Phase Separation of Wheat Flour Dough Studied by Ultracentrifugation and Stress Relaxation. I. Influence of Water Content

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ABSTRACT

Separation of starch and gluten into two aqueous phases is an important aspect of wheat flour dough development. The fact that starch can easily be washed from dough, leaving behind a gluten gel, illustrates the phase separation of dough very well. To study this phase separation, dough was subjected to high centrifugal forces of 100,000 × *g*. The tendency of starch and gluten to separate into two phases is reported for six wheat cultivars at water contents ranging from 38 to 50%. At high water contents, a separation of the dough into liquid, gel, gluten, and starch phases was observed. When the water content was reduced, unseparated dough was found at the bottom of the test–tube. At low water contents no separation was observed. The influence of dough water content on the stress-relaxation modulus was considerable at water contents where no separation took place. For doughs prepared with increasing water contents, where separation occurred, the modulus was less affected. This was interpreted as being the region where gluten development was complete. Separation properties varied for cultivars of wheat. Three cultivars with the same relative amounts of separated phases produced roughly the same stress-relaxation modulus.

Wheat flour dough has been described as being bicontinuous, i.e., consisting of two continuous aqueous phases (Eliasson and Larsson 1993). The two aqueous phases in developed dough, described as phase-separated, are the water-swelled protein phase (gluten) and the liquid phase (containing dispersed starch granules and solubles) (Eliasson and Larsson 1993). The fact that starch can easily be removed from a wheat flour dough by washing leaving behind a gluten gel, illustrates the phase separation of dough very well.

The importance of the liquid phase in wheat flour dough was pointed out by MacRitchie (1976). Submitting the dough to ultracentrifugation resulted in separation into a liquid phase and a solid phase. Conductivity measurements showed that the liquid phase in dough was continuous and not dispersed. The water contents of the separated phases were 86.0% (liquid phase) and 34.5% (solid phase). A summary of the composition of dough liquors, including other studies, is given in MacRitchie (1986).

Attempts have been made through rheological investigations to relate the dynamic modulus to the gluten and starch components of wheat flour dough (Hibberd 1970b, Smith et al 1970, Navickis et al 1982). In other studies, the modulus was related to the loaf volume of baked bread (He and Hoseney 1991, Mani et al 1992). The liquid phase of bicontinuous dough is interesting from a rheological point of view, as the rheological behavior at small deformations is determined by the properties of the two aqueous phases. In this study, the phase separation of wheat flour dough is illustrated by the phases obtained after separation by ultracentrifugation. Our objective was to relate this separation behavior to rheological measurements at small deformations, i.e., stress relaxation. The influence of water content was studied for six wheat flours.

MATERIAL AND METHODS

Material

Six wheat flours of different origin and with different baking performance were selected for this study. The protein (Kjeldahl, N × 5.7), starch (Åman et al 1994), ash (650°C, 16 hr), and damaged starch (method 76–30A, AACC 1983) contents are given in Table I. The baking performance of the flours was obtained according to Olered (1979) and is given in Table II. Wheat flours included in the study were: Kosack and Sport (Swedish winter and spring wheat, respectively); Rouquin (a spelt [dinkel] wheat); wheat cv. Prairie; a feed wheat; and Prego (a triticale). The flours were all milled in a Quadrumat Senior Mill to extraction rates of 59–69% (Svalöf Weibull AB, Svalöv, Sweden).

Dough Mixing

Wheat flour doughs were mixed in a 10-g farinograph bowl. Flour (10 g) was mixed with distilled water to give doughs with water contents of 38–50% by dough weight. The investigated water contents differed by cultivar, depending on where in the water interval the doughs were convenient to handle. At least six doughs with different water contents were prepared from each flour to study the influence of water content on the rheological behavior and separation properties. Mixing was done at 30°C, 60 rpm. Mixing time was fixed to 5 min because an optimal mixing time was hard to determine with the 10 g mixing bowl, particularly at low water contents. To estimate the farinograph water absorption using the 10-g mixing bowl, the mixing resistance was calibrated against the mixing resistance resulting from mixing in the 300-g mixing bowl. Method 54–21 (AACC 1983) was used. Estimated farinograph water absorptions are given in Table II.

Dough Rheology

The rheological behavior of the doughs was studied using a Bohlin VOR rheometer (Metric Analys, Stockholm, Sweden) in the stress-relaxation mode. Tests were performed with cone and plate geometry (diameter 30 mm, cone angle 5.4°). Measurements were made at 25°C, with an initial equilibrium time of 600 sec between the application of the sample and the imposition of the strain (0.006). The strain rise time was 0.1 sec. To prevent water loss, dough surfaces exposed to air were covered with silicone oil. The initial value of the stress-relaxation modulus (G_0) and two relaxation times ($t_{0.5}$ and $t_{0.1}$) were chosen to characterize the rheological behavior. Relaxation times were taken as the times where G was reduced to 50 and 10% of G_0, respectively. The stress was allowed to relax during at least 1,000 sec. The water dependence of the modulus was fitted to power-law relationships ($R > 0.99$). The relaxation times were fitted either to second order ($R > 0.98$) or linear relationships ($R > 0.68$). As the measurements were performed in a range of water contents, and only a few

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measurements were performed at the same water content, a deviation given by replicates can not be calculated. The reproducibility of the stress-relaxation parameters studied is in general within 10% for flour-water doughs (Larsson and Eliasson 1996).

Ultracentrifugation of Dough

Doughs with the same water contents as those used in the stress-relaxation measurements were centrifuged for 1 hr at 100,000 × *g* in an MSE superspeed 65 centrifuge (MSE Ltd., Manor Royal, Crawley, Sussex, England). The dough sample (≈5 g) was placed in a test tube with a diameter of 8.5 mm and a height of 71 mm. The volume fraction of the separated phases was estimated by measuring the height in the test tube with a slide caliper. Larsson and Eliasson (1996) determined the volume fraction of the separated phases with an accuracy of ±6%, except for some of the unseparated fractions and the smallest liquid phases, which deviated up to 15%. To determine the water content, the separated phases were dried for 1 hr at 130°C. Drying the samples for an extra 30 min did not reduce the water content further. The water content of the separated phases was obtained with an accuracy of ±1.5 % for triplicates (Larsson and Eliasson 1996). In the present study, the same reproducibility was obtained for Sport and Prego, run in duplicate. For the other cultivars, four single samples in the range of water contents were studied.

RESULTS AND DISCUSSION

Chemical Composition and Baking Performance

The chemical compositions of the flours are listed in Table I. The flours cover a wide range of protein contents from 8.8 to 16.7%. Baking performance (Table II) was related to the protein content of the flour. A high-damaged starch content was obtained for Prairie wheat, indicating a hard endosperm. In contrast, the spelt wheat, Rouquin had a low-damaged starch content.

Farinograph Measurements

Farinograph water absorptions are listed in Table II. The lowest water addition was required for Rouquin and Prego. Prairie and Sport required the highest amounts of water. According to Tipples et al (1978), the water absorption of a wheat flour can be attributed to the amount of damaged starch, as well as the protein content. Consequently, the high-damaged starch content in Prairie would explain its high water absorption. For Sport, the high water absorption is probably the result of high protein content.

TABLE I
Chemical Composition of Flours[a]

	Protein	Starch	Ash	Damaged Starch
Kosack	11.1	81.8	0.39	6.1
Sport	16.7	76.6	0.42	4.5
Prairie	12.7	81.0	0.48	8.6
Rouquin	12.8	81.2	0.46	2.3
Feed	8.8	84.7	0.44	6.2
Prego	9.0	83.3	0.42	3.6

[a] % On dry, total basis.

TABLE II
Baking Performance and Water Absorption of Flours

	Baking Performance	Farinograph Water Absorption[a]	Estimated Water Content[a,b]
Kosack	Medium	45.3	43.5
Sport	Good	47.1	45.3
Prairie	Medium	47.5	46.5
Feed	Poor	44.4	43.7
Rouquin	Medium	43.3	43.2
Prego	Poor	43.6	42.7

[a] % By dough weight.
[b] At the intersection of the two linear parts in Figure 5a.

Ultracentrifugation

The phase separation of gluten and starch was studied by centrifugation of dough at high speeds. The separation of Kosack doughs at 39.8–48.6% water content is illustrated in Figure 1. At the highest water content (48.6%), the dough separated into a liquid phase, a gel layer, a gluten phase, and a starch phase. When the water content was reduced (47.1% water for Kosack), the degree of the separation was lower, and unseparated dough appeared below the starch phase in the test tube. The liquid phase probably contained soluble sugars and proteins, according to a summary of analytical data pertaining to the liquid phase (MacRitchie 1986). The gel layer was distinguished from the liquid phase by the fact that it had a more solid appearance and could not be poured. A sharp boundary between the two phases was also observed after separation. The water content of the liquid phase (88%) was higher than that of the gel phase (81%). The gel layer was studied with a microscope under polarized light. Small starch granules were observed in higher concentrations towards the bottom of the gel layer. On top of the gluten phase a thin white layer of small starch granules was observed. To determine the location of damaged starch, the separated fractions were stained with Congo red (0.1% solution). The damaged starch was found in the thin white layer on top of the gluten phase and in the unseparated fraction, close to gluten pieces. Some starch granules, both small and large ones, were also observed in the gluten phase.

The degree of the separation was dependent on the water content of the dough (Fig. 1). The amount of unseparated dough increased at lower water contents. Below a certain water content (43.7% for Kosack) the separation was strongly hindered and no liquid phase separated. At the lowest water content, no separation was obtained at all. This trend was observed for all the flours studied. Prego, the triticale, was only studied at three water contents: low (38.3%), farinograph water absorption (43.6%) and high (46.9%). Although the relative amounts of the separated phases varied, as well as the water content at which the separation was observed, the water content occurred somewhere just before the farinograph water absorption (Table II).

In Figure 2, the relative amounts of separated dough phases at the farinograph water absorption are shown for Kosack, Sport, Prairie, feed, Rouquin wheat and Prego (the triticale). The Prairie

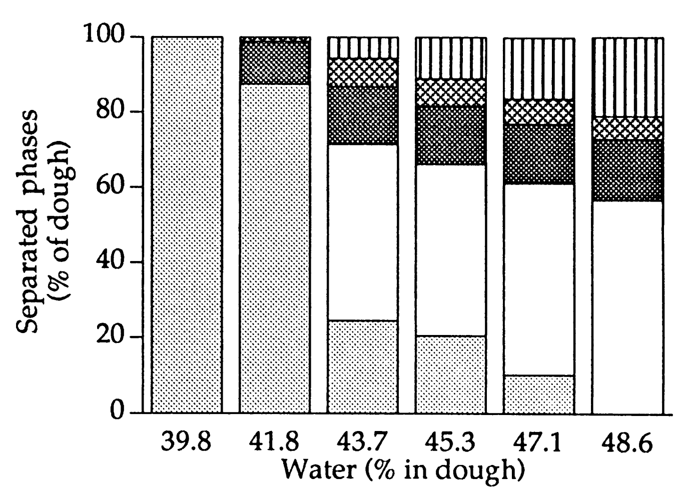

Fig. 1. Separated dough made from Kosack flour with water contents of 39.8–48.6%. Symbols for separated phases are: liquid (striped), gel (crossed), gluten (dark gray), starch (white), and unseparated dough (light gray).

wheat differed from the other cultivars, in that complete separation of starch and gluten was obtained at the farinograph water absorption. Rouquin and Sport on the other hand, showed little tendency to separate, with the largest volume fraction of unseparated dough observed. A lower degree of separation was also indicated for Sport by diffuse boundaries between the separated phases, and the gluten and starch phases appearing less separated from each other at any water content investigated. The most distinct boundaries were obtained for the triticale, feed and Prairie wheats. Kosack separated almost as well as the latter cultivars. Larsson and Eliasson (1996) showed that the separation was impaired when the polar lipid lecithin was added to the dough.

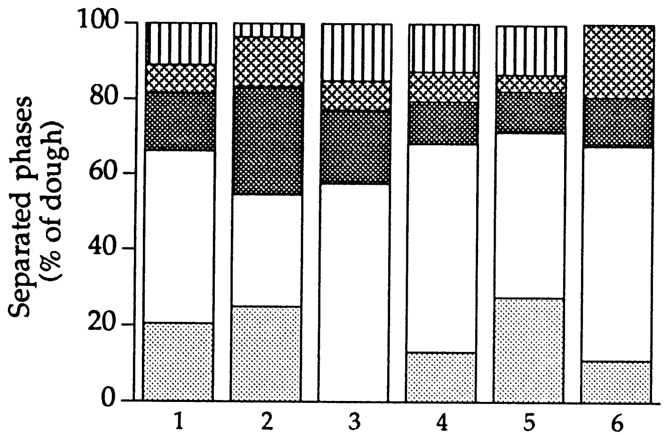

Fig. 2. Separated dough at the farinograph water absorption for Kosack (1), Sport (2), Prairie (3), feed wheat (4), Rouquin (5), and Prego (6). Symbols as in Fig. 1.

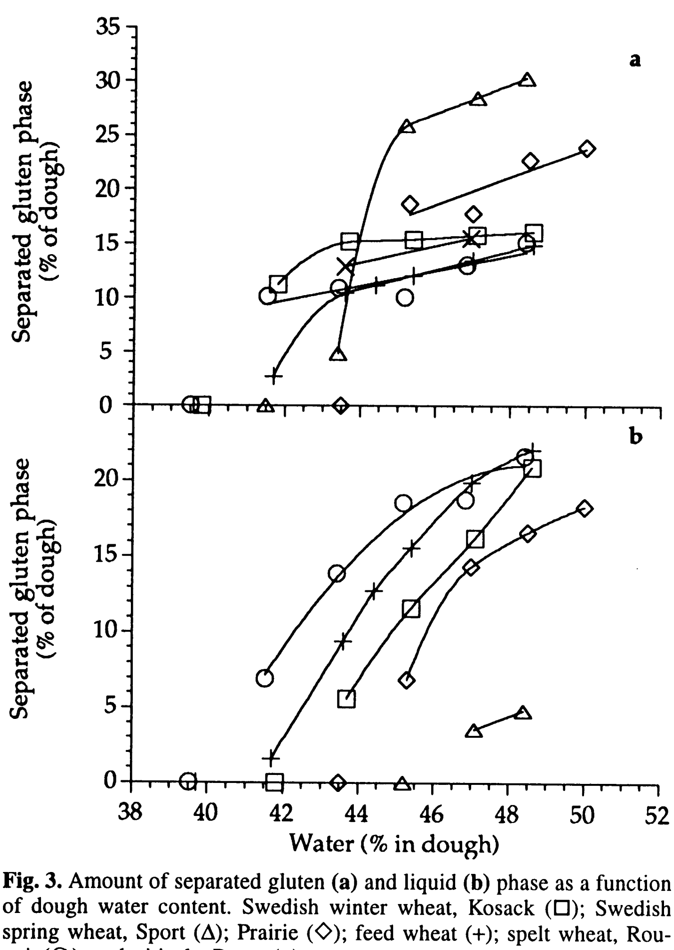

Fig. 3. Amount of separated gluten **(a)** and liquid **(b)** phase as a function of dough water content. Swedish winter wheat, Kosack (□); Swedish spring wheat, Sport (Δ); Prairie (◇); feed wheat (+); spelt wheat, Rouquin(○); and triticale, Prego (×).

These results imply the possibility that the lower degree of separation observed for Sport doughs was caused by a high content of native polar lipids. On the other hand, based on the good phase separation for the Prairie and the feed wheats and Prego (the triticale) one might speculate that their amount of polar lipids was lower. For the feed wheat, the separation of a thin oil film was observed on the top of the liquid phase. The largest volume fraction of gluten phase observed for the Sport dough was consistent with its high protein content (Table I). The gluten phase recovered from Rouquin dough was remarkably small in relation to its protein content (Table I). Apart from a considerable gluten phase, a large amount of gel phase was observed for Sport. The investigation of the phases by microscopy, showed that a large amount of damaged starch was present towards the bottom of the gel phase. However, the large amount of gel phase could not be correlated with a high amount of damaged starch, as the damaged starch was low for Sport (Table I). The separated liquid phase was small for Sport, which may be due to the high protein content of Sport flour (Table I). No liquid phase at all was observed for Prego (the triticale), instead a large amount of gel phase was found. The sum of protein, starch, and ash in Table I is lower for Prego, which indicates a higher content of some component(s) not analyzed. A higher content of total pentosans has been reported for triticales compared with wheat (Bushuk and Larter 1980, Saini and Henry 1989). It seems reasonable that pentosans contribute to the large amount of gel phase observed for Prego in the present study. The fact that no liquid phase was observed, even at the highest water content investigated, implies that the second aqueous phase of Prego consists only of the gel and starch phases.

According to Figure 1, the gluten phase of Kosack developed before any liquid phase separated out. The amounts of gluten and liquid phases which separated out in the water range studied, are shown for all flours in Figure 3a and b. The lowest dough water content required to produce a gluten or liquid phase may be estimated by extrapolation of the amount of the separated phases. The highest dough water contents, where no phase separated out, are included in Figure 3a and b. Comparing these two figures shows a separation of a gluten phase before a liquid phase for Sport. This was also most likely the result for Prairie. No liquid phase at all was obtained for Prego. For these four cultivars, the amount of water required for the separation of a liquid phase seemed to be higher than the amount required for the gluten phase to separate. This indicates that the properties of developed gluten induce the phase separation in accordance with an earlier discussion of the phase separation (Eliasson and Larsson 1993). The different separation properties of the gluten and the liquid phases may also be studied in Figure 3a and b. When the water content of the doughs was increased, the amount of liquid phase increased significantly, while the gluten phase increased moderately. This increase leveled off to various degrees. In some cases, the amount of gluten remained fairly constant at higher water contents. This behavior indicates that gluten is only able to swell to a certain degree and is not capable of incorporating more water at higher water contents. Instead, excess water was recovered in the liquid phase (Fig. 3b). However, a slight increase in the amount of separated gluten phase was observed when the dough water content was increased. The degree of the increase varied by cultivar and was greatest for Sport and Prairie. This increase appears to indicate further gluten swelling, but this was ruled out by determination of the water content of the separated phases. Less unseparated dough was obtained for Kosack as the water content was increased (Fig. 1), which showed that additional gluten was released from the unseparated fraction. This was also the case for the other cultivars. As expected, the separation of gluten and starch was favored by higher dough water contents.

In a study by MacRitchie (1976), dough was separated by ultracentrifugation at 100,000 × g. No liquid phase was observed at a dough water content of 34.5%. This water content was consistent with the appearance of a second continuous aqueous phase in dough, as indicated by conductivity measurements. Results from the present investigation differ from those of MacRitchie in a number of ways. An improved separation of the solid phase into starch and gluten phases was maintained in the present study. The liquid phase was also observed first at higher water contents (41–46%, Fig. 3b). Despite the difference in these results, the unseparated fraction in the present investigation contained approximately the same amount of water (33.5–35.5%) as the solid phase and the dough where no liquid phase separated in the study by MacRitchie. The reason for the divergence of the results is not clear, however, the improved separation in the present investigation may be due to the fact that longer test tubes were used. The mixing procedure also differed between the two studies. In the work of MacRitchie the doughs were mixed to peak consistency in a mixograph (46% water), while in the present investigation the doughs were mixed 5 min in a farinograph. The separation may be improved by a longer mixing time (Larsson and Eliasson 1996).

The water contents of the separated phases (total weight basis) are given for the flours studied in Figure 4a–f. The gel layer contained ≈80% water, the gluten phase 50–55%, and the starch phase ≈30%. The unseparated dough observed below the starch phase contained 33.5–35.5% water (results not shown). The water contents of the gluten and the gel phases of the triticale, were somewhat higher than those of the other flours. Although the amount of developed gluten increased moderately at higher dough water contents (Fig. 3a), the amount of water associated with the separated gluten phase was almost independent of the water content of the doughs (Fig. 4a,c–f). The observation that an increase in dough water content did not increase the water content of gluten is in agreement with the findings of Willhoft (1973). Neither was the water content of the separated gel and starch phases affected by the water content of the dough. This was true for all the cultivars, except for Sport, where the amount of water associated with the separated gel and gluten phases increased when the dough water content increased (Fig. 4b). Compared with the other cultivars, a higher water content of both the gel layer and the gluten phase was observed for the separated Sport doughs. The findings for Sport seemed to be explained by the lower tendency of the dough to separate. The diffuse boundary lines of Sport were not observed for any of the other cultivars. For Sport, an improved degree of separation was observed at higher water contents; first as a larger amount of water in the gel phase, second, as an increase in the water content of the gluten phase, concomitant with less starch in the gluten phase; and third, a decrease in the water content of the starch phase, i.e., less gluten was recovered in the starch phase.

The distribution of water in dough has been estimated from water absorption measurements of the different components (Bushuk 1966). It was found that the total amount of water associated with starch, protein, and pentosans was 45.5, 31.2, and 23.4%, respectively. For the phase separation properties, this implies that the whole liquid phase is occupied by starch and pentosans so there is no free water. The water distribution in dough may also be evaluated from the phases separated by ultracentrifugation. These values can be obtained if the approximation that the density of the separated phase is equal to the density of dough is accepted. Calculated for the Kosack dough containing 45.3% water (Fig. 2), the fractions of the total amount of water, associated with the phases obtained were: 16% (unseparated), 30% (starch), 19% (gluten), 13% (gel), and 22% (liquid phase). Considering the large differences for cultivars in the amount of separated gluten phase, the amount of water in the separated phases of the different cultivars would also vary. At farinograph water absorption, the amount of water associated with the gluten phase of the cultivars was: 12% (Rouquin), 13% (feed wheat), 17% (Prego), 19% (Kosack), 22% (Prairie), and 34% (Sport). It can be concluded that the high protein content of Sport gave the largest amount of water associated with the gluten phase. On the other hand, Rouquin, with a rather large amount of unseparated dough and a protein content similar to that of the Prairie (Table I), exhibited a small amount of water associated with the gluten phase, i.e., a small amount of separated gluten phase (Fig. 2). Consequently, the fraction of the available water in dough that was incorporated into the gluten phase was strongly dependent on cultivar.

Stress-Relaxation Modulus

Figure 5a shows the relationship between stress-relaxation modulus (G_0) and dough water content for the flours. G_0 was strongly dependent on water content; the modulus decreased with increased water content. The strong dependence of the dynamic modulus on water content is well established (Hibberd 1970a, Smith et al 1970, Hibberd and Parker 1975, Navickis et al 1982, Abdelrahman and Spies 1986, Dreese et al 1988). In this study, flours from six wheat cultivars with water contents of 38–50% (by dough weight) were investigated. All the flours, except the

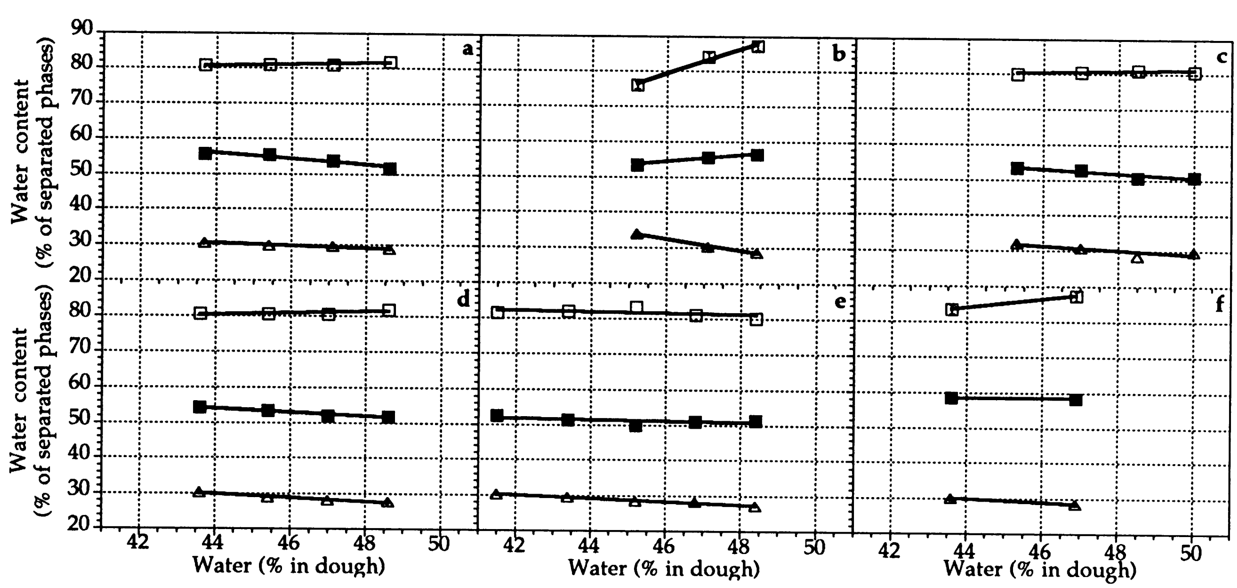

Fig. 4. Water content of gel (□), gluten (■), and starch (Δ) phases. **a–f,** Kosack, Sport, Prairie, feed wheat, Rouquin, and Prego, respectively.

Prairie, exhibited approximately the same modulus at a given water content (Fig. 5a). The differences between cultivars were greatest at lower water contents. At higher water contents, a plateau where G_0 was less dependent of water content was observed for all cultivars. The water content at which the plateau started differed for the cultivars. For Prairie, the value was 47%, whereas, for the other flours, it was 44–45%. The farinograph water absorption (Table II) was found at approximately the same water content as the onset of the plateau in the G_0 versus water content plot (Fig. 5a). This was observed for all cultivars except Sport, where the water content corresponding to the farinograph water absorption was on the plateau at 47.1%. This elevated water absorption may be due to the high protein content of Sport. In general, the plateau (where G_0 is less dependent on water content) may be considered as the range of water contents where gluten swelling was complete and the two continuous aqueous phases are developed. This was confirmed by the fact that the phase separation occurred in the same range of water contents (Fig. 3a and b).

The stress-relaxation modulus can be related to gluten strength

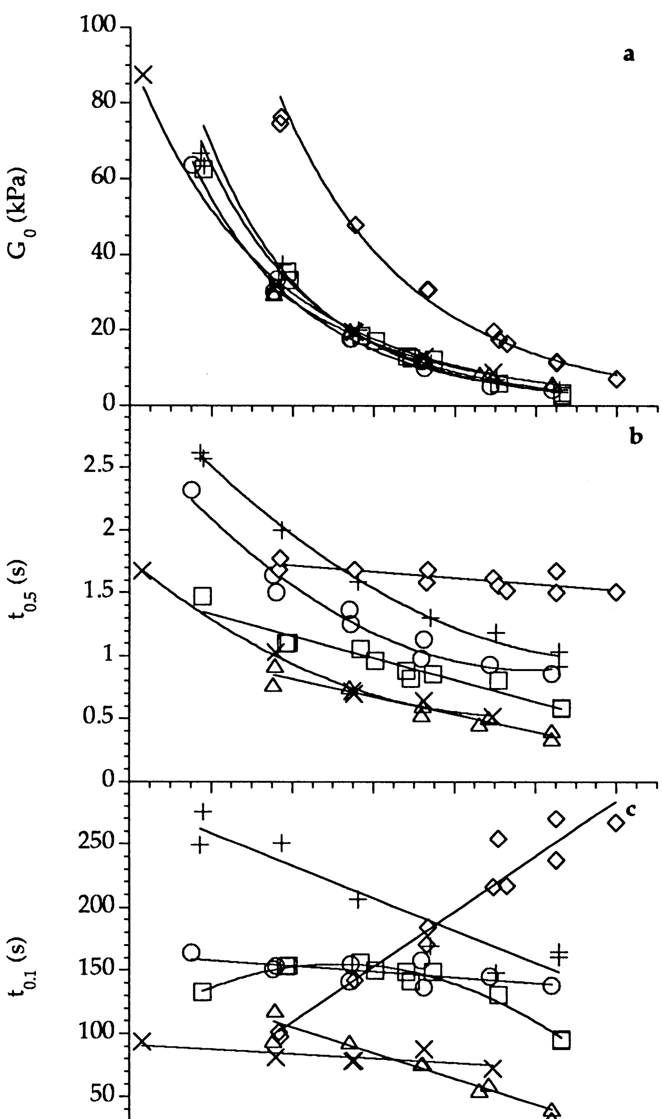

Fig. 5. Influence of dough water content on **a,** stress-relaxation modulus (G_0); **b** and **c,** stress-relaxation times ($t_{0.5}$ and $t_{0.1}$, respectively). Symbols as in Fig. 3.

and amount, the amount of liquid phase (fraction of starch in the liquid phase), and the amount of unseparated dough. We concluded that no simple comparison between cultivars could be undertaken as both the amounts and the properties of the separated phases differed between cultivars. However, some observations can be made.

When values of G_0 were compared at the highest water contents, which was expected to relate to the greatest influence of gluten on G_0, the highest moduli were observed for Prego (8.8 kPa), Prairie (7.1 kPa), and Sport (6.6 ± 0.5 kPa). With a reproducibility of 10%, it seems possible to distinguish between these higher values and the lower ones obtained for Rouquin (4.1 kPa), feed wheat (3.5 ± 0.3 kPa), and Kosack (3.0 ± 0.3 kPa). Approximately the same amount of each dough phase separated for Rouquin, feed wheat, and Kosack. Figure 3a and b shows the separated gluten and liquid phases. The same amount of all the separated phases for Rouquin, feed wheat, and Kosack correlates well with the small difference in modulus obtained on the plateau. For Prego, Prairie, and Sport, different reasons for the high modulus may be distinguished. Prairie was expected to have a stronger gluten than the Swedish cultivars. Sport, the Swedish spring wheat, with a high protein content (Table I), had a small liquid phase densely packed with starch. On the other hand, the gluten phase was large and the amount of starch low. The greatest influence on the stress-relaxation modulus of Sport was probably due to the lower tendency to separate. The behavior of Sport seemed to agree well with the fact that starch cannot be considered as an inert filler (Hibberd 1970b, Smith et al 1970). In the case of Prego (triticale), the high value of G_0 (8.8 kPa) was attributed to the lack of a liquid phase, causing reduced flow properties to the second aqueous phase of the dough.

The phase separation characteristics of doughs made of different types of flours are affected by the varying amounts of gluten, starch, or gel phase in the system. The amount of liquid phase seemed to play an important role in the determination of the effect on the modulus at small deformations. For example, when gluten was added to the system (at constant water content), as in the study by Smith et al (1970), a reduction in the amount of separated liquid phase was observed, due to the high water absorption of gluten. Depending on the original amount of the liquid phase, the increase in modulus can vary from one cultivar to another. On the other hand, when the water content was increased at different protein-to-starch ratios, as in the study by Hibberd (1970b), the fraction of starch in the liquid phase decreased at a constant protein-to-starch ratio. This is consistent with a reduced volume fraction of starch in the liquid phase and, hence, a decrease in the modulus. The modulus was less affected by the protein-to-starch ratio at lower water contents. At increasing water contents, the protein-to-starch ratio became more and more important because of the amount of liquid phase present in the phase-separated system.

Relaxation Times

The stress-relaxation of dough can be separated into two flow processes (Bohlin and Carlsson 1981). The first relaxation process is strongly cooperative and occurs at shorter times (0.1–10 sec), whereas the second process is weakly cooperative and takes place at longer times (10–10,000 sec). In this study, the stress-relaxation times, $t_{0.5}$ and $t_{0.1}$, were chosen to represent the two flow processes. The first relaxation process was characterized by the short relaxation time ($t_{0.5}$), the time when half of G_0 had relaxed. When the second relaxation process started, G_0 was reduced to 20–30 % of its initial value. This indicates that $t_{0.1}$ (the time when 10% of G_0 remained) took place during the second relaxation process. The second relaxation process has been related to the continuous gluten phase in dough (Carlsson 1981).

The dependence of the relaxation of the first flow process, represented by $t_{0.5}$ on water content, is shown in Figure 5b. Two different types of relationship were observed, depending on the wheat cultivar. In one type, the relaxation time was only slightly influenced by water content (Kosack, Sport, and Prairie). The $t_{0.5}$ for Prairie was high. The relaxation time of the feed wheat, Rouquin, and Prego appeared to be sensitive to water content, particularly at low water contents. The influence of water content on these cultivars was reminiscent of the effect on G_0.

The largest differences between cultivars for the influence of water content were observed for the long relaxation time, $t_{0.1}$ (Fig. 5c). A faster relaxation of the second flow process (lower $t_{0.1}$) was found for Kosack, Sport, and the feed wheat at increasing water contents. The reduction in $t_{0.1}$ may be interpreted as a dilution of the second flow process or possibly a decrease in the amount of the unseparated fraction at higher water contents, which was pronounced for the feed wheat and only moderate for Sport, Rouquin, and Prego. The most conspicuous behavior was observed for Prairie, with an increasing $t_{0.1}$ at increasing water contents. This unexpected behavior prompted a study on the influence of mixing time at the farinograph water absorption on Prairie doughs. Even after 20 min of mixing, there was no significant influence on $t_{0.1}$ (results not shown). When Kosack and Sport were mixed for 20 min, an increase in $t_{0.1}$ was observed for both cultivars (Larsson and Eliasson 1996). The different effects of prolonged mixing on $t_{0.1}$ indicates that the Prairie gluten was stronger than that of the other cultivars. One explanation of the effect of water content on $t_{0.1}$ may be related to the influence of starch on the second relaxation process at lower water contents. Below the farinograph water absorption (47.5% water), the data reproduced very well, while at higher water contents large scattering was observed (Fig. 5c). The separation of starch and gluten was already complete at the farinograph water absorption (Fig. 2). At the highest water contents a spontaneous separation (lumpy dough) was observed. These inhomogeneities of the samples may explain the large scattering of Prairie data in Figure 5c. For Rouquin (the spelt wheat) and Prego (the triticale), the relaxation times of the second flow processes were not affected by the water content in the interval studied.

Relationship Between Phase Separation and Stress-Relaxation Behavior

Considering dough as bicontinuous with two interpenetrating aqueous phases (Eliasson and Larsson 1993), the liquid phase serves as the medium in which starch and solubles (the gel phase) are located. To correlate the phase separation properties with the stress-relaxation measurements, the modulus was plotted as a function of the amount of separated liquid phase for the five cultivars (Fig. 6). As soon as phase separation had taken place, a linear relationship between the modulus and the liquid phase was observed for all cultivars. The linear relationship in Figure 6 seemed to indicate that starch suspended in the liquid phase conferred viscoelasticity to the dough, also at the highest water contents studied. This may either be caused by the dilution of the second aqueous phase or originate in the presumed washing effect of the liquid phase, i.e., reducing the starch content of gluten. The latter suggestion is consistent with the behavior of Kosack (Fig. 1) as a decreasing amount of unseparated dough was observed at increasing dough water contents. Prairie deviated slightly from the linear relationship and was only linear at the three highest water contents corresponding to the plateau in Fig. 5a, where complete separation was obtained.

In the water content range studied, it is tempting to divide the dependence of G_0 into two regions. In the first region (at lower water contents), G_0 was strongly dependent on the water content and no separation occurred when the dough was submitted to ultracentrifugation. In the second region, at higher water contents, separation had taken place, and the modulus was less dependent on the water content. If these two regions are approximated to linear, the intersection of them seems to coincide with the area of water contents where the second aqueous phase appeared. The water content here was estimated at ≈43–44% for Kosack, the feed wheat, Rouquin, and Prego, and ≈46% for Sport and Prairie (Table II). This estimated water content also seems to be related to the farinograph water absorption (Table II).

It may be expected that the influence of starch on the stress-relaxation modulus would be almost eliminated at a certain ratio of starch-to-liquid phase. To further evaluate the effect on the stress-relaxation modulus of the amount of starch in the liquid phase, the modulus was plotted as a function of the total starch content (Table I) divided by the amount of separated liquid phase (Fig. 7). This involved the assumption that all the starch was suspended in the liquid phase, i.e., no unseparated fraction. This was true for Prairie at the farinograph water absorption and for Kosack, the feed wheat, and Rouquin only at the highest water contents investigated. A complete separation of gluten and starch was not observed for Sport. Figure 7 shows that when the stress-relaxation modulus approached 0, the ratio of starch to the amount of liquid phase was ≈2. Figure 6 shows that the influence of the liquid phase on the stress-relaxation modulus decreased linearly in the water range studied where separation occurred. Figure 7 shows that the amount (volume) of liquid phase needed to reduce the influence of the aqueous starch phase on the stress-relaxation modulus must be more than half the amount (weight) of starch present in flour. At such high water contents, the dough

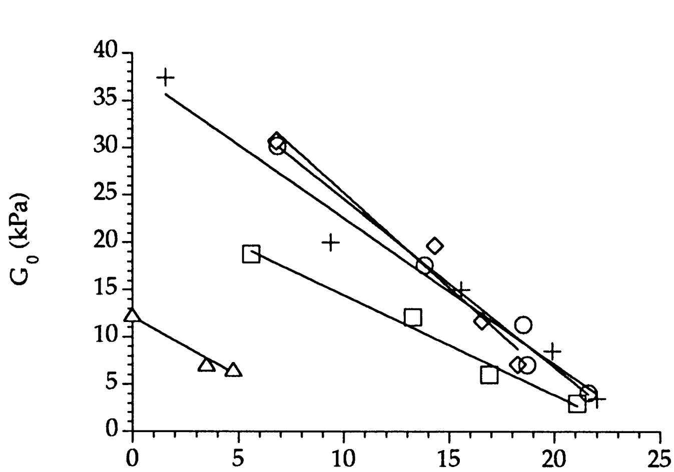

Fig. 6. Stress-relaxation modulus (G_0) as a function of amount of separated liquid phase. Symbols as in Fig. 3.

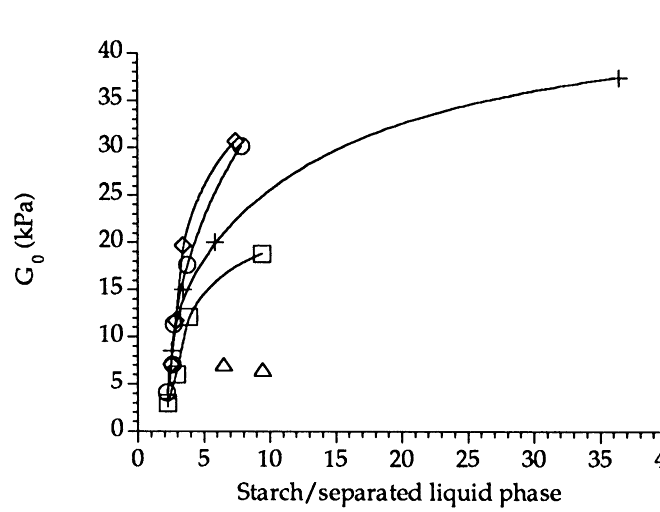

Fig. 7. Stress-relaxation modulus (G_0) as a function of starch content in dough (weight %) divided by the amount of separated liquid phase (volume %). Symbols as in Fig. 3.

was judged to be impossible to handle, perhaps because the liquid phase was present in amounts large enough to enable noninteracting starch to spontaneously leave the dough.

CONCLUSIONS

The bicontinuous aqueous phases in wheat flour dough were characterized. Phase separation occurred at high water contents where gluten was fully developed. Because of the gluten development, the remaining water was able to form a second aqueous phase with starch. This caused a greater influence of the gluten phase on the stress-relaxation modulus at higher water contents. In the water content range studied, where separation occurred and a dough of acceptable handling properties could be prepared, starch or the amount of starch relative to the diluting system still influenced the modulus slightly. No phase separation occurred at low water contents. A high stress-relaxation modulus was caused by the highly concentrated suspension resulting at lower water concentrations where gluten development was limited.

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