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Mass transfer limitations during starch gelatinization

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Abstract

A relatively large volume of maize starch suspension (2.5–20% w/w solids) is heated to above its gelatinization temperature by two means: a microwave oven meant to provide a uniform global thermalization of the sample, and a conventional local electrical heater which, depending on agitation, yields different heating patterns on the sample. The progress of gelatinization is registered on-line by an electrical conductance technique. Contrary to what is observed in the conventionally heated samples, gelatinization is not completed in the microwave irradiated samples although the temperatures reached are as high as with conventional heating. This is attributed to poor mass transfer of water molecules during microwave irradiation as a result of the short processing period and the absence of mixing of water with starch components. The latter is in line with observations regarding the role of agitation in gelatinization under conventional heating.

Keywords: Microwaves; Electrical conductance; Starch gelatinization; Maize starch; Heating patterns

1. Introduction

Gelatinization can be described as a sequence of changes in starch granules upon heating: starch granules first absorb large amounts of water, then swell to many times their original size and finally leach their starch components. Depending chiefly on the starch/water ratio but also on a number of processing parameters such as temperature, heating rate, strain history, etc. a viscous solution or a gel is formed. Many aspects of starch gelatinization have been extensively studied in the past since it is a phenomenon encountered in many food processes, e.g. baking, production of cereals and soups, etc. However, relatively little work has been focused on investigating the time-dependent role of water in granule disordering and dissolution at a fundamental physicochemical level (Slade & Levine 1991).

Recently, Karapantsios, Sakonidou, and Raphaelides (2000) have reported that the mixing pattern inside a test vessel where a starch/water suspension heats-up can pose an additional barrier for gelatinization. In particular, it has been observed that in the absence of an appropriate low-shear agitation to promote gentle mixing of every part of the starch/water suspension and also transfer heat throughout the entire sample mass, there are sections in the test vessel where gelatinization is greatly retarded. However, that work could not distinguish whether it was the insufficient mixing of water with starch components or the poor heat transfer in these sections of the vessel that was the cause of this. This is because both mixing and heat transfer are promoted by the agitating action of the mixer.

A practical way to separate the two contributions is to compare gelatinization performed under conventional local heating, where agitation is necessary to transport heat in the rest of the sample, with gelatinization under microwave global heating where no agitation is needed. Microwave heating of foods has been traditionally considered as an effective way to achieve fast and uniform thermal treatment of large sample volumes Buffler 1993; Knutson, Mart, & Wagner, 1987; Mudgett, 1989). Strictly speaking, this is not entirely true since parameters such as the geometry and dielectric properties of the sample as well as characteristics of the microwave source and cavity substantially influence the development of the thermal profile (Chen, Singh, Hagghighi, & Nelson, 1993; Fakhouri & Ramaswamy, 1993; Fleischman, 1996;

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Kirk & Holmes, 1975; Lin, Anantheswaran, & Puri, 1995; Ohlsson & Risman, 1978; Ramaswamy & Pillet-Will, 1992; Vilayannur, Puri, & Anantheswaran, 1998a,b; Zhou, Puri, & Anantheswaran, 1995).

A number of studies have investigated the role of geometry and size of foodstuffs regarding the spatial temperature profiles that develop inside the irradiated samples. In general, corner and edge heating effects in rectangular geometries and core heating effects in cylindrical and spherical geometries have been investigated both in modeling and experimental studies. More related to our starch/water systems are studies with whole potatoes of roughly cylindrical geometry (Chen, Collins, Mccarty, & Johnston, 1971; Chen et al., 1993; Huang, Hess, Weber, Purcell, & Hube, 1990; Ohlsson & Risman, 1978; Vilayannur et al., 1998b; Zhou et al., 1995).

To our knowledge there is no prior work studying the spatial temperature non-uniformities in water/starch suspensions gelatinizing under microwave heating. Goebel, Grider, Davis, and Gordon (1984) and Zylema, Grider, Gordon, and Davis (1985) working with suspensions of different wheat starch content recorded the transient temperature profiles but only at the centre of their samples. Goebel et al. (1984) worked with 1:1, 1:2, 1:4 and 5:95 starch/water ratios while Zylema et al. (1985) worked with 1:1, 1:2, 1:4 and 1:8 starch/water ratios. Despite the different sample sizes both studies found that their most dilute suspensions needed about 40% more time to heat-up compared to the most concentrated ones, an observation opposite to what might be intuitively expected (Buffler, 1993).

A number of papers examine the variation of the dielectric properties—permittivity and loss factor—of foodstuffs, e.g. Nelson (1973). These properties dictate the penetration depth and absorption of microwaves in the sample and are appreciably dependent on temperature. The volumetric power absorption by the sample depends chiefly on the electric field intensity inside the sample which in turn is the result of a complex interplay among the cavity configuration, the geometry of the sample and the dielectric properties of the sample (Buffler, 1993).

Studies of dielectric properties of starch/water suspensions in the commercial microwave oven frequencies (915 and 2450 MHz) are scanty in literature. Miller, Gorden, Davis, (1991) using a cavity perturbation technique investigated the role of temperature on the dielectric properties of starch/water mixtures (1:1 and 1:2) prepared from waxy and normal potato, chemically modified tapioca and waxy and normal corn with different degrees of substitution. For normal corn these authors reported that in the range 30–90 °C the permittivity is approximately constant while the loss factor reduces by approximately 40% with temperature but, interestingly, in the range of starch thermal transitions (~60–75 °C) it remains invariant. Regarding differences between the two concentrations, the maximum deviations observed were less than 15% for the permittivity, with a tendency to diminish for temperatures above gelatinization, whereas the loss factor was virtually constant throughout. Ndife, Sumnu, and Bayndirli (1998) also found a progressive attenuation of the loss factor of starch suspensions with temperature.

The only study known to us dealing with dielectric properties of dilute suspensions like ours is the one by Ryynänen, Risman, and Ohlsson (1996). The temperature dependence of the dielectric properties of potato, wheat, corn and waxy corn starch suspensions with 1:2.3 and 5:95 starch/water ratios was examined. For temperatures 60–95 °C, the range of interest to this work, these authors reported a decrease in permittivity by ~25% and in loss factor by ~50%, roughly the same for all concentrations. The deviation in permittivity among concentrations was approx. 20% (the higher values attained by the more dilute suspensions) while the loss factor was essentially invariant.

The present study examines the distinct heating patterns that are produced by conventional and microwave heating of maize starch suspensions of different solids content (2.5–20% w/w). Temperature recordings at two positions in the test vessel provide information about any spatial temperature distributions in the sample. The role of agitation in the case of conventional heating is investigated. The electrical conductance technique recently reported by Karapantsios, et al. (2000) is used to monitor the progress of gelatinization. Comparison of electrical conductance records from suspensions heated by the two heating methods offers the opportunity of separating the role of mass and heat transfer on gelatinization phenomena, thus clarifying an issue originally introduced in the latter publication. The comparison is further supported by use of classical methods to examine the degree of gelatinization.

2. Materials and methods

2.1. Materials

Commercial maize starch with a moisture content of 13.5% was purchased from Group Amylum S.A., Greece. Thermal measurements performed on maize starch/water samples using a Perkin-Elmer DSC-6 instrument showed that the temperature of the onset of gelatinization was 67.5 ± 0.6 °C and the peak gelatinization temperature was 72.4 ± 0.4 °C. More about the physicochemical characterization of the starch employed can be found in an earlier publication (Karapantsios et al., 2000).

The liquid used was filtered tap water with a specific conductivity between approximately 600 and 700 μS/cm. A series of starch/water mixtures with solids concentration of 2.5, 5, 7.5, 10, 12.5, 15 and 20% w/w was tested. The volume of the sample was always above 0.5 l (it varied slightly between heating methods due to experimental constraints), which was sufficiently large for heat and mass transfer phenomena to occur on a macroscopic scale.

2.2. Test vessel with conductance probe

All gelatinization experiments were performed inside a cylindrical Plexiglas test vessel with ID = 6.8 cm, OD = 7.8 cm and height = 23 cm. A detailed description of the test vessel is given elsewhere (Karapantsios et al., 2000) and only some essential information is provided here. Conductance measurements were made by means of two parallel stainless steel ring electrodes (strips) with a width of 3 mm, placed 3 cm apart, and located at the mid-height of the test vessel. The electrodes were flush mounted onto the inner surface of the test vessel to avoid disturbing the cross section of the vessel. The separation distance between electrodes was based on the volume-averaging approach of modeling porous media (e.g. Carbonell & Whitaker, 1984). Preliminary tests have shown that the size of the probe selected was large enough to average temperature variations (hot and cold spots in the sample) yet small enough to preserve the local character of the measurements.

An alternating current (a.c.) carrier voltage of 0.5 V peak-to-peak was applied across the electrodes at a frequency of 25 kHz in order to eliminate capacitive impedance. Such high frequency electrical measurements are most representative of the free water in the intergranular spaces and in the pores of the material since strongly bound water has a resonance at much lower frequencies (Hardman, 1985). So, as the granules take up the surrounding free water during gelatinization the electrically accessible free water decreases reflecting the structural changes accompanying starch swelling and solubilization. Exploratory tests have shown that the electronic interaction between the employed microwave field (2450 MHz) and the diagnostic signal (25 kHz) is negligible due to the several orders of magnitude difference in scale and the strongly dissipative nature of the former.

The response of the electrodes was fed to a special electronic analyzer. The direct current (d.c.) voltage output of the analyzer was converted to apparent conductance of the medium between the electrodes, K_{app}, using a calibration curve based on precision resistors. To differentiate between actual structural changes (due to gelatinization) and temperature effects but also eliminate errors due to liquid conductivity measurement, the apparent conductance signal was normalized with the conductance of the test section full of water, K_{app}/K_{max}. The degree of starch granule swelling during gelatinization was independently assessed by polarizing light microscopy and application of Congo red dye whereas the residual crystallinity was occasionally checked by DSC thermograms (Perkin–Elmer DSC-6).

2.3. Heating methods

Microwave heating. The starch/water dispersion (~540 g) was initially preheated with stirring to below its gelatinization temperature (<60 °C), in a glass beaker placed over a heating plate. This initial heating step was adopted because the continuous stirring in the beaker prevented the sedimentation of the ungelatinized starch granules. At the end of preheating, the suspension was poured into the test vessel and immediately inserted in the microwave oven (Miele, M696) and then its magnetron was energized at full power (700 W nominal at 2450 ± 50 MHz). No means of agitation was employed. During the runs the ambient air inside the microwave oven was kept at ~70 °C using a hot air-blower in order to suppress heat losses from the test vessel. Calorimetric tests showed that the hot air in the cavity did not affect the power supplied by the magnetron (although it resulted in its cooling fan being kept on continuously). All experiments ended the moment the suspension started to boil at the surface of the test vessel. This was happening while the temperature measured at the center of the conductance probe (mid-height of the vessel) was still a few degrees below 100 °C showing longitudinal variations of temperature inside the test vessel.

Conventional heating. Runs were conducted under a constant heat supply provided by a submerged heating coil placed at the top of the test vessel. The heating power was 150 W yielding a heating rate of $\Delta T_{liq}/\Delta t_{20°-60°} \cong 3$ °C/min. To assure homogeneity of the liquid mixture (~600 g) during heating, a center stirrer was employed with four impellers mounted along an electrically insulated shaft. Among them, specific reference must be made to a custom-made grid paddle assembly, specially designed to achieve mixing in every part of the gelatinized viscous material. For concentrations 12.5, 15 and 20% w/w mixing was performed at 260 rpm throughout the whole runs. For concentrations 2.5, 5, 7.5 and 10% w/w mixing was initially done with 260 rpm until the conductance signal reached a maximum (always at around 70 °C) and then it was adjusted to a final value of 40, 160 or 260 rpm.

2.4. Temperature measurement

Measurement of temperature inside a microwave cavity is a matter of concern. Thermocouples, the most common means of temperature measurement, were used by many studies in the past but readings were taken only with the radiation being turned off momentarily (Fakhouri & Ramaswamy 1993; Khraisheh, Cooper, & Magee, 1997; Kirk & Holmes, 1975; Ramaswamy & Pillet-Will, 1992). This was because conventional thermocouple probes exhibit powerful electrical discharges and signal perturbations inside a microwave field (Buffler, 1993; Randa, 1990; Wickersheim, Sun & Kamal, 1990).

In this study, temperature was measured on-line by specially constructed thermocouples following the work of Kingston and Jassie (1986) and Ramaswamy, Van de Voort, Haghava, Lightfoot, and Timbers (1991). The thermocouples (type K, AGW 28 wire, Omega Ltd) were housed in aluminum sleeves with their leads shielded using nickel coated copper overbraid. Moreover the thermocouples were never operated in the cavity without a load but always immersed in the starch suspension which offered an additional protection from the microwave field. Temperatures were measured up to 100 °C, the maximum working temperature, without noticing problems such as arcing or transient temperature spikes.

Another issue of concern when using thermocouples to measure the temperature inside a microwave cavity is that the thermocouple wires transmit energy out of the cavity (Kingston & Jassie, 1986; Ramaswamy et al. 1991;). Such phenomena, however, are prominent with small size samples where multiple reflection of microwaves occurs within the cavity before the microwaves are absorbed by the sample. With our large volume samples, simply grounding the thermocouple shield to the oven metallic frame proved a satisfactory leakproof precaution. Indeed, simple calorimetric tests showed that energy leaks were always below 0.5% of the nominal power.

Temperatures were measured in the center of the conductance probe at two radial positions across the vessel: one at its center axis and the other 2–3 mm from the inside vessel wall. For the conventional heating scheme the readings of the two sensors were in such close proximity (± 0.3°) that no differentiation was made in presenting them but instead single mean values were calculated. On the contrary, for the microwave heating, measurements from the two sensors are presented separately.

3. Results and discussion

3.1. Macroscopic observations

The appearance of the samples at the end of the microwave experiments was as homogeneous as with the agitated samples in conventional heating. In both cases the starch suspension was transformed to an opaque paste of an even consistency. This was a proof that first preheating the sample outside the cavity and then gelatinizing it under microwave radiation was effective in avoiding settling of starch granules at temperatures below gelatinization. However, our observations are in contrast with what has been reported in literature, e.g. Goebel et al. (1984), Zylema et al. (1985) and Ndife et al. (1998). In those studies, regions with different macrostructural characteristics (gelled, pasted, chalky, etc.) were identified in the microwave gelatinized samples, depending largely on the starch/water ratio. Such phenomena were repeatedly observed also in preliminary tests in this work when our samples were heated inside the microwave oven all the way from room temperature to complete gelatinization. So, despite the rapid heating of the sample by microwave irradiation there must always be concern with problems of granule stratification when the sample has appreciable fluidity (before gelatinization). Such concerns were expressed in the discussion with reviewers in Goebel et al. (1984) and were accepted by these authors.

Another persistent feature of all the microwaved samples was the existence of numerous tiny gas bubbles ($d < 0.5$ mm) dispersed evenly in the sample volume. This was never observed in the conventionally heated samples. Such bubbles probably originate from the diminishing solubility of air in the water of the samples as their temperature increases. In the absence of agitation these bubbles do not have the chance to come together and coalesce and get finally removed from the sample. These bubbles may play a small role in promoting natural convection in the very low viscosity mixtures (low solids content + high temperatures) and may also alter to some extent the electrical properties of the sample.

3.2. Temperature profiles

Microwave heating. Fig. 1(a)–(d) presents the temperature time records measured by the two thermocouples during microwave heating for solids concentrations 2.5, 7.5, 12.5 and 20% w/w. The circles correspond to the temperatures at the center of the measuring volume while the triangles to the temperatures adjacent to the wall.

In all plots in Fig. 1, the readings from the two sensors increase at a comparable rate from a value below 60 °C (temperature at which the dispersion is poured into the test vessel) to a value around 70–75 °C. At this point the temperature at the center starts to climb faster than the temperature near the wall (the latter increases at a roughly constant rate throughout the entire heating period). A similar thermal 'runaway' at the center of the sample was observed by Zylema et al. (1985) for more concentrated suspensions. At the end of the experiments temperature differences between sensors as high as 10–15 °C are observed. An exception to that is the suspension with 2.5% solids where the two readings approach again each other at high temperatures probably due to natural convection effects in the low viscosity mixture.

As already mentioned it is rather common that microwaves create hot and cold spots in a sample depending on a number of physical and geometrical characteristics. With a cylindrical sample like ours the key phenomenon is the *focusing* of the microwave energy at the central region of the sample that occurs when microwave signals of random orientation impinge on the sample (Buffler, 1993). Similar arguments regarding divergent thermal gradients with microwave heating due to test vessel shape and size were also reported in the past, e.g. Chen et al. (1971, 1993), Collins and McCarty (1969), Huang et al. (1990), Ohlsson and Risman (1978), Vilayannur et al. (1998b), and Zhou et al. (1995).

The real issue for this work is whether these gradients prevent uniform heating of the samples by microwaves. As mentioned earlier the separation distance between the ring electrodes (3 cm) is large enough to average temperature variations (hot and cold spots) in the sample. So, electrical measurements can effectively represent an evenly heated

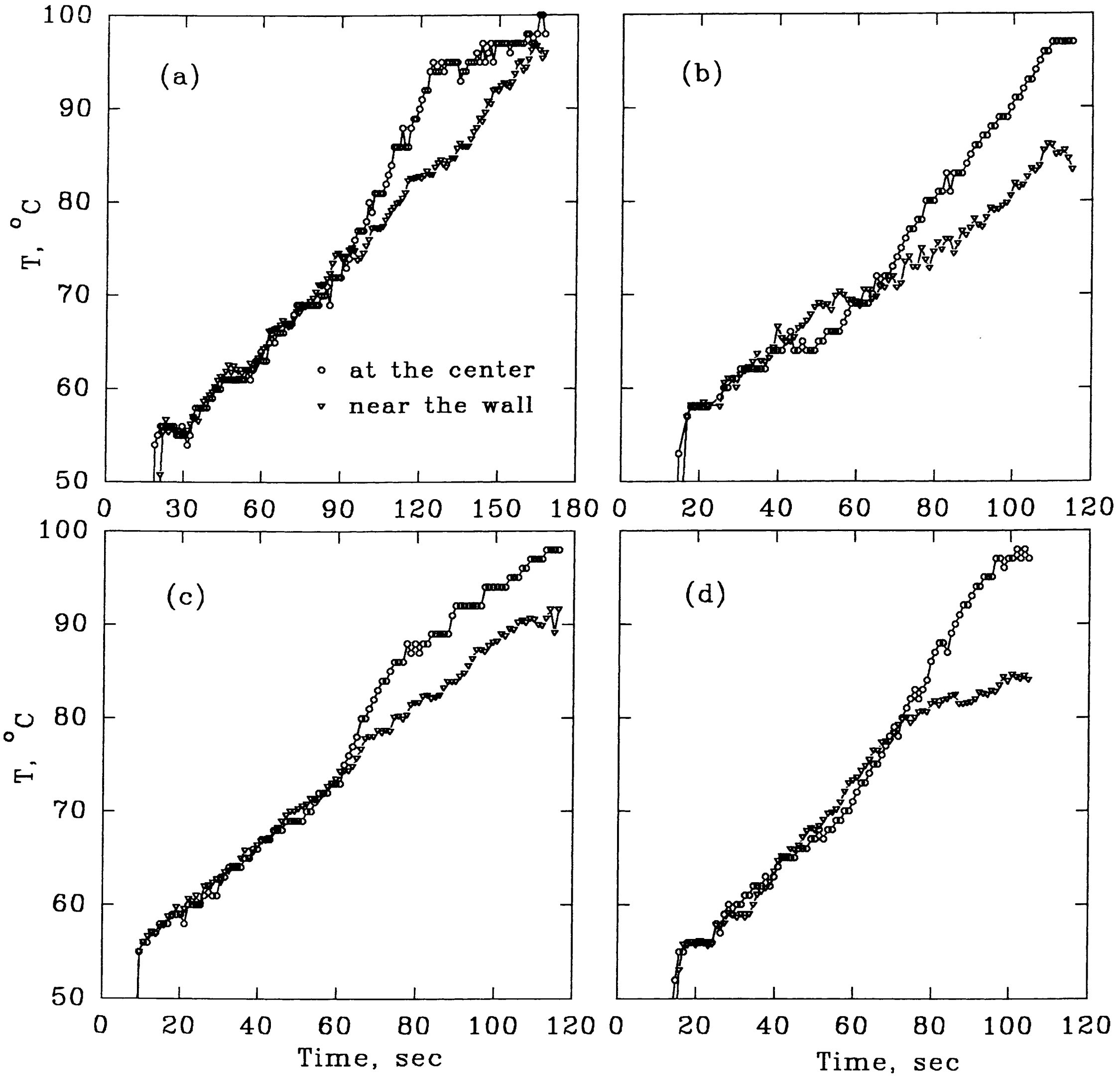

Fig. 1. Temperature versus time for starch/water mixtures with solids concentration of (a) 2.5%, (b) 7.5%, (c) 12.5% and (d) 20% w/w, heated in a microwave oven. The circles represent the temperatures recorded at the center of the measuring volume while the triangles the temperatures recorded near the wall of the test vessel.

volume of the sample. In Fig. 2, average temperature values calculated from instantaneous values of the recorded signals of the two thermocouples are compared to each other for suspensions with 2.5, 7.5, 12.5 and 20% w/w solids. These temperatures will be referred henceforth as the volume-average values corresponding to the electrical measurements. The different scales between Figs. 1 and 2 are because the curves in Fig. 2 are reduced to a common time scale for the shake of comparison. This is done by taking as time zero the time the calculated average temperature reads 60 °C. One should recognize that the largest uncertainty concerning the average values of Fig. 2 exists at the end of the experiments where the divergence of the original signals in Fig. 1 is larger.

As can be seen in Figs. 1 and 2 the temperature rises faster in the dispersions with higher starch content. Such observations have been made previously, e.g. Goebel et al. (1984) and Zylema et al. (1985), and are opposite to the basic principle that microwave heating rates reduce as the water becomes more immobilized (Buffler, 1993). These earlier studies attributed this behavior to an increased energy conversion in starch granules accompanying the structural transformations upon gelatinization. However, such effects should in principle be reflected in changes in the dielectric properties, gelatinization enthalpy and specific heat but this is not the case. As has been mentioned in Section 1 (Ryynänen et al., 1996), in the examined temperature range the loss factor is practically unchanged and the permittivity changes by at most 20%. Moreover, the effect of the gelatinization enthalpy is negligible whereas the heat capacity of the more dilute suspensions is above the values of the denser ones by no more than ~10% (Ndife et al., 1998; Noel & Ring, 1992). Some heat transferred by convection and conduction, perhaps inevitable in our large samples, would have the opposite effect (faster heating of the less viscous dilute suspension) on the observed heating rates due

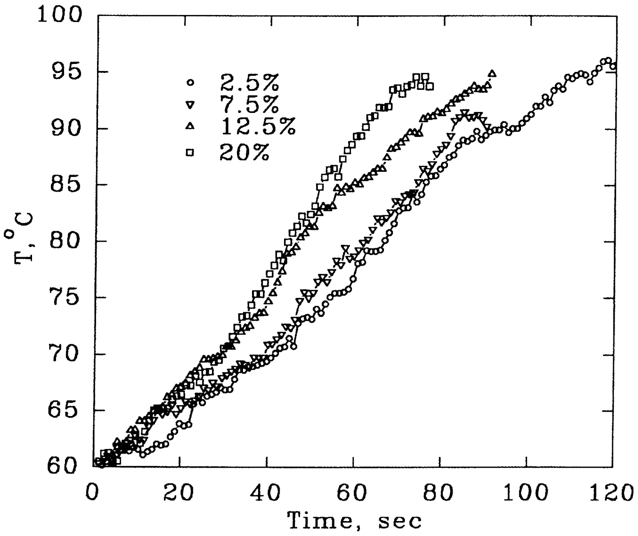

Fig. 2. Average-temperature versus time for starch/water mixtures with solids concentration of 2.5, 7.5, 12.5 and 20% w/w, heated in a microwave oven.

to the limited processing time. So, a convincing explanation is still needed and more work is required on this.

Conventional heating. Fig. 3 displays the temperature profiles recorded for a series of starch/water dispersions, in the range 2.5–20% w/w solids content, under conventional heating at 260 rpm throughout. The almost constant (among concentrations) initial heating rate below 70 °C is reduced at temperatures above 70 °C to an extent depending on solids content. The inverse phenomenon from that in Fig. 2 takes place, i.e. the temperature rises faster in the dispersions with lower water content. This agrees again with the temperature profiles obtained by Goebel et al. (1984) under conventional heating. The different trends in Figs. 2 and 3 clearly show that the mechanism of heat transport in conventional heating is different to microwave heating. In the former, heat is transported chiefly by the convective currents imparted by agitation and that is why heat diffuses faster in the low viscosity dilute suspensions. It must be noted that for dispersions with 15 and 20% solids, heat diffusion is so slow after the onset of gelatinization that the material in contact with the heating element burnt and the experiments had to be terminated much earlier. In conventional heating, the role of the mixing rate is crucial regarding heat transport. Fig. 4 compares the temperature profiles obtained with three different final mixing rates, namely 260, 160 and 40 rpm, for a dispersion with 10% solids. While the curves for 260 and 160 rpm increase above 70 °C at comparable rates, this is different for the 40 rpm curve which exhibits a plateau indicating a roughly isothermal situation. The same qualitative behavior was observed for all dispersions with less than 10% starch content.

3.3. Conductance measurements

Fig. 5 displays the apparent conductance K_{app} of various starch/water suspensions versus the corresponding volume–average temperature under microwave heating. The curve for pure water is included for comparison. The conductance curves are quite smooth; some low noise level is attributed to averaging unsmoothed instantaneous temperature values from the two thermocouples. At higher starch concentrations the initial K_{app} (at 60 °C) is lower due to the smaller water fraction in the suspension. Upon heating the conductance curves tend to rise

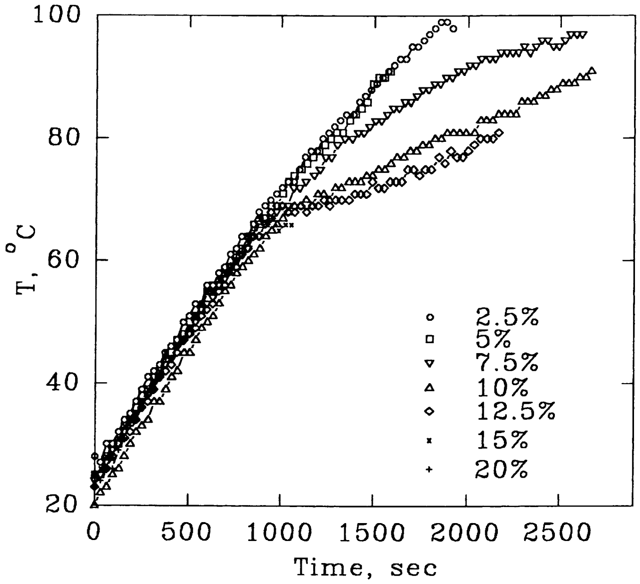

Fig. 3. Temperature versus time for starch/water mixtures with solids concentration of 2.5–20% w/w, heated by a conventional local electrical heater. For the presented runs mixing was performed at 260 rpm throughout.

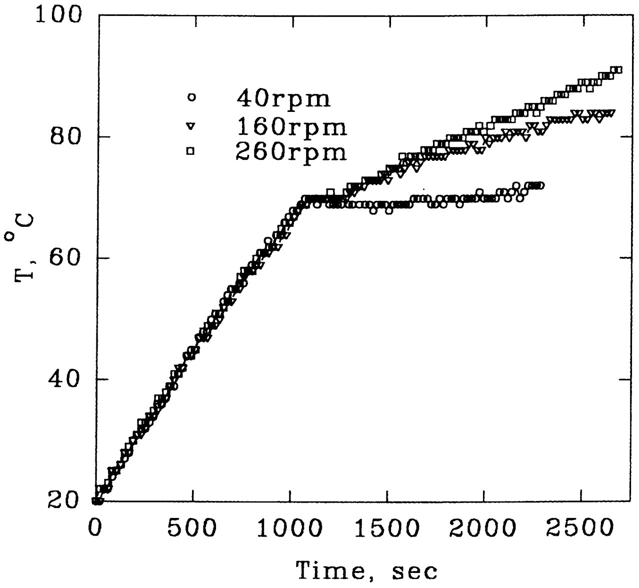

Fig. 4. Temperature versus time for starch/water mixtures with solids concentration of 10% w/w, heated in a microwave oven. Mixer speed initially at 260 rpm and at 70° C adjusted to final values of 40, 160 and 260 rpm.

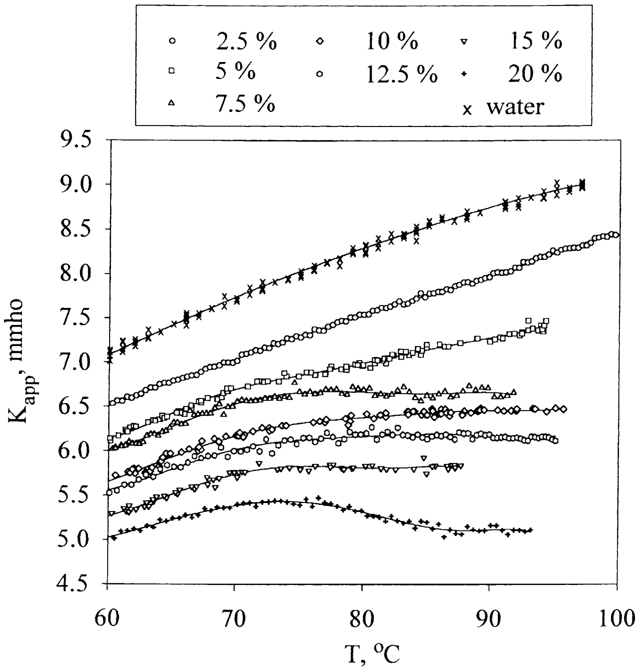

Fig. 5. Apparent conductance K_{app}, versus average-temperature for starch/water mixtures of various solids concentration heated in a microwave oven.

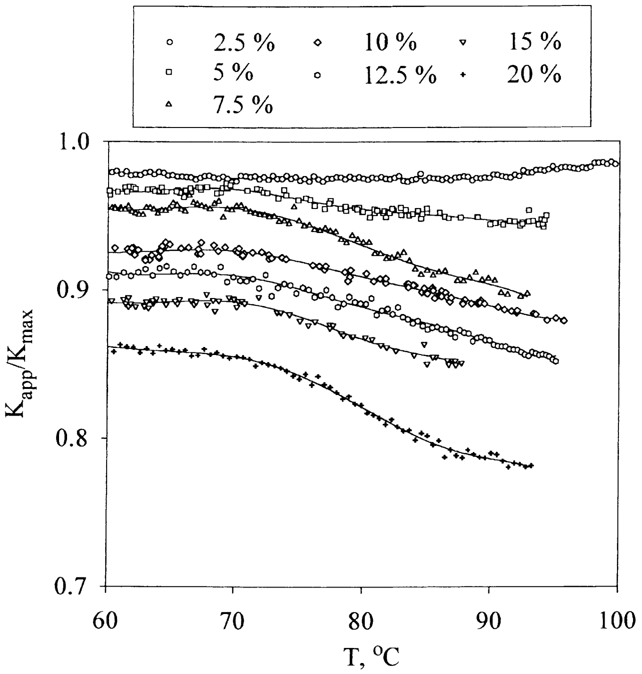

Fig. 6. Reduced apparent conductance, K_{app}/K_{max}, versus average-temperature for starch/water mixtures with solids concentration of 2.5–20% w/w, heated in a microwave oven.

according to the temperature dependence of water conductivity. Karapantsios et al. (2000), performing conventional heating experiments, observed that around 70 °C the starch/water suspensions attain a maximum electrical accessibility to free water (corresponding approximately to a minimum viscosity). At temperatures higher than 70 °C, as the samples progressively gelatinize, part of their water becomes gradually immobilized by the swelling granules and gets less and less electrically accessible. Similar trends are also observed under microwave heating. Indeed, in Fig. 5 the conductance curves for concentrations above 2.5% reduces their ascending slope above ~70 °C and even shifts toward lower values for 20% solids. The above behavior is in line with ideas advanced by Callaghan, Jolley, Lelievre, and Wong (1983) who argued about the different interaction of free water molecules with starch granules within two distinct regions in the paste: a mobile liquid-like and an immobile crystalline one (at stages where crystallinity is not fully lost due to gelatinization). These ideas were further supported by the gelatinization experiments of Goebel et al. (1984).

Fig. 6 presents the normalized apparent conductance of the mixture, K_{app}/K_{max}, versus the volume–average temperature. These curves reflect better the drastic change in the physical condition of the sample upon gelatinization discarding the temperature effect on electrical conductance. In case of a mixture with 2.5% solids the curve is practically horizontal showing that for such high availability of water, electrical conductance is not sensitive enough to capture the progress of gelatinization. At such low solids concentration starch does not swell sufficiently to allow the granules to close pack. In this case most of the water probably stays outside the swollen granules particularly if gelatinization is not complete (see below). The small increase in conductance at T > ~90 °C is possibly due to convection within the test vessel for a such low viscosity sample which brings hotter masses inside the measuring volume of the ring electrodes. For higher concentrations and temperatures above ~70 °C the curves decline towards lower values. Therefore, electrical conductance measurements give a good picture of water availability and consequently of the progress of gelatinization. This was also the case for the experiments of Karapantsios et al. (2000) under conventional heating.

The aforementioned authors found that whenever their relative conductance signal reached a plateau (at the end of the heating-up period), gelatinization was completed. According to this, the microwave heated suspensions of this study did not complete gelatinization since the respective K_{app}/K_{max} curves exhibit a declining trend, instead of a plateau, at the end of the experiments. Fig. 7 compares the normalized conductance traces obtained under microwave and conventional heating for a suspension of 10% solids. While the curve corresponding to conventional heating reaches a final plateau value, the curve corresponding to microwave heating does not do so although it reaches even higher temperatures. That is, the sample is fully gelatinized under conventional heating but not under microwave heating. It appears, therefore, that gelatinization is more than a temperature or energy absorption phenomenon.

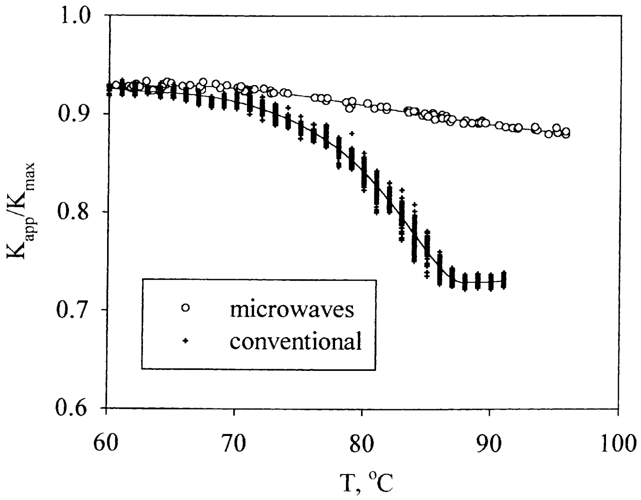

Fig. 7. Reduced apparent conductance, $K_{\text{app}}/K_{\text{max}}$, versus average-temperature for starch/water mixtures with solids concentration of 10% w/w, heated in a microwave oven and by a conventional local electrical heater, respectively.

3.4. Degree of gelatinization

To shed more light on the above comparisons, samples taken at the end of experiments from both heating schemes were examined with the classical methods of polarizing microscopy, application of Congo red dye and occasionally by DSC analysis. Repeated tests demonstrated that whenever a plateau was reached under conventional heating, gelatinization was always completed. However, in the case of microwave irradiated samples where a plateau was never reached gelatinization was not fully concluded.

Observations of incomplete gelatinization under microwave heating were also made in the past, e.g. Goebel et al. (1984) and Zylema et al. (1985), and were attributed to the limited starch–water interaction during the short microwave heating period. Inspection of Figs. 1–4 reveals that while for the conventional heating the sample needs approximately half an hour to go from ~60 °C to the end of the experiment, 2–3 min were enough for microwave heating. Thus, although microwaves bring the sample to equally high temperatures with conventional heating, the short time involved hinders the full disruption of starch granules by water molecules. Such time-dependent phenomena over and above any temperature or energy barriers, indicate the existence of a mass-transfer controlling step in the gelatinization process. This notion was further supported by a few tests where microwave processed samples examined one hour after the termination of the treatment showed that gelatinization was completed.

The major differences between the microwave and conventional heating schemes was the duration of the processing time and agitation. Karapantsios et al. (2000) found that in cases of poor agitation the conductance curves at the end of the conventional heating experiments showed a gradual decline instead of a plateau. This was assumed to reflect slow water diffusion phenomena from the bulk to the starch granules. By introducing a better agitation scheme (different mixer configuration) these authors managed to completely gelatinize their samples. However in their case, as it is also demonstrated in Fig. 4, the agitation influenced both heat and mass transfer within the sample volume and a distinction could not be made between these two effects. Microwave heating made it possible to overcome the heat transfer problem without agitating the sample and so illustrated that mass transfer limitations may be the rate-controlling step in gelatinization.

4. Conclusions

Specially constructed thermocouples located at two radial positions across a cylindrical test vessel provided information on the heating pattern of maize starch suspensions under microwave irradiation. Temperature profiles under microwave heating were compared with temperature profiles obtained under a conventional (electrical) heating method. In the latter the role of mixing on heat and mass transport was investigated. A recently reported electrical conductance technique was successfully employed to assess on-line the extent of gelatinization during processing. The geometrical design of the conductance probe was such that could provide information on a relatively large volume of the sample averaging local temperature non-uniformities. Despite the high temperatures achieved by microwave irradiation, gelatinization was never complete in the short microwave processing times. A comparison between electrical conductance records acquired under microwave and under conventional heating showed that mass transfer dominates starch gelatinization rather than heat transfer.

References

Buffler, C. R. (1993). *Microwave cooking and processing*. Reinhold: Van Nostrand.

Callaghan, P. T., Jolley, K. W., Lelievre, J., & Wong, R. B. K. (1983). Nuclear magnetic resonance studies of wheat starch pastes. *Journal of Colloid and Interface Science*, *92*, 332–337.

Carbonell, R. G., & Whitaker, S. (1984). Heat and mass transfer in porous media. In J. Bear, & Y. Corapcioglu (Eds.), *Fundamentals of transport phenomena in porous media* (pp. 121–198). The Hague: Nijhoff.

Chen, S. C., Collins, J. L., Mccarty, I. E., & Johnston, M. R. (1971). Blanching of white potatoes by microwave energy followed by boiling water. *Journal of Food Science*, *36*, 742–743.

Chen, D.-S. D., Singh, R. K., Hagghighi, K., & Nelson, Ph. E. (1993). Finite element analysis of temperature distribution in microwaved cylindrical potato tissue. *Journal of Food Engineering*, *18*, 351–368.

Collins, J. L., & Mccarty, I. E. (1969). Comparison of microwave energy with boiling water for blanching whole potatoes. *Food Technology*, *23*, 337–340.

Fakhouri, M. O., & Ramaswamy, H. S. (1993). Temperature uniformity of microwave heated foods as influenced by product type and composition. *Food Research International*, *26*, 89–95.

Fleischman, G. J. (1996). Predicting temperature range in food slabs undergoing long term/low power microwave heating. *Journal of Food Engineering*, *27*, 337–351.

Goebel, N. K., Grider, J., Davis, E. A., & Gordon, J. (1984). The effects of microwave energy and convection heating on wheat starch granule transformations. *Food Microstructure*, *3*, 73–82.

Hardman, M. T. (1985). In Birch G.G., & Lindley M.G. (Eds.), *Interaction of food components* (pp. 31–74). New York: Elsevier.

Huang, J., Hess, W. M., Weber, D. J., Purcell, A. E., & Hube, C. S. (1990). Scanning electron microscopy: tissue characteristics and starch granule variations of potatoes after microwave and conductive heating. *Food Structure*, *9*, 113–122.

Karapantsios, T. D., Sakonidou, E. P., & Raphaelides, S. N. (2000). Electrical conductance study of fluid motion during starch gelatinization. *Journal of Food Science*, *65*(1), 144–150.

Khraisheh, M. A., Cooper, T. J. R., & Magee, T. R. A. (1997). Microwave and air drying I. Fundamental considerations and assumptions for the simplified thermal calculations of volumetric power absorption. *Journal of Food Engineering*, *33*, 207–219.

Kingston, H. M., & Jassie, L. B. (1986). Microwave energy for acid decomposition at elevated temperatures and pressures using biological and botanical samples. *Analytical Chemistry*, *58*, 2534–2541.

Kirk, D., & Holmes, A. W. (1975). The heating of foodstuffs in a microwave oven. *Journal of Food Technology*, *10*, 375–384.

Knutson, K. M., Mart, E. H., & Wagner, M. K. (1987). Microwave heating of food. *Lebensmittel Wissenschaft und Technologie*, *20*(3), 101–110.

Lin, Y. E., Anantheswaran, R. C., & Puri, V. M. (1995). Finite element analysis of microwave heating of solid foods. *Journal of Food Engineering*, *25*, 85–112.

Miller, L. A., Gordon, J., & Davis, E. A. (1991). Dielectric and thermal transition properties of chemically modified starches during heating. *Cereal Chemistry*, *68*(5), 441–448.

Mudgett, R. E. (1989). Microwave food processing. *Food Technology*, *43*, 117–126.

Ndife, M., Sumnu, G., & Bayndirli, L. (1998). Differential scanning calorimetry determination of gelatinization rates in different starches due to microwave heating. *Lebensmittel Wissenschaft und Technologie*, *31*, 484–488.

Nelson, S. O. (1973). Electrical properties of agricultural products—a critical review. *Transactions of the ASAE*, *16*(3), 384–400.

Noel, T. R., & Ring, S. G. (1992). A study of the heat capacity of starch/water mixtures. *Carbohydrate Research*, *227*, 203–213.

Ohlsson, T., & Risman, P. O. (1978). Temperature distribution of microwave heating-spheres and cylinders. *Journal of Microwave Power*, *13*(4), 303–310.

Ramaswamy, H. S., & Pillet-Will, T. (1992). Temperature distribution in microwave-heated food models. *Journal of Food Quality*, *15*, 435–448.

Ramaswamy, H., Van De Voort, F. R., Haghava, G. S. V., Lightfoot, D., & Timbers, G. (1991). Feed-back temperature control system for microwave ovens using a shielded thermocouple. *Journal of Food Science*, *56*(2), 550–555.

Randa, J. (1990). Theoretical considerations for a thermo-optic electric field strength probe. *Journal of Microwave Power*, *25*, 133–140.

Ryynänen, S., Risman, P. O., & Ohlsson, T. (1996). The dielectric properties of native starch solutions—a research note. *Journal of Microwave Power and Electromagnetic Energy*, *31*(1), 50–52.

Slade, L., & Levine, H. (1991). Beyond water activity: Recent advances based on an alternative approach to the assessment of food quality and safety. *Critical Reviews in Food Science and Nutrition*, *30*(2/3), 115–360.

Vilayannur, R. S., Puri, V. M., & Anantheswaran, R. C. (1998a). Size and shape effect on nonuniformity of temperature and moisture distributions in microwave heated food materials: Part I simulation. *Journal of Food Engineering*, *21*, 209–233.

Vilayannur, R. S., Puri, V. M., & Anantheswaran, R. C. (1998b). Size and shape effect on nonuniformity of temperature and moisture distributions in microwave heated food materials: Part II. *Journal of Food Engineering*, *21*, 235–248.

Wickersheim, K., Sun, M., & Kamal, A. (1990). A small microwave E-field probe utilizing fiberoptic thermometry. *Journal of Microwave Power*, *25*(3), 141–148.

Zhou, L., Puri, V. M., Anantheswaran, R. C., & Yeh, G. (1995). Finite element modeling of heat and mass transfer in food materials during microwave heating—model development and validation. *Journal of Food Engineering*, *25*, 509–529.

Zylema, B. J., Grider, J. A., Gordon, J., & Davis, E. A. (1985). Model wheat starch systems heated by microwave irradiation and conduction with equalized heating times. *Cereal Chemistry*, *62*(6), 447–453.