



Kinetics of lactose crystallization and the effect on physical changes and reconstitution of milk powders

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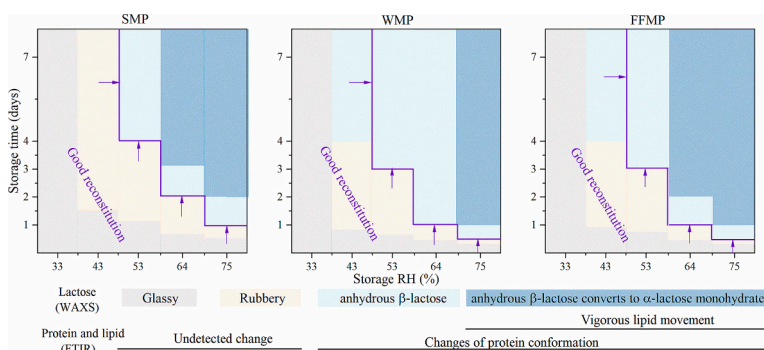
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HIGHLIGHTS

- Lactose crystallizes always as anhydrous β -lactose first.
- Anhydrous β -lactose converts to α -lactose monohydrate at RH above 64 %.
- The rate turns faster with increasing RH, and changes across powder types.
- Formation of crystalline phases affects protein conformation and lipid movement.
- Formation of a crystalline phase does not always affect reconstitution.

GRAPHICAL ABSTRACT

Impact of lactose crystallization on protein and lipid changes, and on the reconstitution behavior of milk powders during storage. SMP: skim milk powder; WMP: whole milk powder; FFMP: fat-filled milk powder.



ARTICLE INFO

Keywords:

Lactose crystallization
Kinetics
Reconstitution
Milk powder
Wide-angle X-ray scattering
Infrared spectroscopy

ABSTRACT

Lactose crystallization in dairy powders during storage is associated with impaired reconstitution performance. While this transformation from amorphous to crystalline lactose is known to accelerate above the glass transition temperature, its detailed kinetics and impact on reconstitution, particularly in complex milk powders, remain unclear. This study explores the crystallization behavior of lactose and its effect on reconstitution in three commercial milk powders (skim, whole, and fat-filled) stored at 33–75 % relative humidity (RH) and 25 °C for 50 days. Results show that anhydrous β -lactose forms between 43 % and 53 % RH, while at 64 % and 75 % RH, it further converts to α -lactose monohydrate. Crystallization rate increased with RH. Protein changes correlated to both the formation and phase conversion of crystalline lactose, whereas lipid movement became more pronounced when α -lactose monohydrate formed. Notably, and contrary to common assumptions, lactose crystallization at conditions near the glass transition (43 % RH at 25 °C) did not impair reconstitution, as determined by particle size distribution. This finding reveals a critical window for optimizing milk powder storage without

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<https://doi.org/10.1016/j.powtec.2025.121195>

Received 29 March 2025; Received in revised form 27 May 2025; Accepted 30 May 2025

Available online 4 June 2025

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compromising functionality, offering new insights for quality control and shelf-life management in the dairy industry.

1. Introduction

Milk powders, a good alternative for fresh milk, can encounter quality deterioration due to the relative humidity (RH) and temperature during transportation and storage. Quality deterioration of milk powder during storage manifests across multiple dimensions, including changes in physical properties (e.g., particle size and flowability), nutritional losses (e.g., lysine bioavailability), organoleptic defects (e.g., browning and off-flavors), and reduced functional performance (e.g., impaired reconstitution) [1,2]. Regardless of such changes, milk powder is ultimately expected to reconstitute into a colloidal system that closely resembles fresh milk. During the reconstitution of degraded milk powders, a series of physicochemical changes occur across scales - from bulk properties to molecular interactions [3–8]. For instance, lactosylation of casein can lead to protein cross-linking on the surface of milk protein concentrate (MPC), contributing to reduced solubility [3]. Protein conformational changes have been linked to surface wrinkling, which hinders wetting in infant formula powders. In the same study, powders that underwent lactose crystallization exhibited longer wetting times compared to those containing amorphous lactose [5]. Moreover, fat migration was found to increase significantly with the degree of lactose crystallization in infant formula [9]. Previous studies also indicate that the progress of many of these physicochemical changes is interconnected via the molecular constituents of the product. For example, lactose crystallization can contribute to denaturation and/or aggregation of proteins, causing in turn fat coalescence, where fat coalescence results in fat migration [7,9,10]. In milk powders containing considerable amounts of amorphous lactose, lactose crystallization is broadly perceived as the primary molecular change inducing many secondary phenomena related to loss of quality and performance [11–15]. Understanding lactose crystallization and investigating the relationship between lactose crystallization and other macromolecular changes has thus been of interest, with the aim of producing improved formulations with increased control of quality [2].

Amorphous lactose is glassy in fresh spray-dried milk powder [14]. It turns rubbery when the storage temperature is higher than the glass transition temperature (T_g). The higher molecular mobility of rubbery lactose increases nucleation and can thereafter lead to the formation of crystalline lactose [14,16]. In the case of moisture-induced lactose crystallization, a plasticization effect of the absorbed water depresses the T_g, where lactose crystallization is initiated and water is released [17], resulting in decreased water content depending on the storage conditions [16–20]. Different crystalline phases have been reported in stored milk powder. Anhydrous α -lactose, anhydrous β -lactose, and anhydrous α/β -lactose at molar ratios 5:3, 4:1 or 1:4 were mostly observed at low RH (< 57 %), while α -lactose monohydrate mostly was observed at high RH (e.g. 64 % and 75 %) [4,11,15,17–19]. Lactose crystallizes at different rates depending on RH, temperature and formulation [20–22], and lactose thus evolves into different polymorphs during storage. Until now, only Jouppila et al. have studied the kinetics of lactose crystallization in SMP using X-ray diffraction (XRD). A detailed and systematic study of the appearance and disappearance of the various polymorphs of lactose is missing in the literature.

Since the different states of lactose possess quite various physicochemical properties [23,24], the change of lactose state might affect other molecules in the vicinity. Several hypotheses have been proposed on the link between lactose crystallization and changes in the state of proteins and fat in dairy powders. Lactose was hypothesized to partly replace water during spray drying to maintain the solubility of protein [25]. Some researchers support the idea of a sugar-protein interaction via hydrogen bonds [15,26]. Therefore, the segregation of lactose

during crystallization may consequently change protein stability and induce a protein conformational change. Even though both lactose crystallization and conformational changes of protein were observed in the same conditioned powder [15], a direct link between lactose crystallization and conformational changes of protein has not been reported.

For a variety of fat-containing powders, a widely shared view on the effect of lactose crystallization on fat is that crystalline lactose creates capillary interstices and ruptures particle structure, allowing lipid to be expelled to the surface [4,12,14,27], i.e., lipid migration. Qi et al. recently verified this hypothesis and further showed that the amount of expelled fat depends on the size of the lactose crystals [28]. Most studies investigating the component distribution of lipid, protein, and lactose used X-ray photoelectron spectroscopy (XPS), which probes only elements at the outer surfaces, i.e., at nanometer depth. Under these conditions, measuring on fresh powder, lipids are over-represented [25,29,30], and the detection of changes in other components is thus depressed [4]. Fourier-transformed infrared spectroscopy (FTIR) can probe all components at larger depths of micrometer scales, and has shown its ability in depicting the changes of major components in milk powders [5,28,31]. The link between lactose crystallization and lipid migration at a greater depth in milk powders can be explored by FTIR.

Overall, there is still a gap in the characterization of the kinetics of lactose crystallization in complex milk powders, and consequently, how that affects other milk powder components critical for reconstitution. This paper aims to enhance the understanding of the kinetics of lactose crystallization in commercial milk powders, specifically in terms of rates and the development of polymorphs during lactose crystallization. We investigate the impact of lactose crystallization on the physical properties of milk proteins and lipids, as well as the reconstitution performance of milk powders. Water sorption, glass transition, and formation of crystalline lactose in three commercial dairy powders stored at different RHs were followed. The changes of protein and lipids in milk powder were monitored using FTIR and accompanied by characterization of the reconstituted milk using laser diffraction particle size analyses.

2. Materials and methods

2.1. Materials

To incorporate compositional differences in our investigation, three spray-dried milk powders containing a considerable amount of lactose were obtained from Arla Foods (Viby, Denmark): Skim Milk Powder (SMP), Whole Milk Powder (WMP), and Fat Filled Milk Powder (FFMP). The composition of the powders is summarized in Table 1. All samples were packed under low-oxygen conditions and stored at 20 °C until use. FFMP is a relatively recent product category compared to SMP and WMP. It is produced by supplementing skim milk with vegetable fat prior to homogenization and spray drying [32]. In industry, FFMP is recognized to exhibit different storage stability than WMP. However, limited scientific literature is available on FFMP in comparison to SMP and WMP.

Table 1
Composition of SMP, WMP and FFMP (g/100 g).

	SMP	WMP	FFMP
Protein	32	24	22
Lactose	54	39	36
Sucrose	n.a.	n.a.	5
Milk fat	0.5–0.9	28	n.a.
Vegetable fat (palm oil)	n.a.	n.a.	28

2.2. Storage of milk powder and determination of water sorption

Powders were weighed into small vials with a bed height of ~ 3 mm. The vials were placed in vacuum desiccators over P_2O_5 for 3 weeks to dry at room temperature, i.e., approximately 23°C . The dehydrated powders were transferred to desiccators, where the RHs were controlled by saturated salt slurries. Magnesium chloride (VWR Chemicals, Belgium), potassium carbonate (VWR Chemicals, Belgium), magnesium nitrate (Thermo Scientific, USA), sodium nitrite (Thermo Scientific, Belgium) and sodium chloride (Sigma-Aldrich, Germany) were used to create RHs of 33 %, 43 %, 53 %, 64 % and 75 % respectively, at 25°C . Each type of milk powder had 6 replicate vials for each RH level, 3 vials to monitor water sorption and 3 vials to do analysis during storage. The sampling points are 4 and 10 h, plus 1, 2, 3, 4, 7, 9, 11, 15, 22, 29, 36, and 50 days (all at 25°C). Observations are coded in the format of powder type/storage RH/storage time; for example, SMP/33/1 represents SMP stored at 33 % RH for 1 day.

2.3. Differential scanning calorimetry (DSC)

Approximately 10 mg of powder were weighed into a DSC crucible (Material No: 00027331, Mettler Toledo, USA) and hermetically sealed. The crucible was positioned in the DSC (Thermal Analysis System DSC1, Mettler Toledo, USA) with an empty crucible as reference. Samples were scanned from -50°C to 100°C at $5^\circ\text{C}/\text{min}$ after an equilibration at -50°C for 1 min.

2.4. Wide-angle X-ray scattering

Wide-Angle X-ray Scattering (WAXS) measurements were performed using a Nano-inXider (Xenocs, France). Powder was packed tightly in a capsule (1 mm thick) and sealed with Kapton tape. Samples were placed on a 16-hole Peltier stage and exposed to a Cu-anode beam (50 kV and 0.6 mA) of $800\ \mu\text{m}$ width for 900 s at room temperature ($\sim 20^\circ\text{C}$).

2.5. Fourier-transform infrared spectroscopy

The infrared spectra of powders were recorded by Attenuated Total Reflection-Fourier Transform-Infrared spectroscopy (ATR-FTIR) using the wavelength region $4000\text{--}600\ \text{cm}^{-1}$ (ABB Bomen MB100, Canada; ATR unit, DuraScopeTM, SensIR Technology). The spectra were collected with a resolution of $4\ \text{cm}^{-1}$.

2.6. Laser diffraction particle size analysis

Powders were reconstituted in deionized water at 25°C for 1 h using a magnetic stirrer at 500 rpm. The particle size distribution (PSD) of the reconstituted milk was then measured using Mastersizer3000 with a Hydro MV module (Malvern Instruments Ltd., Worcestershire, UK). Refractive indices of 1.45 and 1.33 were selected for particles and dispersant, respectively. Each measurement was taken in triplicate at a dilution corresponding to a laser obscuration between 10 and 12 % at a stirrer speed of 2500 rpm.

3. Results and discussion

3.1. Water sorption

Change of mass based on non-fat solid versus storage time is shown in Fig. 1. The moisture is expressed as a function of non-fat solid to account for the fact that fat should not interact with water. Milk powders (dehydrated by controlled preconditioning) rapidly absorb a considerable amount of water in the first 24 h. One can observe that the maximum moisture content is reached within a few days, followed by a decrease in moisture content, i.e., a release of water and subsequent equilibration with the environment. The kinetics of water absorption are

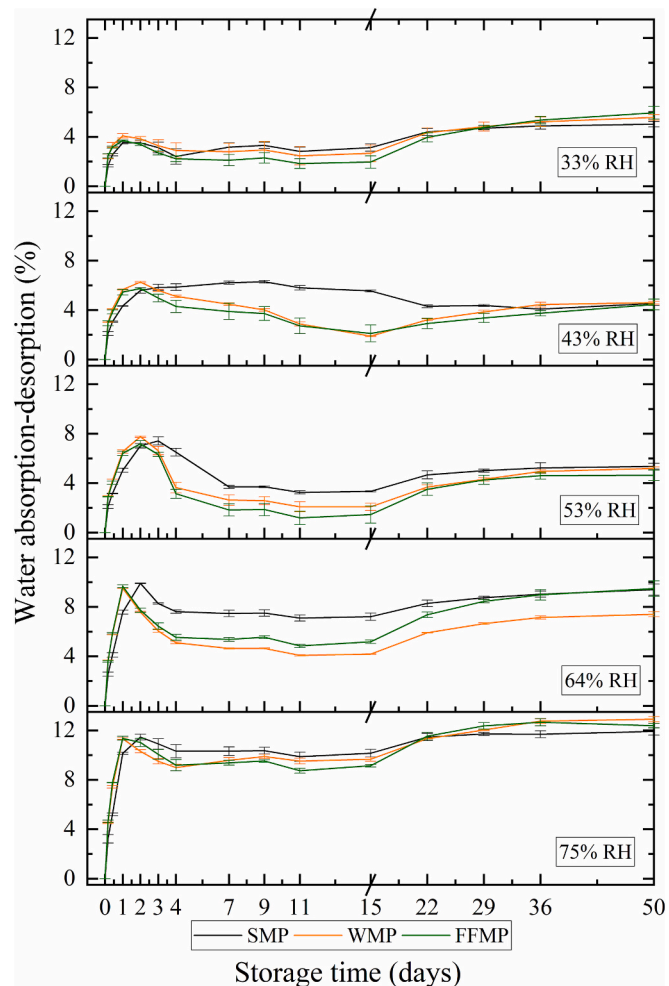


Fig. 1. Water sorption and desorption behavior of skim milk powder (SMP), whole milk powder (WMP), and fat-filled milk powder (FFMP) ($n = 3$) during storage at 25°C under controlled relative humidity (RH) conditions ranging from 33 % to 75 % RH for 50 days. Powders were stored in RH-controlled desiccators, and relative weight changes were monitored to assess moisture uptake and release. Moisture content (y-axis) is expressed based on non-fat solids. The x-axis shows storage time, with a break at day 15 and different time scales before and after the break.

similar in shape for all powders. There is a slight delay in the time at which the peak occurs for SMP powders. It is rather surprising that the amount of water released is smaller for 75 % RH when compared to 53 % and 64 % RH. A small peak was observed for 33 % RH, although this release might be well within experimental error. Similar kinetics for water release have been reported for SMP in the literature, e.g., release within 1–2 days in the RH range of 40 %–54 % and within 24 h at RH greater than 57 % at 25°C [17,19,33–35]. The equilibrium moisture measured is also in broad agreement with the literature, e.g., for SMP stored at 66 % RH, 17 % of moisture content was reported and 7 % at 43 % RH [33]. Despite the simplicity of these experiments, the interaction of water with powders can be complex and can be affected by many factors beyond lactose crystallization, including volume and area-to-volume of the sample, particle size, bed packing [19,33,34,36,37]. A small water release peak at 33 % RH has been reported in a study comparing water release with different bed heights [34]. The specifics of the experimental protocol might also affect results, for example, how many times the desiccators have been opened, and how long the samples are exposed. Release of water is associated with lactose crystallizing, and the rate of water release is used to infer the kinetics of lactose crystallization, e.g., fast water release was attributed to fast

crystallization [38,39]. However, as water can interact in many ways with the powders, it could be an oversimplification to estimate the kinetics of lactose crystallization from water release data only.

3.2. Glass transition

Lactose crystallizes when the system has high enough mobility, i.e., goes into a rubbery state. It is of interest to identify the time and moisture content at which this transition occurs. Fig. 2 shows Tg “onset” for SMP as a function of storage time. As time increases, powders absorb water, resulting in a decrease in Tg, because of the plasticization effect of water. At higher RH, the rate of depression of Tg is larger, as a result of a higher water content reached over storage time. At and above 43 % RH, Tg decreases to below the storage temperature (25 °C), indicating that these samples have gone into a rubbery state. The time it takes to be in the rubbery state depends on the storage RH, e.g., for 43 % RH it is about 1.5 days. To get the time of transition to a rubbery state in SMP, exponential curves (Fig. S1) were fitted to the data presented in Fig. 2. DSC measurement for WMP and FFMP could not be interpreted, as the thermal signal from lipid melting overlaps with the glass transition region. Instead, fitted the functions in Fig. S3 and S4 are used to deduce the time of glass transition in WMP and FFMP by matching the corresponding moisture content where SMP shows its glass transition (Fig. S1 and S2). The range of Tg values obtained via direct observations for SMP are comparable to those observed in the literature, e.g., at 33 % reported glass transition ranged from 22 to 33 °C [40,41]. Once the lactose in powders is in the rubbery state, crystallization could occur at a different rate depending on storage conditions. The kinetics of this lactose crystallization will be investigated in the next section.

3.3. Formation of crystalline lactose observed by WAXS

The kinetics of lactose crystallization were obtained from scattering experiments. An example of WAXS profiles, WMP stored at 75 % RH, as a function of storage time, is shown in Fig. 3a. WAXS profiles of all stored powders, together with fresh powders, are shown in Fig. S5 and S6. At 33 % RH, the profiles are similar to those of the fresh powder, indicating that lactose remained amorphous during 50 days of storage. For RH 43 % to 75 %, peaks evolved during storage, indicating the formation of

crystalline lactose. The polymorphic forms of crystalline lactose were identified by comparing characteristic peaks in the 2 θ range 9.5–18.5° and profile patterns in the range 18.5–22.5° with those of lactose standards analyzed by the same setup of WAXS (profiles of standards not shown). Characteristic peaks, such as anhydrous β -lactose at ~10.5° and α -lactose monohydrate at ~12.5° and ~16.4° [18,28,42] designated in literature, were also considered when identifying polymorphic forms. Quantification of the distinct crystalline phases was done by integrating the area of characteristic peaks at 10.5° and 12.5° for anhydrous β -lactose and α -lactose monohydrate, respectively [28]. The area-counts are normalized between 0 and 100 % for each measurement series, i.e. each identified phase, to compensate for differences in amount of sample during measurements. In Fig. 3b–e the kinetics of the two identified phases are shown for our milk powders.

Lactose crystallizes mainly as anhydrous β -lactose at 43 % and 53 % RH, irrespective of powder type. At 43 % RH (Fig. 3b), the (delayed) growth of anhydrous β -lactose in SMP is clearly faster than that in WMP, followed by FFMP. At 53 % RH (Fig. 3c), the same trend is observed but with much smaller differences in the growth rate. At 64 % RH, it is still anhydrous β -lactose that has formed in WMP, whereas in SMP and FFMP, lactose crystallizes first as anhydrous β -lactose, followed by a decrease of this form and a gradual increase of α -lactose monohydrate. This occurs after 4 days and 3 days for SMP and FFMP, respectively. The growth rate (Fig. 3d) of anhydrous β -lactose seems nearly similar, whereas the rate of α -lactose monohydrate in SMP is faster (albeit delayed) than that of FFMP when comparing the degree of phase conversion within the time of appearance of α -lactose monohydrate. At 75 % RH, lactose in all three powders crystallizes as anhydrous β -lactose first, then α -lactose monohydrate appears after 3, 2, and 2 days for SMP, WMP, and FFMP, respectively. Subsequently, anhydrous β -lactose disappears after 7, 29, and 7 days in SMP, WMP, and FFMP, respectively. The growth rates of both phases in SMP seem similar to that of FFMP, but considerably faster than that of WMP (Fig. 3e).

Our identification of the polymorphs corroborates that lactose first crystallizes above 33 % RH at approximately 25 °C [18,43,44]. The identified phases for SMP and WMP align with the general observations from literature that lactose tends to crystallize into anhydrous phases at low RH and α -lactose monohydrate at high RH (where low and high RH are usually split at approximately RH 57 % across different studies) [4,11,15,17–19]. Our study further reveals that lactose crystallizes as an anhydrous polymorph and then converts to a hydrate phase when sufficient water is present, representing a sequential time dependency in lactose crystallization. An X-ray-based study of the kinetics of lactose crystallization in milk powder with a composition similar to that of a commercial product was reported only once. In that study freeze-dried SMP was stored at 53.8 % to 85.8 % RH at 24 °C for 24 days [18]. They reported the faster development of crystalline peaks at higher RHs, in agreement with our results.

It was speculated in literature that the fat in milk powder delays lactose crystallization because fat-containing powder is hydrophobic, which limits the diffusion of water [40,45]. However, our results reveal that fat-containing powders (WMP and FFMP) start having crystalline lactose earlier than SMP, albeit at a slower growth rate compared to SMP. The delayed moisture diffusion (Fig. 1) and higher protein content in SMP (Table 1) may inhibit the plasticization effect of water on the glass transition of lactose in SMP, and thus the delayed appearance of crystalline lactose, compared to WMP and FFMP. Even though fat overrepresentation on the surface [29] could theoretically delay absorption of water due to its hydrophobic nature, in our two fat-containing powders, agglomeration and lecithination are expected to mitigate the hydrophobic influence of fat. The general slower growth of crystalline lactose in fat-containing powders across different RHs could be attributed to the impingement and constriction of fat on lactose [12].

When comparing the two types of fat-containing powder, the faster growth of α -lactose monohydrate in FFMP compared to WMP might result from the presence of sucrose in the formulation of FFMP. Edrisi

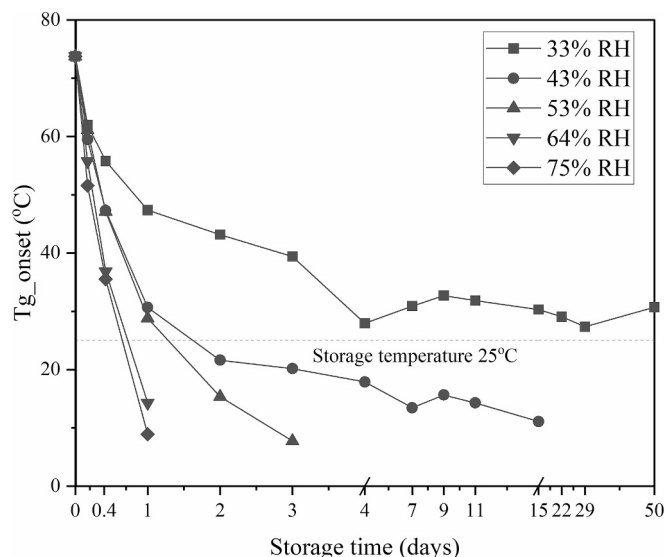


Fig. 2. Onset glass transition temperature (Tg) of skim milk powder (SMP) over time during storage at 25 °C under various relative humidities (33 % to 75 % RH). Measurements were performed using DSC ($n = 1$). Tg onset is plotted against storage duration to indicate the evolution of powder state stability under different RH conditions.

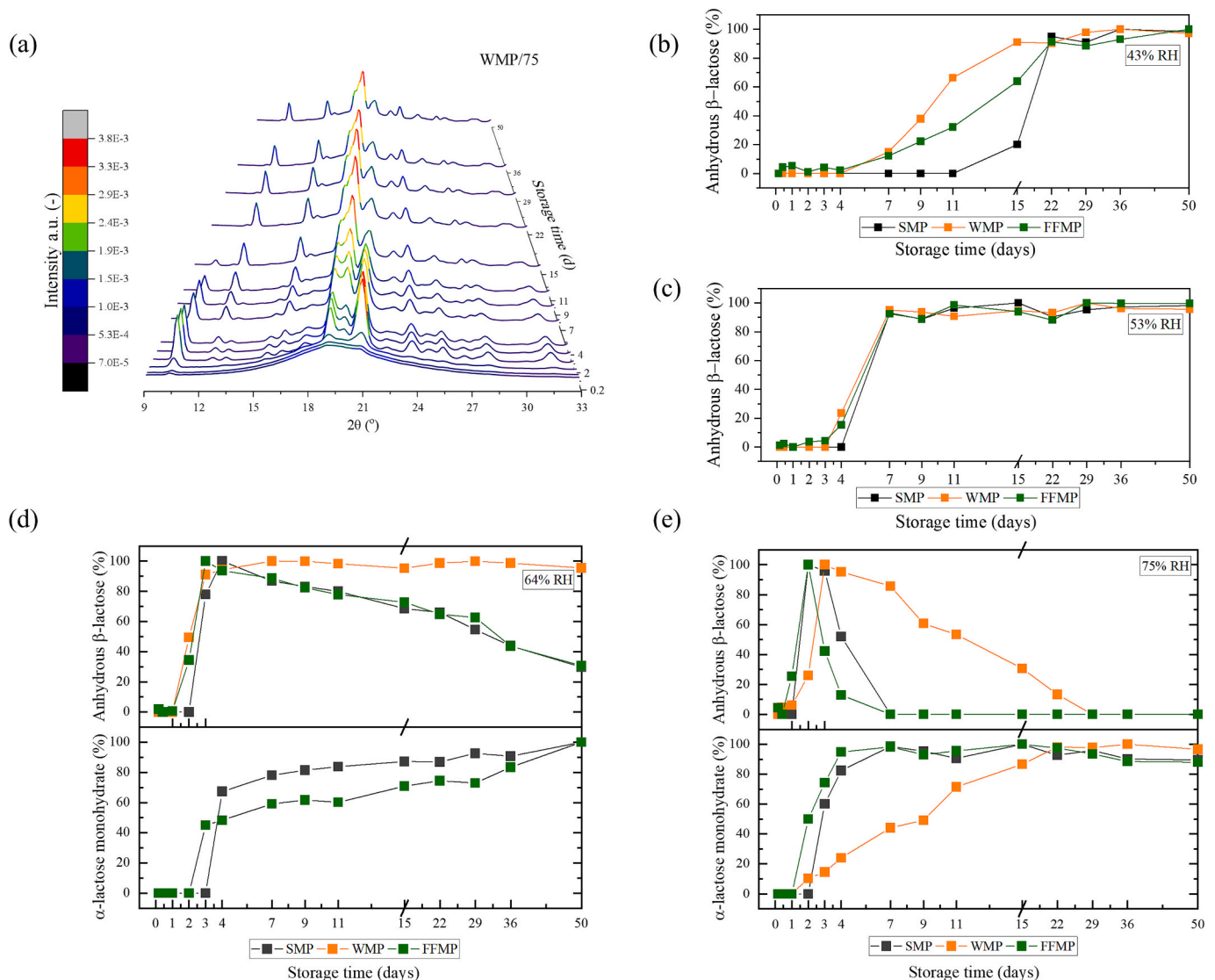


Fig. 3. Kinetics of lactose crystallization in skim milk powder (SMP), whole milk powder (WMP), and fat-filled milk powder (FFMP) during storage at 25 °C under RH conditions where crystalline lactose was found, characterized by wide-angle X-ray scattering (WAXS):

(a) WAXS intensity profiles of WMP over time at 75 % RH;

(b-c) Quantification of anhydrous β-lactose from peak intensities at 43 % and 53 % RH;

(d-e) Quantification of both anhydrous β-lactose and α-lactose monohydrate at 64 % and 75 % RH.

Quantification results are expressed by normalizing the integrated peak areas from 0 to 100 %. The x-axis shows storage time, with a break at day 15 and different time scales before and after the break.

et al. found that at 75 % RH, increasing the sucrose content in lactose/sucrose powders induced faster crystallization of lactose [37]. Adding sucrose might create a faster saturated environment for lactose [46] in FFMP, thus accelerating lactose recrystallization from anhydrous β-lactose to α-lactose monohydrate.

3.4. Lactose crystallization and release of water in SMP, WMP and FFMP

The time required for the powders to move from a glassy to a rubbery and eventually a crystalline stage at different RH is indicated in Fig. 4. The time of glass transition from 43 % to 75 % was estimated based on fitted profiles (section 3.2). The formation of crystalline lactose was estimated as the time right before crystalline lactose was detected in Fig. 3b-e. A similar method was used for identifying the time for water release using data from Fig. 1. The tendencies in Fig. 4 can be separated into three zones. In Zone I, samples are glassy; in Zone II, the material is rubbery; and in Zone III, there is crystallization and polymorphic

conversion of crystalline lactose. Water release seems to happen around the crystallization points, albeit it would be difficult to identify the order of events at low RH values. The water measurements, though simple in nature, are very sensitive to perturbations, and as such, one should be careful with the interpretations of the results. Jouppila et al. found that the release of water occurred after the formation of crystalline lactose (specifically α-lactose monohydrate) at and above 66 % RH, possibly since proteins could also absorb water. Our results demonstrate that there is a delay between powders turning glassy and crystallization, where the delay time decreases with increasing RH.

3.5. Effect of lactose crystallization on macromolecules measured by FTIR

The effect of lactose crystallization on milk proteins and lipids was examined in FTIR spectra. Fig. 5a,b show the evolution of spectra in the regions assigned to protein (1710–1488 cm^{-1}) and lactose (930–854 cm^{-1}) for two sample series. Fig. 5c,d show the evolution of spectra

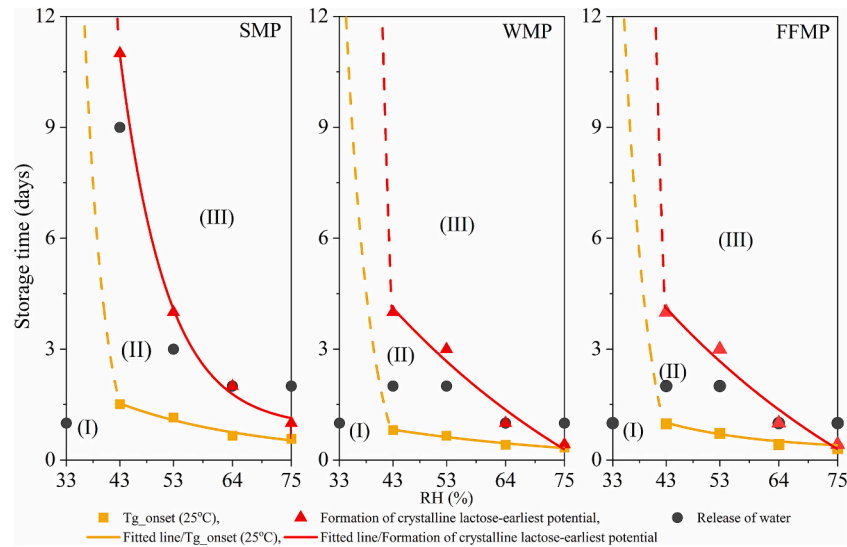


Fig. 4. Isothermal (25 °C) state diagram of lactose in milk powders (SMP, WMP, FFMP) with corresponding water release profiles during storage at 33 %–75 % RH. State zones (I, II, and III marked in figures) were delineated based on the onset and progression of lactose crystallization, indicating transitions between glassy (Zone I), rubbery (Zone II), and crystalline (Zone III) states.

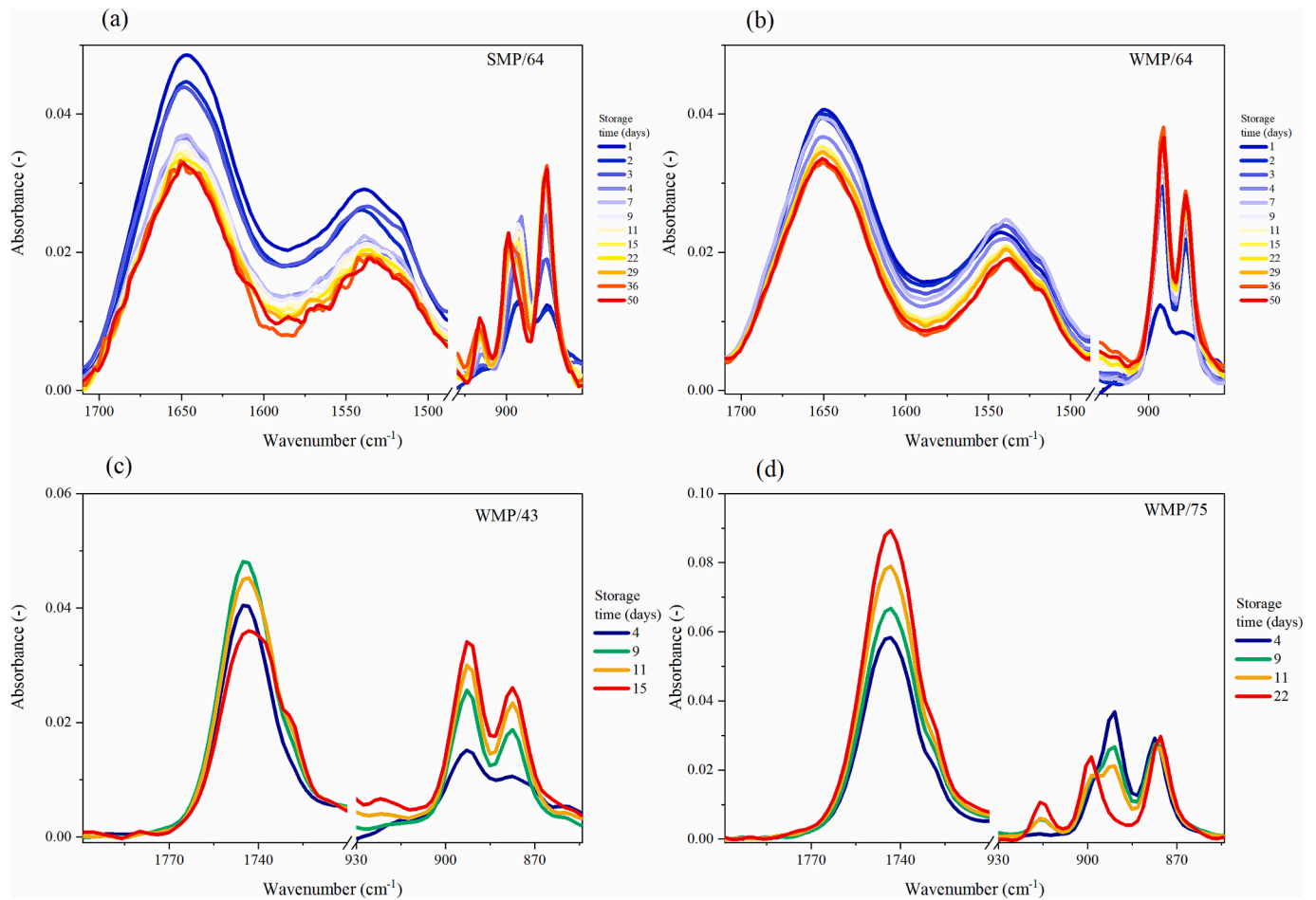


Fig. 5. Selected FTIR spectra showing molecular changes in milk powders during storage at 25 °C: (a) Lactose (930–854 cm^{-1}) and protein (1710–1488 cm^{-1}) regions for SMP at 64 % RH; (b) Same regions for WMP at 64 % RH; (c) Lactose and lipid (1799–1710 cm^{-1}) regions for WMP at 43 % RH; (d) Same regions for WMP at 75 % RH.

assigned to lipids ($1799\text{--}1710\text{ cm}^{-1}$) and lactose ($930\text{--}854\text{ cm}^{-1}$) from two samples. For quantitative comparison, the spectral baseline was removed and intensity was normalized. With the increase of the storage time, peaks assigned to lactose [42] turn sharper and become more defined ($930\text{--}854\text{ cm}^{-1}$), and a new one appears (e.g., 916 cm^{-1}), indicating the formation of crystalline lactose. This agrees with our lactose crystallization kinetics determined by WAXS. The smooth, broad band appearance of Amide I ($1710\text{--}1590\text{ cm}^{-1}$) and Amide II ($1590\text{--}1488\text{ cm}^{-1}$) [47,48] also changes over storage time (Fig. 5a,b). These changes of initial smoothness for the Amide regions indicate a change in the secondary conformation. Shoulder peaks appear at around 1737 and 1727 cm^{-1} (Fig. 5c,d) in the region assigned to lipid [49]. These FTIR spectral observations collectively suggest that changes in the secondary structure of the proteins and the conformational changes of lipids coincide with the evolution of crystalline lactose. More spectral change over storage time and the state of lactose can also be seen in supplementary Figs. S7, S8, and S9.

Based on Beer-Lambert's law, the intensities in FTIR correspond to the concentration or abundance of species. For solid samples like our milk powders, there are known challenges in direct quantification, specifically sample presentation on the ATR interface and effective light penetration, which is both wavelength dependent and a function of the

refractive indices of different matrices [50]. Working with relative ratios can partly alleviate these artifacts. We therefore limit our interpretation to relative ratios of integrated intensities (i.e. areas) of selected spectral bands for three components, namely lactose ($930\text{--}854\text{ cm}^{-1}$), protein Amide I ($1710\text{--}1590\text{ cm}^{-1}$), and lipid ($1799\text{--}1710\text{ cm}^{-1}$).

Fig. 6 shows relative ratios between integrated lactose and protein bands for each type of powder. The ratio before the glass transition (33% RH) is relatively constant. For SMP the ratio increases over time for RH at 43% and above. The increase in ratio starts at the time point where anhydrous β -lactose was detected and increases with the progress of crystallization (compare Fig. 6 with Fig. 3b-e). A similar increase is seen for WMP and FFMP at 43% RH and above. Exceptions are WMP at 75% RH and FFMP at 64% and 75% RH, where a decrease in the ratio can be seen. The increase coincides with the formation of anhydrous β -lactose, while the decrease matches with the polymorphic conversion from anhydrous β -lactose to α -lactose monohydrate (compare Fig. 6 with Fig. 3b-e).

Different lactose species might change the intensities of our selected FTIR spectral region. One thus needs to recognize that the observed ratio change is not only driven by lactose crystallization. Integration of the bands of the individual components shows that (relative) intensities of protein decrease while those of lactose increase over storage time

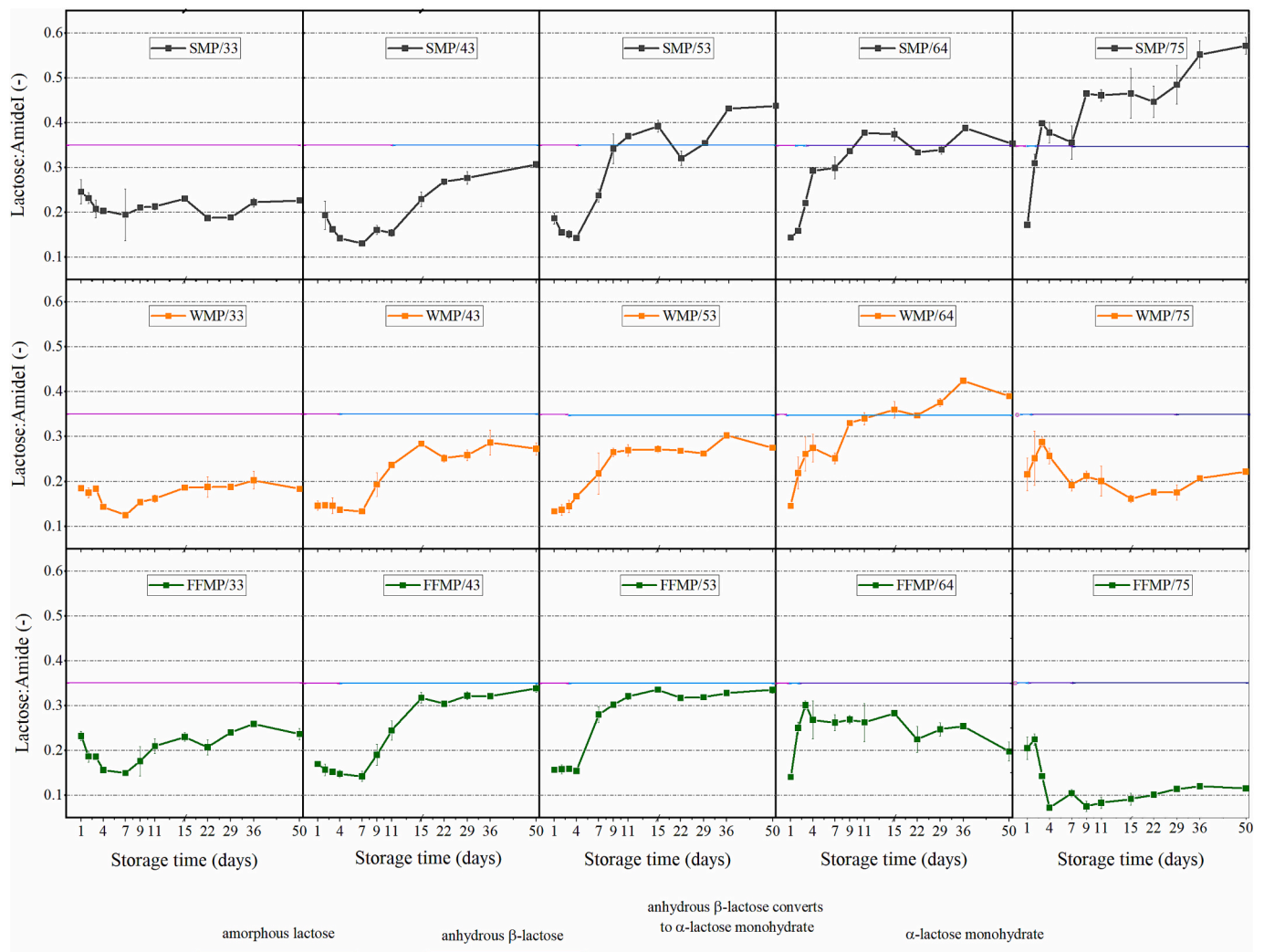


Fig. 6. Relative spectral intensity ratios of lactose to protein in skim milk powder (SMP), whole milk powder (WMP), and fat-filled milk powder (FFMP) during storage at $25\text{ }^{\circ}\text{C}$ under 33% to 75% relative humidity (RH). Ratios were calculated by dividing the integrated peak areas of the lactose band ($930\text{--}854\text{ cm}^{-1}$) by those of the protein Amide I band ($1710\text{--}1590\text{ cm}^{-1}$) in FTIR spectra. The x-axis represents storage time, with a break at day 15 and different time scales before and after the break. Information on the state of lactose, obtained from WAXS anal, is superimposed using lines.

(Fig. S10). This does suggest that the relative ratio change is not merely driven by the change in chemical species of lactose. It suggests a change in the physical state of lactose and protein.

The same approach of evaluating relative FTIR band ratios was also carried out for lipid over lactose (Fig. S11). Two notable patterns are an increasing trend for 75 % RH, when anhydrous β -lactose converts to α -lactose monohydrate, and a gradual reduction when crystallization reaches a plateau, indicating that polymorphic conversion affects the abundance of lipid that FTIR can detect. Using FTIR allows this study probing concurrent changes of proteins and fat in relation to lactose crystallization because of the greater penetration depth (micrometers) of FTIR compared to that of XPS (nanometers) in previous studies [29,30]. Fat movement was not observed when lactose was in the amorphous state, which is different from the previous finding in a study that employed XPS [51].

3.6. Particle size distribution in reconstituted milk

Fig. 7a shows the particle size distribution (PSD) in reconstituted milks for FFMP stored for 7 days at the five investigated RHs, which is an example of showing how storage RH affects PSD. The reconstituted milks showed multiple peaks, one around 0.5 μm , one around 10 μm , and one around 100 μm . The particle sizes of powders stored at 33 % and 43 % RH are nearly identical, being dominated by relatively small particle sizes, while the particle size increased with increasing RH from 53 % to 75 % RH. The PSD of powders stored at 53 % RH showed deviations from that of powders stored at 33 % RH, indicating deterioration of quality. For the powders stored at 64 and 75 % RH, one can see very large particle sizes, e.g., in the order of 100 μm , indicating that the powders would not be of acceptable reconstitution quality. The PSDs of powders stored at 33 % RH are comparable with those of fresh SMP, WMP, and FFMP from literature [7,10,30,52]. This is expected since those powders were stored below the T_g and little change is anticipated. Interestingly, our results show that the powders stored at 43 %, even though stored above T_g , appear to remain unchanged.

In Fig. 7b, PSDs of FFMP stored at 64 % RH for different durations are shown, which is a representative example showing the effect of storage time at the same storage RH. After day 1, the PSD changes with particles having a size of more than 10 μm appearing, indicating a decrease in reconstitution quality. These changes become dramatic after 3 days of storage. Unfortunately, due to challenges with the measurement

equipment, there are no reliable measurements available for storage times longer than 7 days for this investigation. However, the progress of PSDs going from good to bad powders had developed within these first 7 days.

3.7. Impact of lactose crystallization on the reconstitution quality

To elucidate the impact of lactose crystallization on the reconstitution quality of milk powders, we integrated data on glass transition, lactose crystallization behavior, and particle size distribution of the reconstituted samples. By superimposing reconstitution performance onto the progression of lactose phase transitions, we constructed Fig. 8, which visually illustrates the relationship between lactose state and reconstitution outcome. This figure is a companion to Fig. 4. The x-axis is the storage RH and the y-axis is the time (in days) required for the system to reach a certain state. A schematic line (purple) is added, indicating the powder turning to have bad reconstitution quality after this line. Reconstitution quality was assessed by comparing the particle size distribution (PSD) profiles with those of powders stored at 33 % RH, which served as the reference for good reconstitution powder (Section 3.6).

Fig. 8 shows that powders stored at higher RH have little difference in the time required to transition to a rubbery state, to transit further to anhydrous β -lactose and for reconstitution quality to flip from good (left of the line) to bad (right of the line). With a reduction of storage RH, we notice a significant deviation in the rate of lactose crystallization and reduction of reconstitution quality. For example, for powders stored at 53 %, it takes approximately 1 day for the powders to reach a rubbery state, and 3 days for lactose to crystallize. A reduction in reconstitution quality was observed as early as day 3 at 53 % RH, coinciding with the formation of anhydrous β -lactose. Similar correlations between lactose crystallization and reduced reconstitution were found at 64 % and 75 % RH. Interestingly, this pattern did not hold at 43 % RH. Despite the formation of anhydrous β -lactose, the reconstitution quality of fat-containing powders at this condition remained comparable to that of powders containing amorphous lactose within the time frame of our study.

Our group previously reported that milk powder which had undergone lactose crystallization during storage was still able to reconstitute into a homogeneous milk system. In that study, FFMP was stored for four months, during which lactose crystallization was completed [28].

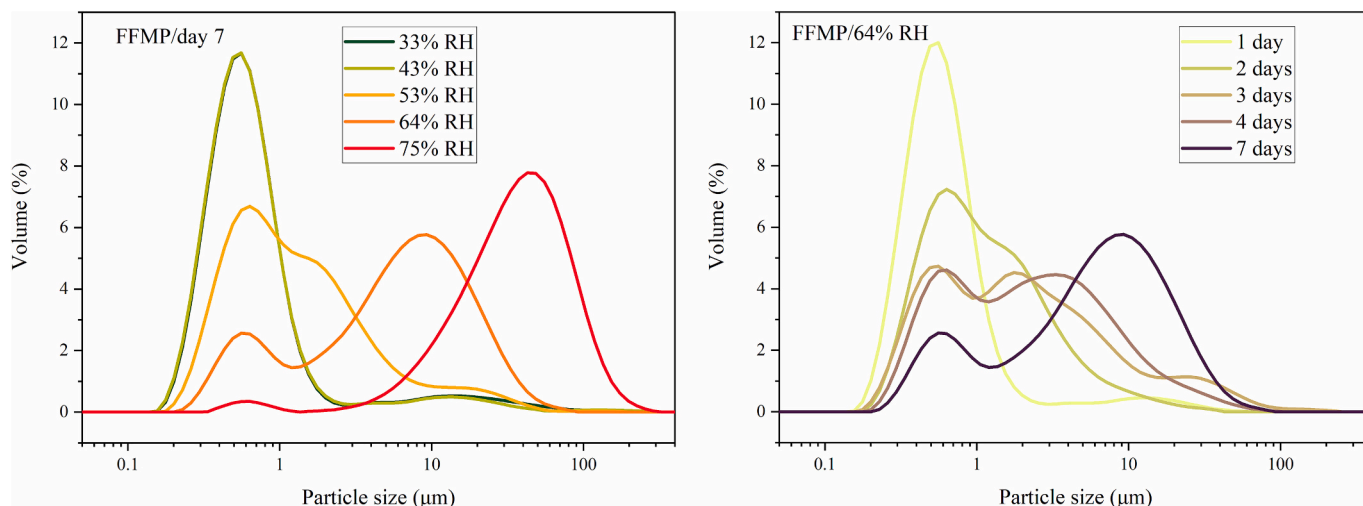


Fig. 7. Particle size distributions (PSD) of reconstituted milk from stored FFMP powders: (a) PSD profiles of samples stored for 7 days under 33 % to 75 % RH at 25 °C; powder under 33 % RH is an example of powder with good reconstitution quality; (b) PSD evolution from day 1 to day 7 at 64 % RH and 25 °C. These two plots are examples showing how storage RH and time affect reconstitution quality. The changes in PSD were used to evaluate the impact of storage conditions on reconstitution quality.

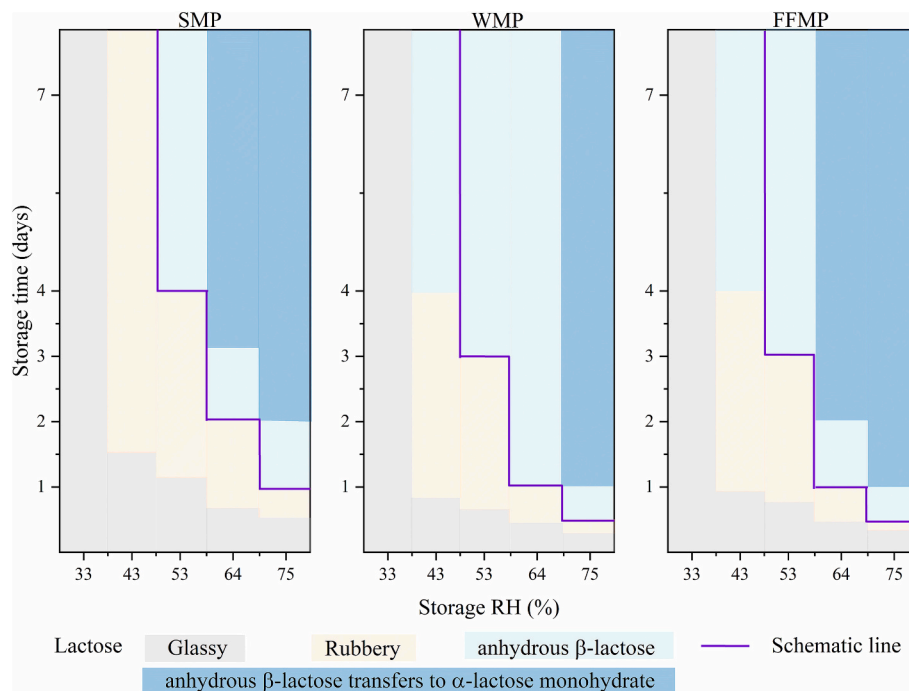


Fig. 8. Schematic representation of the impact of lactose crystallization on reconstitution quality. The diagram integrates storage RH conditions, storage time, and crystalline states of lactose in milk powders with particle size distribution (PSD) data of reconstituted milks. A conceptual boundary line (purple) distinguishes powders with good (left side of the line) versus impaired (right side of the line) reconstitution performance. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Lactose crystallization is typically associated with a general decline in milk powder quality. However, both the present and our previous study [28] demonstrate that its formation does not necessarily impair reconstitution quality. In powders stored at relatively low humidity (e.g., 43 % RH), lactose crystallization and reconstitution deterioration do not coincide. Regardless of its physical state, lactose will eventually dissolve during reconstitution. Therefore, the observed reduction in reconstitution quality, reflected by an increase in PSD, is more likely attributed to protein aggregation, lipid coalescence, and/or the formation of protein-lipid complexes [7]. FTIR analysis at 43 % RH did indicate changes in protein conformation and lipid mobility associated with lactose crystallization. However, these structural modifications appear to be minor and insufficient to cause visible protein aggregation or fat globule coalescence in the reconstituted milk.

4. Conclusion

The current work reports, for the first time, isothermal kinetics of lactose crystallization for complex milk powders within a broad storage RH range from 33 % to 75 % RH at 25 °C. Crystallization rates increased with an increase in RH. Lactose crystallized as an anhydrous phase, mainly anhydrous β -lactose between 43 % RH and 53 % RH, and crystallized mainly as anhydrous β -lactose first and converted to α -lactose monohydrate at 64 % and 75 % RH. Lactose crystallization varies among milk powders with different fats and sugars, indicating formulation-specific event. The crystalline lactose appeared earlier but grew more slowly in WMP and FFMP compared to SMP. α -lactose monohydrate crystallized faster in FFMP than WMP. Concurrent monitoring of macromolecular changes by FTIR showed that protein changes and lipid movement correlated with the formation of crystalline lactose and the phase conversion. However, these changes (formation of crystalline lactose, protein conformation, and lipid movement) did not impact the reconstitution quality of powders stored at 43 % RH, as evaluated by the particle size distribution of reconstituted milks. These findings indicate that lactose crystallization does not universally impair the reconstitution

quality of milk powders during storage. Overall, this study provides enhanced knowledge in the kinetics of lactose crystallization in complex milk powders as well as insights on the effect of formulation. The finding that milk powders stored above their glass transition (25 °C, 43 % RH) can still reconstitute into good-quality milk provides valuable insights for optimizing quality control practices in the dairy industry.

CRediT authorship contribution statement

Xiàowēi Qí: Writing – review & editing, Writing – original draft, Visualization, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Frans W.J. van den Berg:** Writing – review & editing, Visualization, Validation, Supervision, Methodology, Investigation, Data curation, Conceptualization. **Jacob Judas Kain Kirkensgaard:** Writing – review & editing, Validation, Methodology. **Kirsten Gade Malmos:** Writing – review & editing, Supervision, Resources, Conceptualization. **Jens Risbo:** Writing – review & editing, Methodology. **Serafim Bakalis:** Writing – review & editing, Visualization, Supervision, Resources, Project administration, Funding acquisition, Data curation, Conceptualization.

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.

Acknowledgements

The authors thank Innovation Fund Denmark and Arla Amba for funding this project (1063-00031B), and Mariam Mahmoud from the Department of Food Science at the University of Copenhagen for measuring particle size distribution. Data were generated by accessing research infrastructure of the University of Copenhagen, including FOODHAY (Food and Health Open Innovation Laboratory, Danish

Roadmap for Research Infrastructure).

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.powtec.2025.121195>.

Data availability

Data will be made available on request.

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