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Credit author statement

David J. McSweeney: Conceptualization, Methodology, Formal Analysis, Investigation, Visualization, Writing – Original Draft

Valentyn Maidannyk: Methodology, Investigation

James A. O'Mahony: Funding acquisition, Supervision, Writing – Review and Editing

Noel A. McCarthy: Funding acquisition, Project Administration, Conceptualization, Supervision, Writing – Review and Editing

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Rehydration properties of regular and agglomerated milk protein concentrate powders produced using nitrogen gas injection prior to spray drying

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Abstract

This study evaluated the effect of high-pressure nitrogen (N_2) gas injection prior to spray drying on the subsequent rehydration properties of regular and agglomerated milk protein concentrate (MPC) powders. Conductivity measurements demonstrated a slower release of ions for powders produced using N_2 injection (NI) as they took longer to wet and sink due to their lower density. However, analysis of particle size distribution on reconstitution at both 23 and 50 °C showed an improvement in powder dispersion with NI. Powder solubility, when measured at 23 °C, was higher for the NI powders, while agglomeration negatively impacted solubility. Confocal laser scanning microscopy analysis showed a faster diffusion of dye into regular MPC powder particles produced using NI. The improvement in powder dissolution with NI was attributed to higher porosity and the presence of air voids which facilitated increased water transfer and accelerated the breakdown of primary powder particles.

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1. Introduction

The rehydration of high-protein, casein-dominant dairy powders, including milk protein concentrate (MPC), is currently a significant challenge encountered by the food and beverage industry. These value-added ingredients provide unique nutritional (i.e., high-protein and calcium, low lactose content) and functional (i.e., heat stability, gelation) properties. To exploit the functionality of dry dairy ingredients, rapid and complete powder rehydration is generally required. However, this is impaired by reduced water transfer due to non-covalent protein-protein interactions (Havea, 2006) and high micellar casein content (Schuck et al., 1998, 2002), with dispersion of primary powder particles regarded as the rate-limiting stage of rehydration due to the presence of a network of casein micelles at the powder particle surface (Mimouni et al., 2009).

A water temperature of approximately 50 °C, in combination with high-shear treatment and extended mixing times, are normally required to accelerate the rehydration of casein-dominant powders (Gaiani et al., 2006b; McCarthy et al., 2014), but this is not desirable for ingredient manufacturers and end-users. Ideally, rehydration should take place within a short time period at ambient temperature (~20 °C) and low shear to minimise manufacturing time and production costs (Saggin and Coupland, 2002). Previous research has proposed several processing and formulation strategies to promote the rehydration of casein-dominant powders, including cold microfiltration during micellar casein concentrate manufacture (Crowley et al., 2018), high-pressure treatment (Udabage et al., 2012), acidification of skim milk before membrane filtration (Liu et al., 2019), and the incorporation of monovalent salts (e.g., sodium chloride) into the concentrate before spray drying (Schuck et al., 2002; Sikand et al., 2013).

Gases have been used in dairy processing to alter the functional properties of a range of products. For example, carbon dioxide (CO_2) has been used to improve the shelf life and quality of milk, cheese and fermented beverages (Hotchkiss et al., 2006). However, only a limited number of studies have reported their impact on the rehydration properties of dairy powders. Marella et al. (2015) investigated the effect of CO_2 injection into skim milk before and during membrane filtration and reported the subsequent characteristics of the MPC powder. An improvement in cold water (10 °C) solubility was observed, which was attributed to the solubilisation of colloidal calcium phosphate during membrane fractionation due to the decrease in pH and a reduction in micellar casein interactions. However, the incorporation of CO_2 into dairy streams during processing may change product composition, presenting challenges for some existing applications. Bouvier et al. (2013) used CO_2 during extrusion-porosification to manufacture MPC powders with enhanced dispersibility compared to MPC produced using conventional spray drying. The achievement of a sub-micron particle size distribution after only 2 h of rehydration was attributed to the partial dissociation of casein micelles as well as increased porosity of powder particles. Kosasih et al. (2016a, 2016b) investigated the addition of dry ice (i.e., solid CO_2) to whole milk concentrate prior to spray drying and showed an improvement in the dispersibility of powder particles. Nitrogen (N_2) gas has also been used in dairy processing to modify ingredient functionality (Adhikari et al., 2018). One apparent benefit of using N_2 gas is that, unlike CO_2 and compressed air, it is inert, so is unlikely to alter the pH of the dairy concentrate or promote oxidation in the final product. Hanrahan et al. (1962) reported an improvement in whole milk powder dispersion when N_2 gas was incorporated into the concentrate before spray drying. Similarly, Bell et al. (1963) enhanced the dispersibility of skim milk powder by injecting compressed air into the concentrate between the high-pressure pump and atomisation nozzle.

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Aside from the incorporation of gas into dairy streams, powder particle structure and physical properties can be modified *via* a process known as agglomeration. It can be performed by returning fine powder particles from the cyclone to the top of the drying chamber during droplet dehydration or by combining the spray dried powder with water or a binder in the fluidised bed (Gianfrancesco et al., 2008). The process of intentionally mixing the atomised spray with small, dry powder particles is known as forced secondary agglomeration (Pisecky, 2012). The effects of fluid bed agglomeration on the physicochemical properties of milk protein isolate powders have been reported by Ji et al. (2015, 2016, 2017), whereby improvements in powder wettability were achieved, with no improvement in solubility. Gaiani et al. (2007) reported that agglomeration using fines return was effective in accelerating rehydration of whey protein powder, while it resulted in impaired rehydration performance for casein-dominant powder. Furthermore, the rehydration characteristics of MPC powders produced using both agglomeration and N_2 gas injection have not been established. A previous study (McSweeney et al., 2021) by the current authors investigated the influence of N_2 gas injection directly prior to spray drying, agglomeration by fines return, and a combination of these approaches, on the physical and bulk handling properties of MPC powders. The MPC powder produced using N_2 gas injection had lower density and flow index values, with higher air content, specific surface area, porosity and surface fat, compared to the powders produced without N_2 gas injection, while agglomeration also decreased powder density but improved flowability. Given the significant changes to the structure of the powder particles, the current study was designed to investigate the rehydration properties of these MPC ingredients. Several techniques were employed to elucidate the impact of these processing modifications on the performance of the powders throughout the main stages of rehydration (i.e., wetting, dispersion and dissolution).

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2. Materials and methods

2.1. Powder manufacture

The manufacture, composition and basic physical properties (e.g., density, morphology, porosity, powder particle size) of the regular (R), regular with nitrogen gas injection (RN), agglomerated (A) and agglomerated with nitrogen gas injection (AN) milk protein concentrate (MPC) powders used in this study were described by McSweeney et al. (2021). Briefly, concentrate (21.2% total solids, w/w) was first prepared from MPC powder using high-shear treatment and hydrodynamic cavitation. Prior to spray drying, the concentrate was pre-heated to 70 °C and pumped to the atomisation nozzle using a high-pressure pump (HPP). Regular (R) MPC powder was produced using a conventional spray drying process. Agglomerated (A) powders were manufactured by returning all fines collected in the cyclone to the atomisation zone of the spray dryer main chamber. For MPC powders produced with nitrogen (N_2) gas injection (i.e., RN- and AN-MPC), N_2 was injected (3.5 kg/h) at a pressure of ~190 bar into the feed line, after the HPP and prior to atomisation, using a pressurised injection unit (Carlisle Process Systems, Farum, Denmark).

2.2. Immersional and capillary rise wetting behaviour

Immersional wetting was measured using the GEA Niro method (GEA Niro, 2009) with one modification; 4 g of each powder sample was added to the beaker of water (250 mL; 25 °C). Capillary rise wetting was measured using a modified Washburn method with 2 g of each powder sample added to a cylindrical stainless-steel tube (diameter = 2.4 cm) with an open base covered by filter paper and parafilm (Ji et al., 2015). The analysis was first carried out with no powder to determine the quantity of water absorbed by the filter paper and parafilm (i.e., control), and subsequently this value was subtracted from the test values. The weight of the tube was recorded before and after the addition of powder. The top of the tube

was submerged in 25 °C ultrapure water and the wettability was quantified by measuring the additional mass of the wetted powder after 20 min, with results presented as the mean of three independent measurements.

2.3. Confocal laser scanning microscopy and liquid phase water diffusion

A Leica TCS SP5 confocal laser scanning microscope (CLSM; Leica Microsystems CMS GmbH, Wetzlar, Germany) was used for the real-time visualisation of dye penetration into powder particles, as described by Power et al. (2020). Liquid phase water diffusion in MPC powders was measured using the novel method presented by Maidannyk et al. (2019). Rhodamine B was added to anhydrous powders which allowed diffusion of the dye molecules into the particles without changing particle morphology and preventing solubilisation, thereby providing an indicator of powder hydration. The CLSM images were obtained at fixed time intervals and represent real-time water diffusion. Diameters of particles were detected using Leica TCS SP5 software in the size range 6-142 µm. The areas of individual powder particles were measured using spherical approximation and this information, combined with the time of dye penetration, enabled the local effective diffusivity of the liquid phase in individual powder particles to be calculated. Initially, powder particles appear as dark particles with a dark green background. However, during the water diffusion process, the fluorescent dye penetrates the particles and changes their colour to bright green.

2.4. Water sorption isotherms

Water sorption analysis was carried out as described by Maidannyk et al. (2020), with one modification: powders were weighed at intervals of 0, 2, 4, 6, 8, 10, 24, 48, 72, 96 and 120 h. The water content in each system was plotted as a function of time, and the

Guggenheim-Anderson-de Boer (GAB) relationship was fitted to data to relate water activity and water content of anhydrous powders, as shown in equation (1):

$$\frac{m}{m_0} = \frac{Cka_w}{(1-ka_w)(1-ka_w+Cka_w)} \quad (1)$$

Where m is the water content (g of water/100 g of dry solids), m_0 – the monolayer value of water content, C, k – constants, which can be calculated from m_0.

2.5. Measurement of mineral release using conductivity

Conductivity of MPC dispersions (1.5% protein, w/w; 300 mL of ultrapure water in a 400 mL beaker) was measured using a Titrando autotitrator equipped with a five-ring conductivity measuring cell and accompanying Tiamo v2.3 software (Metrohm Ireland Ltd, Athy Road, Co. Carlow, Ireland). The probe was calibrated at 25 °C with a KCl solution of known conductivity (12.9 mS/cm) and a temperature coefficient of 2.07 was used (Crowley et al. 2015). Before the addition of powder to the beaker of water, 1 min was allowed to elapse to establish a baseline reading of conductivity and powder was then added over a period of 1 min. It is expected that cations and anions found in the serum phase, (e.g., H^+ and Cl^-) would contribute most to conductivity measurements rather than minerals found in the colloidal phase, e.g., calcium (Zhuang et al., 1997; Schuck et al., 2007).

2.6. Particle size distribution of milk protein concentrate dispersions

The particle size distribution of MPC dispersions was measured using a laser-light diffraction unit (Malvern Mastersizer 3000; Malvern Instruments Ltd, Worcestershire UK) equipped with a 300 RF lens, as described by McSweeney et al. (2020). Additionally, powders were reconstituted using low-speed mixing for 1 h at 23 °C. Size measurements

were recorded as the volume-weighted mean particle diameter ($D_{[4,3]}$), median diameter (D_{50}) and cumulative diameters (D_{90} and D_{10}), whereby 10, 50 and 90% of the powder volume is represented by powder particles smaller than the size indicated. Particle size measurements were recorded when the laser obscuration reached 3-4%.

2.7. Powder solubility

The solubility of MPC powders was measured as described by McSweeney et al. (2020). Powder solubility was given by the total solids content of the supernatant (obtained following centrifugation at 3000*g* for 10 min) expressed as a percentage of the total solids content of the initial dispersion.

2.8. Statistical analysis

Measurements of the powder rehydration characteristics were performed in triplicate and results presented as mean ± standard deviation. Analysis of variance (one-way ANOVA; Tukey's HSD) was performed using the IBM SPSS (Version 24; Armonk, NY, USA) statistical analysis package. The level of significance was set at $P < 0.05$.

3. Results and discussion

3.1. Wetting behaviour of milk protein concentrate powders

Wettability analysis by the GEA method showed that all milk protein concentrate (MPC) powders did not completely wet or sink below the surface of the water within 10 min. However, the water became increasingly turbid for the regular (R) and agglomerated (A) nitrogen (N_2) injection powders (i.e., RN- and AN-MPC) compared to R- and A-MPC, in which the water remained relatively clear (data not shown). Bouvier et al. (2013) reported that extrusion-porosification, which created MPC powder particles with high porosity, did not

improve wetting time as it had the same wettability index (>120 s) as a conventionally spray dried MPC powder. The capillary rise wetting behaviour, observed using the modified Washburn method, is shown in Fig. 1. A-MPC absorbed the most water (1.0 g) and R-MPC absorbed the least (0.43 g). The N_2 injection (NI) process appeared to improve capillary rise wetting for the regular powders as RN-MPC absorbed 0.74 g of water. However, for the agglomerated powders, AN-MPC absorbed a lower quantity of water (0.61 g) than A-MPC despite having a higher porosity. The difference in capillary rise wetting between R- and A-MPC may be explained by the differences in powder particle size; A-MPC had a $D_{[4,3]}$ of 142 μm while R-MPC had a $D_{[4,3]}$ of 79 μm. Similarly, Ji et al. (2016) reported water absorption levels of 0.24 and 1.0 g for non-agglomerated and agglomerated milk protein isolate (MPI) powders, respectively. It has been previously reported that agglomeration improved the wetting behaviour of a native phosphocaseinate (NPC) powder due to the large powder particle size (mean = 285 μm) and high porosity (Gaiani et al., 2005, 2007). One of the main factors influencing powder wettability is the surface composition (Gaiani et al., 2006a), and the presence of fat specifically on the surface of spray dried powders would be expected to influence the wetting behaviour by increasing surface hydrophobicity. The surface composition of the powders in the current study were established previously by McSweeney et al. (2021), where it was reported that the NI powders had significantly higher amounts of surface fat (e.g., 2.02 for R-MPC and 11.1 for RN-MPC), while these samples still performed relatively well in powder wetting experiments. Kim et al. (2002) reported that surface fat had a strong, negative impact on the wettability of several dairy powders (e.g., cream, skim and whole milk powder), while Gaiani et al. (2006a) did not find a clear relationship between the surface fat of NPC powders and wetting times.

3.2. Visualisation of liquid phase water diffusion and effective diffusivity

Confocal laser scanning microscopy images showing the movement of the rhodamine B dye into R- and RN-MPC powder particles are displayed in Fig. 2. Complete diffusion of rhodamine dye into R-MPC powder particles took 1563 s (Fig. 2A) compared to 196 s for RN-MPC (Fig. 2B). This was likely caused by the significantly higher occluded air (R-MPC = 24.2 mL/100 g, RN-MPC = 36.3 mL/100 g) and porosity (R-MPC = 0.69, RN-MPC = 0.88) values reported by McSweeney et al. (2021) for this powder.

Large differences were observed in effective diffusivity between agglomerated and regular MPC powders (Fig. 3). AN-MPC had the highest effective diffusivity value of 8.09^{-12} m^2/s compared to 4.09^{-12} m^2/s for A-MPC. Conversely, the movement of rhodamine dye into R-MPC occurred at the slowest rate among all powders at 3.29^{-13} m^2/s, with RN-MPC slightly higher at 4.18^{-13} m^2/s. It is apparent that NI prior to spray drying assisted the transfer of the aqueous dye into the powder particles. The rate of diffusion was most likely higher for agglomerated MPC powders due to the larger powder particle size compared to regular MPC powders. The link between higher effective diffusivity and increasing powder particle size has been reported previously by Power et al. (2020) for enzymatically crosslinked MPC powders.

3.3. Water sorption isotherms

Water sorption profiles for MPC powders are displayed in Fig. 4. It is evident that water content and the time to reach equilibrium increased with increasing relative humidity (RH). Lactose crystallisation did not occur as this is generally indicated by a sudden decrease in water content, while Kelly et al. (2015) also reported the absence of lactose crystallisation in MPC powders containing ~80% protein (w/w). NI promoted a faster uptake of moisture during the early stages of RN-MPC powder storage compared to R-MPC powders (Fig. 4A and B). For example, after 8 h at 85% RH, R-MPC had a water content of 7.2 g/100 g

compared to 9.5 g/100 g for RN-MPC. However, after 144 h, R-MPC (18.2 g/100 g) and RN-MPC (17.8 g/100 g) powders had similar water contents. Agglomerated powders (Fig. 4C and D) absorbed more water overall than their non-agglomerated counterparts. After 8 h at 85% RH, AN-MPC had a water content of 14.9 g/100 g compared to 11.8 g/100 g for A-MPC. This trend was also evident for the effective diffusivity analysis presented in Section 3.2. whereby larger powder agglomerates favoured the movement of water into the particles. Particle size distribution has been previously identified as an important determinant of a materials water sorption behaviour. Mathlouthi and Roge (2003) reported that smaller particles of sugar were capable of absorbing more water than larger particles, while Murrieta Pazos et al. (2014) observed a similar trend for durum wheat semolina. However, Ji et al. (2017) reported that MPI powders agglomerated using fluidised bed granulation showed similar water sorption, despite differences in particle size. In the current study, the surface composition of powders may have been a contributing factor as the surface of agglomerated powder particles was significantly higher in lactose than that of regular powders (McSweeney et al., 2021).

3.4. Measurement of mineral release using conductivity

The release of minerals from powder particles was complete by approximately 3000 s (Fig. 5). It is evident that R- and A-MPC released ions at a faster rate than both RN- and AN-MPC powders. The R- and A-MPC powders underwent wetting and sinking after approximately 600 s (time to reach steady state), which can be inferred from the beginning of the plateau on the graph. However, a surface barrier was evident during stirring for powders produced using NI and it took ~1400 s for this plateau to be reached. This result is likely related to the physical properties of the NI powders as they had lower bulk and particle density values and higher air contents. Masters (1985) reported that sinking of powder

particles is supported by high particle density and low occluded air, while a low particle density will cause the powder to float on the surface of the water. Fitzpatrick et al. (2016) attributed the poor wettability (>1 h) of an MPI powder to its low apparent density (0.81 g/cm^3), which was similar to the particle density values for RN-MPC (0.96 g/cm^3) and AN-MPC (0.88 g/cm^3). It has been previously reported by Mimouni et al. (2010) that minerals (non-micellar material) are freely released during rehydration of MPC but that protein dispersion is the rate-limiting stage.

3.5. Particle size distribution and solubility of milk protein concentrate dispersions

NI significantly enhanced the dispersion of MPC powder particles following reconstitution (Table 1). When powders were mixed for 30 s at 23 °C, sub-micron particles were not present and all samples had monomodal volume-based distributions, suggesting that casein micelles were not released from primary powder particles (Fig. 6A); however, a significantly smaller particle size was observed for RN-MPC ($D_{[4.3]}$ = 32.6 µm) compared to R-MPC ($D_{[4.3]}$ = 79.6 µm). For agglomerated powders, the $D_{[4.3]}$ was significantly lower for AN-MPC (41.8 µm) in comparison to A-MPC (119 µm) under these conditions. The improvement in dispersion of RN and AN-MPC is likely accounted for by the powders lower loose bulk density and the higher porosity and interstitial space. The more porous structure of NI powder particles and the presence of large air voids between these particles would facilitate increased water transfer, while also increasing the physical space between casein micelles and reducing protein-protein interactions. This would appear to promote the structural collapse of powder particles when added to water, as interactions between poorly dispersible micellar casein, particularly at the particle surface, are considered to be responsible for the slow rehydration of MPC (Anema et al., 2006; Mimouni et al., 2009, 2010).

The water temperature used during reconstitution significantly affected the particle size distribution, with a higher temperature enhancing the fragmentation of MPC powder particles (Fig. 6B). When the temperature of the reconstitution water was 50 °C, the $D_{[4.3]}$ values were 18.4 µm and 1.59 µm for the R- and RN-MPC powders, respectively (Table 1). All powders had a bimodal volume-based distribution, with a peak <1 µm and size range of 8-300 µm. However, the volume of sub-micron particles was higher for RN- and AN-MPC compared to R- and A-MPC. This implies that a large quantity of particles present in the NI powder dispersions were casein micelles, suggesting higher levels of dissolution were achieved.

A-MPC powder had poorer dissolution properties as indicated by the larger particle size ($D_{[4.3]}$ = 119 µm) after reconstitution at 23 °C compared to R-MPC ($D_{[4.3]}$ = 79.6 µm). Therefore, the agglomeration of high-protein powders during spray drying appears to be counter-productive for improving rehydration, unlike its use in the production of skim and whole milk powders (Pisecky, 2012). Gaiani et al. (2005, 2007) reported similar results whereby agglomeration increased the overall rehydration time of NPC powders as it delayed the dispersion process. However, in the current study, reconstitution at 50 °C resulted in no significant differences between agglomerated and regular (non-agglomerated) powders, with $D_{[4.3]}$ values of 20 and 18.4 µm for A-MPC and R-MPC, respectively (Table 1). This suggests that increasing the water temperature may moderately alleviate this issue with A-MPC dispersion.

MPC powders were also analysed after magnetic stirring for 1 h in ultrapure water at 23 °C, with bimodal particle size distributions obtained for NI powders compared to monomodal size profiles for non-NI powders (Fig. 6C). This corresponded to $D_{[4.3]}$ values of 13.6 µm for RN-MPC and 14.4 µm for AN-MPC compared to 83.7 and 66.3 µm for R- and A-MPC, respectively (Table 1). This result further highlights the improved dispersibility of

the NI powders, at a relatively low reconstitution temperature and agitation rate, compared to non-NI powders. Mimouni et al. (2009) reported that 480 min of stirring at 24 °C was required to fully solubilise a MPC powder (85%, w/w, protein). Similarly, Gaiani et al. (2007) reported that 807 and 572 min of stirring at 24 °C were required to dissolve agglomerated and non-agglomerated micellar casein powders, respectively.

Aside from the particle size data of reconstituted powders, solubility results confirmed the superior dissolution of NI powders, particularly at 23 °C (Table 2), with R- and RN-MPC having 83.6 and 96.2% solubility, respectively. The lower loose bulk density and higher porosity for RN-MPC are also likely to be responsible for the higher levels of solubility (McSweeney et al., 2021). Similarly, A-MPC had a solubility value of 62.6% compared to 92.1% for the AN-MPC powder (Table 2). However, when powders were reconstituted at 50 °C, no significant differences in solubility were observed. It has been previously reported by Mimouni et al. (2009) that an increase in solvent temperature accelerates the release of constituent materials from MPC powder particles into the aqueous phase. Overall, these results indicate that physical and structural properties (e.g., density, air content and porosity) play a significant role in the rehydration of high-protein MPC powders.

4. Conclusion

This study demonstrated that the dissolution of MPC powder is enhanced by the injection of N_2 gas into the concentrate prior to spray drying. It is proposed that higher powder particle porosity and lower density are responsible for the improvement in rehydration performance. Water transfer during reconstitution was promoted by the presence of large air voids and pores throughout the powder particles, resulting in a large volume of small, dispersed particles (i.e., <1 µm). Agglomeration alone favoured powder wetting, water uptake and particle hydration; however, it had a negative impact on powder particle

dissolution. Combining N_2 gas injection with agglomeration resulted in further improvements in diffusion and wetting behaviour but did not confer any additional improvement in dispersion and solubilisation of powder particles compared to N_2 gas injection alone. The injection of N_2 gas into high-protein concentrate prior to spray drying is a relatively simple and effective processing technology to enhance powder particle dispersibility and solubility, while avoiding the use of chemical additives which may disrupt casein micelle integrity. The impact of N_2 gas injection on bulk handling and other functional properties of MPC powders (e.g., density and dissolution) after storage and transport should be considered in future research.

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References

Adhikari, B.M., Truong, T., Bansal, N., Bhandari, B., 2018. Use of gases in dairy manufacturing: A review. Crit. Rev. Food Sci. Nutr. 58, 2557-2569. https://doi.org/10.1080/10408398.2017.1333488.

Anema, S.G., Pinder, D.N., Hunter, R.J., Hemar, Y., 2006. Effects of storage temperature on the solubility of milk protein concentrate (MPC85). Food Hydrocolloids 20, 386-393. https://doi.org/10.1016/j.foodhyd.2005.03.015.

Bell, R.W., Hanrahan, F.P., Webb, B.H., 1963. Foam spray drying methods of making readily dispersible nonfat dry milk. J. Dairy Sci. 46, 1352-1356. https://doi.org/10.3168/jds.S0022-0302(63)89280-2.

Bouvier, J.M., Collado, M., Gardiner, D., Scott, M., Schuck, P., 2013. Physical and rehydration properties of milk protein concentrates: comparison of spray-dried and extrusion-porosified powders. Dairy Sci. Technol. 93, 387-399. https://doi.org/10.1007/s13594-012-0100-7.

Crowley, S.V., Burlot, E., Silva, J.V.C., McCarthy, N.A., Wijayanti, H.B., Fenelon, M.A., Kelly, A.L., O'Mahony, J.A., 2018. Rehydration behaviour of spray-dried micellar casein concentrates produced using microfiltration of skim milk at cold or warm temperatures. Int. Dairy J. 81, 72-79. https://doi.org/10.1016/j.idairyj.2018.01.005.

Crowley, S.V., Desautel, B., Gazi, I., Kelly, A.L., Huppertz, T., O'Mahony, J.A., 2015. Rehydration characteristics of milk protein concentrate powders. J. Food Eng. 149, 105-113. https://doi.org/10.1016/j.jfoodeng.2014.09.033.

Fitzpatrick, J.J., van Lauwe, A., Coursol, M., O'Brien, A., Fitzpatrick, K.L., Ji, J., Miao, S., 2016. Investigation of the rehydration behaviour of food powders by comparing the

Journal Pre-proof

behaviour of twelve powders with different properties. Powder Technol. 297, 340-348. https://doi.org/10.1016/j.powtec.2016.04.036.

Gaiani, C., Banon, S., Scher, J., Schuck, P., Hardy J., 2005. Use of a turbidity sensor to characterize micellar casein powder rehydration: Influence of some technological effects. J. Dairy Sci. 88, 2700-2706. https://doi.org/10.3168/jds.S0022-0302(05)72948-9.

Gaiani, C., Ehrhardt, J.J., Scher, J., Hardy, J., Desobry, S., Banon, S., 2006a. Surface composition of dairy powders observed by X-ray photoelectron spectroscopy and effects on their rehydration properties. Colloids Surf. B: Biointerfaces 49, 71-78. https://doi.org/10.1016/j.colsurfb.2006.02.015.

Gaiani, C., Scher, J., Schuck, P., Hardy, J., Desobry, S., Banon, S., 2006b. The dissolution behaviour of native phosphocaseinate as a function of concentration and temperature using a rheological approach. Int. Dairy J. 16, 1427-1434. https://doi.org/10.1016/j.idairyj.2005.12.004.

Gaiani, C., Schuck, P., Scher, J., Desobry, S., Banon, S., 2007. Dairy powder rehydration: influence of protein state, incorporation mode, and agglomeration. J. Dairy Sci. 90, 570-581. https://doi.org/10.3168/jds.S0022-0302(07)71540-0.

GEA Niro, 2009. A 5 b – Wettability IDF method. GEA Process Engineering A/S, Gladsaxevej, Denmark.

Gianfrancesco, A., Turchiuli, C., Dumoulin, É., 2008. Powder agglomeration during the spray-drying process: measurements of air properties. Dairy Sci. Technol. 88, 53-64. https://doi.org/10.1051/dst:2007008.

Hanrahan, F.P., Tamsma, A., Fox, K.K., Pallansch, M.J., 1962. Production and properties of spray-dried whole milk foam. J. Dairy Sci. 45, 27-31. https://doi.org/10.3168/jds.S0022-0302(62)89321-7.

Journal Pre-proof

Havea, P., 2006. Protein interactions in milk protein concentrate powders. Int. Dairy J. 16, 415–422. https://doi.org/10.1016/j.idairyj.2005.06.005.

Hotchkiss, J.H., Werner, B.G., Lee, E.Y.C., 2006. Addition of carbon dioxide to dairy products to improve quality: A comprehensive review. Comp. Rev. Food Sci. Food Safety 5, 158-168. https://doi.org/10.1111/j.1541-4337.2006.00008.x.

Ji, J., Cronin, K., Fitzpatrick, J., Fenelon, M., Miao, S., 2015. Effects of fluid bed agglomeration on the structure modification and reconstitution behaviour of milk protein isolate powders. J. Food Eng. 167, 175-182. https://doi.org/10.1016/j.jfoodeng.2015.01.012.

Ji, J., Fitzpatrick, J., Cronin, K., Fenelon, M.A., Miao, S., 2017. The effects of fluidised bed and high shear mixer granulation processes on water adsorption and flow properties of milk protein isolate powder. J. Food Eng. 192, 19-27. https://doi.org/10.1016/j.jfoodeng.2016.07.018.

Ji, J., Fitzpatrick, J., Cronin, K., Maguire, P., Zhang, H., Miao, S., 2016. Rehydration behaviours of high protein dairy powders: The influence of agglomeration on wettability, dispersibility and solubility. Food Hydrocolloids, 58, 194-203. https://doi.org/10.1016/j.foodhyd.2016.02.030.

Kelly, G.M., O'Mahony, J.A., Kelly, A.L., Huppertz, T., Kennedy, D., O'Callaghan, D.J., 2015. Influence of protein concentration on surface composition and physico-chemical properties of spray dried milk protein concentrate powders. Int. Dairy J. 51, 34-40. https://doi.org/10.1016/j.idairyj.2015.07.001.

Kim, E.H.J., Chen, X.D., Pearce, D., 2002. Surface characterization of four industrial spray-dried dairy powders in relation to chemical composition, structure and wetting property. Colloids Surf. B Biointerfaces 26, 197-212. https://doi.org/10.1016/S0927-7765(01)00334-4.

Journal Pre-proof

Kosasih, L., Bhandari, B., Prakash, S., Bansal, N., Gaiani, C., 2016a. Effect of whole milk concentrate carbonation on functional, physicochemical and structural properties of the resultant spray dried powder during storage. J. Food Eng. 179, 68-77. https://doi.org/10.1016/j.jfoodeng.2016.02.005.

Kosasih, L., Bhandari, B., Prakash, S., Bansal, N., Gaiani, C., 2016b. Physical and functional properties of whole milk powders prepared from concentrate partially acidified with CO_2 at two temperatures. Int. Dairy J. 56, 4-12. https://doi.org/10.1016/j.idairyj.2015.12.009.

Liu, D., Zhang, J., Yang, T., Liu, X., Hemar, Y., Regenstein, J.M., Zhou, P., 2019. Effects of skim milk pre-acidification and retentate pH-restoration on spray-drying performance, physico-chemical and functional properties of milk protein concentrates. Food Chem. 272, 539-548. https://doi.org/10.1016/j.foodchem.2018.08.094.

Maidannyk, V., Lutjes, E., Montgomery, S., McCarthy, N., Auty, M.A.E., 2019. Measurement of effective diffusion coefficients in dairy powders by confocal microscopy and sorption kinetic profiles. Food Struct. 20, 100-108. https://doi.org/10.1016/j.foostr.2019.100108.

Maidannyk, V., McSweeney, D.J., Hogan, S.A., Miao, S., Montgomery, S., Auty, M.A., McCarthy, N.A., 2020. Water sorption and hydration in spray-dried milk protein powders: Selected physicochemical properties. Food Chem. 304, 125418. https://doi.org/10.1016/j.foodchem.2019.125418.

Marella, C., Salunke, P., Biswas, A.C., Kommineni, A., Metzger, L.E., 2015. Manufacture of modified milk protein concentrate utilizing injection of carbon dioxide. J. Dairy Sci. 98, 3577-3589. https://doi.org/10.3168/jds.2014-8946.

Masters, K., 1985. Analytical methods and properties of dried dairy products. In: Hansen, R. (Ed), Evaporation, Membrane Filtration and Spray Drying in Milk Powder and Cheese Production. North European Dairy Journal, Denmark, pp. 393-403.

Journal Pre-proof

Mathlouthi, M., Rogé, B., 2003. Water vapour sorption isotherms and the caking of food powders. Food Chem. 82, 61-71. https://doi.org/10.1016/S0308-8146(02)00534-4.

McCarthy, N.A., Kelly, P.M., Maher, P.G., Fenelon, M.A., 2014. Dissolution of milk protein concentrate (MPC) powders by ultrasonication. J. Food Eng. 126, 142-148. https://doi.org/10.1016/j.jfoodeng.2013.11.002.

McSweeney, D.J., Maidannyk, V., O'Mahony, J.A., McCarthy, N.A., 2021. Influence of nitrogen gas injection and agglomeration during spray drying on the physical and bulk handling properties of milk protein concentrate powders. J. Food Eng. 110399. https://doi.org/10.1016/j.jfoodeng.2020.110399.

McSweeney, D.J., Maidannyk, V., Montgomery, S., O'Mahony, J.A., McCarthy, N.A., 2020. The influence of composition and manufacturing approach on the physical and rehydration properties of milk protein concentrate powders. Foods 9, 236. https://doi.org/10.3390/foods9020236.

Mimouni, A., Deeth, H.C., Whittaker, A.K., Gidley, M.J., Bhandari B.R., 2009. Rehydration process of milk protein concentrate powder monitored by static light scattering. Food Hydrocolloids 23, 1958-1965. https://doi.org/10.1016/j.foodhyd.2009.01.010.

Mimouni, A., Deeth, H.C., Whittaker, A.K., Gidley, M.J., Bhandari, B.R., 2010. Investigation of the microstructure of milk protein concentrate powders during rehydration: Alterations during storage. J. Dairy Sci. 93, 463-472. https://doi.org/10.3168/jds.2009-2369.

Murrieta-Pazos, I., Galet, L., Patry, S., Gaiani, C., Scher, J., 2014. Evolution of particle structure during water sorption observed on different size fractions of durum wheat semolina. Powder Technol. 255, 66-73. https://doi.org/10.1016/j.powtec.2013.10.049.

Journal Pre-proof

Pisecky, I.J., 2012. Dried milk products. In: Westergaard, V., Refstrup, E. (Eds.), Milk Powder Manufacture. GEA Process Engineering A/S, Copenhagen, Denmark, pp. 121-139.

Power, O.M., Maidannyk, V., McSweeney, D.J., Fenelon, M.A., O'Mahony, J.A., McCarthy, N.A., 2020. Water sorption and hydration properties of high protein milk powders are influenced by enzymatic crosslinking and calcium chelation. Powder Technol. 364, 680-688. https://doi.org/10.1016/j.powtec.2020.01.075.

Saggin, R., Coupland, J.N., 2002. Ultrasonic monitoring of powder dissolution. J. Food Sci. 67, 1473-1477. https://doi.org/10.1111/j.1365-2621.2002.tb10308.x.

Schuck, P., Davenel, A., Marietta, F., Briard, V., Mejean, S., Piot, M., 2002. Rehydration of casein powders: effects of added mineral salts and salt addition methods on water transfer. Int. Dairy J. 12, 51-57. https://doi.org/10.1016/S0958-6946(01)00090-5.

Schuck, P., Mejean, S., Dolivet, A., Gaiani, C., Banon, S., Scher, J., Jeantet, R., 2007. Water transfer during rehydration of micellar casein powders. Lait 87, 425-432. https://doi.org/10.1051/lait:2007016.

Schuck, P., Roignant, M., Brulé, G., Davenel, A., Famelart, M.H., Maubois, J.L., 1998. Simulation of water transfer in spray drying. Dry. Technol. 16, 1371-1393. https://doi.org/10.1080/07373939808917466.

Sikand, V., Tong, P.S., Walker, J., 2013. Effect of adding salt during the diafiltration step of milk protein concentrate powder manufacture on mineral and soluble protein composition. Dairy Sci. Technol. 93, 401-413. https://doi.org/10.1007/s13594-013-0110-0.

Udabage P., Puvanenthiran A., Yoo J.A., Versteeg, C., Augustin, M.A., 2012. Modified water solubility of milk protein concentrate powders through the application of static

Journal Pre-proof

high pressure treatment. J. Dairy Res. 79, 76-83. https://doi.org/10.1017/S0022029911000793.

Zhuang, Y, Zhou, W., Nguyen, M.H., Hourigan, J.A., 1997. Determination of protein content of whey powder using electrical conductivity measurement. Int. Dairy J. 7, 647-653. https://doi.org/10.1016/S0958-6946(97)00059-9.

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Figure captions

Fig. 1. Mean weight of water absorbed for regular (R), regular with N_2 injection (RN), agglomerated (A) and agglomerated with N_2 injection (AN) milk protein concentrate (MPC) powders after submerging in ultrapure water (25 °C) for 20 min using a modified Washburn method.

Fig. 2. Confocal laser scanning microscopy images showing the movement of rhodamine B dye into (A) regular and (B) regular with N_2 injection milk protein concentrate powders.

Fig. 3. Effective diffusivity (m^2/s) for regular (R), regular with N_2 injection (RN), agglomerated (A) and agglomerated with N_2 injection (AN) milk protein concentrate (MPC) powders.

Fig. 4. Water sorption isotherms for (A) regular, (B) regular with N_2 injection, (C) agglomerated and (D) agglomerated with N_2 injection milk protein concentrate powders at relative humidity values of 11 (□), 23 (■), 33 (○), 44 (●), 55 (Δ), 65 (▲), 76 (◊) and 85% (♦) over 144 h.

Fig. 5. Conductivity profiles of regular (■), regular with N_2 injection (▲), agglomerated (□) and agglomerated with N_2 injection (Δ) milk protein concentrate powders measured while stirring in ultrapure water (25 °C) for 90 min.

Fig. 6. Particle size distribution profiles of regular (■), regular with N_2 injection (▲), agglomerated (□) and agglomerated with N_2 injection (Δ) milk protein concentrate powders measured after reconstitution in ultrapure water for (A) 30 s at 23 °C, (B) 30 s at 50 °C and (C) 1 h at 23 °C.

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Table 1. Particle size distribution parameters for regular (R), regular with N_2 injection (RN), agglomerated (A), and agglomerated with N_2 injection (AN) milk protein concentrate (MPC) dispersions after high-speed (HS) mixing at 23°C and 50°C for 30 s using a solubility index meter and after low-speed (LS) mixing at 23°C for 1 h using a magnetic stirrer.

Rehydration conditions	MPC	D_{50}	D_{90}	$D_{[4,3]}$
			μm	
HS mixing 30 s at 23°C	R	$68.4^{a} \pm 3.16$	$155^{a} \pm 6.83$	$79.6^{a} \pm 3.38$
	RN	$25.6^{b} \pm 0.85$	$66.0^{b} \pm 2.19$	$32.6^{b} \pm 0.81$
	A	$108^{c} \pm 4.09$	$224^{c} \pm 5.87$	$119^{c} \pm 3.67$
	AN	$35.5^{d} \pm 1.27$	$79.8^{d} \pm 1.75$	$41.8^{d} \pm 1.07$
HS mixing 30 s at 50°C	R	$0.20^{a} \pm 0.07$	$66.0^{a} \pm 10.9$	$18.4^{a} \pm 4.69$
	RN	$0.09^{b} \pm 0.00$	$0.42^{b} \pm 0.02$	$1.59^{b} \pm 0.13$
	A	$0.33^{c} \pm 0.15$	$56.7^{c} \pm 5.29$	$19.6^{a} \pm 3.10$
	AN	$0.10^{b} \pm 0.00$	$0.52^{b} \pm 0.06$	$2.21^{b} \pm 0.36$
LS mixing 1 h at 23°C	R	$76.6^{a} \pm 4.67$	$156^{a} \pm 6.46$	$83.7^{a} \pm 4.01$
	RN	$0.16^{b} \pm 0.05$	$51.4^{b} \pm 11.3$	$13.6^{b} \pm 3.97$
	A	$55.2^{c} \pm 7.67$	$129^{c} \pm 17.3$	$66.3^{c} \pm 8.44$
	AN	$0.19^{b} \pm 0.04$	$51.7^{b} \pm 5.78$	$14.4^{b} \pm 2.45$

[a-d] Values within columns not sharing common superscripts differ significantly ($P < 0.05$)

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Table 2. Powder solubility (%) of regular (R), regular with N_2 injection (RN), agglomerated (A), and agglomerated with N_2 injection (AN) milk protein concentrate (MPC) samples taken after mixing at 23°C and 50°C for 30 s using a solubility index meter and centrifuged at 3000*g* for 10 min.

MPC	Solubility (23°C)	Solubility (50°C)
R	$83.6^{a} \pm 1.9$	$96.7^{a} \pm 0.9$
RN	$96.2^{b} \pm 1.3$	$98.2^{a} \pm 1.1$
A	$62.6^{c} \pm 6.9$	$96.6^{a} \pm 0.7$
AN	$92.1^{a} \pm 3.1$	$98.1^{a} \pm 0.3$

[a-c] Values within columns not sharing common superscripts differ significantly ($P < 0.05$)

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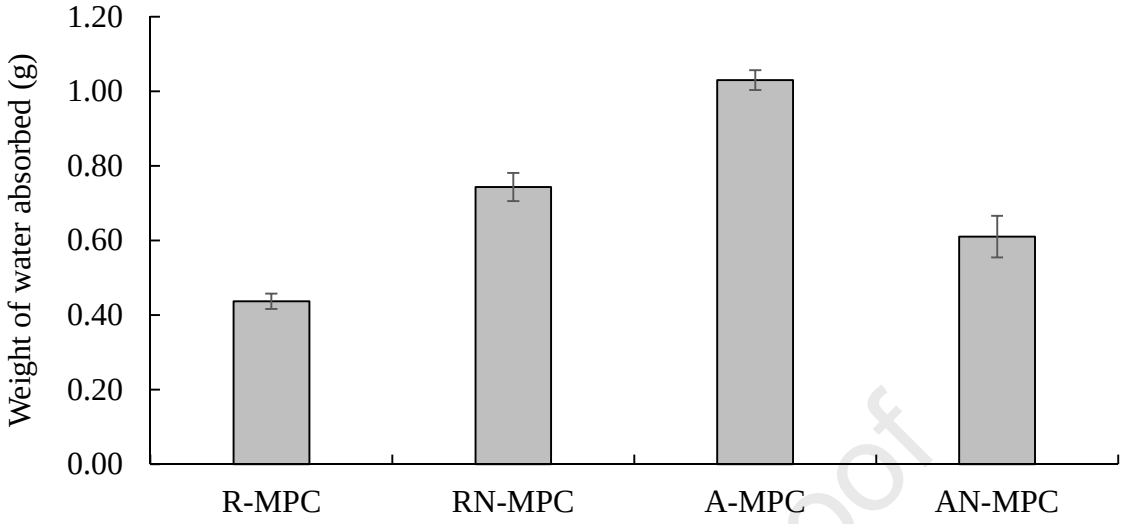

Fig. 1.

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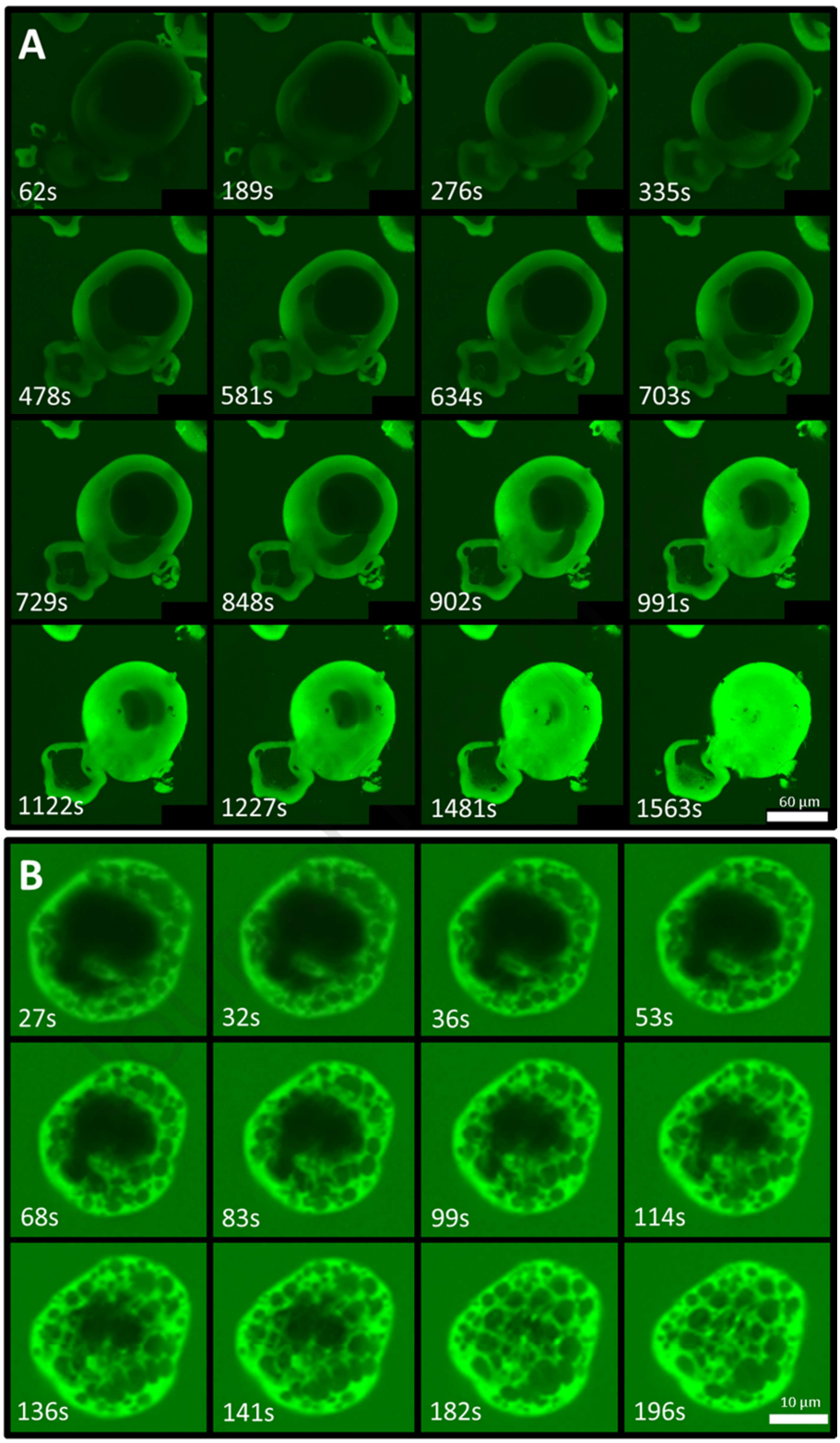
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276s
335s
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634s
703s
729s
848s
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991s
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1227s
1481s
1563s
60 µm
B
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Fig. 2.

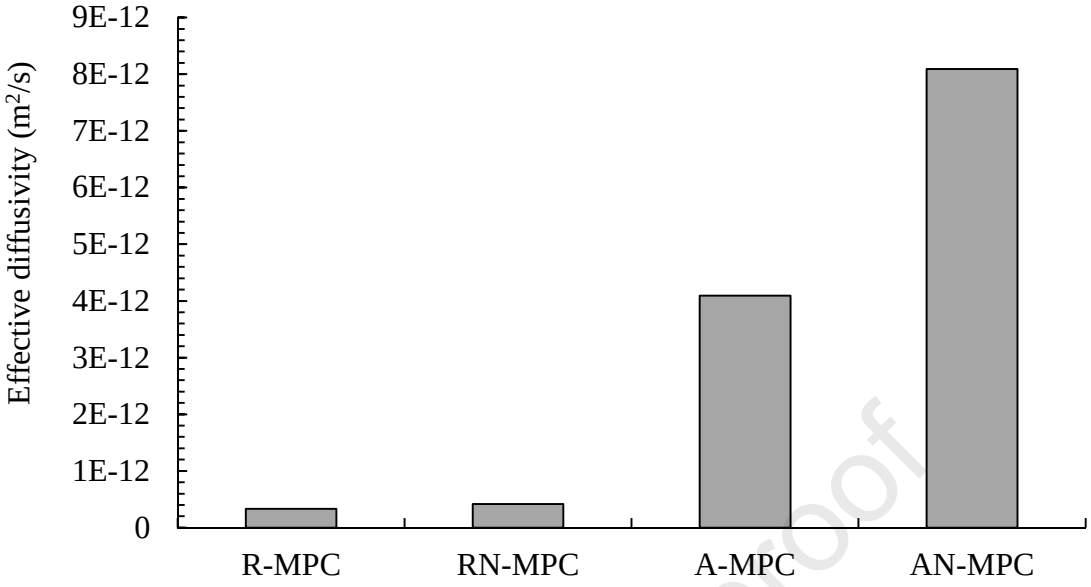

Fig. 3.

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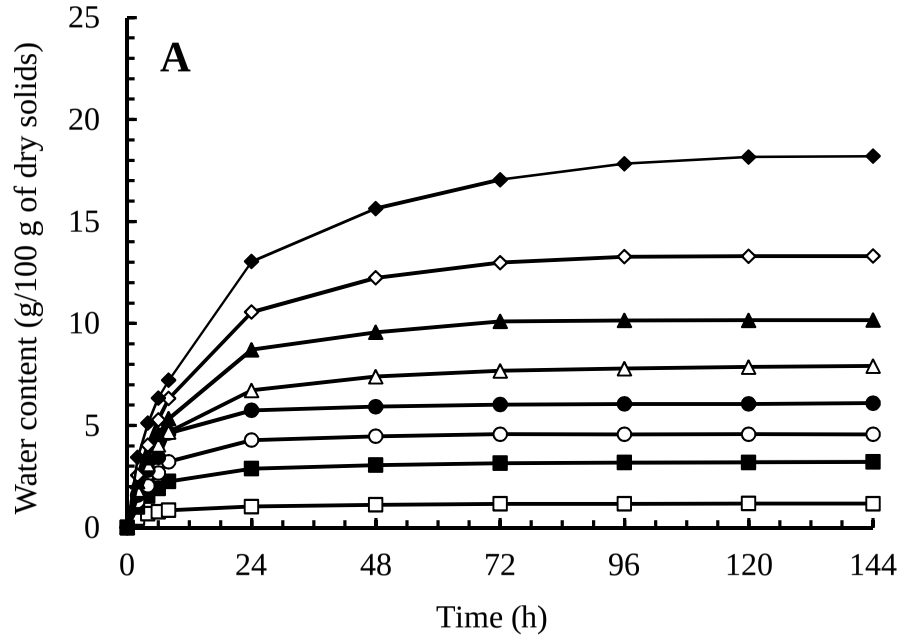

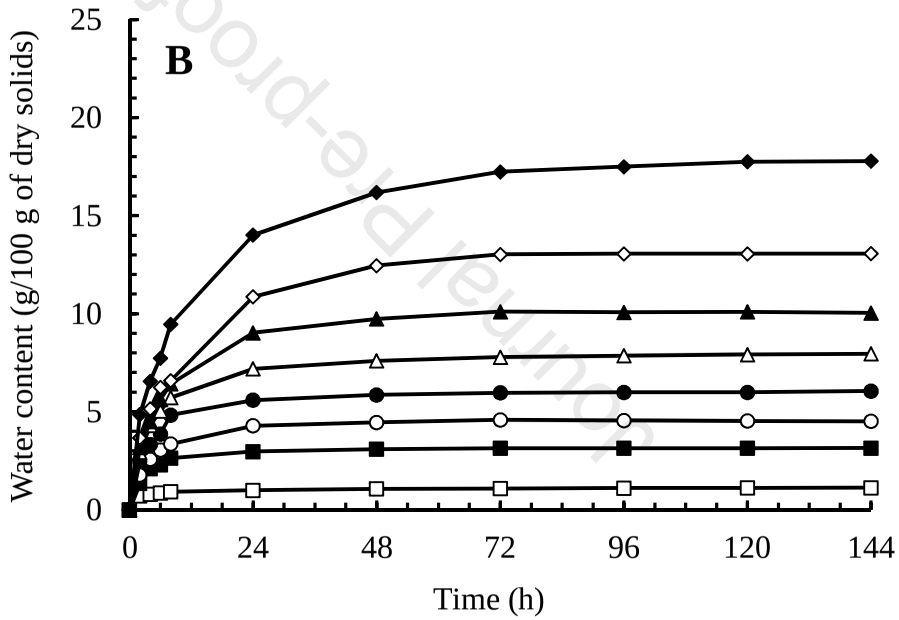

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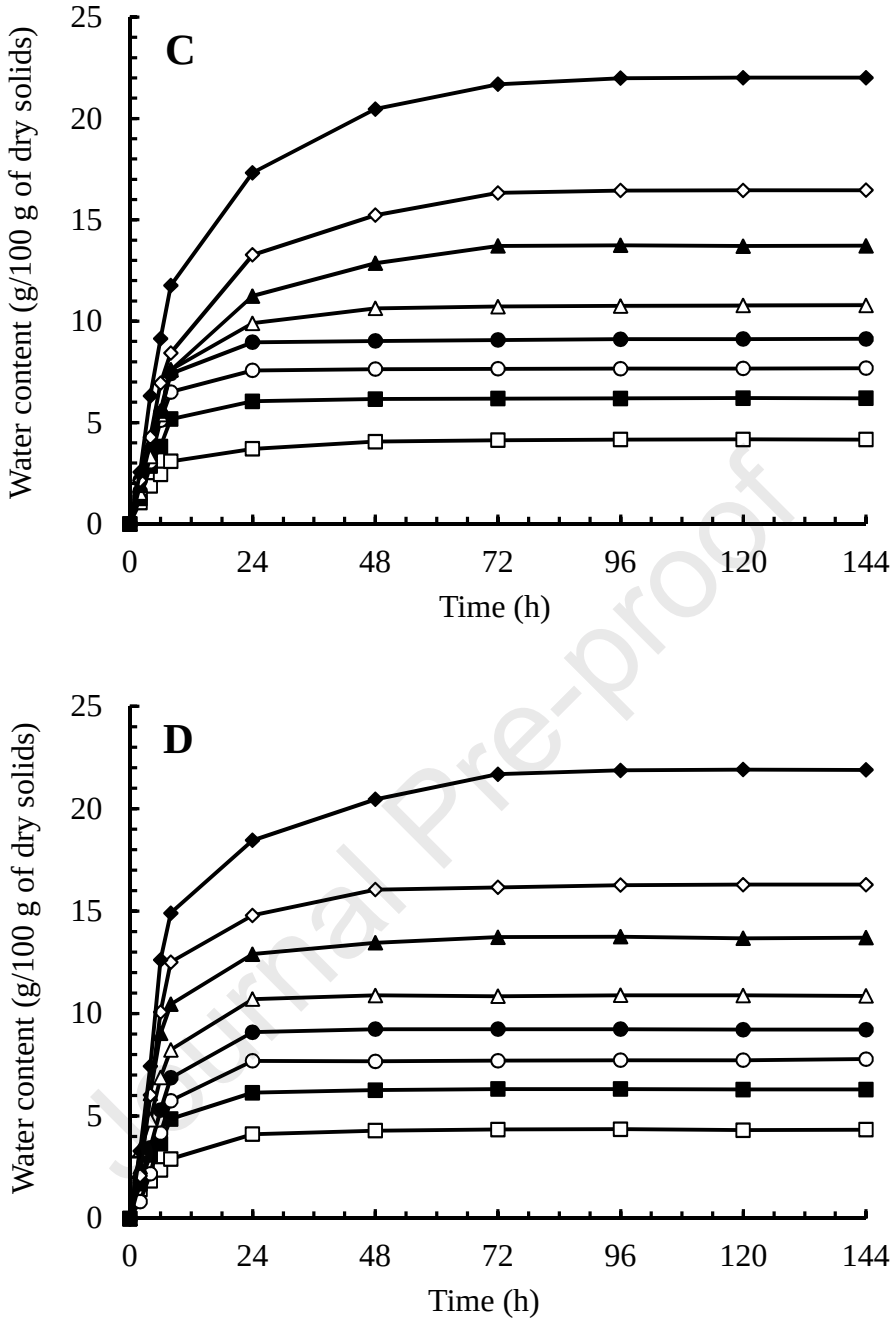

Fig. 4.

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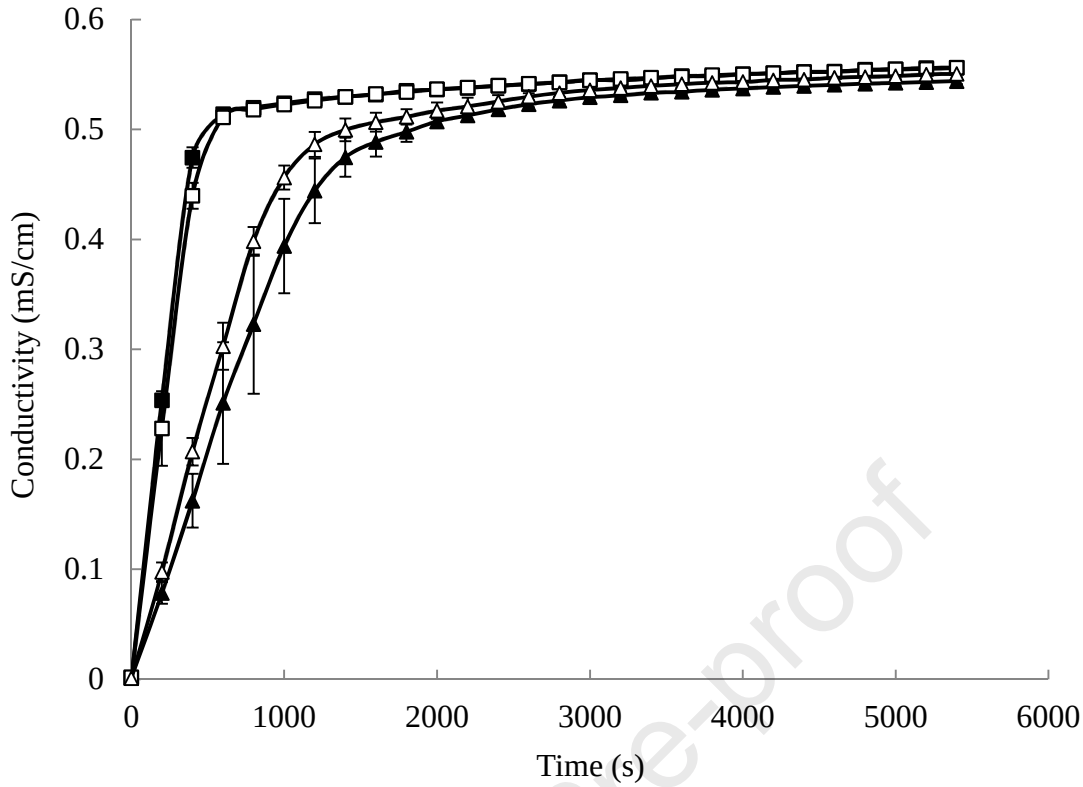

Fig. 5.

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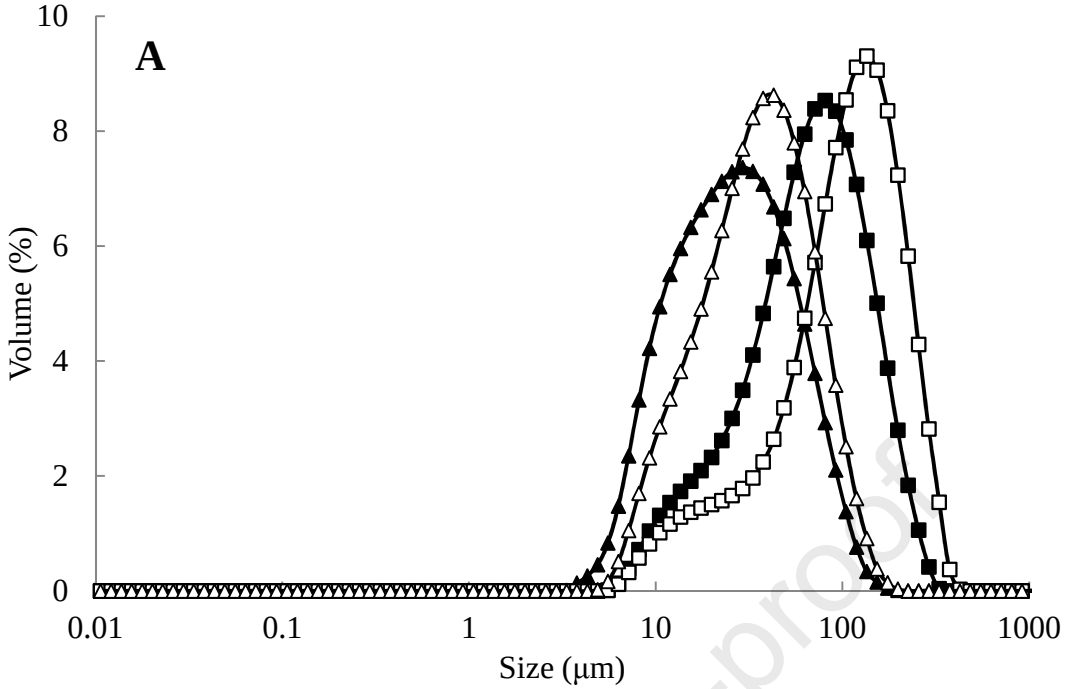

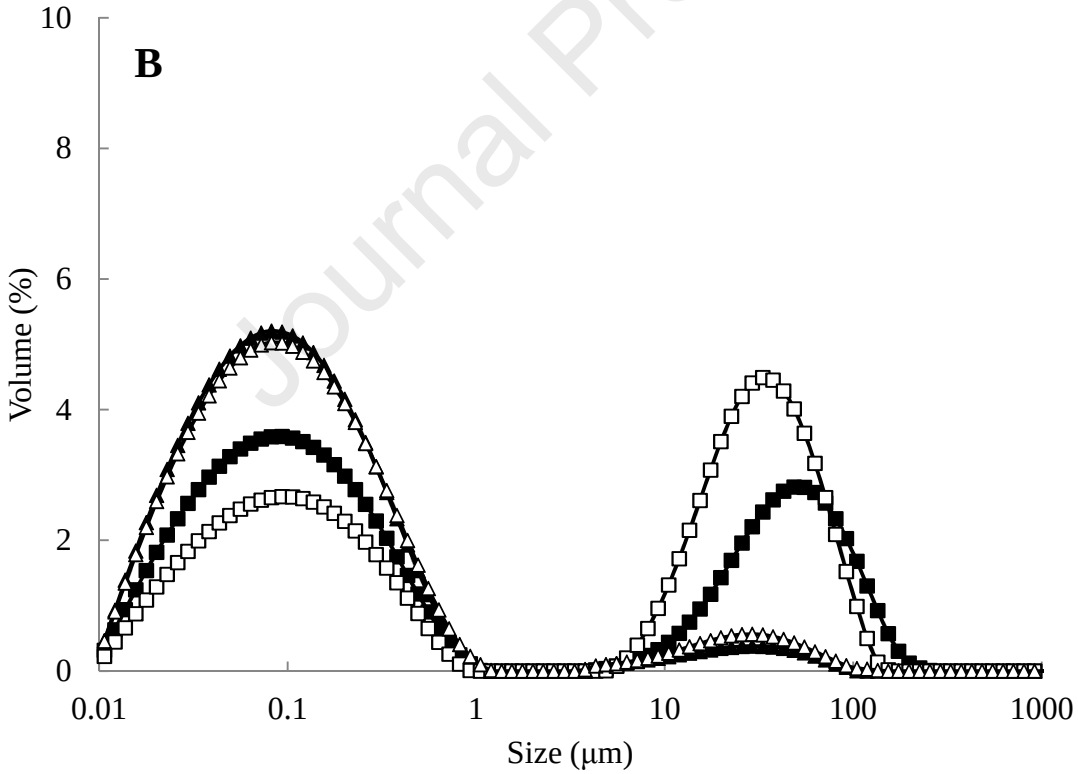

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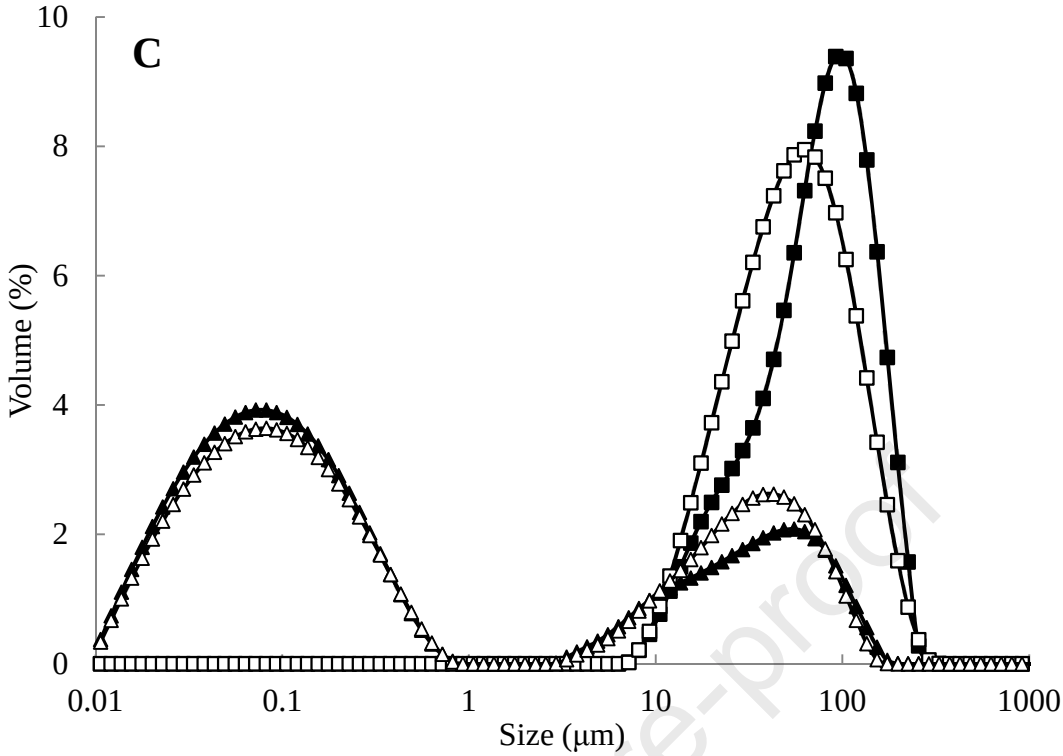

Fig. 6.

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- Diffusion was higher for powders produced using nitrogen (N_2) gas injection.
- Agglomerated powders absorbed more water than non-agglomerated samples.
- N_2 injection enhanced the dissolution of regular and agglomerated powders.
- Agglomeration alone reduced powder solubility.

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Declaration of Interests

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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