

A New Treatment of Sewage Sludge by Direct Thermochemical Liquefaction

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A new treatment of sewage sludge was developed. When sewage sludge was treated with sodium carbonate under pressure of nitrogen at above 300 °C, heavy oils were obtained in sufficiently high yields. Energy balance is briefly discussed, showing this method is feasible in terms of input/output energy ratio.

Sewage sludge is an unavoidable by-product of municipal wastewater treatment and it is estimated that about 50 million tons of sewage sludge (based on 98% moisture content) are produced annually in Japan. Though about 80% of this sludge is disposed of by landfilling or ocean dumping, such a landfill disposition poses a serious problem with respect to pollution control and due to the difficulty of finding disposal sites within reasonable distances from large population areas. In large urban area, incineration is commonly employed for sludge volume reduction. However, incineration is a high cost disposal technique because it needs a great deal of energy inputs for evaporation of water in the sewage sludge. In order to cope with these difficulties, research and development for utilization of sewage sludge as usable energy source have been done, as a result, new processes such as pyrolysis, starved-air combustion, and autogenous combustion have been proposed.^{1,2)} However, these processes are still in a development stage and do not promise to be a net energy producer at the present time.

Recently direct thermochemical liquefaction has been studied for liquid fuel production from various materials such as wood and other organic wastes.^{3,4)} In our previous paper,⁵⁾ it was demonstrated that fermentation wastes can be converted efficiently to heavy oils by heating them with alkali metal salt as a catalyst in an aqueous phase under pressurized inert gas. In the present study, this direct thermochemical liquefaction was applied to sewage sludge in an attempt to develop a new treatment method of sludge disposal.

Raw sludge containing a mixture of primary and waste activated sludge from a sewage treatment facility was used for this study. The sludge is characterized in Table 1. This sludge was dewatered to a 75% moisture content by a belt press dehydrator after a polyelectrolyte coagulant was added to the raw sludge at sewage plant. The term "volatile solid" means the content of organic material. The amounts of moisture content, volatile solid, crude protein, crude fat, and crude

fiber were measured by weight loss on drying at 105 °C, ignition loss of dry solid at 600 °C, Kjeldahl method, ether extraction, and filtration method, respectively. The analyses of elemental composition and heating value were done according to JIS method.

Table 1. Analysis of the sewage sludge

Sludge property			
Moisture content/wt%	75.0	Volatile solid ^{a)} /wt%	84.0
		Heating value ^{a)} /MJ kg ⁻¹	17.2
Chemical composition ^{b)} /wt%			
Crude protein	30.0	Crude fat	13.0
		Crude fiber	33.0
		Non-fibrous Carbohydrate ^{c)}	24.0
Elemental composition ^{b)} /wt%			
C	49.0	H	8.0
		O ^{c)}	39.3
		N	3.7

a) On a moisture free basis.

b) On a moisture and ash free basis.

c) Calculated by difference.

The experiments were carried out using a 300 ml autoclave by changing the reaction temperature, since other operating variables such as pressure and holding time were not substantially dominant in this liquefaction. Figure 1 schematically shows the experimental apparatus with a pressure control circuit.

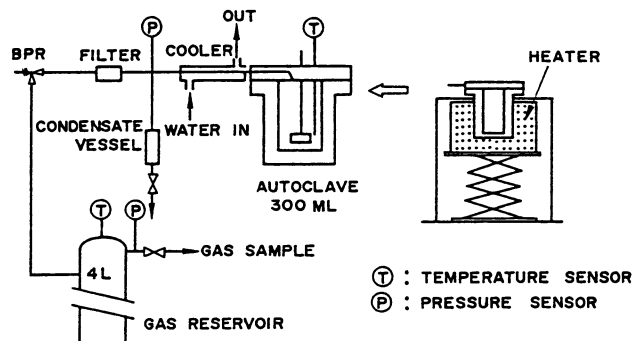


Fig. 1. Experimental apparatus.

After charging 100 g of sewage sludge and 1.25 g of sodium carbonate (anhydrous) into the autoclave, the autoclave was deaerated and pressurized with nitrogen to a required pressure. Then, the temperature was raised to a given reaction temperature within 50 min, while the pressure was kept at a constant pressure by releasing the overpressure through a back pressure regulator (BPR) into a gas reservoir. Once the temperature reached to the given temperature, the

autoclave was immediately cooled down to room temperature. The volume and composition of gas evolved were determined by a gas meter and a gas chromatograph. The autoclave was then opened and the reaction mixtures were removed and stored.

The reaction mixtures were separated into three phases: the heavy oil, the solid residue and the aqueous phase. After the reaction mixtures were extracted with dichloromethane as a solvent, the solid residue was separated by filtering both of the extract and the raffinate. The dichloromethane was evaporated from the extract using a rotary evaporator. The heavy oil, which was the solvent soluble material, was obtained as a dark brown viscous material in this manner. The solid residue was defined as the material insoluble in both water and the solvent. The aqueous

phase, which contained water soluble and the solvent insoluble material, was the filtrate of the raffinate.

The conversion yield, energy yield, and energy consumption ratio (ECR) were defined as:

$$\text{Conversion yield/\%} = \frac{\text{weight of volatile materials in each phase}}{\text{weight of volatile materials in sewage sludge}} \times 100 \quad (1)$$

$$\text{Energy yield/\%} = \frac{\text{energy in heavy oil produced}}{\text{energy in sewage sludge}} \times 100 \quad (2)$$

$$\begin{aligned} \text{ECR} &= \frac{\text{energy required for liquefaction reaction}}{\text{available energy in heavy oil produced}} \\ &= \frac{\Delta Q}{\gamma \times Y_0 \times (1-W) \times VS \times H_0} \quad (3) \end{aligned}$$

where, ΔQ means the energy required to heat the sewage sludge up to a given reaction temperature, γ is an efficiency of available combustion energy, Y_0 is a heavy oil yield, W and VS are moisture content and volatile solid, respectively, of a starting material, and H_0 is the lower heating value of the heavy oil produced. A ratio (ECR value) greater than unity means the overall system consumes more energy than it produces, while a ratio of less than unity indicates the system is a net energy producer.

The effects of the reaction temperature on the conversion yield are demonstrated in Fig. 2. In this Figure, the heavy oil yield increased with the increase

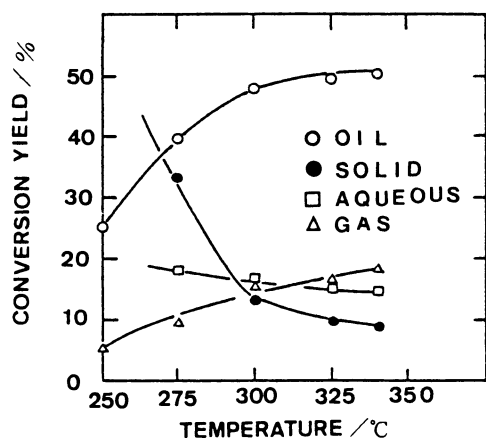


Fig. 2. Effects of the reaction temperature on the conversion yield.

in the reaction temperature up to 300 °C at which the heavy oil yield reached almost a maximum. By contrast, the conversion yield of solid residue decreased as the temperature increased, and approached to a minimum value at above 300 °C. It is obvious from these data that most of the unreacted materials remained in the solid residue below 275 °C. On the other hand, the conversion yield of gas increased, but the yield of aqueous phase material slightly decreased with the rise in temperature. The decrease of the yield of aqueous phase material may be attributed to the decomposition of acidic compounds which were formed during the liquefaction, because the pH of

aqueous solution became more basic as the temperature increased.

Heavy oils obtained in this study contained 67-70% carbon, 7-9% hydrogen, and 21-25% oxygen. The heating values were 31-33.5 MJ kg⁻¹ and the average molecular weights were about 370 over the reaction temperature range of 250 to 340 °C. Accordingly, the reaction temperature does not have any pronounced effect on the properties of heavy oil.

Figure 3 shows an example of material balance of liquefaction process obtained at 340 °C. After the liquefaction and the separation, 50% of the starting volatile materials was converted to heavy oil, 9% was to the solid residue, 15% was to the aqueous phase material, and 18% was converted to gas, mainly CO₂. The remainder 8% was regarded as an unavoidable loss due to vaporization of lighter fractions during

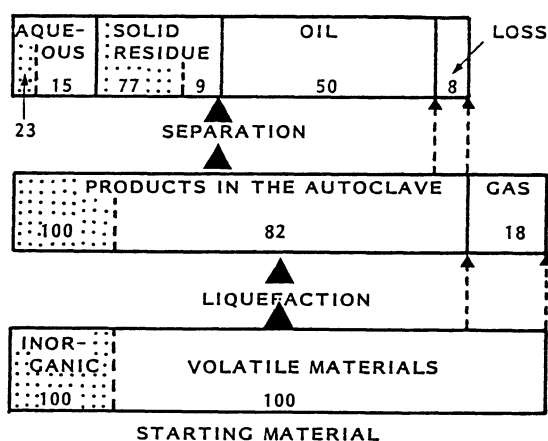


Fig. 3. Material balance of liquefaction process.

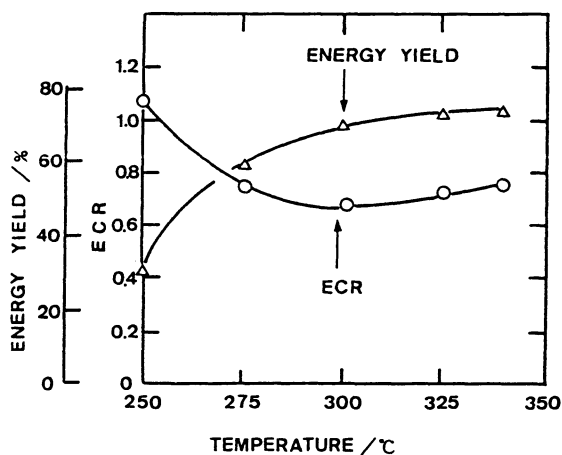


Fig. 4. Relationship between ECR, energy yield and reaction temperature.

the removal operation of dichloro-methane. On the other hand, 77% of inorganic materials was in the solid residue and 23% was in the aqueous phase. The amount of inorganic materials in the aqueous phase nearly corresponds to the weight of sodium carbonate added as a catalyst.

Figure 4 shows the relationship between ECR, energy yield and reaction temperature. On the calculation of ECR by Eq. 3, data obtained in this study were used; Y_0 is heavy oil yield, W is 0.75, and H_0 is 31 MJ kg^{-1} , respectively. It was assumed that VS was 0.65 because VS of the sludge in this experiment was fairly high (0.84) compared with the VS of the standard sewage sludge. It seems also reasonable to assume that the efficiency (γ) is 0.6 and the specific heats of water and dry solid, both of which are involved in ΔQ , are 4.18 and 1.25 $\text{kJ kg}^{-1} \text{ } ^\circ\text{C}^{-1}$. From Fig. 4, it can be seen clearly that the liquefaction process operated at above 275 $^\circ\text{C}$ is a net energy producer, because the ECR is less than unity above that temperature. The ECR has a minimum value at 300 $^\circ\text{C}$, while the energy yield increases as the reaction temperature increases. Accordingly, the process operated at 300 $^\circ\text{C}$ is most efficient.

In conclusion, the treatment of sewage sludge by direct thermochemical liquefaction could be expected to be sufficiently profitable. Though further studies including the availability to various kinds of sewage sludge and the survey of more suitable catalyst are needed, this method may possibly prove to be a good alternative means for sludge disposal.

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