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Conversion of Sewage Sludge and Livestock Manure into Valuable Materials by Subcritical Water Treatment Technology

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Abstract: Sewage sludge is treated by various technologies before disposal to avoid environmental risks that are caused by pathogens, hazardous chemicals and heavy metals. However, many of conventional treatment methods are costly, using significant levels of energy. To solve this problem, a subcritical water treatment (SCWT) technology was developed. The characteristics of the SCWT technology is that it can process any kinds of wastes with a wide range of water content by a liquid mixed reaction using steam at a high temperature and pressure. In this study, demonstration experiments with sewage sludge and livestock manure were undertaken using the SCWT reactor by varying temperature, pressure, and time. As for the results, the appropriate conditions regarding treating sewage sludge were found to be at a temperature of 100 °C - 200 °C and a pressure of 1.0 MPa - 2.0 MPa. In addition, the content analysis of the treated samples gave the excellent results relevant for a fertilizer. For example, the content of the treated sewage sludge were: water content 60.97 %, pH 4.16, organic content 87.33 %, T-N 0.69 %, P2O5 0.705 %, K 3,150 mg/kg , Ca 70,500 mg/kg , Mg 3,090 mg/kg , and Na 1,150 mg/kg . These results show good elements for a general organic fertilizer, and it was verified to be pathogen free, as well as hazardous chemicals with heavy metals low enough to satisfy standards for continuous applications on agricultural lands.

Keywords: *fertilizer; organic wastes; recycling system; subcritical water*

I. INTRODUCTION

The world's communities have entered the recycling era with people's awareness of safeguarding the global environment. In recent years, using treated sewage sludge biosolids for agriculture was also considered as a new direction for sewage treatment policy. Especially, collecting phosphorus which is a valuable resource included in sewage is implemented in collaboration with the Ministry of Agriculture, Forestry and Fisheries of Japan because the price of fertilizers is rising due to the shortage of phosphorus on a world-wide scale [1]. The sewage system discharges massive amount of sewage sludge. Sewage sludge is treated by various technologies before disposal to avoid environmental risks that are caused by pathogens, hazardous chemicals and heavy metals. When the safety of biosolids from treated sewage sludge is ensured it becomes a valuable organic resource for recycling systems. However, many of conventional treatment methods are costly, using significant levels of energy with many procedures.

With this importance being placed on recycling systems, a subcritical water treatment (SCWT) technology was developed (Fig. 1, Fig. 2). The characteristics of the SCWT technology is that it can process any kind of organic wastes with a wide range of water content by a liquid mixed reaction using steam at a high temperature and pressure. There is no incineration process; therefore this technology does not generate carbon dioxide, dioxin, or nitrous oxide. In addition, an offensive odor is not generated because of the sealed process.

In this study, demonstration experiments with sewage sludge and livestock manure were undertaken using the SCWT reactor by varying temperature, pressure, and time. Then, the appropriate conditions regarding treating them were found and the content of the treated samples were analyzed.

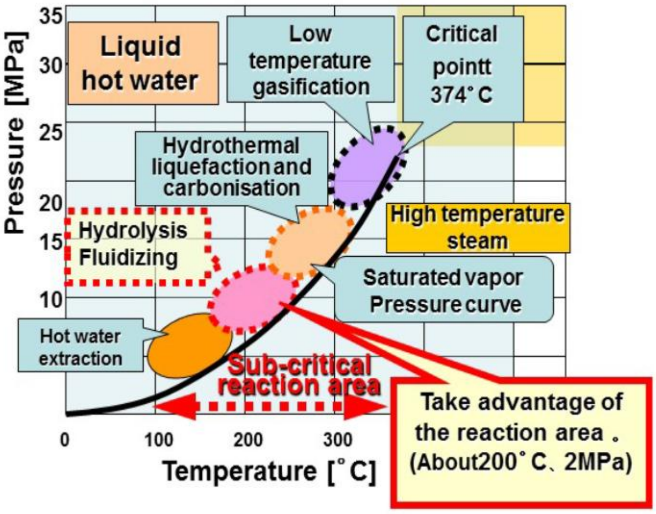

Fig. 1 Principal of the subcritical water treatment technology

Fig. 2 Subcritical water treatment reactor

II. SUBCRITICAL WATER TREATMENT (SCWT) TECHNOLOGY

The key principle of the SCWT is that it only uses high pressure steam to process organic waste. When the temperature and the pressure of water are raised to 374 °C and 22 MPa (220 atmospheres), it becomes an uniform fluid that is neither steam nor water. This condition is called the critical point. The hot water where temperature and pressure are lower than the critical point is called subcritical water, and a hydrolysis reaction can occur with the subcritical water. Through this reaction, the organic molecules such as cellulose, starch, protein and lignin are decomposed into respectively, polysaccharides, glucose, amino acids and phenols as well as being changed into a liquid form from solids (Fig. 1).

Fig. 2 shows the batch style SCWT reactor that processes combustible waste by using high pressure and high temperature in a pressurized vessel. It has an advantage of being able to process various types of organic waste to valuable materials so that the process could be called a multipurpose recycling system of resources. The SCWT reactor has low construction costs as well as low operation costs which are about 1/5 of that of the incineration process. The choice of location is easy because there is no requirement for waste water treatment. Drained steam water is a liquid fertilizer. Drained water from the reactor is also a liquid fertilizer.

The batch type SCWT reactors with a volume of 2 m^3 installed in China and Taiwan were used for the demonstration experiments. The boiler capacity was 500 kg/h. The sewage sludge and livestock manure used in the demonstration experiments were sewage sludge and pig manure in China and chicken excreta in Taiwan.

III. EXPERIMENTAL PROCEDURE

For most organic material treatment, the temperature for the hydrolysis reaction in the sub-critical water domain is around 200°C and the pressure of the saturated vapor is 1.2~1.6 MPa. If the temperature and pressure conditions are higher than these, a degradation of organic material and loss of organic nutrients occurs. Therefore, the temperature was set between 180 ~ 200°C. The SCWT was processed automatically with a program of steam injection to satisfy the pre-determined temperature and pressure conditions. The reaction time ranged from 15 to 30 minutes. After the reactor steam vapor was degassed, it was stabilized to normal pressure and the processed materials were discharged. The disposal of wet biomass as well as sewage sludge basically depends on their original water content. In the case of sewage sludge treatment and disposal, dewatering is a key element for the SCWT reactor process. Dewatered sewage sludge was processed in a batch type SCWT reactor, in which it was found that there was no change in water content through the experiment. After placing sewage sludge in the reactor, it was mixed with rice chaff to adjust the appropriate water content for preventing liquefaction. The rice chaff also plays an important role to supply silicate material enabling binding of heavy metals. Therefore, the appropriate amount of the rice chaff was investigated. Then the content analyses of the sewage sludge and livestock manure used for the demonstration experiment were performed before and after the SCWT.

IV. RESULTS AND DISCUSSION

The sewage sludge, pig manure, and chicken excreta before and after the SCWT are shown in Fig.3. The appropriate conditions regarding treating sewage sludge were found to be at a temperature of 100 °C - 200 °C and a pressure of 1.0 MPa - 2.0 MPa. The sewage sludge shown in Fig. 3 was treated with a temperature of 185 °C, a pressure of 1.2 MPa, and the reaction time was set to 30 minutes. Pig manure and chicken excreta were treated around the similar conditions.

Table 1 shows the chemical contents of the sewage sludge, before and after the treatment, respectively. For example, the content of the treated sewage sludge were: water content 60.97 %, pH 4.16, organic content 87.33 %, T-N 0.69 %, P_2O_5 0.705 %, K 3,150 mg/kg , Ca 70,500 mg/kg , Mg 3,090 mg/kg , and Na 1,150 mg/kg .

Table 2 and 3 show chemical content of pig manure and chicken excreta after treatment. These results show good elements for a general organic fertilizer as shown in Table 4, Table 5, Table 6 (Table 5 also shows the sewage sludge use standards for farmland in Japan). In addition, it was verified to be pathogen free, as well as hazardous chemicals with heavy metals low enough to satisfy the Chinese standards for continuous applications on agricultural lands. In China, heavy metal contamination is regulated and managed by sewage authorities. The concentrations of heavy metals in sewage sludge by the SCWT reactor, were reduced to less than the designated safety levels.

There are food chemical residue regulations for antibiotics, antibacterial agents, growth hormones, pesticides, and herbicides in Japan. The Chinese government has commenced having residual pesticide controls for food as well. The Chinese government is promoting organic farming to enable the reduction of pesticides and now recommends the production of safe organic fertilizers. Although it is thought that these chemical substances are degraded in the process of making animal fecal composts, it is still being investigated.

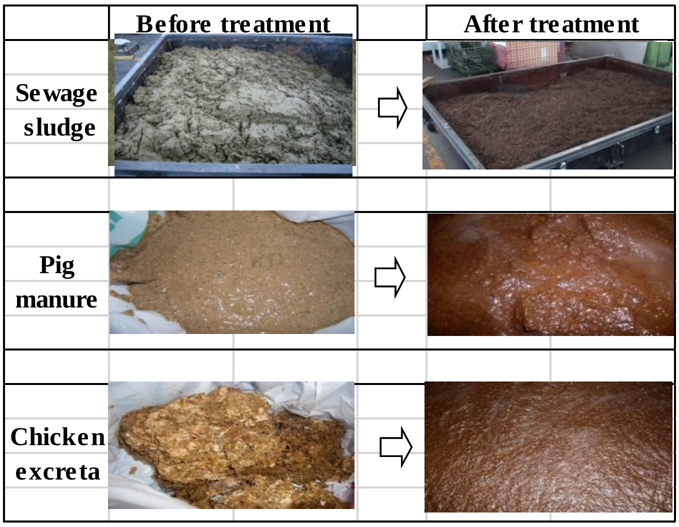

Fig.3 Sewage sludge, Pig manure, and Chicken excreta before and after the treatment by the subcritical water treatment technology

Table 1 Analyses of sewage sludge before and after treatment

Examination items	Before Treatment	After Treatment
Water content (%)	84.83	60.97
p H	6.36	4.16
Organic Matters (%)	70.87	87.33
Ash (%)	29.13	12.67
Nitrogen (%)	0.88	0.69
Phosphorus（P_2O_5) (%)	0.923	0.705

Potassium (K_2O) (mg/kg)	1.16×10^4	3.15×10^3
Sodium (mg/kg)	3.22×10^3	1.15×10^3
Calcium (mg/kg)	2.04×10^4	7.05×10^4
Magnesium (mg/kg)	1.12×10^4	3.09×10^3
Iron (mg/kg)	1.10×10^4	4.19×10^3
Copper (mg/kg)	262	76
Zinc (mg/kg)	976	300
Aluminum (mg/kg)	1.60×10^4	4.90×10^3
Beryllium (mg/kg)	＜0.10	＜0.10
Lead (mg/kg)	30.4	20.4
Arsenic (mg/kg)	6.77	1.6
Mercury (mg/kg)	7.75	0.88
Cadmium (mg/kg)	0.52	0.097
Chromium (mg/kg)	42	22.4
Bacillus species （cfu/g）	2.13×10^9	Undetected
Fungi （cfu/g）	1.56×10^5	Undetected
Actinomycetes species （cfu/g）	4.56×10^6	Undetected

Table 2 Analyses of pig manure after treatment

Examination items	After Treatment
Water content (%)	27
pH	8.2
Organic Matters (%)	82
Nitrogen (%)	2.2
Phosphorus (P_2O_5) (%)	4.3
Potassium (K_2O) (mg/kg)	3.8
Lead (mg/kg)	14
Arsenic (mg/kg)	7
Mercury (mg/kg)	0.4
Cadmium (mg/kg)	0.1
Chromium (mg/kg)	2
NPK content (%)	10.3

Table 3 Analyses of chicken excreta after treatment

Examination items	After treatment
Water content (%)	15.6
pH	7.5
Organic Matters (%)	89.3
Nitrogen (%)	4.8
Phosphorus (P_2O_5) (%)	2.7
Potassium (K_2O) (mg/kg)	0.9
Calcium (mg/kg)	3.1
Magnesium (mg/kg)	0.3
Copper (mg/kg)	< 25
Zinc (mg/kg)	< 137
Lead (mg/kg)	< 10
Nickel (mg/kg)	< 5.0
Arsenic (mg/kg)	< 10.0 mg/kg
Mercury (mg/kg)	< 0.2 mg/kg
Cadmium (mg/kg)	< 0.5 mg/kg
Chromium (mg/kg)	< 10 mg/kg
C/N Ratio	10.7

Table 4 Soil Environmental Standards of China (GB15618-1995)

No.	Element	Unit	1st class	2nd class(farmland)			3rd class(forest land)
			Natural Background	pH<6.5	pH=6.5-7.5	pH>7.5	pH>6.5
1	Cadmium	mg/kg	0.2	0.3	0.3	0.6	1
2	Mercury	mg/kg	0.15	0.3	0.5	1	1.5
3	Arsenic(paddy field)	mg/kg	15	30	25	20	30
4	Arsenic (field)	mg/kg	15	40	30	25	40
5	Copper (farmland)	mg/kg	35	50	100	100	400
6	Copper (orchard)	mg/kg	—	150	200	200	400
7	Lead	mg/kg	35	250	300	350	500
8	Chromium (paddy field)	mg/kg	90	250	300	350	400
9	Chromium (field)	mg/kg	90	150	200	250	300
10	Zinc	mg/kg	100	200	250	300	500
11	Nickel	mg/kg	40	40	50	60	200

Table 5 Sewage Sludge Use Standards for farmland of China (CJ/T 309-2009) and for that of Japan

No.	Element	Standard value in China (mg/kg)		Standard value in Japan (mg/kg)	
		A class	B class	Fertilizers Regulation Act	Soil Environmental Standard
1	Cadmium	<3	<15	<5	150
2	Mercury	<3	<15	<2	15
3	Lead	<300	<1,000	<100	150
4	Chromium	<500	<1,000	<500	250
5	Arsenic	<30	<75	<50	150
6	Nickel	<100	<200	<300	–
7	Zinc	<1,500	<3,000	–	–
8	Copper	<500	<1,500	–	–

Table 6 Sewage Sludge standard for land disposal in China

No.	Element	Standard value (mg/kg)
1	Cadmium	＜20
2	Mercury	＜25
3	Lead	＜1,000
4	Chromium	＜1,000
5	Arsenic	＜75
6	Nickel	＜200
7	Zinc	＜4,000

8	Copper	＜1,500
9	Total cyanides (mg/kg-DS)	＜10

V. SOLIDIFICATION/STABILIZATION CHARACTERISTICS OF HEAVY METALS

Many techniques have been developed for soil remediation by stabilization of the heavy metals in soil. One of the most popular methods is rinsing the soil with acid and leaching the heavy metals and collecting the leachate then entrapping with cement. The SCWT method can remediate the contaminated soil by solidifying heavy metals in soil with lime. After the SCWT, it was found that the soil forms new crystalline structures that are calcareous materials and siliceous materials, called tobermorite ($5CaO \cdot 6SiO_2 \cdot 5H_2O$) as shown the crystal model in Fig. 4. During this process, heavy metal ions are replaced with Ca ions so that heavy metals are captured in the crystal. With processing by the SCWT, heavy metals in the sludge are solidified and then become difficult to leach out by water because there are relatively large amounts of Ca and SiO_2 in the sludge[2].

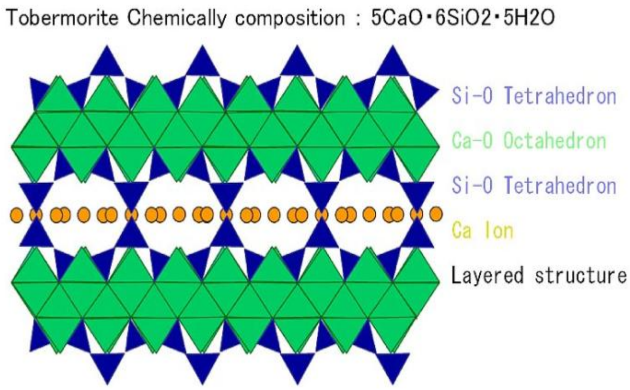

Fig. 4 Crystalline structure of tobermorite

In Japan, there are two types of standards for soil in terms of environmental safety. They are the Leaching Standard and the Content Standard [3]. The Leaching Standards are set for the control of pollution of heavy metals in ground water used for drinking water. This standard for heavy metal concentrations are used to compare the results of the leaching experiment. Table 7 shows the results of the leaching experiment performed for the dehydrated sludge and treated sludge by the SCWT method compared to the soil environmental standard. It can be seen that heavy metal concentrations by the SCWT method were below the values of soil environmental standard in Japan.

In order to supply silicate for the material balance of forming the tobermorite in sludge, it is necessary to add materials such as rice chaff. The rice chaff plays an important role not only to adjust the appropriate water content for preventing liquefaction but also to supply silicate material enabling binding of heavy metals.

Table 7 Heavy metal concentration in the dewatered sludge and in the subcritical water treated sludge compared to the soil environmental standard in Japan

Element	Dewatered sludge	Treated sludge	Environmental standard for soil,Japan
Lead (mg/kg)	0.003	0.001	0.01
Arsenic (mg/kg)	0.025	0.009	0.01
Mercury (mg/kg)	ND	ND	0.0005
Cadmium (mg/kg)	0.0004	ND	0.01
Chromium (mg/kg)	ND	ND	0.05
Total cyanides (mg/kg)	ND	ND	Undetected

CONCLUSIONS

In this study, demonstration experiments with sewage sludge and livestock manure were undertaken using the subcritical water treatment (SCWT) reactors by varying temperature, pressure, and time. As for the results, it was found that the aerobic fermented compost was easily obtained by setting the weight ratio of the rice chaff to the wastes at approximately 30 % to adjust the appropriate water content for preventing liquefaction. The appropriate conditions regarding treating sewage sludge were found to be at a temperature of 100 °C - 200 °C and a pressure of 1.0 MPa - 2.0 MPa. Pig manure and chicken excreta were treated around the similar conditions. The content analysis of the treated samples gave the excellent results relevant for a fertilizer. The results show good elements for a general organic fertilizer, and it was confirmed to be pathogen free, as well as hazardous chemicals with heavy metals low enough to satisfy standards for continuous applications on agricultural lands.

Abbreviation

Subcritical water treatment (SCWT)

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