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Physical and structural properties of honey crystallized by static and dynamic processes

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Physical and structural properties of honey crystallized by static and dynamic processes

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ABSTRACT

The aim of the present study was to investigate the main physical and structural properties of honey during static and dynamic induced crystallization. For this aim, three honey samples, chosen with different F/G (fructose/glucose) ratios, were analysed for content of glucose crystals, colour, texture, viscosity and microscopic observations during the two processes. Correlation of data was analysed through Pearson coefficient and Principal Component Analysis. Results showed that the dynamic process allowed to reduce by 5–6 folds the time necessary to reach full crystallization. Moreover, while during static crystallization crystals progressively grew in size, the constant stirring of the mass allowed to obtain products characterized by low hardness and viscosity, due to the formation of micro-crystals. The dynamic process led to final crystallized honeys with very similar colour and structural characteristics although starting from very different initial values.

1. Introduction

Crystallization of honey is a natural process that occurs spontaneously during storage and leads to important changes in the product structural and rheological characteristics. Its rate depends on many parameters, both compositional (sugars and water content, presence of impurities and of micro-crystals) and environmental (temperature and thermal history) (Venir et al., 2010).

However, the most used index for classifying honey in beekeeping sector is the F/G (fructose/glucose) ratio according to which products are defined as fast (F/G < 1.11), medium (1.11≤ F/G ≤ 1.33) and slow or absent (F/G > 1.33) crystallization.

Crystallization, or granulation, is often considered undesirable from honey consumers and processors, both from a sensorial and technological point of view, causing difficulties in handling, fractionating and pouring.

Affecting its textural properties, it makes honey less appealing to the consumer, who generally prefers it liquid and/or transparent (Tosi et al., 2004). Indeed, although it does not affect its objective quality parameters, it affects the consumers' subjective perception of honey quality during purchase (Šánová et al., 2017). Moreover, spontaneous crystallization may lead to the formation of coarse crystals that may cause phase separation, sedimentation and water activity increase up to levels which may be congruous with microbial fermentative processes (Venir et al., 2010). However, a process known as induced or guided crystallization, is gaining popularity because it leads to a crystallized product with specific rheological properties. This process, patented by Dyce (1,987,893, 1935) is obtained by adding a small amount of finely crystallized honey, giving to the system mechanical energy by mixing. In particular, according to Chen (Chen et al., 2009), by adding 5–10% of ground, previously granulated honey, which provides the nuclei (seed crystals) for new crystal growth, it is possible to obtain an homogeneously crystallized product characterized by fine crystals, that cannot be detected by the tongue, and by low viscosity. This type of product represents an interesting innovation, having defined rheological characteristics and being stable during storage, because fully crystallized. However, it has been only poorly characterized. Moreover, the optimization of the crystallization process requires a deep knowledge of the modification induced on qualitative characteristics of the final products.

In a previous study by our group (Dettori et al., 2018), the influence of induced granulation on some quality characteristics of honey, such as colour, texture and crystal size, was evaluated. Results showed that increasing the F/G ratio, a considerable increase of hardness during crystallization occurred. However, the investigated process was carried out only in the traditional static manner. In a following research (Tappi et al., 2019), the crystallization kinetic of the same honey type carried out in static and dynamic process was compared with specific focus on the changes of water state. The constant movement of the

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honey mass allowed to reach complete crystallization 5–6 times faster. Results showed that water activity, starting from low values, increased but never reached the threshold for microbial development. However, in this work the main quality parameters influencing consumer perception (colour, texture and rheological properties) were not investigated. Hence, the aim of the present study was to investigate the main physical and structural properties of honey during static and dynamic induced crystallization. For this aim, three honey samples, chosen with different F/G ratios, were analysed for content of glucose crystals, colour, texture, viscosity and micro-structural observations during the two processes.

2. Materials and methods

2.1. Raw material

The honey samples considered in the present study were selected by Piana Ricerca e Consulenza S.r.l. laboratory (Italy), with the aim of reproducing specific F/G ratios of 1.05, 1.25 and 1.45. Samples were named respectively: fast crystallization (FC), medium crystallization (MC) and slow crystallization (SC). For each, half was used for static crystallization (s) and half for the dynamic one (d).

Before starting the crystallization trials, samples were gently heated up to 50 °C in order to melt any preformed crystal. The absence of glucose crystals was evaluated by optical microscopy. Each sample was added with 5% (w/w) of glucose crystal seed obtained by citrus fresh liquid honey that was processed by continuous stirring at 14 °C for 5 days. The same seed was used for both static and dynamic processes. Prior to use, the seed was stored at 14 °C. The precise composition (after crystal nucleus addition) in sugars and water content has been reported by Tappi et al. (2019).

2.2. Static crystallization

In order to not break the structure of honey during the crystallization process, liquid honey was poured into samples holders ready for each analytical determination. Samples were kept in a climatic chamber at 14 °C for the duration of the crystallization process.

Storage duration and sampling intervals were determined for each sample by means of preliminary tests aimed to evaluate the total duration of each crystallization process and to consequently plan the related sampling times. The adopted storage durations and sampling intervals were: 0, 2, 7, 9, 22, 43 and 50 days for the fast crystallization (FCs) samples; 0, 10, 15, 34, 51 and 62 days for the medium crystallization (MCs) samples; 0, 7, 14, 21, 28, 34, 41, 48, 63, 83 and 102 days for the slow crystallization (SCs) samples.

2.3. Dynamic crystallization

Samples were subjected to dynamic crystallization with an in-house made steel stirrer (already described in Tappi et al., 2019). The internal volume of the chamber was 1.2 L and the propeller was kept at a constant speed of 14 rpm.

The mixing chamber was externally cooled with a water/ethylene glycol fluid, so to grant a stirred sample temperature of 14 °C. Considering that samples were poured at room temperature, the initial analysis point was considered when the sample reached 14 °C (about 3–4 h). At selected intervals of time, honey samples were collected for the analytical determination without interrupting the process. Care was taken that the level of honey was always over the edge of the cochlea to avoid air being incorporated into the mass. The length of the process and the intervals of analysis were determined on the basis of preliminary trials carried out to evaluate the total duration of each crystallization process and to consequently plan the related sampling times, that for each honey sample were of 0, 1, 2, 4, 7, 8, 10 and 11 days for the fast crystallization (FCd) samples; 0, 1, 2, 3, 5, 7, 9, 12, 14 for the medium crystallization (MCd) samples; and 16 and 0, 2, 3, 4 7, 9, 11, 14, 18, 21, 25, 28 and 32 days for the slow crystallization (SCd) samples.

2.4. Analytical determinations

2.4.1. Differential scanning calorimetry (DSC)

Thermal analysis was carried out by differential scanning calorimetry using a DSC mod. Q20 (TA Instrument, Germany) equipped with a cooling unit (TA-Refrigetated Cooling System90). Heat flow and temperature calibration were performed with distilled water (T_m 0.0 °C), indium (T_m 156.60 °C) and zinc (T_m 419.47 °C) under a dry nitrogen flow of 50 mL min^{-1}.

After honey was added with a crystal nucleus, it was weighed in 50 μl aluminum DSC capsules and sealed. Samples were stored at 14 °C and at each sampling time three replicates were analysed.

Honey samples were scanned at 5 °C/min from 14 to 100 °C.

Peaks were integrated with the software TA Universal Analysis (TA Instrument, Germany) determining temperature (T_m) and enthalpy (ΔH) of melting of granulated honeys. Glucose monohydrate (Sigma-Aldrich, MA, USA) was used to evaluate the specific melting enthalpy (J/g) of crystals.

2.4.2. Colour

Colour has been determined by using a spettro-photocolorimeter mod. Color-FlexTM-A60-1010-615, (Hunter Lab, Reston, Virginia) equipped with a sample holder port size of 50''. The instrument was calibrated with black and white standard tiles before each set of measurements. The CIE 1976 L*a*b* system was used, the parameters of L* (lightness) and a* (red/green index) were considered on three replicate measurements.

2.4.3. Texture

For textural measurements, liquid honey was poured in small glass vases (internal volume of 36 mL, filled by half). For each sample, three replicates were analysed at each sampling time.

A penetration test was carried out by using a Texture Analyser TA.HDi500 (Stable Micro Systems, Surrey, UK) according to Conforti, Lupano, Malacalza, Arias and Castells (2006) with some modifications. A cylindrical stainless-steel probe with a flat section (d = 10 mm) at a displacement speed of 0.5 mm/s was used. Penetration was carried out for 7 mm into the product. By the acquired curves of Force (N) versus time (s), the following parameters were extrapolated: hardness (N) as the force registered at the peak value, adhesivity (N s) as the negative force area obtained after compression.

2.4.4. Rheological measurements

Rheological measurements on honey samples were carried out at 25 °C using a controlled stress–strain rheometer mod. MCR 300 (Physica/Anton Paar, Ostfildern, Germany) equipped with a plate-plate (PP50) and a Peltier system. The rheological behaviour was analysed in steady state conditions. After a pre-shearing of 500 s at 0.1 s^{-1}, viscosity was measured increasing shear rate from 0.1 to 10 s^{-1} within 250 s, taking 25 measurements points. Ranges were chosen after preliminary trials and according with studies of Nayik et al. (2018) and Gómez-Díaz et al. (2009) with some modifications. Each measurement was replicated three times in two different samples (1 mL each). Moreover, in order to better compare results, viscosity values of all samples were also compared at the references shear of 0.1 and 10 (1/s), according with Boussaid et al. (2015).

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2.4.5. Microstructural analysis

Microstructure of honey was qualitatively evaluated by microscopic observation, performed with a Nikon upright microscope mod. Eclipse Ti–U (Nikon Co, Japan) equipped with a Nikon digital video camera mod. digital sight DS-Qi1Mc (Nikon Co, Japan).

A small amount of honey was placed on a microscope slide and covered with another cover slide. Observations were carried out against a dark background with a crossed polarizer. Images were acquired at a magnification of 20x.

2.5. Statistical analysis

For colour, texture and rheological parameters, significant differences (p-level < 0.05) between mean values at the beginning and at the end of the crystallization process were explored by means of the analysis of variance (ANOVA with post-hoc Tukey HSD). Pearson's analysis, (p-level < 0.05) was performed to evaluate the correlation among the evaluated parameters. Principal component analysis (PCA) was used as explorative technique to discriminate the samples and to display the correlation between the parameters of colorimetric, textural and rheological parameters. All the statistical elaboration was made by Statistica 8 (StatSoft, INC, USA).

3. Results and discussion

3.1. Differential scanning calorimetry (DSC)

The specific melting enthalpy of glucose crystals calculated by DSC measurements was 176.8 ± 4.3 J/g. This value is considerably higher compared to the ones reported in the scientific literature (Al-Habsi et al., 2013; Venir et al., 2010) that were in the range of 106–108 J/g, although the shape of the observed peaks were very similar, the first one related to water released and the second one to the melting of the resulting crystal. The difference may be due mainly to a different scanning rate used (10 °C/min compared to the 5 °C/min of the present work) and to the DSC sensitivity. However, through this value, the amount of crystals in 100 g of honey were determined and the values obtained at the end of the storage for each sample were consistent with the expected ones (data not reported), considering the initial glucose content and its solubility in water.

Crystal content was monitored for a longer period (data not reported) but there was no significant increase in any of the samples, confirming that crystallization reached its final point.

Fig. 1 shows the evolution of crystal content in honey samples crystallized statically (A) or dynamically (B). Crystal contents at the end of the processes did not show significant differences ($p < 0.05$) for the same honey type but subjected to different conditions of crystallization. For FC samples, final crystal content was in the range of 21–22 g/100 g, for MC samples in the range of 17–18 g/100g and for SC in the range of 12–13 g/100g. However, the rate of crystallization was considerably different as observed by the figure. Indeed, static samples reached complete crystallization after 50, 90 and 102 days for respectively FC, MC and SC samples, while only after 11, 16 and 32 when they were constantly stirred in guided system.

Dynamic crystallization is known to increase crystallization rate and to affect honey rheological properties. However, to our knowledge, this process has been investigated so far only in our previous research (Tappi et al., 2019), addressing the changes in water state. By using both DSC and TD-NMR, two main phases were observed during static crystallization, while only one fairly linear phase was observed during dynamic crystallization. As also observed by Venir et al. (2010) and Dettori et al. (2018), the biphasic behaviour can be attributed to the alternation of the nucleation phase and the crystal growth phase. The constant stirring of the honey mass during dynamic crystallization may have hindered the growth of the crystals favouring a constant process of nucleation.

To calculate the rate of crystallization of the different samples, the slope of the straight lines (for static samples before the change of slope as suggested by Venir et al. (2010)) were calculated.

Results are reported in Table 1.

As expected, the initial crystallization rate decreased by increasing the ratio F/G, in both crystallization modes. The energy input provided by stirring promoted an increase of the crystallization rate of 1.3–2 folds compared to the respective static sample. According to Hartel and Shastry (1991), the forced migration of molecules promoted by agitation counterbalances the nucleation impediment given by increased viscosity.

3.2. Colour

Colour of honey is mainly related to its botanical origin and to the content of some minor components as minerals and polyphenols; however, it may be subjected to changes due to processing operations and storage. Fig. 2 shows the evolution of colour parameters of L* and a* during static (A; C) and dynamic (B; D) crystallization of the three types of honeys. Generally, an increase of L* was observed in all samples, ranging from 10 to 20 points values in static honey, and from 20 to 40 in dynamic honeys. The a* parameter, on the contrary, showed only very little variations during static crystallization, while showed a significant decrease during the dynamic one, in particular in sample FCd, reducing the differences among samples related to this parameter.

In a previous study by our research group, Dettori et al. (2018), the appearance of glucose crystals during storage showed to promote colour changes proportionally to the crystallization rate, leading to higher differences when the F/G ratio was lower. In the present research, it appears that the dynamic crystallization causes a more pronounced variation of colour characteristics, both in terms of L* and a*, that leads to a more uniform final appearance of the samples.

3.3. Texture

Fig. 3 shows the textural parameters of respectively hardness (A) and adhesivity (B) measured in the different honey samples during crystallization. Initial values of hardness were very low and similar for all samples (0.01–0.03 N) without significant differences ($p < 0.05$).

During static crystallization, hardness, that can be considered as an index of the internal forces within a crystallized honey structure, increased significantly and proportionally to the crystallization rate, as already observed by Dettori et al. (2018), reaching values of 48.9, 27.3 and 2.8 N for respectively FC, MC and SC samples. Similarly, adhesivity, starting from an average value of −0.03 N reached values of −12.7, −9.9 and −2.6 N for respectively the FC, MC and SC samples. This parameter depends on interatomic and intermolecular forces and is the sum of surface and cohesive energy.

The obtained results showed that, as the crystalline phase increased, a solid structure was formed where crystals created a matrix characterized by a cohesion that increased with time. Moreover, it is possible to state that the composition influenced not only the rate of crystallization, but the amount of glucose that crystallizes also affects the final structure and consequently the textural properties.

When the honey mass was constantly stirred, a slight increase of hardness and adhesivity was also observed. At the end of the process, hardness values were in the range of 0.02–0.04 N and adhesivity values in the range of −0.03–0.06 N. In general, the magnitude of the changes was considerably lower compared to the statically crystallized samples and only for FCd sample values were significantly ($p < 0.05$) higher compared to the initial ones.

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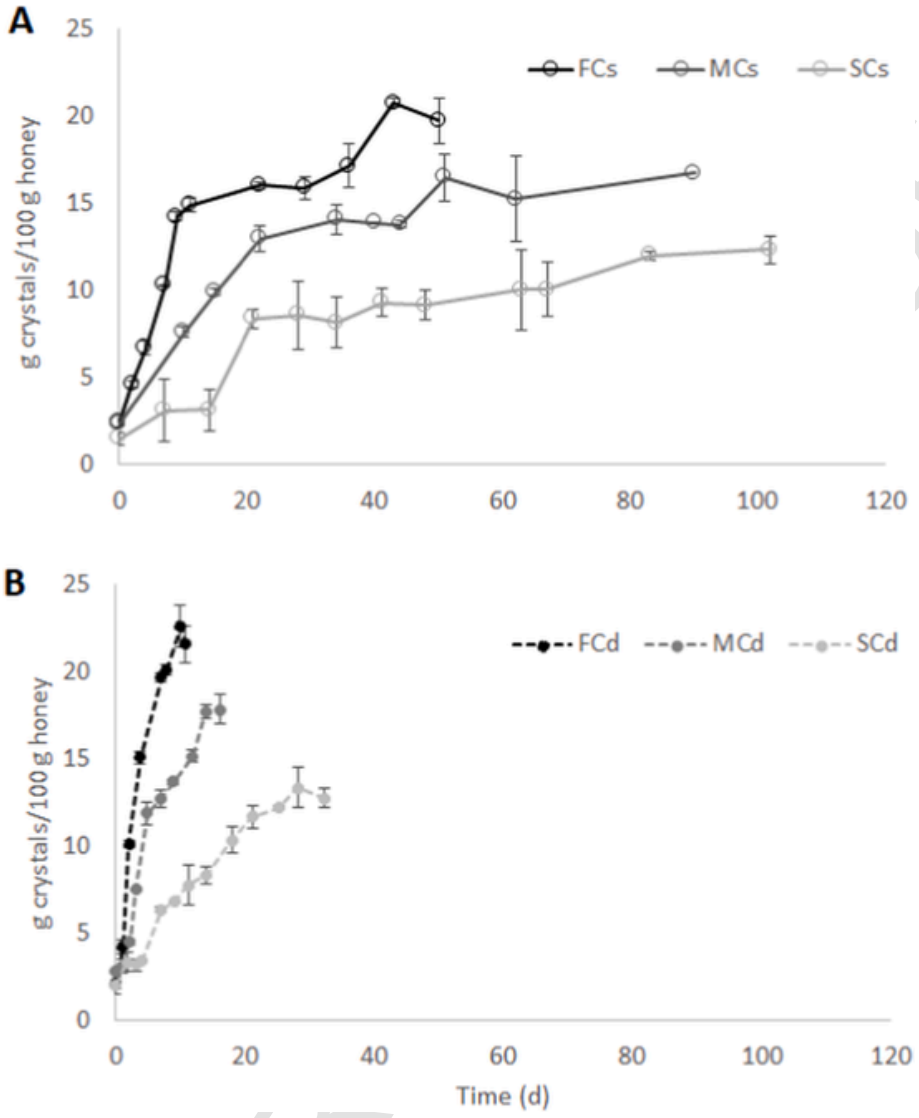

Fig. 1. Crystallization kinetic of honey stored statically (A) and dynamically (B) at 14 °C.

Table 1
Crystallization rate ((g crystals/100 g honey) day $^{-1}$).

	Static Crystallization mode		
Sample		R^2	Days
FC	1.271	0.985	9
MC	0.486	0.791	21
SC	0.296	0.998	21
	Dynamic Crystallization mode		
Sample		R^2	Days
FC	2.074	0.932	10
MC	1.086	0.916	14
SC	0.405	0.975	28

3.4. Rheological measurements

Table 2 reports the values of viscosity measured in the three honey samples, just after seeding and at the end of the crystallization processes, both static and dynamic. Despite the differences in composition, no differences were observed in the initial viscosity values that were in the range of 7–9 Pa s.

During static crystallization, as expected, a significant increase was observed in all samples proportionally to the rate of the process and inversely proportional to their F/G ratio.

A similar result was observed at the end of dynamic crystallization. However, absolute values were 3-5-fold lower compared to the one obtained through static crystallization. This is consistent with the expected results according to Gonnet (1994).

Moreover, differences were also observed in the flow behaviour of the samples as a consequence of the crystallization method. Fig. 4 reports an example of the obtained flow curves, under the form of the dependence of shear stress on shear rate for FCs samples during static (A) and dynamic (B) crystallization.

In the liquid state, at the beginning of the process, all samples showed a Newtonian behaviour, as already reported by various authors (Bakier et al., 2016; Bhandari et al., 1999; Mora-Escobedo et al., 2007).

Honeys obtained by static crystallization, except that for t_0, followed a similar pattern of flow (Fig. 4). According to literature, at low shear rate (first region), Brownian movements predominate over hydrodynamic forces and in sample t_0 a lower Newtonian plateau was observed. In the other samples, at different times of storage and at low

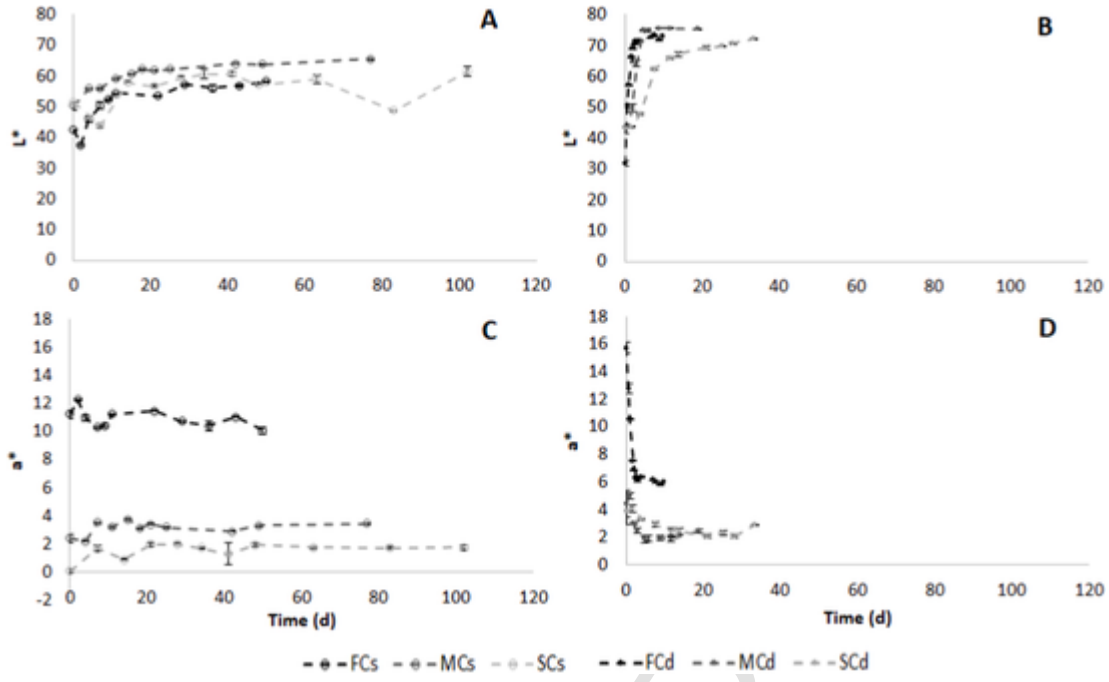

Fig. 2. Evolution of luminosity (L*) and red index (a*) in honey samples during static (A and C) and dynamic (B and D) crystallization.

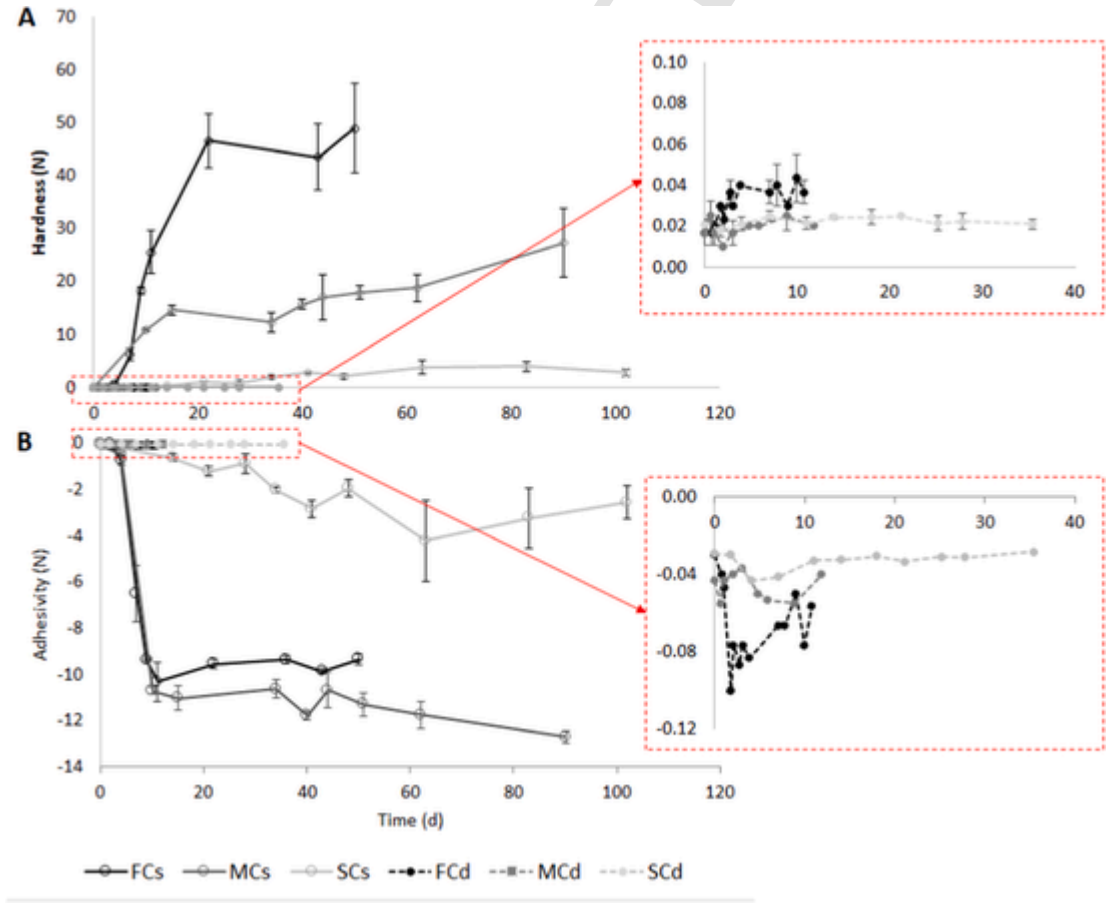

Fig. 3. Textural parameters of hardness (A) and adhesivity (B) of honey samples during static and dynamic crystallization.

shear rates, an apparent yield value was highlighted, becoming higher as storage time increased, probably in relation to the growth of the crystals. In the intermediate shear rate region, hydrodynamic forces prevail over structural and Brownian motion forces and particles are deformed and orientated in flow, involving a reduction in viscosity values, as observed in Fig. 4. This behaviour was evident for all the sam-

Table 2
Viscosity (Pa s) of honey samples at the beginning and at the end of the static or dynamic crystallization process.

Sample	initial value	final value	
		static	dynamic
FC	7.14 ± 0.89 [a]	182.04 ± 9.59 [a]	37.67 ± 3.22 [a]
MC	8.87 ± 2.28 [a]	61.44 ± 8.49 [b]	17.50 ± 0.34 [b]
SC	7.58 ± 1.06 [a]	45.58 ± 17.02 [c]	14.77 ± 4.24 [c]

Different letters indicate significant differences along the same column at $p < 0.05$.

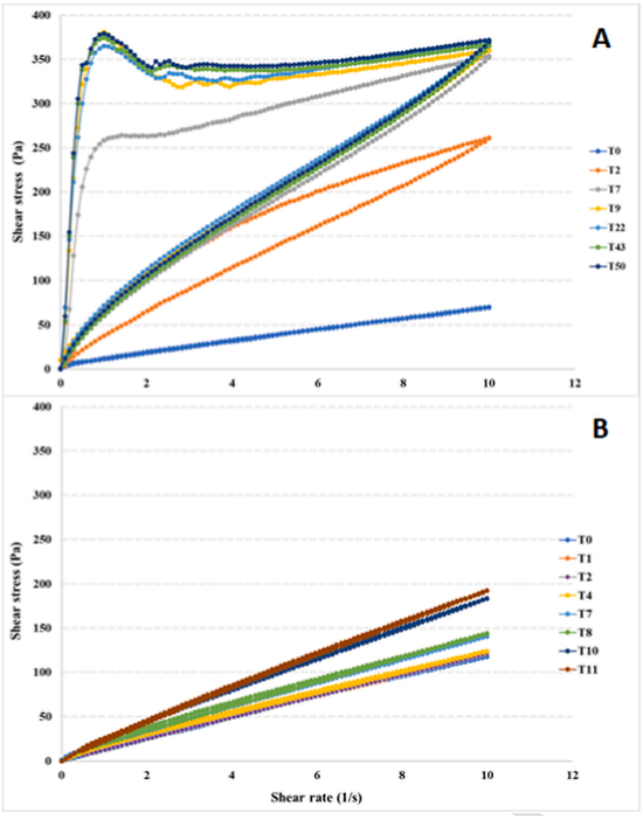

Fig. 4. Example of the flow curves for fast crystallization (FCs) samples during static (A) and dynamic (B) crystallization.

ples except t_0, that showed a Newtonian behaviour (a constant trend) for all shears. In the third region, it was possible to observe an upper Newtonian region in which the viscosity ($\eta\infty$), called the limiting viscosity at infinite shear rate, was constant with the changing of the shear rate (Aguilera and Stanley, 1999; Banerjee et al., 2010; Galindo-Rosales et al., 2010).

On the contrary, as shown inf Fig. 4B, during dynamic crystallization, although the value of viscosity increased with time, all samples maintained a Newtonian behaviour until the end of the process.

Crystallized honey is a two-phase structure of a semi-solid type, whose rheological behaviour is determined by the mass fraction of both the solid phase and the morphology of the crystalline structure Mora-Escobedo et al. (2007). Considering the DSC results, both types of crystallization lead to the same amount of crystallized glucose at the end of the process; however, the constant stirring of the honey mass probably avoided the formation of large size crystals and of a crystalline structure.

It is important to underline that viscosity is a fundamental characteristic for honey, both from a technological point of view and form a consumer acceptance point of view (Bakier et al., 2016).

3.5. Microstructural analysis

Fig. 5 shows, as an example, the images, obtained using an optical microscope with a polarising filter, of FC honey samples, at the beginning (A) and at the end of the crystallization process carried out in static (B) and dynamic mode (C).

The crystals visible at the beginning belongs to the crystal nuclei used for inducing the granulation process. In the samples crystallized in static mode (B), crystals grew in size and appeared to have formed, at the end of the process, a solid crystalline structure with large, irregular and sharp hedged crystals, that reached length up to 200 μm.

However, in the sample kept under constant agitation (C), up to three dimensional classes can be observed: only few big crystals (length > 50 μm) are visible and can be assumed to derive from the original crystal nuclei; a second class is represented by 1–50 μm size crystals that have a needle or a plate shape and finally, in the background, it is possible to observe a large number of very small crystals (<1 μm).

This result confirms the previously observed properties of the different honeys. In particular, shape and size of the crystals may influence, according to the scientific literature (Negueruela and Perez-Arquillue, 2000; White J. W., 1976), the colour parameters of honey, probably on account of a different light refraction. Moreover, the absence of a structure and the presence of micro-crystals are probably the cause of the textural and rheological properties observed.

3.6. Data correlation

The Pearson correlation matrix among colorimetric, textural and rheological parameters evaluated at the beginning and at the end of the crystallization process is reported in Table 3. Results showed that only textural and rheological parameters were significantly correlated, specifically viscosity was positively correlated to hardness ($r = 0.945$) and negatively to adhesivity ($r = -0.740$). On the contrary, the colorimetric parameters of luminosity (L*) and red index (a*) were not correlated between themselves, nor with the other parameters.

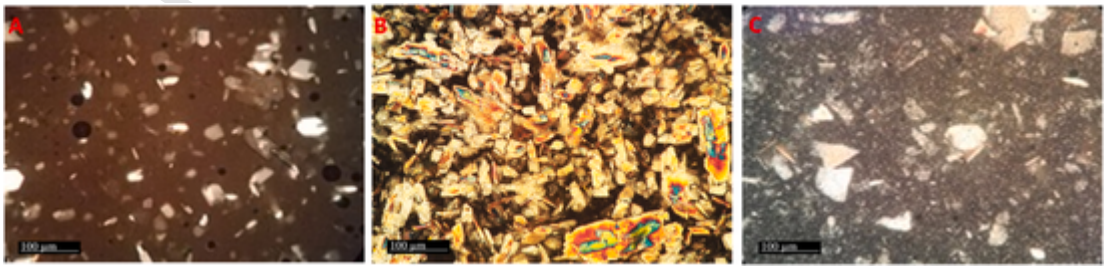

Fig. 5. Crystal images, acquired with optical microscope with a polarising filter, of MC samples at the beginning of the process (A) and at the end of the crystallization performed with the static (B) and dynamic (C) methods.

Table 3
Pearson correlation among colorimetric, textural and rheological parameters.

	L*	a*	Hardness	Adhesivity	Viscosity
L*	–				
a*	−0.369	–			
Hardness	0.178	0.215	–		
Adhesivity	−0.243	−0.050	−0.885*	–	
Viscosity	0.225	0.261	0.945*	−0.740*	–

* Significant at $p < 0.05$.

PCA was developed considering all parameters evaluated in this study and the obtained score plot is reported in Fig. 6. Along PC1 (56.44%), samples are separated over textural and rheological properties. Samples resulted arranged from left to right with increasing hardness and viscosity and decreasing adhesivity. Samples FC and MC at the end of the crystallization (t_f) were clearly separated from the others when the process was static, while for SC samples the distance was minimal, similar to the distance between all the samples crystallized with the dynamic process.

PC 2, that explained 27.67% of the variance, discriminated samples according to the L* and a* colorimetric parameters. Generally, all samples moved towards the lower part of the graph during crystallization, however the highest distance was observed for SC samples.

With regards to both colour and structural parameters, samples crystallized with the dynamic process resulted very close together, showing how the dynamic process tends to lead to a similar quality even if starting from different initial levels.

4. Conclusions

Results of the present research showed that the constant movement of the honey mass during induced crystallization leads to increase its rate, but also to very different qualitative parameters, in terms of colour, texture and rheology. The most noticeable difference is the strong hardness and viscosity reduction of the final crystallized honey obtained by stirring, and the maintenance of the Newtonian behaviour due to the formation of very small crystals and the absence of a strong crystallized matrix. Moreover, while the traditional static method leads to pronounced differences in the crystallized product based on initial compositional differences, the dynamic method allows to obtain much more similar final products.

Different textural and rheological characteristics can be very important for consumer perception and for new product development. For these reasons, the modulation of the crystallization method through the addition of crystal nuclei and the use of stirring could be very promising. However, the influence of the crystal nuclei amount and of the stirring rate should be further evaluated, in order to clarify the effect of these processing parameters on the final honey quality and stability.

CRediT authorship contribution statement

Silvia Tappi: Conceptualization, Methodology, Investigation, Formal analysis, Visualization, Writing - original draft. **Virginia Glicerina:** Investigation, Methodology, Writing - original draft. **Luigi Ragni:** Resources, Writing - review & editing. **Amanda Dettori:** Investigation, Conceptualization, Data curation. **Santina Romani:** Writing - review & editing. **Pietro Rocculi:** Supervision, Funding acquisition, Writing - review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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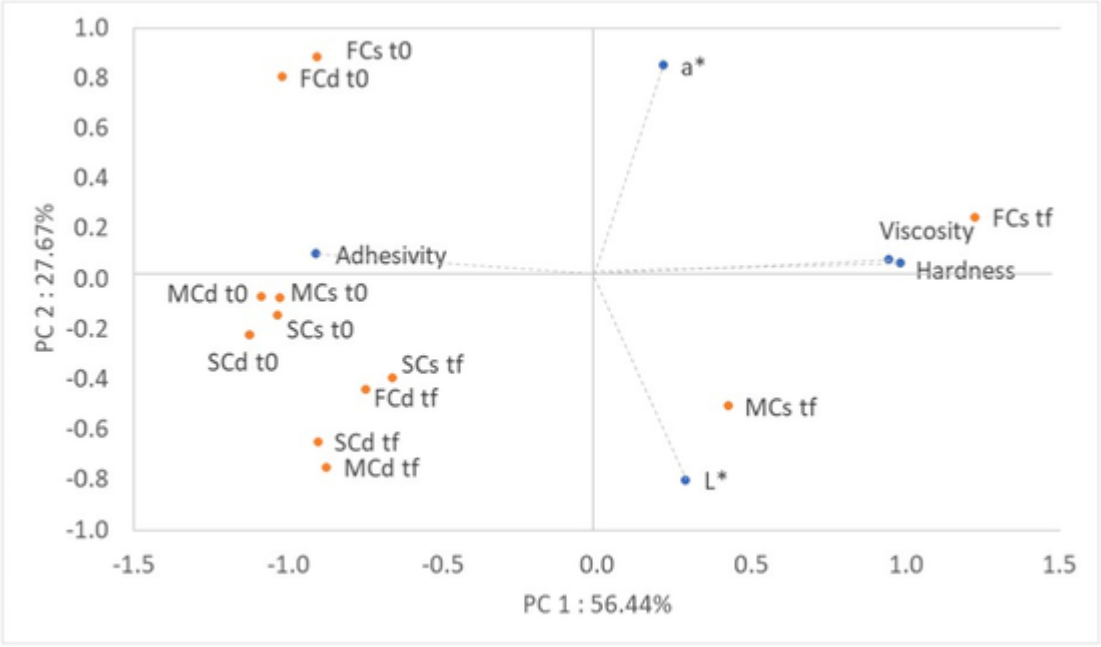

Fig. 6. Scores and loadings biplot of measured variables and considered samples at the beginning (t0) and at the end (tf) of the crystallization processes.

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