High-power Ultrasound to Control Honey Crystallisation

This report details the successful development of a process for liquefying candied honey based on the use of ultrasound technology. This research is important due to the concern within the honey industry that the present heating regime used to liquefy candied honey is reducing the quality of honey, particularly its flavour. Therefore, it was necessary to undertake this research project to determine if it was feasible and cost effective to replace the present heat treatment used by the honey industry for the liquefaction of naturally crystallised or candied honey, with an ultrasound treatment. The experiments undertaken used a laboratory scale ultrasound processor to gather critical data that could be used to support industrial scale-up trials within a honey packing company, or by beekeepers. In addition, the Dyce creamed honey process was examined with respect to the factors that affect the crystallisation of D-glucose monohydrate and thus the quality of creamed honey, and whether ultrasound treatment could improve the quality of creamed honey.

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Background

It is a common problem within the honey industry for heat-treated liquefied honey to crystallise during storage, particularly during cold weather. Since liquid honey is preferred by Australian consumers, and by food companies (for ease of handling), then an alternate method to expensive and time-consuming heating is required to retard the crystallisation process in honey. Creamed honey production is a difficult process to control, particularly related to crystallisation, the size of the D-glucose monohydrate crystals, and thus the quality of the final product related to hardness and spreadability. The process used by the honey industry to produce creamed honey is based on the Dyce method (Dyce, 1931a,b; Dyce, 1976). Ultrasound treatment has the potential to control this crystallisation process. However, before any effects of ultrasound can be determined, it is necessary to be able to produce high quality creamed honey in the laboratory using the Dyce method, and to be able to determine the amount of crystalline D-glucose monohydrate present in creamed honey. Ultrasound treatment was examined to determine if it can improve the creamed honey process and the stability of the final product.
Who is the information targeted at?

This report is targeted at beekeepers and honey packing companies.

Aims and Objectives

Aims:

(1) To reduce the amount of expensive heating and loss in quality during liquefaction of candied honey by developing an alternate, cost-effective ultrasound based method for the partial or complete liquefaction of candied honey, with a view to ultrasound having direct application for beekeeper control of honey crystallisation, or for liquefying candied honey prior to decanting in a honey packing plant.

(2) To better control the texture of creamed honey spread by developing an ultrasound based method that enhances the nucleation rate and produces uniform crystal growth in a creamed honey system, with a view to it being used by beekeepers and honey processors for producing consistent and high quality creamed honey.

Objectives:

(1) To investigate the effect of ultrasound treatment on candied honey, including individual glucose monohydrate crystals

(2) To determine the ultrasound conditions for liquefying candied honey and for controlling crystallisation in honey

(3) To investigate the effect of ultrasound treatment on the creamed honey production process

Methods Used

1. Effect of Ultrasound Treatment on the Liquefaction of Candied Honey

The main study of the effect of ultrasound on candied honey was divided into three experiments.

In the first ultrasound liquefaction experiment, it was necessary to determine which laboratory scale ultrasound sonotrode, out of the available 7 mm, 12 mm or 40 mm diameter sonotrodes, better liquefied candied honey. Candied honey (~250 g) was treated with ultrasound energy interrupted after each of six 10,000 J intervals, for predetermined input energy levels, using three ultrasonic sonotrodes, with the temperature profile in the honey being monitored during each interruption. Input energy, treatment time and power measurements were also recorded from the ultrasonic processor.

The optimum sonotrode (40 mm diameter) and amplitude (12 µm) were selected in Experiment 1. The first aim of Experiment 2 was to determine the minimum input energy required for complete liquefaction. The use of too high an input energy not only wastes energy but will unnecessarily increase the temperature and treatment time. The second aim was to determine if the ultrasound treatment adversely affected the quality of the honey. The third aim was to determine the specific energy input (kWh) required to liquefy one kilogram of candied honey, in order for the developed novel ultrasound liquefaction method to be useful for the honey industry. In the second ultrasound liquefaction experiment, candied Salvation Jane honey samples were treated with six different ultrasound input energy levels using this optimum sonotrode and amplitude. These liquefied honey samples were analysed for their hydroxymethylfurfural (HMF) concentrations, and diastase and invertase activities, since these three quality parameters are normally used by the honey industry and regulators to gauge the heating history and quality of honey.

To complete the ultrasound liquefaction study, a third experiment was carried out to determine the effect of ultrasound treatment, relative to heat treatment, on the stability of liquid honey with respect to subsequent crystallisation on storage. In the third ultrasound liquefaction experiment, candied reworked mixed honey (~200 g) was completely liquefied by ultrasound...
treatment. Reworked mixed honey was selected for this trial as it is a very fast crystallising honey that produces large crystals. This permitted a crystallisation study to be completed in a short time-frame. Crystallisation of ultrasound-treated honey under optimum conditions of 14 °C was monitored (using a microscope as part of an image analyser) and compared with crystallisation in honey samples initially treated with a standard heat treatment.

2. Evaluation of the Effect of Ultrasound Treatment on the Creamed Honey Production Process

A study was carried out to optimise a method to determine the amount of crystalline D-glucose monohydrate present in creamed honey, and to produce a laboratory creamed honey with a similar level of D-glucose monohydrate crystals to that of the commercial Capilano Honey Ltd. creamed honey. Various honey blends were used to produce creamed honey using the Dyce method, and the amount of crystalline D-glucose monohydrate present in these laboratory creamed honeys was determined using a differential scanning calorimeter (DSC).

Results/Key findings

1. Effect of Ultrasound Treatment on the Liquefaction of Candied Honey

The main finding from the first experiment was that the 40 mm diameter sonotrode operated at the 12 µm amplitude was optimum for completely liquefying candied honey. While it has a lower maximum net power for any 1 s period during treatment than does the 22 mm diameter sonotrode, the maximum net power for the 40 mm diameter sonotrode increased steadily after each of the six interrupted 10000 J energy inputs as the honey liquefied, while the maximum net power for the 22 mm diameter sonotrode initially increased, but decreased markedly from the fourth 10000 J energy treatment onwards. As the candied honey liquefies, the power output from the sonotrode increases until the honey is liquid, at which point there is little increase in maximum net power. The decrease in net power after an initial increase observed for the 22 mm diameter sonotrode indicates that there was poor efficiency in the emission of energy from the 22 mm diameter sonotrode into the candied honey. The 7 mm diameter sonotrode produces a lower maximum net power than the other two sonotrodes, again indicating poor output efficiency of energy from this sonotrode into the candied honey. Cumulative treatment times were lower for the 40 mm sonotrode (324 s to 383.3 s; lowest for the 12 µm amplitude) relative to those for the 7 mm (358.3 s to 681.3 s) and 22 mm (394.7 s to 871.0 s) sonotrodes. In addition, the variation in treatment times among replications was also lower for the 40 mm diameter sonotrode. The more efficient the emission of energy from the sonotrode to the honey, the shorter the treatment times. Finally, the maximum temperature reached after the six interrupted 10000 J of energy input was significantly (P<0.05) lower for the 40 mm diameter (66.2 °C to 67.8 °C) sonotrode relative to the 7 mm (78.2 °C to 84.4 °C) and 22 mm (76.4 °C to 82.8 °C) sonotrodes. This reflects the shorter treatment time for the 40 mm diameter sonotrode, which was possibly due to the high maximum net power produced by it.

Key Finding:

Since treatment times need to be as short as possible and temperatures as low as possible, then the 40 mm diameter sonotrodes operated at an amplitude of 12 µm is the optimum condition for complete liquefaction of honey on a laboratory scale.

In the next experiment, a preliminary trial showed that a range of input energies from 50000 J to 70000 J would produce a range of liquefaction efficiencies from partially liquefied to completely liquefied. During a replicated trial involving six input energies between 50000 J and 70000 J, only an input energy of 70000 J completely liquefied candied Salvation Jane honey. The other energy inputs only partially liquefied the candied honey. In addition, the time needed to emit each of the fixed energies from the sonotrode increased from 304 s for 50000 J of input energy to 434.0 s for 70000 J of input energy, since it takes longer for a sonotrode to emit more energy. However, there was no significant (P>0.05) difference in the maximum temperature (which ranged 69 °C to 77.3 °C) after each of the six fixed energy treatments. Further, the maximum net power recorded at any 1 s interval during each energy level treatment was not different from each other.

Key Finding:

A 70000 J ultrasound energy treatment can be used to completely liquefy candied honey in a relatively short time of 434 s, without it adversely affecting the maximum temperature generated in the honey relative to lower energy treatments. There was no significant (P>0.05) difference in the HMF concentration in honeys treated with between 50000 J and 62500 J of input energy and honeys that were heat-treated. However, the HMF concentrations in the honeys treated with 65000 J and 70000 J of input energy were
significantly (P<0.05) lower than the HMF concentration in the heat-treated honeys. This is primarily due to the honey being at the maximum temperature reached of 77.3 °C for a much shorter time (434.0 s) than for a heating regime (55 °C for 16 h and 72 °C for 2 min) which is similar to that presently used by the honey industry. The effect of the energy treatments on enzyme activity was negligible since there were no significant (P>0.05) differences in the diastase activity between honeys treated with any of the six energy inputs and those that were heat-treated, while the invertase activity of most of the ultrasound treated honey was higher than the heat-treated honeys, with this difference not always being significant.

Key Findings:
Use of an ultrasound input energy of 70000 J from a 40 mm sonotrode operated at an amplitude of 12 µm is sufficient to liquefy candied Salvation Jane honey (~250 g) without compromising honey quality. For example, this ultrasound treatment results in the production of a lower concentration of HMF from honey sugars, and no decrease in diastase and invertase activities, relative to a heating regime (55 °C for 16 h and 72 °C for 2 min) similar to that used by industry. The specific energy input needed to completely liquefy candied Salvation Jane honey is 0.126 kWh/kg. Therefore, 10 kg of candied honey will require 1.26 kWh, while 300 kg will require 37.9 kWh.

The first finding from the third experiment was that the D-glucose monohydrate crystallised differently in each type of treated honey. In the heat-treated honey samples, the initial plate crystals that formed at between 14 and 28 days were long thin, spiral-shaped plate crystals. In contrast, in the ultrasound-treated honey samples, most of the initial crystals that formed at between 14 and 49 days were large pentagon-shaped plate crystals. In addition, more needle crystal masses were formed in the heat-treated honeys than were produced in the ultrasound-treated honeys at the end of the monitoring period of 112 days (16 weeks). Moreover, in heat-treated honeys, plate crystals grew underneath these needle crystal masses in the later stages of crystallisation. In contrast, in ultrasound-treated honeys, plates formed after the needles, with subsequent needles growing on these initial plates.

Key Finding:
Ultrasound treatment delays D-glucose monohydrate crystallisation more than does a heat treatment similar to that used by the honey industry. This occurs at both the microscopic level (in a drop of honey) and in bulk samples. In addition, there is a difference in the crystal formation process at the microscopic level in ultrasound-treated honey relative to that in heat-treated honey. The reason for this is not clear, and requires further study. Thus, ultrasound treatment will not only liquefy candied honey without the need for long exposure to high temperatures, but may make the liquefied honey more stable to subsequent crystallisation on storage.

2. Evaluation of the Effect of Ultrasound Treatment on the Creamed Honey Production Process

Samples of commercial Capilano Honey Ltd. creamed honey were initially analysed and found to have an average crystalline D-glucose monohydrate content of 39.6-40.1 g/100 g honey. In addition, two blends consisting of 70% alfalfa honey/30% blue gum honey and 70% canola honey/30% red gum honey were chosen for a subsequent
Finally, ultrasound treatment was then applied to the seed honey prior to it being added to the honey blend. The hypothesis is that if the size of the crystals in the seed honey can be reduced by ultrasound treatment, then the creamed honey process could be enhanced (producing a higher level of crystals) and the final honey would have smaller crystals. This was not the case, and the level of crystals was not significantly (P>0.05) different from the control creamed honey produced with seed honey that was not ultrasound treated.

Included in this experiment was the use of ultrasound treatment of the seeded honey blends at one day and two days after the addition of the seed honey. Again, such treatments were hypothesised to reduce the crystal size and enhance subsequent crystallisation during storage at 14 °C. However, there was no significant (P>0.05) difference in the crystalline D-glucose monohydrate content relative to the control untreated creamed honey. None of the ultrasound treatments enhanced the level of D-glucose monohydrate crystals relative to the untreated creamed honey.

Key Finding:
The untreated canola/red gum creamed honeys (47.1 g/100 g honey) had similar crystal contents to the ultrasound-treated canola/red gum creamed honeys (44.5 g/100 g honey to 47.1 g/100 g honey), while the untreated alfalfa/blue gum creamed honeys (33.1 g/100 g honey) had similar crystal contents to ultrasound-treated alfalfa/blue gum creamed honeys (32.2 g/100 g honey to 33.1 g/100 g honey).

Finally, conditioning of the creamed honey product was investigated. The reason for this is that some of the creamed honey samples produced were creamy in texture and some were semi-solid in texture. There did not seem to be any particular treatment that led to one type of product over the other. In fact, replicates of the same treatment often had both types of texture. As part of the Dyce process (Dyce, 1931a,b; Dyce, 1976), a conditioning step is used prior to the creamed honey being sent to supermarkets for sale. Conditioning is where the creamed honey is stored at 30 °C for a number of days. This study found that such conditioning did produce consistency in texture with all product having a creamy texture. The storage at
30 °C for 14 days produced a small reduction in the crystalline D-glucose monohydrate content, with the final content being similar to that found for commercial Capilano Honey Ltd. creamed honey.

**Key Finding:**

The conditioning process dissolves some of the D-glucose monohydrate crystals leading to an increase in the amount of liquid honey, and an overall softening of the creamed honey, with a consequent improvement in spreadability. While ultrasound treatment did not produce a product that was different to untreated creamed honey, conditioning the final product before sale is very important for producing a consistent product from one production run to another.

**Implications for relevant stakeholders for industry**

Once the data from an industrial trial have been obtained, an ultrasound equipment manufacturing company such as Dr Hielscher GmbH can then provide specifications for liquefying larger amounts of honey such as 300 kg in 200 L drums. However, while the time required to liquefy 10 kg or 300 kg of candied honey will depend on the input power of the ultrasound processor and the capacity of the sonotrode/sonotrodes, it will be less than the time now used to liquefy candied honey in hot rooms. The newer, large plastic drums, which have a completely removable lid, would be ideal for use with the proposed ultrasound processing system. However, the one industrial scale problem likely to be encountered relates to the design of the commonly used 200 L galvabond drums. These drums have only small openings which, while permitting limited insertion of the sonotrode, would not permit the moving of the drum up and down and sideways in a predetermined pattern so as to expose all the candied honey to the ultrasonic waves. However, as part of the industrial trials and the subsequent design of the processor system by an ultrasound equipments manufacturer such as Dr Hielscher GmbH, such a limitation in the galvabond drums may be able to be overcome. It is recommended that part of the Australian honey industry such as major honey packing companies should undertake these types of industrial trials to ensure technology transfer from this project to industry.

The results of this study of the Dyce creamed honey process will aid beekeepers and honey packing companies to better understand their creamed honey process and improve the quality and consistency of their product from batch to batch.

**Recommendations**

Honey packers must make use of this collected data, by taking up the challenge (and rental costs) of participating in a scaled-up industry trial involving an industrial ultrasound processor (much more powerful than used in this project) for liquefying 10 kg candied honey in commonly used plastic containers (e.g., diameter of 270 mm and height of 240 mm). This would be done in consultation with an ultrasound equipment manufacturer such as Dr. Hielscher GmbH and the project’s research team. As part of the recommended system design, the ultrasound sonotrodes will have to be inserted into the drum to a particular depth in order to liquefy the honey down to the bottom of the drum. Initially, the sonotrode will be in contact with the hard candied honey near the top of the container. As liquefaction of the surface candied honey proceeds, the honey container will need to be moved upwards and sideways in a predetermined pattern, so that the sonotrode is brought in contact with as much of the candied honey as possible, so as to minimise the treatment time required. In addition, or alternatively, a stirrer could be inserted in the semi-melted honey to mix the liquid honey with the remaining candied honey, thereby creating a flow in the container past the treatment region around the sonotrode. Ultrasound waves dissipate quickly at a short distance from the sonotrode, so some mixing is required.
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