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論 文 内 容 要 旨

EFFECTS OF SUBSTRATE LOADING RATE AND TEMPERATURE ON METHANO- GENESIS PROCESS IN ANAEROBIC DIGESTION

The anaerobic digestion process is one of the major biological waste treatment processes in use today. This process has been popular in the waste treatment field because it has many advantages such as a high treatment efficiency and methane producing ability.

Although the energy recoverable anaerobic digestion process has been used in organic waste treatment for a century, the potential waste treating ability of this process has not yet been demonstrated. Conventional single-phase digestion restricts physiologically different anaerobic organisms to a reactor and consequently causes the digestion to need a longer retention time. A two-phase digestion process has been recently suggested to improve the anaerobic treatment efficiency with a shorter digestion time which satisfies the increasing waste volume and lower

investment requirements. To develop this new waste treatment technology, an understanding to the process kinetics is necessary. But the product from an acid-forming digester consists of many kinds of volatile fatty acids instead of single ones. Besides, the organisms responsible for degradation of volatile fatty acids (VFA) are metabolically dependent upon each other for survival, i. e., they are ecologically syntrophical. The syntrophical associations among these anaerobic organisms must be emphasized. Moreover, it is doubtful that whether kinetic information using single substrate can be used for predicting methanogenesis from a mixture of VFA. Therefore, to elucidate the process kinetics of methanogenesis further research is needed by using a mixture of VFA with a composition approaching to plant-scale digestion.

In this study a mixture of the major intermediate products of anaerobic digestion, i. e., acetic, propionic and butyric acids were used as the carbon source in methanogenic experiments. The study described in this thesis was undertaken using chemostat-type digesters to investigate the effects of loading rate and temperature on the methanogenesis process in anaerobic digestion.

This thesis includes the following six chapters.

CHAPTER I INTRODUCTION

The purpose of this study was described in this chapter.

CHAPTER II THE METHANOGENESIS PROCESS IN ANAEROBIC DIGESTION

A review on the methanogenesis process was undertaken to develop a framework for this study on the process.

CHAPTER III EFFECTS OF LOADING RATE AT VARIOUS RETENTION TIMES

This chapter elucidated the effects of the loading rate on the methanogenesis process in which a mixture of acetic, propionic and butyric acids was used as the substrate at various retention times. The effects of the loading rate on the parameters such as substrate utilization, distribution of component volatile fatty acids in effluent, methane production, and morphological types of microorganisms were investigated by using chemostat-type anaerobic digesters. The characteristics of the methanogenesis process using a mixture of volatile fatty acids were compared kinetically with that using a single component of volatile fatty acids. High concentration, 20000 mg COD/l, of multisubstrate was used. The digesters were main-

tained at 35 °C. The following conclusions can be drawn from the experimental results.

1. Methanogenesis occurred normally up to an HRT of 2.91 days with a maximum specific substrate utilization rate of 17.1 l/day, a saturation constant of 166 mg COD/l, a growth yield of 0.30, a decay coefficient of 0.099 1/day, and a minimum SRT of 2.42 days.
2. Methanogenic digesters using a mixture of volatile fatty acids can operate stably and effectively at shorter hydraulic retention times than digesters using single components of volatile fatty acids individually. The stability and effectiveness can be proved kinetically.
3. An increase in substrate loading rate by shortening retention time will affect the acetate and propionate degradation significantly and adversely.
4. The methane production is independent of the retention time and has a value of 0.89 g COD or 0.313 l at STP per g COD substrate utilized. The average percentages of methane produced from acetic, propionic, and butyric acids were 39.2, 27.3, and 33.5 %, respectively.
5. Bacilli are the predominant microbial species. While coccoid and sarcinae appear in digester with short retention times. The substrate-specific fraction microbial masses were 74, 10, and 16 % for acetic, propionic, and butyric acids, respectively.

CHAPTER IV EFFECT OF LOADING RATE AT VARIOUS INFLUENT SUBSTRATE CONCENTRATIONS

This chapter elucidated the effects of substrate loading rate on the methanogenesis process at various concentrations. The retention time was controlled at 4.5 days and the influent substrate concentration ranged from 10000 to 90000 mg COD/l.

Specific conclusions from the investigations presented in this chapter include:

1. At an HRT of 4.43 days, methanogenesis occurred normally up to an influent substrate concentration of 70000 mg COD/l or a high loading rate of 15.8 g COD/l-day.
2. Methanogenesis process in anaerobic digestion by using a mixture of volatile acids can progress stably at higher influent substrate concentrations than that by using single component of volatile fatty acid individually.
3. An increase in influent substrate concentration will adversely affect the propionate degradation strikingly.
4. The methane production is independent of the influent substrate concentration and has a value of 0.80 g COD or 0.281 l at STP per g COD substrate utilized.
5. Bacilli are the predominant microbial species. While sarcinae appear in a digester with

high influent substrate concentrations.

CHAPTER V EFFECT OF TEMPERATURE

The effects of temperature were studied at the temperatures ranging from 20 to 50 °C, and the data were used to model the temperature dependent characteristics of the methanogenesis process. Temperature effects on the other parameters of the process were also studied. The following conclusions can be drawn from the experimental results:

1. The optimum operating temperature for the mesophilic methanogenesis process is 35°C.
2. Propionate-degrading acetogens drop in activity markedly at 40°C at the mesophilic digestion temperature.
3. The methane conversion efficiency from substrate is independent of temperature and has a value of 0.313 liter (STP)/g COD of volatile fatty acid.
4. Bacilli are the predominant microbial species in the methanogenesis process using a mixture of volatile fatty acids. The predominance is independent of digestion temperature. While sarcinae and coccoid appear in digesters with short retention times.
5. The substrate utilization of sarcinae and coccoid are less adversely affected by a decrease in temperatures than are those of bacilli.
6. At the mesophilic range, the kinetic constant K_s decreases with increasing temperatures, but constants ν_{max} and Y_g increase with increasing temperatures. The exponential expressions correlating their temperature characteristics are as follows:

$$(K_s)_T = 230(0.944)^{(T-25)} \quad (20^\circ\text{C} \ll T \ll 35^\circ\text{C})$$

$$(\nu_{max})_T = 7.4(1.069)^{(T-25)} \quad (20^\circ\text{C} \ll T \ll 35^\circ\text{C})$$

$$(Y_g)_T = 0.02(1.306)^{(T-25)} \quad (25^\circ\text{C} \ll T \ll 40^\circ\text{C})$$

7. At the retention times longer than 8 days, the methanogenesis can progress normally and satisfactorily even at 25°C, and the substrate removal efficiency (more than 96 %) is the same as that at 35 °C. At the temperature range of 25 to 35 °C, the temperature dependent model is:

$$\frac{1}{SRT} = \frac{(0.160)(1.107)^{(T-25)} \cdot S}{230(0.944)^{(T-25)} + S}$$

CHAPTER VI SUMMARY AND CONCLUSIONS

The conclusions obtained from all the experimental results were summarized in this chapter.

審 査 結 果 の 要 旨

嫌気性消化は有機物の加水分解，揮発性脂肪酸生成およびメタン生成という多段的反応過程で構成されており，各段階の反応を最適に制御できる条件を知ることが，二相嫌気性消化法の設計および管理上必要である。本論文は酢酸，プロピオン酸および酪酸を混合して基質として用いた場合に，基質負荷速度および消化温度がメタン生成プロセスに及ぼす影響を明らかにしたもので，全編6章よりなる。

第1章は総論である。

第2章では，従来の研究を詳細に文献的に検討し，問題点を明確にしている。

第3章では，滞留時間がメタン生成プロセスに及ぼす影響について検討している。著者は，混合基質を用いる場合には，単一基質を用いる場合にくらべて短い滞留時間で安定かつ効率的な運転が可能となること，滞留時間を短くすることが酢酸およびプロピオン酸の分解にかなり悪影響を及ぼすこと，桿菌が優占菌種であるが，短い滞留時間の場合には球菌や連球菌が出現することなどの新しい知見を得ている。

第4章では，流入水基質濃度がメタン生成プロセスに及ぼす影響について検討している。著者は，混合基質の場合には，単一基質の場合にくらべて高い流入水基質濃度で安定的にメタン生成が行われること，流入水基質濃度の増大がプロピオン酸の分解に著しく悪影響を及ぼすこと，桿菌が優占菌種であるが，高い流入水基質濃度では連球菌が出現することなどの興味ある知見を得ている。

第5章では，消化温度がメタン生成プロセスに及ぼす影響について検討している。著者は，中温メタン生成プロセスの最適操作温度が35℃であること，消化温度40℃においてプロピオン酸分解酢酸生成菌の活性が著しく低下すること，基質からのメタン生成が消化温度と独立に揮発性脂肪酸のCODの1g当り標準状態で0.313ℓであること，温度と独立に桿菌が優先菌種であること，連球菌および球菌の基質利用が桿菌にくらべて温度低下の影響をあまりうけないことを明らかにしている。更に，著者は，基質除去に関する温度依存性モデルを提言している。これらは有用な知見である。

第6章は結論である。

以上要するに本論文は，二相嫌気性消化のメタン生成相について研究し，動力学的ならびに微生物学的特性を明らかにし，更に，基質除去に関する温度依存性モデルの提言を行ったもので，衛生工学の発展に寄与するところが少なくない。

よって，本論文は工学博士の学位論文として合格と認める。