the ash component is shown in Table 1.

2.2 Extraction Method

We compared 4 types of methods as follows (each procedure is shown in Fig. 1).

(1) Acid treatment at room temperature [8]

The ash or charcoal is mixed with diluted sulfuric acid and the phosphorus is extracted in acidic condition (blow pH 2) at room temperature, and extracted phosphorus was precipitated by neutralization in conditions around pH 6 to pH 8, and recovered by filtration.

(2) Alkali treatment at room temperature

The ash is mixed with aq. solution of sodium
Phosphorus transfer in the ash through acid or alkali extraction processes

Hydroxide at room temperature, and dissolved phosphorus is recovered as a form of sodium phosphate by vaporization.

(3) Alkali heat treatment [9]
An aqueous solution of the sodium hydroxide is mixed with ash or charcoal, and treated at 750 °C, later, washing water is added to the treated mixture (already be changed to ash) for solid/liquid separation. The phosphorus is recovered by the vaporization of the filtrate.

Table 1  Chemical composition of the raw material (ash).

| Component | Na₂O | Al₂O₃ | SiO₂ | P₂O₅ | K₂O | CaO | Fe₂O₃ | Others |
|-----------|------|-------|------|-------|-----|-----|-------|--------|
| Weight (%)| 3.58 | 13.06 | 26.04| 21.52 | 1.63| 10.02| 14.84 | 9.31   |

Fig. 1  Phosphorus recovery methods.
(4) Hydrothermal method [10]
Aq. solution of sodium hydroxide is added to the ash or charcoal of sewage sludge, and treated at 100 °C to 120 °C. In 1 h, later, washing water was added to the treated mixture for the solid/liquid separation. The phosphorus is recovered by the vaporization of the filtrate.

3. Results and Discussions

3.1 Phosphorus Extraction Rate

The combination of the raw materials (ash and charcoal) and recovery methods (4 types) are shown in Table 2.

In order to make a sufficient reaction, the excessive amounts of the reagents (H\textsubscript{2}SO\textsubscript{4} or NaOH) which are considered to make a complete reaction, were added to the raw materials (ash or charcoal), and the experiments were carried out on mixing rate also shown in Table 2.

The chemical composition of the raw materials, the recovered phosphorus and the residue were analyzed using an X-ray analyzer.

The phosphorus recovery rate was calculated by comparing the P\textsubscript{2}O\textsubscript{5} concentration between the raw material and residue using Eq. (1). The acidic treatment shows the highest recovery performance which reaches around 90%. On the contrary, the alkali treatment indicates the lowest recovery performance (the recovery rate about 60%), however, the recovery rate improved almost 80% by the introduction of the incineration or hydrothermal treatment (Fig. 2).

\[
\text{R-rate} = \frac{(\text{P}_{-\text{raw}} \times \text{W}_{-\text{raw}} - \text{P}_{-\text{treated}} \times \text{W}_{-\text{treated}})}{\text{P}_{-\text{raw}} \times \text{W}_{-\text{raw}}} \quad \text{Eq. (1)}
\]

\begin{itemize}
    \item R-rate: Phosphorus recovery rate;
    \item P\textsubscript{-raw}: P\textsubscript{2}O\textsubscript{5} content in the raw material (charcoal or ash);
    \item W\textsubscript{-raw}: Weight of the raw material (charcoal or ash)
\end{itemize}

Table 2  Combination of the raw material and applied method.

| Mark      | Raw material | Method                      | Amount of the raw material | Amount of the reagent |
|-----------|--------------|-----------------------------|----------------------------|-----------------------|
| Acid-RT   | Ash          | Acid treatment at room temperature | Ash 5 g                   | H\textsubscript{2}SO\textsubscript{4} 3 g |
| Acid-RT   | charcoal     | Acid treatment at room temperature | Charcoal 5 g               | H\textsubscript{2}SO\textsubscript{4} 4 g |
| Alkali-RT | Ash          | Alkali treatment at room temperature | Ash 5 g                   | NaOH 4 g              |
| Alkali-750| Ash          | Alkali heat treatment        | Ash 5 g                   | NaOH 4 g              |
| Alkali-750| Charcoal     | Alkali heat treatment        | Charcoal 5 g               | NaOH 3 g              |
| Hydro     | Charcoal     | hydrothermal method (120 °C) | Charcoal 50 g              | NaOH 30 g             |

Fig. 2  The comparison of the phosphorus recovery rate between some recovery methods.
3.2 Composition of the Recovered Phosphorus

The composition of the recovered phosphorus and the residue are shown in Figs. 3 and 4. The recovered phosphorus by the acid treatment was mainly composed of $\text{P}_2\text{O}_5$ and $\text{Al}_2\text{O}_3$, and considered to be made of aluminum phosphate. In the alkali treatment at room temperature, recovered phosphorus contains $\text{Na}_2\text{O}$ and $\text{P}_2\text{O}_5$, which is considered to be composed of sodium phosphate, however, small amount of aluminum component in the recovered phosphorus. Using the heat treatment or hydrothermal method, the remaining aluminum components became lower [11, 12], which indicates that another reaction happened in these treatments, and further investigations are needed.

Finding the usage of the recovered phosphorus is a very important matter. The phosphorus which was recovered by the acid treatment is mainly made of aluminum phosphate, which has some kinds of usage [13, 14]. However, the purity of the recovered
aluminum phosphate is not good enough for these usages, and also purification of them is not considered to be cost effective. The phosphorus recovered by the alkali treatment is mainly composed of sodium phosphate, which is easily purified by the application of the crystallization method. As mentioned, the heat treatment or hydrothermal method can recover lower aluminum contents, which will be useful when taking account of the usage of the recovered phosphorus.

The significant amount of the residue is also discharged by the phosphorus recovery, and finding a usage of the residues is also an important matter.

4. Conclusions

In order to find an optimal condition of the phosphorus recovery, some recovery methods were investigated using the same raw material. The highest recovery rate was found in the acid treatment, however, the recovered phosphorus was mainly composed of aluminum phosphate. The alkali method can recover the phosphorus as a form of sodium phosphate, however, the recovery rate is low compared to the acid treatment, and also contains a small amount of aluminum component. However, the recovery rate and purity of the recovered phosphorus is improved by introduction of the incineration or hydrothermal method. These performances differ according to some recovery conditions or raw materials [15], hence, much further research is necessary.

Acknowledgements

The authors wish to express their appreciation to Dr. Eric Bray (Professor of Yokkaichi University) for his advice on making the article. The authors also express their appreciation to the Hinaga Wastewater Treatment Center (Yokkaichi City Government, Waterworks and Sewage Bureau) offering us the ash.

References

[1] Hiroshisa, S., and Masaaki, T. 1997. Gekkan Water, 36-40. (in Japanese)
[2] Keiko, Y., Takahashi, Y., and Hatano, M. 2001. “The Experiment on the Recovering Sodium Phosphate from Incinerated Ash of Sewage Sludge.” In Proceedings of 12th Annual Conference of the Japanese Society of Waste Management Expert, 280-1.
[3] Ingham, J., Ryan, J., Keyakida, E., and Ri, J. 1996. “Phosphorus and Metal Recovery from Sewage Treatment Sludge.” In Proceedings of the 7th Annual Conference of the Japan Society of Waste Management Expert, 280-2. (in Japanese)
[4] Ryukou, S., Akihito, S., Takashi, A., Yukinori, S., Yusuke, M., and Yutarou, H 2018. “Phosphorus Recovery Test and Running Cost Estimation Using A Test Plant and Waste Alkalil." Accessed Oct. 5th 2018, https://www.jstage.jst.go.jp/article/jsmcwm/23/0/23_311_pdf.
[5] Masaaki, T., Susumu, K., Hiroshisa, S., Eiji, S., Takao, I., Seiji, H., and Hideo, M. 2004. “Technique for Recovering Phosphorus Salt from Incinerated Ash of Sewage Treatment Sludge.” Transactions of Materials Research Society of Japan 29 (5): 2149-52.
[6] Masaaki, T., Yukimasa, T., and Ken, O. 2015. “Phosphorus Recovery from Carbonized Sewage Sludge by Hydrothermal Processes.” Journal of Material Science and Engineering B 5 (1-2): 58-62.
[7] Masaaki, T., Yukimasa, T., and Eiji, Y. 2019. “Phosphorus; Phosphorus Transfer in the Ash of Sewage Sludge by Acid or Alkali Extraction.” In Proceedings of 30th Annual Conference of the Japanese Society of Waste Management Expert, C4-7P.
[8] Masaaki, T., Susumu, K., Hiroshisa, S., Eiji, S., Takao, I., Seiji, H., and Hideo, M. 2001. “Technology for Recovering Phosphorus from Incinerated Wastewater Treatment Sludge.” Chemosphere 44: 23-9.
[9] Masaaki, T., Kunihiko, S., and Keisuke, N. 2005. “Recovery of Phosphate Salt from Charcoal of Sewage Sludge.” In Proceedings of the 15th Annual Conference of the Japan Society of Waste Management Expert, 386-8. (in Japanese)
[10] Kunihiko, ., Masaaki, T., Yasuo, O., Susuymu, K., and Hideo, E. 2004. “A Technique for Recovering Sodium Phosphate from Incinerated Ash of Sewage Treatment Sludge by Hydrothermal Synthesis.” Transaction of the Material Research Society of Japan 29 (5): 2021-4.
[11] Masaaki, T., Yukimasa, T., and Eiji, Y. 2019. “Phosphorus Recovery from Incinerated Ash of Sewage Sludge by Heat Treatment.” Journal of Material Science and Engineering A 9 (1-2): 13-6.
[12] Masaaki, T., Yukimasa, T., Eiji, Y., and K, S. 2018. “Phosphorus Recovery from Charcoal of Sewage Sludge by Incineration Treatment.” Journal of Material Science and Engineering A 8 (3-4): 59-64.
[13] Yoneyama, K. K. 2019. *Products Information*. Accessed 29th Nov. 2019. https://www.yoneyama-chem.co.jp/en/.

[14] Katsuya, K., Tomoo, S., Michito, T., and Yutaka, D. 2013. “Lead Stabilization Mechanisms of AlPO₄ Prepared from Waste Acid Etchant in Municipal Solid Waste Incinerated Fly Ash.” *Journal of the Japan Society of Material Cycle and Waste Management* 24 (3): 25-34.

[15] Yousuke, N., Msanori, O., Sinsuke, H., and Daisuke, O. 2018. “Chemical Composition of Night Soil Sludge Incineration Ash and the Elution of Phosphate.” *Journal of Environmental Chemistry* 28 (4): 127-39.