AIChE Letter: Separations: Materials, Devices and Processes

Ambient-Temperature Fusible Filter: a Preliminary Experiment and a Proposal for a Filtration Process

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Significance

A new filtration apparatus is proposed using a fusible filter medium (FF). The advantages of FF are (1) materials captured by the filter can be recovered by melting the filter and subsequent gravity separation, (2) the filter medium can be recycled, and (3) the latent heat of solidification removed to produce FF is significantly lower than that of vaporization. The filtration can be further simplified and made more cost-efficient using an "ambient-temperature" fusible filter (ATFF).

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Introduction

To the best of our knowledge, there have been no reports of filtration apparatus using fusible materials apart from our previous proposal.[1] We have previously investigated the use of ice crystals as an *FF*.[2] In that study; emulsified-squid oil included in the visceral organ was separated from the organ with a high efficiency (87%). Herein we briefly report the use of trimethylolethane hydrate (TME · $3H_2O$) as an ambient-temperature *FF* (*ATFF*) to separate rapeseed oil from the oil emulsion. The important characteristics of filtration with an *ATFF* is that melting or solidification of the filter can be carried out at ambient temperature, resulting in a simpler and more cost-efficient (running and capital) *FF* apparatus.

Characteristics of TME · $3H_2O$

TME · $3H_2O$ has been studied and used as a heat storage material.[3–6] TME · $3H_2O$ is a polyalcohol that is both nontoxic and noncollusive,[5] and its chemical reactivity is 0 (NFPA).[7] TME · $3H_2O$ does not promote esterification reaction when used as a fusible filtration substance because filtration is performed at ambient temperature or lower.[8] The use of TME · $3H_2O$ as an *ATFF* will exhibit the following features.

a. The melting/solidification point is 303 K (30°C), which is close to ambient temperature. Thus, the melting or solidification can be carried out at ambient temperature, requiring no artificial input of thermal energy.
b. The latent heat of melting is 218 kJ/kg, which is about 2/3 of that of water (335 kJ/kg).
c. The specific heat is less than that of water (TME · $3H_2O$ is 2.75 kJ/kg/°C at 10°C and 3.58 kJ/kg/°C at 50°C, and water is 4.19 kJ/kg/°C at 0°C and 4.18 kJ/kg/°C at 50°C). Thus, the energy for sensible heat required to increase or decrease temperature for the melting or solidification is also less than that of water.

Furthermore, the density of TME · $3H_2O$ is greater than that of water (TME · $3H_2O$ is 1120 kg/m^3 at 10°C and 1090 kg/m^3 at 50°C, and water is 1000 kg/m^3 at 0°C and 990 kg/m^3 at 50°C). The densities of most biomaterial oils are less than 1000 kg/m^3 (e.g., the density of rapeseed oil at 23°C is 973 kg/m^3), [9] facilitating gravity separation of the captured material (oil) and capturing substance (the melted fusible filter).

Separation of Liquid/Liquid Mixtures

A preliminary experiment demonstrating the use of TME · $3H_2O$ as an ATFF

The following experiments were carried out to separate oil from a biomaterial using TME · $3H_2O$ as an *ATFF*.

A. TME · $3H_2O$ crystal masses (Figure 1b) was made by adding tap water to TME powder (Sigma-Aldrich co., St. Louis, USA) (Figure 1a). The weight ratio of the

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Figure 1. (a) TME powder, (b) Generation of TME · 3H2O crystal masses by addition of water to TME powder, (c) Dried TME · 3H2O crystal masses.

Drying was carried out at ambient temperature (around 20°C), and (d) crushed TME · 3H2O crystals used to make the filter medium. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

TME powder to the added water was 65:35. The mixture in the tube was settled down for 24 h or longer at the room temperature.

B. Excess water (Figure 1b) in the tube was discharged from the mixture (the crystal masses and the water). The masses in the tube were dried by contact with the atmosphere in the room (around 20°C, 24 h or longer.), see Figure 1c.

C. The dried crystal masses were crushed (Figure 1d) and sieved (crystal diameter of 500 μm or less) to make a filter medium.

D. An inner cylinder (0.022 m inside diameter and 0.083 m high), and an outer tube (0.026 m inside diameter and 0.115 m high) were prepared (Figure 2a). To make a filter bed, the sieved crystals (0.0035 kg) were set in the inner cylinder which contained two 400 μm diameter holes in its bottom. The height of the filter bed was 0.025 m (Figure 2b).

E. An oil emulsion comprised of rapeseed oil (ECOERC Co. Toyokoro Japan, 15 mL), NaCl solution (20 mL) and protein surfactant powder (0.003 kg) was prepared consulting a literature.[10] Whey protein isolate (WPI ~91% protein, Meiji Seika Pharma Co., Ltd., Tokyo, Japan) was used as the protein surfactant. NaCl (The Salt Industry Center of Japan, Tokyo, Japan) was added to the solution yield a concentration of 0.9 wt %, to mimic natural creature solution. Solutions were mixed using a Voltex shaker (Voltex Genius3, IKA Werke GmbH & co. KG, Konigswinter, Germany) to make an oil emulsion (Figure 2c). The emulsion was cooled at 0°C; cooling at this temperature results in an increase in the difference in viscosity between the aqueous solution (low concentrate) and the oil.[11]

F. The oil emulsion (6 mL) was poured onto the filter bed in the cylinder.

G. The cylinder was placed in the outer tube (Figure 2b), and subsequently centrifuged using a test-tube centrifuge (Versatile Compact Centrifuge Himax-CF16RX, Hitachi High-Technologies Co., Tokyo, Japan) for 5 min at 0°C and centrifugal effect: 400.

H. The states of both the oil captured by the filter bed and the liquid filtrate that passed through the filter into the triangular pyramid bottom of the outer tube were observed (Figure 3a).

I. The filter bed-/oil-containing cylinder was placed upside down into a second outer tube (same size as

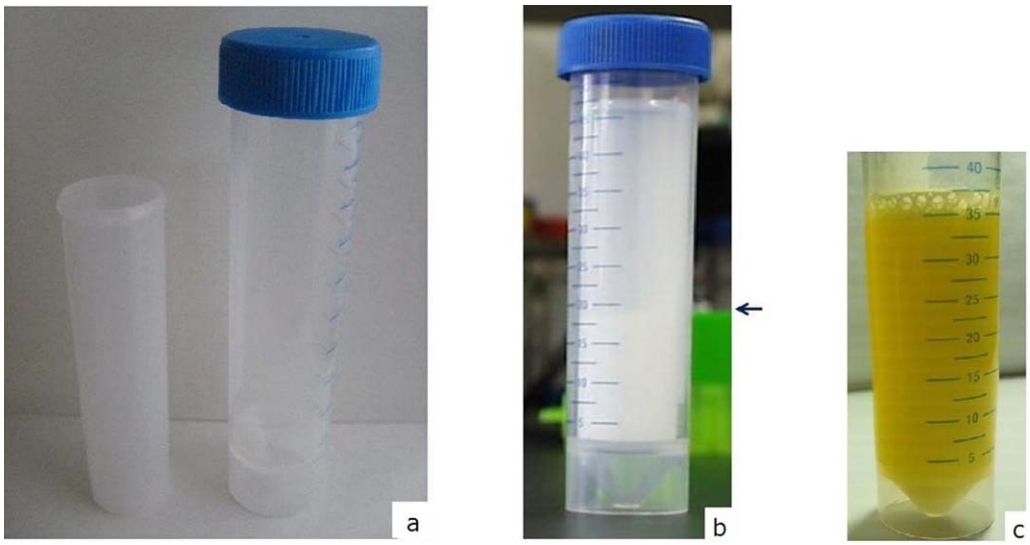

Figure 2. (a) Inner cylinder and outer tube, (b) the inner cylinder containing the sieved crystal filter bed placed inside the outer tube (the arrow indicates the height of the filter bed), and (c) Formation of rapeseed oil emulsion containing WPI.

[Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the first outer tube), and subsequently dipped into a warm-water bottle (start-up temperature about 90°C) for 10 min. The filter bed (TME · $3H_2O$ crystals) was liquidized and the rapeseed oil was separated from the liquidized TME · $3H_2O$ by specific gravity (Figure 3b).

J. The cylinder was removed from the second outer tube. The separated state of the liquidized of TME · $3H_2O$ and rapeseed oil was observed in the tube (Figure 3b).

K. The liquids in step J were cooled at the room temperature (around 20°C for 15 min) until the TME · $3H_2O$ had been solidified. The inclination of the oil and the lower content, whose surface is perpendicular to the longitudinal axis of the tube (Figure 3c), demonstrates that the oil is in its liquidized state and the TME · $3H_2O$ is in its solidified state (crystallized).

L. The amounts of both the captured oil and the liquid filtrate were measured. 2.2 mL of the captured oil (transparent rapeseed oil) (Figure 3d) was obtained (86% recovery rate), while 1.5 mL of the liquid passed through the filter bed.

The recovery rate (R) was calculated using the following equation

$$R = Oc/Oe$$

where Oc is the amount of oil captured by the filter bed (mL), and Oe is the amount of oil in the emulsion supplied to the filter bed (mL).

M. The rest of the sample emulsion (i.e., after removal of 6 mL of the sample) was allowed to sit for 24 h. The emulsion kept the state (cloudy). The emulsion was then centrifuged using the test-tube centrifuge for 5 min at 0°C and centrifugal effect: 400, but still exhibited an emulsion-like state after centrifugation. Moreover, heating to a temperature of 90°C did not have any effect on the sample status.

It should be noted that the rapeseed oil separated in step L was transparent (Figure 3d). Thus, the filtration apparatus exhibits the potential to remove protein from the protein-coated oil emulsion droplets. The experiment was carried out twice; in the second experiment, 2.4 mL of transparent oil (94% recovery rate) and 1.8 mL of the liquid filtrate were obtained.

A proposed filtration process using an ATFF

On the basis of the experiment using TME, the following filtration process (Figure 4) can be proposed to separate oil from a biomaterial using TME · $3H_2O$ as an *ATFF*. The biomaterial is a mixture of an aqueous solution and oil dispersed throughout the solution.

The numbers in Figure 4 indicate the process described below.

1. Addition of water to TME powder to generate the TME · $3H_2O$ crystal masses.
2. Removal of water from the crystal masses by discharging and drying; drying is carried out at an ambient temperature of less than 30°C, which does not require the input of thermal energy.
3. Crystal size adjustment via use of a crush and a sieve.
4. Formation of a filter bed made of the sieved crystals for centrifugal filtration, or other types of filtration such as column filtration.
5. Addition of the mixture to be separated onto the filter bed; the aqueous solution passes through the filter bed, while the oil is captured by the filter bed.
6. Separation of the captured oil and the filter bed in the oil-containing filter bed: the oil-containing filter bed is taken out and heated to a temperature greater than the

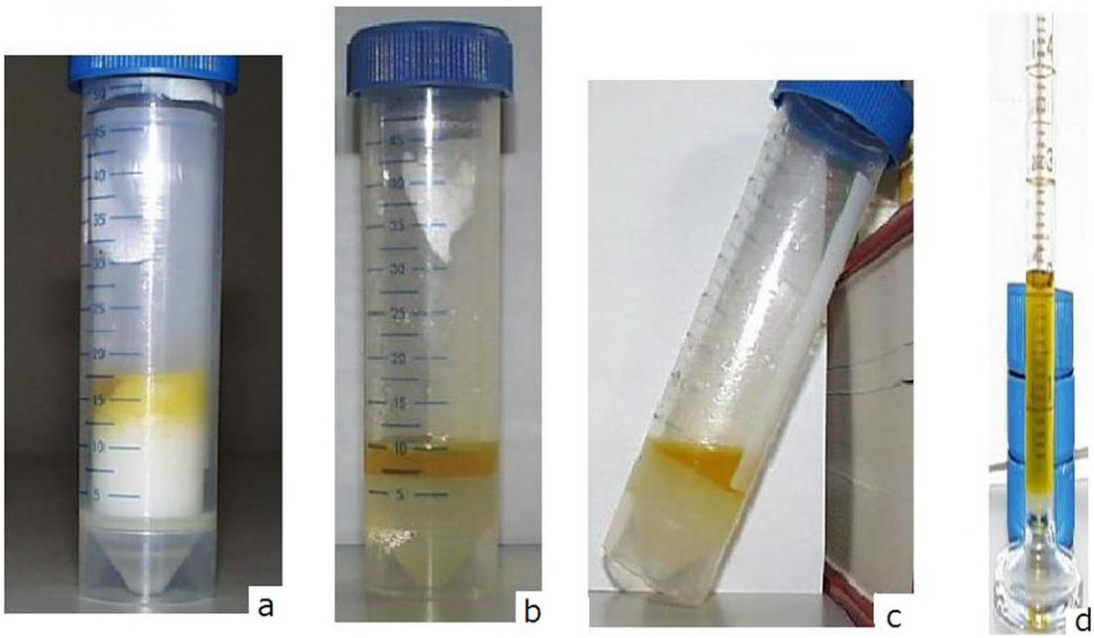

Figure 3. (a) The state just after centrifugation for 5 min at centrifugal effect: 400. Note that the oil was captured in the upper layer of the filter bed, while the aqueous liquid passed through the filter bed and into the triangular pyramid bottom of the outer tube, (b) separation of rapeseed oil and liquidized TME · 3H2O due to the difference in specific gravity, (c) the TME · 3H2O was solidified at ambient temperature (around 20°C), and (d) 2.2 mL of transparent rapeseed oil was obtained (86% recovery rate).

[Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

melting point (30°C) of the filter substance (TME · $3H_2O$), thus, liquidizing the filter substance and allowing for separation of the oil from the filter substance because of the difference in specific gravity.

7. Solidification of the liquidized filter substance: the liquidized filter substance is removed using a connecting pipe and introduced into a tank, where the filter substance is cooled at ambient temperature. This process does not require an input of thermal energy because the melting point of the substance (to allow for solidification) is greater than ambient temperature (e.g., around 20°C). The solidified filter substance is then recycled in step 3.

Application of *ATFF* to Solid/Liquid Mixtures

The *ATFF* was employed for a low-temperature (e.g., under 0°C) separation process in this liquid/liquid experiment. However, it is also useful for the separation of a liquid from either solid/liquid or solid/liquid-liquid mixtures (e.g., biomaterials) by carried out the separation process at an ambient temperature, which separation can be accomplished

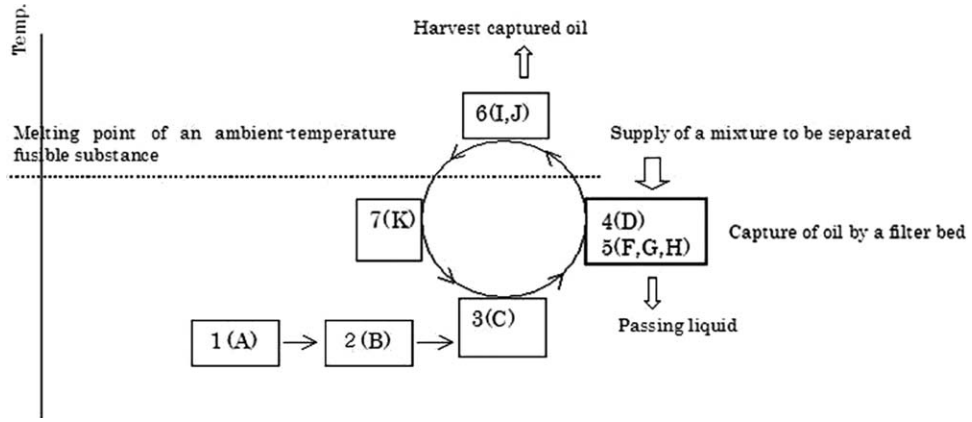

Figure 4. Separation process using an ATFF.

The number and the capital alphabet are each step of the proposed process and the experiment, respectively.

by lowering the viscosity of a liquid in the mixtures by increasing temperature. In this case, the solid phase is captured in the filter bed while the liquid component passes through the bed. The liquid-liquid mixture (in the solid/liquid-liquid mixtures) that passes through the bed can be separated due to the difference in specific gravity. Furthermore, (in this case), the filter medium facilitates separation of the aqueous solution from the oil by coalescence of the suspended fine oil droplets.

Problems Associated with Prior Techniques

One preferred mode of our proposed filtration method is an application to oil-containing biomaterials. With existing techniques, such as sedimentation centrifuges, coalescers with fiber bundles, and solvent extractions, there are frequently difficulties in separating, and they are associated with high costs and/or a low quality of the product. Oil separated using a sedimentation centrifuge faces accumulating of protein-coated oil emulsion droplets,[13] which are stable[10,12] at ambient temperatures or lower. Coalescers with fiber bundles cannot be applied to putrefactive or liquid/solid mixtures. As for solvent extraction, hexane is extensively used even though they are known to be explosive substance. Moreover, solvent extraction have the possibility of reducing the separation efficiencies due to surfactants (e.g., proteins).[10]

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