Original Paper

Coagulation-flocculation treatment of waste milk on dairy farm - Effect of milk composition and pH on coagulation by inorganic coagulant -

Kenji Nekomoto[1], Mitsuyasu Sugizaki[1], Miku Ishihara[1], Shiho Ishikawa[1], Ken-ichi Kondo[2], Tadahiro Ariyoshi[3] and Tsugio Kobayashi[4]

[1] College of Agriculture, Food and Environment Sciences, Rakuno Gakuen University, Ebetsu, Hokkaido, 069-8501, Japan

[2] Vertical Integration Office Co., Ltd., Saitama, Saitama, 339-0045, Japan

[3] Chitose Factory, Orion Machinery Co., Ltd., Chitose Hokkaido, 066-0077, Japan

[4] Chuo-Orion Co., Ltd., Kumagaya, Saitama, 360-0811, Japan

Abstract Milking facilities on dairy farms generate waste milk (i.e., poor-quality milk that cannot be shipped, including colostrum and milk from cows with mastitis). However, it is not practical to treat waste milk using septic tanks, given the substantial capital investment and high costs associated with maintaining them. Therefore, in this study, we investigated a coagulation–flocculation treatment method as an alternative to septic tanks that involves the use of inorganic coagulants for the initial coagulation and polymeric flocculants for the secondary flocculation. Here, we report the results of laboratory experiments on the first coagulation reaction. When polyaluminum chloride (PAC) or polyferric sulfate (PFS) was used as the inorganic coagulant in the first stage, the pH of the waste milk shifted from neutral to acidic depending on the amount of the agent added. Complete clarification of the supernatant occurred between pH 5.6 and 4.4 (median value of 5.0) for PAC and between pH 5.0 and 2.8 (median value of 4.0) for PFS, indicating that flocculation was possible within a limited pH range. Significant correlations were found between the PAC input required to achieve a pH of 5.0 (median of the pH 5.6-4.4 range) and herd performance or milk components for 68 individual milk samples for milk fat percentage (r=0.342**), milk protein (r=0.361**), and non-fat milk solids (r=0.301**). Significant correlations were found between the PFS input required to achieve a pH of 4.0 (median of the pH 5.0-2.8 range) and herd performance or milk components for 72 individual milk samples for individual milk yield (r=−0.348**), days in milk (r=0.296*), milk fat (r=0.384**), milk protein (r=0.598**), and non-fat milk solids (r=0.596**). These results indicate that it is possible to coagulate, flocculate, and clarify waste milk by using an inorganic coagulant and a polymeric flocculant in the first and second stages of treatment, respectively. The amount of inorganic coagulant should be adjusted based on general factors such as milk fat percentage and milk protein while paying careful attention to the coagulation pH range.

Key words: coagulation-flocculation treatment, inorganic coagulant, polymeric flocculant, waste milk, dairy farm

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Corresponding author : Kenji NEKOMOTO (e-mail: nekomoto@rakuno.ac.jp)

Introduction

On dairy farms, large volumes of cleaning wastewater (hereafter referred to as "milking-related wastewater") are generated daily from milking facilities. The concentration of raw milk in wastewater from milking facilities (e.g., wash water from milk pipelines and parlor floors) is relatively low (approximately 1%–2%) [3]. Because the wastewater load is not high, simple activated sludge-based treatment facilities and sedimentation treatment are proposed as treatment methods [2,4,6]. However, when poor-quality milk that cannot be shipped (e.g., colostrum and milk from cows with mastitis) has to be discarded (hereinafter referred to as "waste milk") and is mixed with milking-related wastewater, the biochemical oxygen demand of this wastewater increases dramatically, resulting in a substantially higher wastewater load [4]. It is not practical to treat waste milk using septic tanks, given the substantial capital investment and high costs associated with maintaining septic tanks [5].

In recent years, various methods have been proposed for treating waste milk, including mixing the waste with slurry and applying it to fields or acid coagulation followed by composting [1,5]. However composting may not always be feasible, given that a single cow with mastitis can produce approximately 1 kg of waster milk per day [1], along with other factors such as the shortage of available farmland resulting from the expansion of farm operations [5].

Therefore, in this study, we investigated a coagulation-flocculation treatment method as an alternative to septic tanks and conventional composting. To date, few studies have investigated the use of coagulants to treat poorly biodegradable liquids such as waste milk. In the present study, the authors demonstrated through laboratory experiments that waste milk can be clarified by using inorganic coagulants for the initial coagulation and polymeric flocculants for the secondary flocculation. However, the inconsistency of coagulation on some dairy farms suggested that coagulation-flocculation may be affected by the composition of waste milk and other factors and that parameters such as the amount of coagulant added need to be adjusted to achieve effective treatment.

This study aimed to investigate the effects of milk composition (e.g., milk protein and milk fat) and pH on the initial coagulation reaction using inorganic coagulants and to provide basic data for achieving stable coagulation treatment on dairy farms.

Materials and Methods

1. Coagulation and clarification of waste milk

In the first stage of the treatment investigated in this study, an inorganic coagulant used for water treatment was added to waste milk to cause coagulation. In the second stage, after reaching a certain pH, a polymeric flocculant was added, causing the cloudy components of the waste milk to flocculate and separate. The reaction was judged to be complete when the supernatant became transparent.

2. Coagulant

The coagulants used in the first stage of treatment in this study were Taifloc SR (Taimei Chemicals Co., Ltd.), which is a polyaluminum chloride (PAC), and Polytetsu (Nittetsu Mining Co., Ltd.), which is a polyferric sulfate (PFS). The polymeric flocculant used in the second stage of treatment was Tai Polymer (TA-905, Taimei Chemicals Co., Ltd.)

3. Raw milk

The milk samples used in the laboratory tests consisted of 140 individual and pooled milk samples of different compositions collected during herd examinations conducted on dairy farms in the pasture-based dairy farming area in the Kushiro region of Hokkaido, Japan on October 9, 2023 (68 samples) and August 5, 2024 (72 samples). (Figure 1)

4. Effect of pH on flocculation

When an acidic inorganic coagulant is added to waste milk, the pH shifts from neutral to acidic, affecting subsequent flocculation and clarification when a polymeric flocculant is added. Therefore, we sought to determine the pH range needed to achieve complete flocculation and clarification. This was accomplished by using a digital burette to titrate 50-mL samples of 1:5 diluted milk with an inorganic coagulant (5% PAC or 5% PFS). The milk samples were adjusted to 11 different pHs between 6.0 and 4.0 (0.2 increments) for PAC and 15 pHs between 5.2 and 2.4 (0.2 increments) for PFS. Tai Polymer was subsequently added to all samples at the same rate (0.01 g/L). The reason for using 1:5 diluted milk is that it naturally settles after flocculation, making it easy to confirm complete clarification.

5. Effect of head performance and milk composition on coagulation

To investigate the effect of milk composition on coagulation using inorganic coagulants in the first stage of treatment, we determined the amount of 5% PAC needed to lower the pH of 68 individual milk samples of different composition (50 mL, 1:5 dilutions) to pH 5.0. We also determined the amount of 5% PFS needed to lower the pH of 72 individual milk samples (same as above) to pH 4.0. The target pHs—5.0 for PAC and 4.0

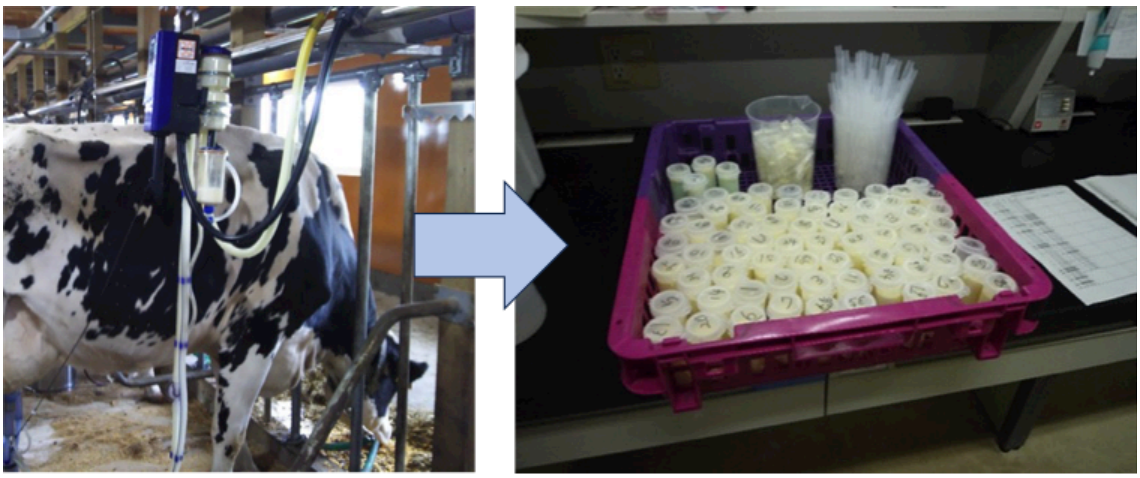

Fig. 1 Milk samples collected for evaluations of dairy head performance

for PFS—were the median values of the pH range over which complete flocculation and clarification were achieved in the previous sub-section.

The parameters examined for their effect on the amount of inorganic flocculant required were as follows: milk yield, days in milk, milk fat percentage, milk protein percentage, non-fat solids, and somatic cell count (linear score).

Results

1. Effect of pH on flocculation

(1) Use of PAC in Stage 1

Figure 2 shows the effect of using PAC as the inorganic coagulant in the first stage and Tai Polymer as the polymeric flocculant in the second stage of treatment on the clarification of comingled milk (50 mL, 1:5 addition of PAC. Complete clarification of the and 4.4 (median value of 5.0), corresponding to the addition of 2.3 to 3.9 mL of 5% PAC to 50 mL of 1:5 diluted milk.

(2) Use of PFS in Stage 1

Figure 3 shows the effect of using PFS in the first stage of treatment in the same manner as (1) on the removal of turbidity. pH decreased with increasing addition of PFS. Complete clarification of the supernatant occurred for pHs between 5.0 and 2.8 (median value of 4.0), corresponding to the addition of 1.9 to 3.8 mL of 5% PFS to 50 mL of 1:5 diluted milk.

2. Effect of herd performance and milk composition on flocculation

(1) Herd performance and milk composition of individual cows

The herd performance and milk composition (mean ± standard deviation) of the 140 individual milk samples used in this experiment were as follows: milk yield, 24.9 ± 6.4 [L/cow]; days in milk, 248 ± 167 [d]; milk fat, 3.99 ± 0.58 [%]; milk protein, 3.59 ± 0.36

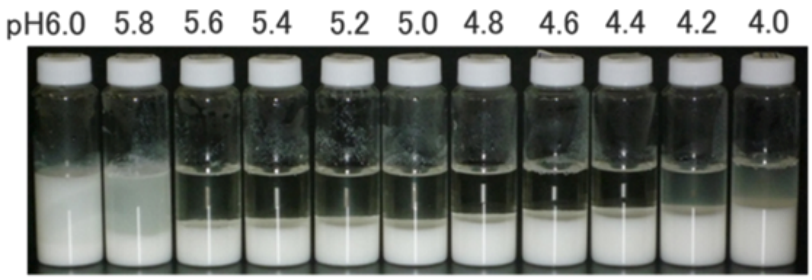

Fig. 2 Relationship between complete clarification and pH in cases using PAC as the first stage

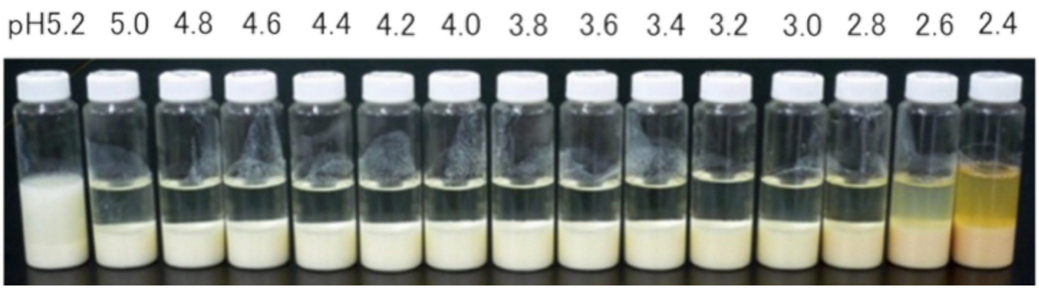

Fig. 3 Relationships between complete clarification and pH in cases using PFS as the first stage

Table 1 The relationship (correlation coefficient) between the amount of 5% PAC or 5% PFS required to achieve individual milk (50mL aliquots of 1:5 dilutions) to pH 5.0 or 4.0 and herd performance or milk components, and the required amount of 5% PAC or 5%PFS for the standard deviation of each component calculated using a linear approximation formula using the least squares method

	milk yield	days in milk	milk fat	milk protein	milk non-fat solids	smatic cell count (linear score)
mean ± standard deviation	24.9 ± 6.4 [L/cow]	248 ± 167 [d]	3.99 ± 0.58 [%]	3.59 ± 0.36 [%]	8.83 ± 0.46 [%]	3.28 ± 2.6
polyaluminum chloride (PAC)						
correlation coefficient	r = -0.200	r = 0.038	r = 0.342**	r = 0.361**	r = 0.301*	r = 0.086
required amount of 5%PAC	※	※	2.0 ~ 4.3 [mL] (2.2-fold)	2.1 ~ 3.7 [mL] (1.8-fold)	1.4 ~ 4.1 [mL] (2.9-fold)	※
polyferric sulfate (PFS)						
correlation coefficient	r = -0.348**	r = 0.296*	r = 0.384**	r = 0.598**	r = 0.596**	r = 0.039
required amount of 5%PFS	3.0 ~ 1.6 [mL] (1.9-fold)	1.5 ~ 3.3 [mL] (2.2-fold)	1.9 ~ 3.2 [mL] (1.7-fold)	2.3 ~ 3.0 [mL] (1.3-fold)	1.9 ~ 3.1 [mL] (1.6-fold)	※

** P<0.01 * P<0.05 ※ Not calculated because no significant correlation was found

[%]; milk non-fat solids, 8.83 ± 0.46 [%]; and somatic cell count (linear score), 3.28 ± 2.6 (Table 1).

(2) Relationship of required PAC input to herd performance and milk composition parameters

Table 1 shows the correlations of 5% PAC required to achieve a pH of 5.0 (median of the pH 5.6–4.4 range resulting in coagulation, obtained from the previous results) with herd performance or milk composition for 68 individual milk samples (50-mL aliquots of 1:5 dilutions).

The components observed to be significantly correlated with PAC input were milk fat percentage (r=0.342**), milk protein (r=0.361**), and milk non-fat solids (r=0.301**) (Figure 4-6).

The 5% PAC input required per 50-mL sample of 1:5 diluted milk, calculated by least-squares linear approximation based on the standard deviation for each component, ranged from 2.0 to 4.3 mL (max/min ratio of 2.2) for milk fat content, 2.1 to 3.7 mL (max/min ratio of 1.8) for milk protein, and 1.4 to 4.1 mL (max/min ratio of 2.9) for milk non-fat solids (Table 1).

(3) Relationship between required PFS input to herd performance and milk composition

Table 1 shows the correlations of 5% PFS required to achieve a pH of 4.0 (median of the pH 5.0–2.8 range resulting in coagulation, obtained from the previous results) with herd performance or milk composition for 72 individual milk samples (50-mL aliquots of 1:5 dilutions).

The components found to be significantly correlated with PFS input were individual milk yield (r=−0.348**), days in milk (r=0.296*), milk fat (r=0.384**), milk protein (r=0.598**) and milk non-fat solids (r=0.596**) (Figures 7–11).

The 5% PFS input required per 50-mL sample of 1:5 diluted milk, calculated by least-squares linear approximation based on the standard deviation for each component, ranged from 3.0 to 1.6 mL (max/min ratio of 1.9) for individual milk yield, 1.5 to 3.3 mL (max/min ratio of 2.2) for days in milk, 1.9 to 3.2 mL (max/min ratio of 1.7) for milk fat, 2.3 to 3.0 mL (max/min ratio of 1.3) for milk protein, and 1.9 to 3.1 mL (max/min ratio of 1.6) for milk non-fat solids (Table 1).

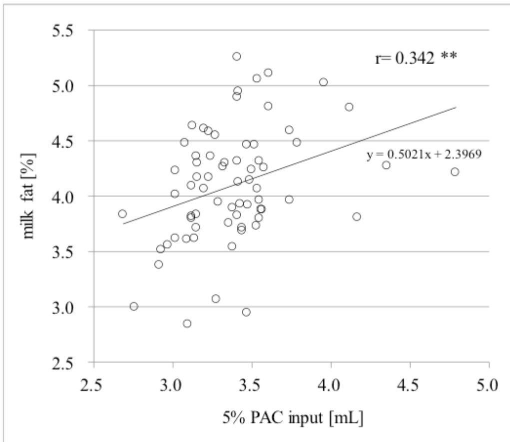

Fig.4 Relationship between PAC input and milk fat

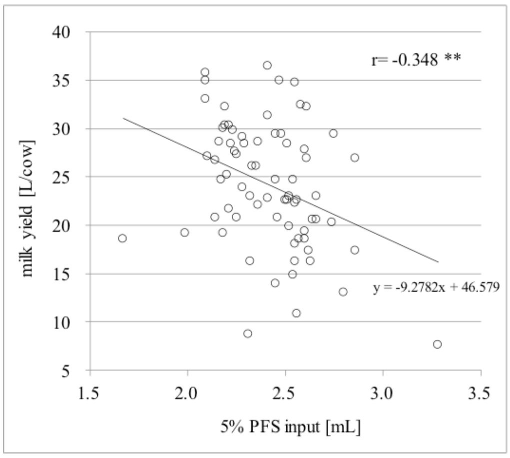

Fig.7 Relationship between PFS input and milk yield

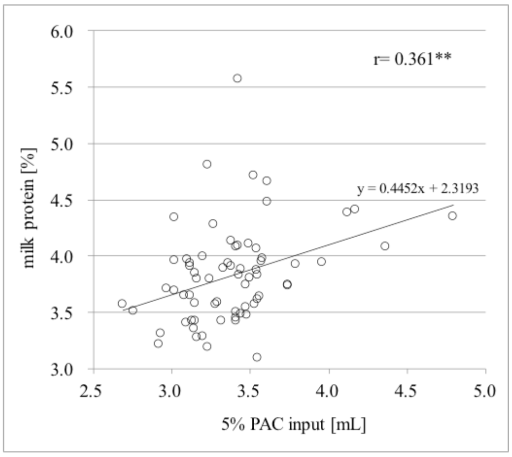

Fig.5 Relationship between PAC input and milk protein

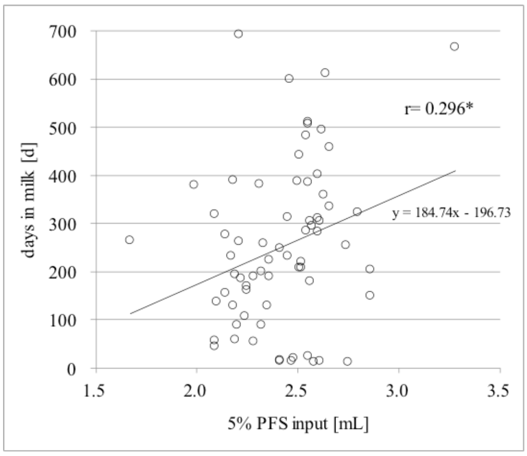

Fig.8 Relationship between PFS input and days in milk

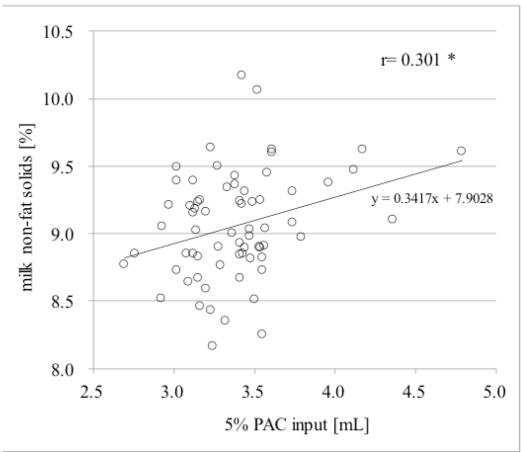

Fig.6 Relationship between PAC input and milk non-fat solids

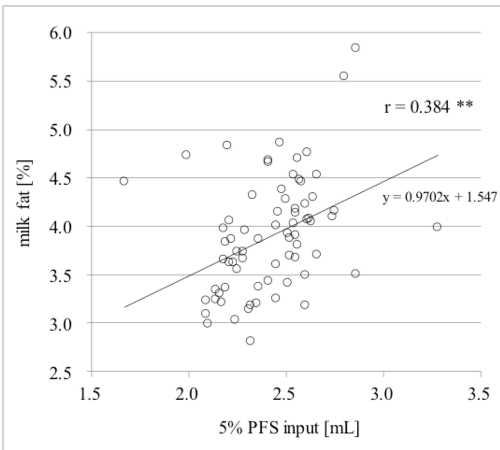

Fig.9 Relationship between PFS input and milk fat

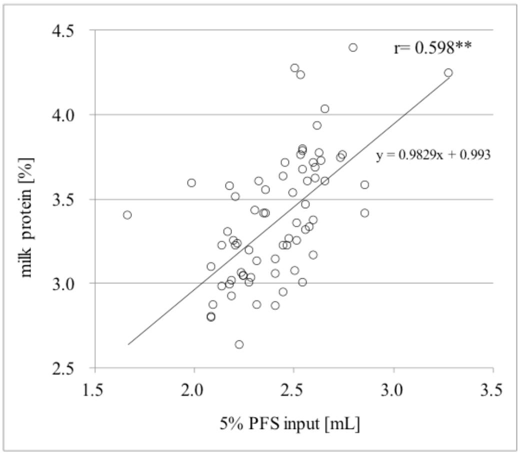

Fig.10 Relationship between PFS input and milk protein

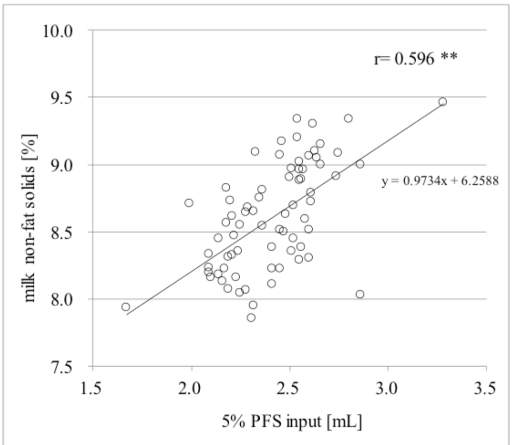

Fig.11 Relationship between PFS input and milk non-fat solid

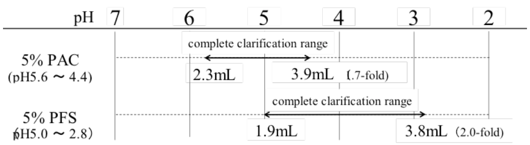

Fig.12 The relationship between the pH range over which coagulation occurs (coagulation pH range) and the amount of PAC or PFS added in the first stage of treatment

Discussion

We demonstrated that it is possible to coagulate, flocculate, and clarify waste milk, using an inorganic coagulant and a polymeric flocculant in the first and second stages of treatment, respectively. This is a promising method for treating waste milk generated by dairy farms. In this context, the causes of inconsistent coagulation in the field (i.e., on dairy farms) were found to be variation in pH after addition of the inorganic coagulant in the first stage of treatment and composition of the waste milk.

The relationship between the pH range over which coagulation occurs (coagulation pH range) and the amount of PAC or PFS added in the first stage of treatment is shown in Figure 12. It is possible that this pH range will not be attained if too much or too little inorganic coagulant is added, given the characteristics of a particular waste milk. In this study, the effective amounts of inorganic coagulant (coagulation range) ranged from 2.3 to 3.9 mL (1.7-fold) for PAC and 1.9 to 3.8 mL (2.0-fold) for PFS. These ranges are likely to vary depending on the composition and characteristics of the inorganic coagulant used; in actual practice, a wider coagulation pH range may be desirable as this is expected to allow greater flexibility in responding to variations in the composition of raw milk.

We also found that it is necessary to adjust the amount of inorganic coagulant added according to the herd performance and composition of the waste milk. A positive correlation was observed between the amount of PAC added in the first stage of treatment and milk fat percentage, milk protein, and milk non-fat solids. Additional correlations were observed for milk yield and days in milk when PFS was used as the coagulant. These results indicate that producers should be able to achieve effective treatment by adjusting the amount of inorganic coagulant added according to the characteristics of the waste milk produced on their farms.

However, although waste milk generated by dairy farms is generally expected to have higher somatic cell counts due to mastitis, no correlation was observed between the required amount of inorganic coagulant and somatic cell count. It can be concluded from these results that the amount of inorganic coagulant should be adjusted based on general factors such as milk fat percentage and milk protein, while paying attention to the coagulation pH range, without consideration of somatic cell count.

The factors influencing flocculation by polymeric flocculants in the second stage of treatment are currently under investigation. The results of these investigations along with those of waste milk clarification trials conducted on-site will be included in future reports.

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原　著

酪農における廃用乳の凝集処理に関する検討

－乳成分や pH が無機系凝集剤による凝結反応におよぼす影響－

猫本健司[1]・杉﨑允康[1]・石原美紅[1]・石川志保[1]・近藤健一[2]・有吉忠広[3]・小林次雄[4]

[1] 酪農学園大学 農食環境学群，北海道江別市，069-8501
[2] 有限会社バイオ，埼玉県さいたま市，339-0045
[3] オリオン機械株式会社 千歳工場，北海道千歳市，066-0077
[4] 中央オリオン株式会社，埼玉県熊谷市，360-0811

酪農場において日々発生する、初乳や乳房炎等による低品質で出荷できずに廃棄する廃用乳は、処理負荷が著しく高いため、浄化槽で処理することは困難である。そこで本研究では、第一段階に無機系凝集剤による凝結反応を、第二段階に高分子系凝集剤を用いた凝集反応によって、浄化槽を用いずに廃用乳を凝集・透明化させる方法を検討した。一連の反応に影響する諸条件について、今回は第一段階に関する室内実験について報告する。第一段階の無機系凝集剤にポリ塩化アルミニウム（PAC）やポリ硫酸第二鉄（ポリ鉄）を用いた場合、その投入量に応じて廃用乳のpHは酸性側に移行した。凝集・透明化が可能なpHの範囲はPACの場合が5.6～4.4（中央値は5.0）、ポリ鉄の場合は5.0～2.8（中央値は4.0）となり、限られた範囲のpHで凝集・透明化が可能であった。PACを使用した際における凝集可能範囲（pH5.6～4.4）の中央値であるpH5.0にするために必要なPAC投入量と、乳牛個体(n=68)の牛群成績および個体乳の成分との間に有意な相関が認められたのは、乳脂率（r=0.342**）、乳蛋白（r=0.361**）、無脂乳固形分（r=0.301**）であった。同様にポリ鉄を使用した際における凝集可能範囲（pH5.0～2.8）の中央値であるpH4.0にするために必要なポリ鉄投入量と、乳牛個体(n=72)の各項目との間に有意な相関が認められたのは、個体乳量（r=−0.348**）、分娩後日数（r=0.296*）、乳脂率（r=0.384**）、乳蛋白（r=0.598**）、無脂乳固形分（r=0.596**）であった。以上の結果から、廃用乳を凝集・透明化することは可能であり、酪農現場で実用化できる有望な処理方法であると考えられるが、無機系凝集剤の投入量は、凝集可能なpHの範囲に留意しながら、乳脂率や乳蛋白などの乳成分に合わせて調整することが必要である。

キーワード：凝集反応、無機系凝集剤、高分子凝集剤、廃用乳、酪農場

連絡者: 猫本健司 nekomoto@rakuno.ac.jp

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