J. Environ. Syst. and Eng., JSCE, No. 734/VII-27, 167-174, 2003 May

TREATMENT OF SEWAGE SLUDGE BY COLD CIRCULATION FREEZE-DRYING EQUIPMENT – WITH A VIEW TO RECOVERING BIOMASS RESOURCES

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Cold Circulation Freeze-Drying Equipment of a pilot plant was produced to freeze-dry sewage sludge with a view to recovering cellulose of a biomass resource. The recovery yields of solids and water by the produced equipment were 90.0–96.0% (w/w) and 170–250% (v/v), respectively. The moisture content of the dewatered sewage sludge was about 12%. The cost of the freeze-drying treatment for sewage sludge was estimated to be approximate for present sewage sludge treatment using liquefied natural gas (LNG) waste cold heat. These results also suggested that freeze-drying treatment for sewage sludge did not occur by-products (dioxin, etc.) derived from incineration.

Key Words : *freeze-drying, sewage sludge, cellulose, biomass, recovered resources*

1. INTRODUCTION

The amount of sewage sludge from sewage treatment plants is increasing year by year with the increase of sewered population [1]. Researches in the dewatering of sewage sludge have been carried out seeking means of utilization because sludge accounts for 40% of all industrial waste in Japan [2]. The dewatering process is one of the most important factors in sewage sludge treatment because of moisture content that can reach 97%. All sewage sludge is dewatered to reduce waste sludge volume. Although about 50% of the dewatered sewage sludge is incinerated then discarded in landfill sites, its remainder is treated as such sludge recycle resources as energy through anaerobic digestion, solid amendments by dewatered and dried sludge, construction materials by incineration ash and melted slag, etc [3].

Sewage sludge comprises protein, carbohydrate, crude fiber (cellulose), polysaccharide, etc [4]. Cellulose in sewage sludge is particularly difficult to remove and degrade because it is the formation of rigid and insoluble microfibrils [5]. We have carried out the following research regarding cellulose in sewage sludge: (1) development of an analytical method for cellulose [6]; (2) a survey of cellulose profiles in sewage treatment plants [7]; (3) development of a recovery method for cellulose in primary sludge [8]. We found that primary sludge contains 17 % (w/w) cellulose as a biomass resource, and that cellulose can be recovered using an autoclaving treatment with diluted sulfuric acid. However, use of dried sewage sludge as a raw material appeared to be necessary for effectively recovering cellulose from sewage sludge [9].

A more efficient treatment for dewatering sewage sludge targets not only reducing the amount of sewage sludge but also reducing the cost and energy of treatment. Typical dewatering treatments comprise mechanical and physical processes such as centrifugal dewatering, beltpress filter, filter press, air-heating treatment and vacuum dewatering [10]. The dewatering method or a combined method is chosen based on the characteristics of the sludge for operations at sewage treatment plants [11]. However, those treatment methods are not useful as the dewatering pretreatment for recovering cellulose because their conditions allow the possibility of denaturation of the cellulose. Therefore, the best dewatering treatment for cellulose recovery was thought to be a freeze-drying treatment comprised as a dewatering process in the food industry.

Freeze-drying means to directly sublimate water from ice to gas in a vacuum after freezing it solid, and this is thought to be an effective process for dewatering solids. The advances of freeze-drying are unchanged appearance, rapid reconstruction, lightweight product, storage stability, etc [12]. In addition, the freeze-drying treatment can retain odors in the solid because the vapor pressure of odors is lower than that of ice [13]. This suggests the ability to imprison offensive odors. The freeze-drying treatment was, therefore, thought the best method as dewatering of sewage sludge for recovering cellulose.

This study proposed Cold Circulation Freeze-Drying Equipment as a pilot plant for dewatering sewage sludge and describes a performance test using its equipment.

2. FREEZE-DRYING THEORY

Three familiar phases of water (ice, liquid and vapor) coexist at only one temperature and pressure called the triple-point [13]. The triple-point conditions of pure water are 0.0075°C and 613.281 Pa. At all other temperatures and pressures, not more than two of the phases can exist simultaneously. During evaporation at such pressures, water passes directly from the solid state to the vapor state. This particular evaporative process is known as sublimation of the basic water-transport phenomenon thought to occur in freeze-drying [14].

The mechanism of freeze-dry conditioning is a physical process whereby water separates from impurities as ice crystals form under vacuum condition. Ice is a solid that consists of a crystallographic arrangement of water molecules, and the impurities are also frozen and form the flocs. Treatment under the triple-point of water can separate water and impurities since this treatment supplies the heat of sublimation under the triple-point [15].

In the case of sludge, the relation between solid and water in sludge is one of the important factors for dewatering by the freeze-drying process. Sludge consists of colloidal sludge and water [16]. The colloidal sludge contains pore water, surface water, free water and other forms [17]. In the case of freeze-drying process, those forms are separated from solids as frozen pieces of water [18]. This means that there are also flocs of only sludge. The surface area of sludge flocs diminishes, and the sludge flocs can decrease surface water that attaches on sludge surface [19]. As a result, sludge is separated from water because the water in the sludge is excluded from floc phases by ice. This theory explains how sludge is dewatered by sublimation in a vacuum condition.

3. PRINCIPLE OF COLD CIRCULATION FREEZE-DRYING EQUIPMENT

Although the freeze-drying treatment in the food industry needs the stable treatment for keeping the high qualities of raw foods without changing shapes, the case of freeze-drying treatment need to grind sewage sludge for reducing the amount of waste sewage sludge. This reason leaded that the freeze-drying treatment for sewage sludge needed continuative cold contact with solid. Therefore, Cold Circulation Freeze-Drying Equipment would be expected the best treatment method for freeze-drying sewage sludge. A continuous reaction between sewage sludge and the high pressure cold would be considered an atmospheric freeze-drying [20], and the equipment for sewage sludge was thought to be possible without a vacuum device. This equipment comprised a circulative sludge-freezing reactor for contact of cold and solid from the above-mentioned reasons.

Cold Circulation Freeze-Drying Equipment as a pilot plant for sewage sludge is shown in **Fig. 1**.

4. MATERIALS AND METHODS

(1) Materials

Primary sludge collected from a sewage treatment plant (separate sewer system) was used as a standard substance. The collected sewage sludge was centrifuged for 10 minutes at 3,000 rpm (H-103N; Kokusan, Tokyo, Japan). It was washed with distilled water like the analysis of suspended solid (SS), then the washed sewage sludge was freeze-dried. The contents of cellulose, SS and ash in the collected sample were analyzed and found to be 22.7% (w/w; dry weight), 10,910 mg/L and 12% (w/w; dry weight), respectively. Cellulose content was analyzed by a novel method of determining cellulosic substances in raw wastewater and sludge [6]. SS and ash were analyzed by the standard

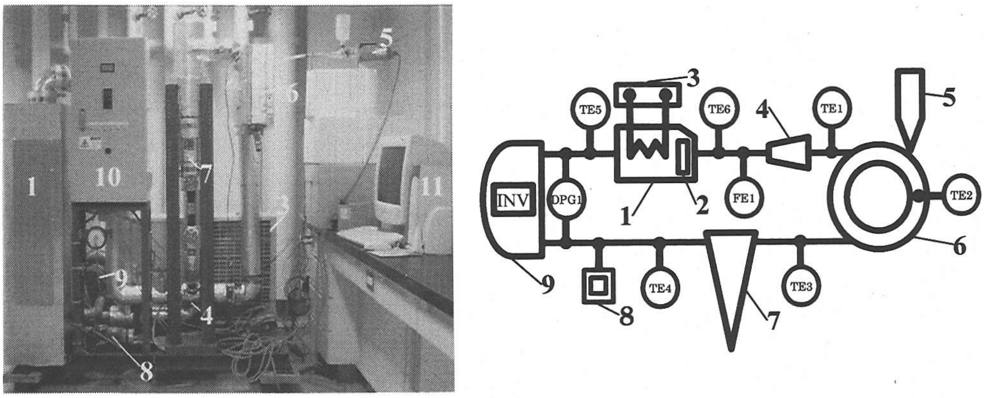

Fig.1 Photograph and schematic of Cold Circulation Freeze-Drying Equipment: (TE) thermo sensor; (FE) flow meter; (DPG) pressure drop sensor; (INV) inverter; (1) Air cooler; (2) Duct heater; (3) Condensing unit; (4) Hopper ejector; (5) Mohno-pump; (6) Sludge-freezing reactor; (7) Cyclone separator; (8) Vacuum pump; (9) Ring blower; (10) Control panel; (11) PC.

methods for the examination of wastewater [21]. The moisture contents (80, 90, 95 and 99%) of the standard substances were prepared by the mixture of the dried primary sludge and distilled water.

(2) Procedure of freeze-drying

Figure 1 shows a photograph and a schematic of Cold Circulation Freeze-Drying Equipment. This equipment comprised a control panel, a condensing unit, a ring blower, an air cooler, a duct heater, a vacuum pump, a mohno-pump (3NY06; Heishin Engineering & Equipment, Hyogo, Japan), a sludge-freezing reactor (volume; 580 mL) and a cyclone separator. A personal computer was used for observing temperature (TE 2–TE 6) and cold flow (FE 1) in real time. The results of experiments displayed only TE 2 temperature on each graph.

This equipment froze and dewatered sewage sludge as following: (1) the flow generated by the high jet blower was continually cycled into the sludge-freezing reactor; (2) the sample injected into the reactor was frozen on the wall of the reactor by the flow; (3) the sample was dried by temperature control in the sludge-freezing reactor.

The vacuum pump was not used because the pre-experiments were confirmed atmospheric freeze-drying [20]. Although the cyclone separator was used as a trap to avoid the entering of solids into the ring blower, it was not used for recovering solids because of the long recovering time compared with other treatment in pre-experiments. The net (mesh: 1 mm, material: non-woven fabric of high density polyethylene) was set up between the cyclone separator and the sludge-freezing reactor to protect the flow of solid.

The freeze-drying processes were divided into two modes of a freezing process and a drying process. The freezing process was as following; (1) the sludge-freezing reactor was pre-cooled down until TE 2 was -10°C at 50 Hz (inverter frequency of ring blower); (2) the sample was injected into the reactor by the mohno-pump at 60 mL / a minute until the pressure at DPG 1 reached 9.7 kPa; (3) the sample was frozen for 30 minutes; (4) for the sample of moisture content 99%, the sample was again injected to repeat processes (1) to (4). In the drying process, after the freezing process (4), the frozen sample was dried at 4°C (TE 1) until the temperatures TE 2 and TE 3 were the same. The pre-experiments showed that the samples were not frozen in the sludge-freezing reactor when TE 1 was adjusted over 5°C.

For the sample of moisture content 80%, the mohno-pump was not used due to structural problems in the pump. The sample was previously put into the sludge-freezing reactor. The pre-cooling and freezing of sludge were carried out at the same time. This process was continued for 30 minutes after TE 2 was -10°C. The drying process was carried out under the same conditions as other samples.

After freeze-drying process, water, which was separated during the freeze-drying process, was recovered by defrosting at the air cooler.

Drying time (h), drying rate (g/h), solid recovery (%), water recovery (%), and moisture content (%) of recovered samples were analyzed for the performance evaluation of the freeze-drying equipment.

5. RESULTS

Figure 2 shows the results of moisture contents 99–80%, and A, B, C and D in **Fig. 2** show the pre-cooling process, the sludge injection process, the freezing process and the drying process, respectively. These results in **Fig. 2** show the latest data that were measured after several experiments by the same sample and the experimental conditions, and the results of the latest data and the former data were almost same and reproducibility was confirmed.

(1) Moisture content 99%

Total amount of sample was 840 mL (dry weight; 8.4 g) at repetitions. There was a gradual decrease in the amount of injected sample process by process because the dried solid was accumulated in the sludge-freezing reactor after each process. As a result, treatment time decreased with each process. Although the 1st process appeared not to drastically decrease the cold flow as the sample was injected, cold flows decreased drastically in the 2nd process. These results indicated that the injected sludge was frozen on the internal walls of the sludge-freezing reactor in the 1st process and was frozen with the previously dried sludge after the 2nd process. This reduction in the cold flow decreased with every process because of the decreasing amount of sample.

(2) Moisture content 95%

The cold flow (FE 1) drastically decreased from 59.5 to 49.0 m^3/h as sample was injected into the sludge-freezing reactor. This was due to flow resistance of clogged sludge in the sludge-freezing reactor. The temperature (TE 2) increased to 2°C (14°C temperature increase) for only 20 minutes after finishing the freezing process. This assumed that the surface ice was more sublimated than the other parts of the frozen sludge.

(3) Moisture content 90%

The temperature (TE 2) was drastically increased from -10 to 1.6°C with cold flow decreased from 59.5 to 36.8 m^3/h for 3 minutes from the sample injection time. The temperature TE 2 was increased from -12 to 3.4°C for about 20 minutes after finishing the freezing process. The sample was dried for 51 minutes from the start of the drying process because TE 2 and TE 3 were the same. The cold flow increased from 35.8 to 50.3 m^3/h at the same time, and the results also assume that the frozen sludge was dried by sublimation.

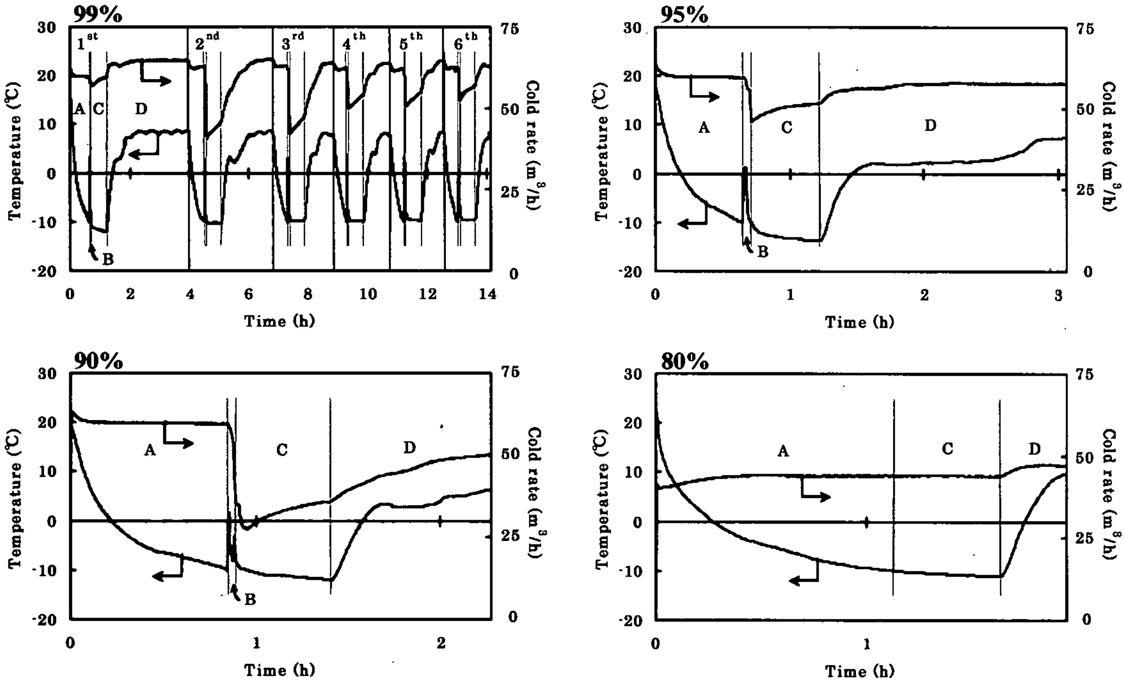

Fig. 2 Results at 99-80% moisture contents: (A) pre-cooling process; (B) sludge injection process; (C) freezing process; (D) drying process.

(4) Moisture content 80%

The pre-cooling process was longer for than the experiments at moisture content 90–99% because the pre-cooling process and the freezing process took place at the same time. The cold flow slowly increased from the start of this experiment. This showed that the sublimation began immediately from the frozen part of sludge. The increasing of TE 2 and FE 1 from the start of the drying process showed that the frozen sludge was sublimated as drastically as for moisture content 90–99% above.

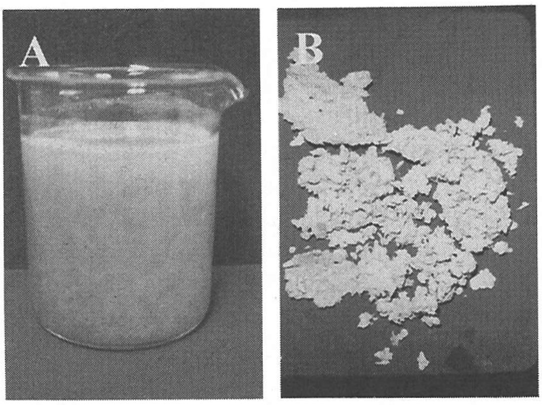

Fig. 3 Photographs before (A) and after (B) freeze-drying treatment. Moisture content: A = 99%, B = 11.7%.

6. PERFORMANCE EVALUATION OF COLD CIRCULATION FREEZE-DRYING EQUIPMENT

Table 1 shows the drying rates and recovery yields for solids and water. The recovery yields for solids were 90.0–96.0% (w/w), and the other solids were trapped in the cyclone separator. Although the net with smaller mesh was thought to be more efficiency for high recovery yields, this was not adapted because the net resisted the flow.

The moisture content of recovered samples was 11.5–12.1% (w/w) (**Table 1**). **Figure 3** shows the photographs of before (A) and after (B) freeze-drying treatment by the sample of moisture content 99%. The photographs visually confirmed that the sample was freeze-dried by the produced equipment because the moisture content of the dried sample was 11.7% (w/w). This showed that the produced freeze-drying equipment could effectively dewater sewage sludge.

The recovery yields for water at the air cooler were 170–250% (v/v) (**Table 1**), and there were about 2 times the amount of the initial sample. This showed that water in the atmosphere was frozen with the sample in the freeze-drying process (pre-cooling and freezing) and recovered by sublimation. The drying rates were faster with decreased moisture content. This result suggested that the low moisture content sludge was effectively freeze-dried.

If the sample (1 g; dry weight) of moisture content 96%, which is the moisture content of most crude sewage, was freeze-dried by this equipment, a drying time and a drying rate was calculated 9 minutes and 6.7 g/h from the drying rates in **Table 1**, respectively.

The separated water at the air cooler by defrosting was thought to be same constituent with distillated water. Because its water became gas by the

Table 1. Drying rates and recovery yields for solid and water.

Experiment conditions				Drying		Recovery		
MC[1]	Process	Sample		Time[2]	Rate	Solid	Water	MC[1]
(%)		(mL)	(g)		(g/h)	(%)	(%)	(%)
99	1st	180	1.80	2h44m	0.7	–	–	–
	2nd	170	1.70	1h45m	1.0	–	–	–
	3rd	145	1.45	59m	1.5	–	–	–
	4th	130	1.30	54m	1.5	–	–	–
	5th	115	1.15	46m	1.5	–	–	–
	6th	100	1.00	31m	1.9	–	–	–
	Total	840	8.40	7h39m	1.1[3]	93.5	170.2	11.7
95	–	180	9.00	51m	10.6	90.0	210.0	12.1
90	–	180	18.00	52m	20.8	91.7	234.6	11.5
80	–	100	20.00	19m	63.2	96.0	250.0	12.0

(1) Moisture content; (2) h = hour (s) and m = minutes; (3) an average (1st to 6th).

sublimation during the freeze-drying process.

7. FREEZE-DRYING TREATMENT COST FOR SEWAGE SLUDGE

The freeze-drying treatment is basically carried out to prepare for the sublimation latent heat applied by some method. Freeze-drying cost is generally higher (10–20 times) than other drying treatments because the freeze-drying treatment required a longer drying time [12]. This is the greatest problem for freeze-drying treatment, and the solution of this problem is thought to point toward more effective freeze-drying treatment. To reduce the freeze-drying cost, the idea was to increase the amount of treatment per process but also using LNG waste cold heat instead of refrigerators. Freeze-drying cost by LNG waste cold heat was 75% lower than treatment by refrigerator [22].

Figure 4 shows a proposal for a freeze-drying system by LNG. LNG is liquefied by reducing its temperature to -162°C at atmospheric pressure [23]. LNG is used mostly for electric power generation [23]. The waste cold heat is produced when the generator turbine is rotated by vaporized LNG and generates electricity [23]. The produced waste cold heat is used as storage refrigerant in food industry [22]. However, only a small amount of waste cold heat is utilized as refrigerant at present. Therefore, the waste cold heat is supposed to utilize for freeze-drying treatment for sewage sludge and to reduce the freeze-drying cost. At sewage treatment plants, the freeze-drying treatment is possible to treat sludge by freeze-drying alone instead in present sludge treatment facilities (dewatering facility, drying facility, incinerator, melting furnace, exhaust gas treatment equipment, waste heat boiler, etc.). The freeze-drying treatment, used in place of present treatments, is assumed to reduce environmental pollution due to the by-products (dioxin, etc.) of incineration.

The treatment cost of usual sewage sludge treatment (dewatering → incineration → melting) is 14,000–44,000 yen per ton in Japan [24, 25, 26, 27]. With freeze-drying treatment using LNG waste cold heat, the cost, in Japan, was calculated at 10,000–20,000 yen per ton [12, 22]. These findings suggest that freeze-drying treatment using LNG waste cold heat could be performed at the same cost as present treatments of sewage sludge. Combination sewage treatment and LNG plants are therefore suggested as a city planning strategy for the future because nearly all are built on landfill sites or near estuaries.

8. CONCLUSIONS

Dewatering and incineration for the reduction of sewage sludge are carried out by mechanical and physical processes. Sewage sludge contains cellulose as a biomass resource, and the recovery of cellulose is also related to the reduction of the sewage sludge. This study investigated whether or not Cold Circulation

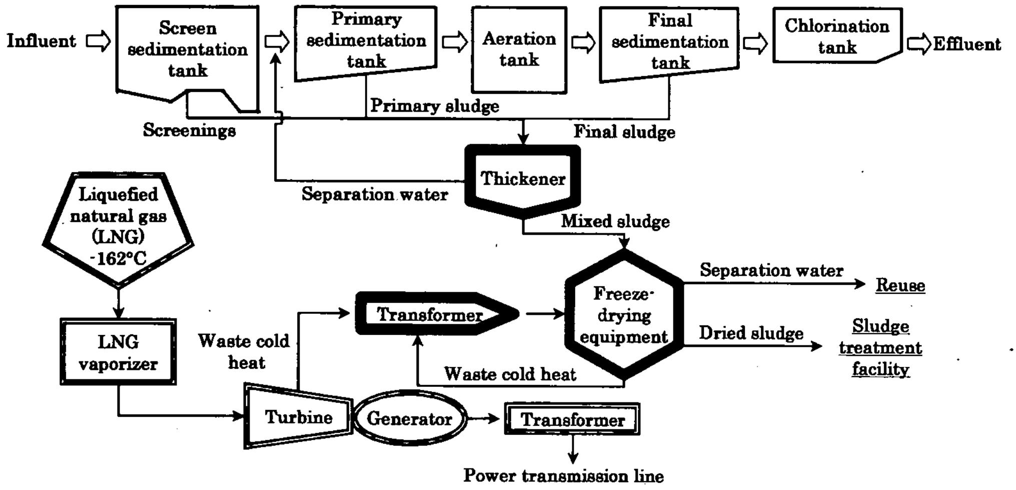

Fig. 4 Proposed of freeze-drying system for sewage sludge using LNG waste cold heat: (Black line) wastewater treatment plant; (double line) LNG power generation plant; (bold line) freeze-drying plant.

Freeze-Drying Equipment could be applied to the dewatering of sewage sludge. Cold Circulation Freeze-Drying Equipment was produced as a pilot plant, based on information from references, and experiments were carried out with the produced equipment.

The produced equipment could freeze-dry sewage sludge to about moisture content 12% with grinding solids. The recovery yields for solid and water were 90.0–96.0% (w/w) and 170–250% (v/v), respectively. The drying rates were increased by decreasing moisture content. The cost of freeze-drying using LNG waste cold heat was calculated to be the same as the present treatment of sewage sludge, and combined sewage treatment and LNG plants were proposed as a city planning strategy for the future.

ACKNOWLEDEMENT: This work was supported by Project Research of Shizuoka Prefecture.

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(Received August 29, 2002)