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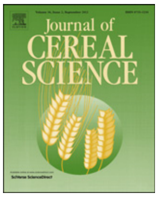

Review

Key issues and challenges in whole wheat flour milling and storage

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ABSTRACT

Whole wheat flour is increasingly popular as research continues to reveal the benefits of whole grains and the food industry offers more whole grain options for consumers. The purpose of this review is to address milling and shelf-life issues that are unique to whole wheat flour. No standard methods are available for whole wheat flour milling, resulting in very different bran particle sizes. Literature suggests that moderate bran particle size is the best for bread production, while small particle size is better for non-gluten applications. Shelf-life of whole wheat flour is shorter compared to white flour due to the presence of lipids and lipid-degrading enzymes. Lipolytic degradation leads to reduction in functionality, palatability and nutritional properties. Strategies to stabilize whole wheat flour have focused on controlling lipolytic enzyme activity and have marginally succeeded.

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1. Introduction

AACC International has defined whole wheat flour as being prepared from wheat (other than durum) such that the proportions of the intact grain – the bran, germ, and endosperm – remain unaltered (AACC International, 1999). Whole wheat flour contains substantially more vitamins, minerals, antioxidants and other nutrients than regular wheat flour, since these compounds are concentrated in the outer portions of the grain (Weaver, 2001). Some of these nutrients are replaced in the enrichment process of wheat flour, which is mandatory in 64 countries around the world (Flour Fortification Initiative, 2012), although many nutritional components are still lower, especially minerals and dietary fiber.

With the advent of modern roller mills during the industrial revolution, whole wheat flour production all but disappeared during much of the twentieth century. In the US, whole wheat flour production was about 2% of total wheat flour production in 2000 (Vocke et al., 2008) and only about 7% of the population consumed at least 3 servings of whole grains per day [US Department of Agriculture (USDA, 2000).

Food companies worldwide have responded to the mounting evidence supporting the benefits of whole grains with a 1960% increase in whole grain product launches in 2011 compared with 2000 (Mintel, 2011). In the US, the increase in whole grain food production nearly tripled whole wheat flour production from 2002 to 2011: 3.13×10^8 kg (6.91×10^6 cwts) in 2002–2003 compared with 9.33×10^8 kg (2.05×10^7 cwts) in 2010–2011 (1.8% and 5% of total wheat flour production, respectively; Sosland, 2011).

Whole wheat flour possesses several unique challenges to the milling and baking industries. For instance, whereas milling procedures for traditional flours have been well-established, whole grain flours are produced by a variety of techniques and result in flours with widely different particle sizes and functionalities (Kihlberg et al., 2004). Furthermore, whole wheat flour contains more enzymatic activity (Every et al., 2006a), lipids (Chung et al., 2009), and antioxidants (Adom et al., 2005) than wheat flour, which can affect end-use (Every et al., 2006a; Galliard and Gallagher, 1988; Tait and Galliard, 1988; Wang et al., 2004) and storage properties (Bell et al., 1979; Galliard, 1986a, 1986b, 1994; Hansen and Rose, 1996; Tait and Galliard, 1988). The purpose of this review is to address these key issues – milling and shelf-life – and discuss strategies to overcome new challenges relative to increased whole wheat flour production.

2. Whole wheat flour milling

2.1. Wheat selection

Wheat kernel physical characteristics, such as uniformity in kernel hardness and size, are important for milling traditional wheat

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flour because they maximize separation of the bran from the endosperm during roller milling (Li and Posner, 1987; Posner and Hibbs, 2005). These parameters may not be important for milling whole wheat flour, since separation of kernel components is not the goal.

Bran color is one kernel attribute that is not often considered when milling wheat flour, but has a substantial impact on whole wheat flour. Wheat kernel pericarp color can vary from white to black or from red to blue, although most commercial wheats are classified as red or white. Most wheat produced in the US is red wheat; therefore, most flour, including whole wheat flour, is milled from red wheat. Although chemically similar (Table 1), whole grain flour produced from white wheats produces bread with a lighter color and less bitter flavor, which is generally favored by consumers, compared with red wheat flours (McGuire and Opalka, 1995). However, there are other factors to consider. For instance, in a sensory analysis of whole grain muffins made from white or red wheat, consumers perceived muffins made from red wheat as more healthy, even though nutritional composition of the two muffins was nearly identical (Camire et al., 2006).

On a genotypic level, selection of varieties of wheat for whole wheat flour production may pose some challenges. End-use quality attributes, such as water absorption and gluten strength, are an important part of wheat selection, although these analyses are typically performed only on wheat flour. Data from these tests may not accurately predict the performance of a variety of wheat in a whole wheat flour application. Bruckner et al. (2001) analyzed mixograph and baking properties in 11 winter and 12 spring wheat varieties grown in four locations using both wheat flour and whole grain flour. They found that, while correlations between flour and whole flour for many variables were significant, correlation coefficients varied widely. For instance, water absorption and loaf volume correlation coefficients ranged from 0.17 to 0.81 and from 0.08 to 0.72, respectively, depending on variety and crop year. Clearly the outer portions of the wheat kernel exert physical and chemical effects on dough properties that vary among different types of wheat (de Kock et al., 1999; Noort et al., 2010; Seyer and Gelinas, 2009).

Table 1
Composition of refined flour, white hard wheat and red hard wheat (USDA, 2011); nutrients that are added during enrichment (US standards) are in bold; other components are included: soluble and bound phenolics (Kim et al., 2006; Liyana-Pathirana and Shahidi, 2006), α-amylase activity (McCrate et al., 1981; McCleary et al., 2002), and water-soluble and enzyme-extractable pentosan (Delcour and Hoseney, 2010; Hong et al., 1989); * Ceralpha units, ** MDU, millidextrinizing units per gram per hour.

Nutrient	Unbleached, enriched, all-purpose wheat flour	Wheat, hard white	Wheat, hard red winter
Macronutrients			
Protein (g)	11.7	11.31	12.61
Fat (g)	1.11	1.71	1.54
Ash (g)	0.534	1.52	1.57
Dietary fiber (g)	3.07	12.2	12.2
Minerals			
Calcium (mg)	17.0	32.0	29.0
Iron (mg)	5.27	4.56	3.19
Magnesium (mg)	25.0	93	126
Phosphorus (mg)	123	355	288
Potassium (mg)	121	432	363
Sodium (mg)	2.27	2.00	2.00
Zinc (mg)	0.795	3.33	2.65
Copper (mg)	0.163	0.363	0.434
Manganese (mg)	0.774	3.821	3.985
Selenium (μg)	38.5	–	70.7
Vitamins			
Vitamin C (mg)	0	0	0
Thiamin (mg)	0.891	0.387	0.383
Riboflavin (mg)	0.561	0.108	0.115
Niacin (mg)	6.70	4.381	5.464
Pantothenic acid (mg)	0.497	0.954	0.954
Vitamin B-6 (mg)	0.050	0.368	0.3
Folate (μg)	207.8	38.0	38.0
Vitamin B-12 (μg)	0	0	0
Vitamin A (IU)	2.27	0	0
Vitamin E (mg)	0.261	1.01	1.01
Vitamin D (IU)	0	0	0
Vitamin K (μg)	0.341	1.9	1.9
Other			
Extractable phenolics (mg/g)	0.354–0.388	0.451–0.566	0.554–0.655
Total phenolics (mg/g)	0.699	3.338–3.807	3.827–3.973
α-Amylase activity	0.10–0.14 units*/g	47–57 MDU**	34–47 MDU**
Water-soluble arabinoxylan (%)	0.4–0.8	0.568	0.648–0.709
Total arabinoxylan (%)	1.5–2.5	4.839	4.79–6.92

Because the outer portions of the wheat kernel affect baking quality by both physical and chemical means, quantifying these attributes may be important in selecting wheat varieties that are most appropriate for whole grain baking. For instance, bran friability, its ability to be reduced to small particle sizes, varies among cultivars (Greffeuille et al., 2006) and cultivars with low friability produce higher quality bread (Seyer and Gelinas, 2009). The outer portions of the kernel also contain various chemical compounds and enzymes that can affect baking properties (Joye et al., 2009), such as glutathione (Every et al., 2006b), phytate (Lehrfeld and Wu, 1991), ferulic acid (Adom et al., 2005), and lipoxygenase (Every et al., 2006a). Because these constituents are concentrated in the outer portions of the wheat grain, a given wheat variety may be acceptable for use in traditional baking but may adversely affect whole wheat baking.

Consumers of whole grain products are generally more health conscious than those that do not consume whole grain products. Therefore, it may be worthwhile to select wheat genotypes for whole grain applications based on desirable nutritional properties such as phytochemical and dietary fiber content (Gebruers et al., 2010; Ward et al., 2008). This is already done for some other grains, such as oats, where millers often select particular varieties of grain with high β-glucan content to support health-claims for heart health and cholesterol reduction.

2.2. *Milling process*

Perhaps the most important consideration in producing whole grain flour is selecting the milling process that will be used. Indeed, milling technique may have a greater impact on whole wheat bread quality than the quality of wheat used for producing the flour or the formulation of the bread itself (Kihlberg et al., 2004). The two predominant techniques for grinding whole grain flours are stone and roller mills. Whole grain flours could also notionally be produced with an impact or hammer mill but this is rarely used (Kent and Evers, 1994).

2.2.1. *Stone milling*

Stone mills are the oldest attrition mills used for making whole grain flours, which simultaneously use compression, shear, and abrasion to grind wheat kernels between two stones and produce a theoretical extraction rate of 100% (Kihlberg et al., 2004). Modern stone mills are metal plates with composition stones attached (Posner and Hibbs, 2005).

Stone mills generate considerable heat due to friction. This can result in considerable damage to starch, protein, and unsaturated fatty acids in comparison with other milling techniques (Prabhasankar and Rao, 2001). Furthermore, in large, continuous milling operations, heat generated from stone milling can pose a fire risk.

Interestingly, there appears to be a marketing advantage by using the term "stone ground" with consumers, as evidenced by the preponderance of whole wheat flour products making this claim in both retail and commercial markets (Posner and Hibbs, 2005). Thus, some mills will "crack" the grain using a stone mill with the plates situated far enough apart to not generate excessive heat. Additional capital costs may be required to equip existing mills with such a set up. The cracked wheat is then reduced to flour on a roller mill.

2.2.2. Roller milling

The process of roller milling involves separation of the endosperm from the bran and germ followed by gradual size reduction of endosperm (Ziegler and Greer, 1971). In this process, wheat is passed through a series of corrugated and smooth rollers accompanied by sifting between stages. Producing flour that fulfills the requirement for being whole grain is achieved by blending bran and germ back with the endosperm flour in the naturally-occurring proportions. Feeding the bran and germ milling streams with the endosperm flour stream is most often achieved in a continuous process, rather than collecting all fractions in separate bins and recombining at the end of milling. In this case, production of whole wheat flour would not involve additional capital expense beyond what is required for regular roller milling. Sometimes whole wheat flour is made by physically separating flour millstreams and then recombining at the end of the milling process. This is usually done when the bran will undergo some post-milling such as ultra fine grinding or heating. In these cases, capital costs would be required for the post-milling, plus equipment for recombining the fractions.

When producing whole wheat flour on roller mills, a number of conditions are different from those used for wheat flour (Kent and Evers, 1994). First, conditioning (tempering) is less important when milling whole wheat flour. While wheat flour relies on proper conditioning to facilitate endosperm and bran separation, this is not required for whole wheat milling. Thus, in theory no conditioning should be required, although many mills will add 1–2% moisture to soften the grain and improve efficiency in terms of the energy required to produce the flour. Efficiency can also be improved by tightening the roll gap and using more open scalp covers to increase the break release, as well as changing some of the smooth rolls to corrugated during reduction. The purifier air valves should also be adjusted so that the bran and germ are not rejected but are returned to the reduction system (Kent and Evers, 1994).

There are several noteworthy advantages of making whole grain flour from roller mills as opposed to stone mills. First, the amount of grinding and reduction at each roll can be adjusted to accommodate variations in raw materials, which makes roller milling both economical and flexible (Posner and Hibbs, 2005). Second, the use of selective corrugations and differential speeds subjects the endosperm fraction to minimal shear and compressive forces during the grinding and reduction, which allows less heat to build on reduction rolls and results in less destruction to chemical components in the flour (Prabhasankar and Rao, 2001). A third advantage of making whole grain flours from roller mills is that wheat bran and germ can be separated from the endosperm fraction and subjected to further processing such as heating or fine grinding to affect the storage or functional properties of the flour (Posner and Hibbs, 2005).

2.2.3. Particle size

Differences in whole wheat milling practices are evident in a survey of 5 national brands of whole wheat flour (Fig. 1). As shown, 43% of whole wheat flour from brand 1 passed through a sieve with a 0.230 mm opening, while <6% passed through this sieve in the other brands; brand 2 contained >10% of particles >0.841 mm, while the other brands had 1% or less of this particle size.

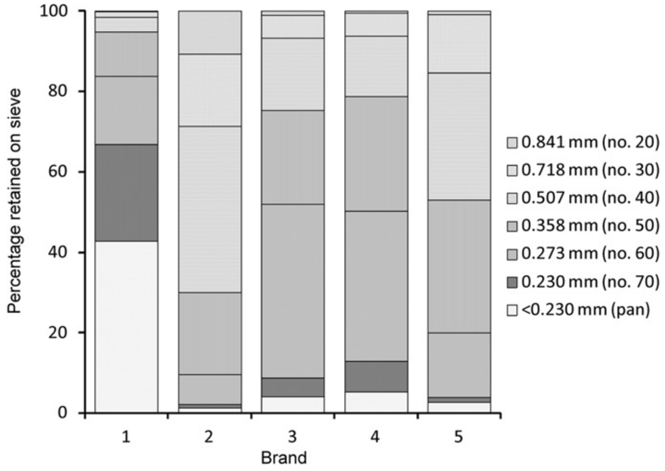

Fig. 1. Particle size distribution of five brands of whole wheat flour obtained from a local market. From the packaging for each flour: brand 1 was "All natural premium whole wheat flour"; brand 2 was "Old-fashioned 100% stone ground all natural whole wheat flour"; brand 3 was "All natural whole wheat flour"; brand 4 was "100% stone ground whole wheat flour"; and brand 5 was "Premium 100% whole wheat flour". Flour (100 g) was separated on a sieve shaker (Model SS-15, Gilson Company, Lewis Center, OH) for 15 min and then the weight retained on each sieve was recorded according to the American Society for Testing and Materials standard methods (Pope and Ward, 1998); values represent the average of three replications.

Particle size of the bran fraction in whole wheat flour has an amazing influence on functional properties of the flour. In general, large wheat bran particles (mean particle size of more than about 500 μm) lead to higher water absorption (Anderson and Eastwood, 1987; Mongeau and Brassard, 1982; Robertson and Eastwood, 1981) and loaf volume (de Kock et al., 1999; Galliard and Gallagher, 1988; Zhang and Moore, 1999) compared with finer bran particle sizes (mean particle size less than about 500 μm). However, if bran particles are too coarse (>600 μm), bread possesses a rough crust appearance and gritty texture (Zhang and Moore, 1999). Small particles have a greater negative impact on bread quality because chemical components in the bran can interact more readily with gluten and inhibit development (Noort et al., 2010). However, from a nutritional standpoint, smaller particles could help in the release of vitamins and other components from the outer cells of the kernel (Kahlon et al., 1986). Thus, a moderate particle size (mean bran particle size of about 400–500 μm) may be the most desirable in whole wheat flour for bread production.

Products that do not require gluten development may have different particle size requirements compared with those that do. In an evaluation of 69 soft wheat cultivars for whole wheat cookie baking quality, cookie spread was influenced by whole wheat flour particle size (Gaines and Donelson, 1985). Small particles produced large cookies (more spread), while larger bran particles produced smaller cookies (less spread). Furthermore, bran particle size influences cake quality. For instance, cakes produced with up to 36% (flour weight) wheat bran of different particle sizes (50, 80, 250 μm) showed the greatest increase in firmness, chewiness, and yellowness when more coarse particle sizes were used. In a sensory evaluation, these changes were not favored by consumers; the best sensory acceptability was reached when finer particle sizes were used (Gomez et al., 2010).

3. Whole wheat flour storage

While not enough data exist to suggest a definitive shelf-life for whole wheat flour, it is well accepted that the shelf-life of whole wheat flour is considerably shorter than regular wheat flour. Flour millers stamp use-by dates of 3–9 months after milling on whole wheat flour packages, while regular wheat flour use-by dates range from 9 to 15 months after milling. Although these dates can be helpful, actual shelf-life could be shorter or longer depending on temperature and humidity during storage and on failure endpoints (i.e., the point at which the flour is deemed unacceptable as determined by the company or the experimenter).

Whole grain flour storage is accompanied by a cascade of biochemical changes that lead to reduced flour functionality. The most unstable components in whole wheat flour are the lipids (Pomeranz, 1988). Lipid degradation is the predominant cause of the loss in flour functionality during whole wheat flour storage. Indeed, Tait and Galliard (1988) demonstrated that exchanging the lipids in fresh and stored whole wheat flour could completely account for the changes in flour functionality as a result of storage. Therefore, this section will emphasize whole wheat flour lipid degradation, with brief discussions on changes in other flour components.

3.1. *Lipid degradation during whole wheat flour storage*

Despite being a minor constituent of wheat flour, endogenous lipids contribute substantially to flour functionality. Bread made from defatted flour is inferior to bread with endogenous lipids, even when shortening is added during the mixing process (Bell et al., 1979; Moore and Hoseney, 1986). Upon mixing flour with water, lipids cannot be extracted from dough with common solvents due to binding with gluten proteins, which is essential for proper gluten development (Goesaert et al., 2005).

Lipids begin to break down in whole wheat flour by hydrolytic rancidity, which can be followed by oxidative rancidity. These changes can occur enzymically or non-enzymically and affect flour quality (Fig. 2).

3.1.1. *Hydrolytic rancidity*

Hydrolytic rancidity in whole wheat flour proceeds through the action of lipase (O'Connor et al., 1992). Lipase (EC 3.1.1.3) hydrolyzes triacylglycerols to non-esterified fatty acids and diglycerides, monoglycerides, and eventually glycerol; thus the release of non-esterified fatty acids in whole wheat flour is related to lipase activity. Wheat lipase activity is mostly located in the bran fraction of the grain (Galliard, 1986a). There is an enzyme termed 'wheat germ lipase' that catalyzes deesterification of triacetin and other artificial water-soluble substrates (O'Connor et al., 1992) and thus is technically an esterase. True lipase activity (i.e., activity on water-insoluble substrates) of wheat germ lipase is likely a result of contamination with lipase from wheat bran (Galliard, 1994). Lipase exhibits maximum activity in wheat at about 17% moisture content; however, at moisture contents commonly observed in flour during storage (10–14%), lipase activity continues at about 50% of maximum (Fig. 3). This property makes lipase unique among hydrolytic enzymes, i.e., lipase only requires a catalytic amount of water to act, whereas excessive amounts of water protect the lipid from being exposed to the catalytic site of the enzyme and reduce activity (Galliard, 1994).

Hydrolytic rancidity in whole wheat flour can lead to a decrease in sensory quality (Hansen and Rose, 1996) and functional properties of whole grain flour (Galliard, 1994; Pomeranz, 1988; Tait and Galliard, 1988). Weekly sensory evaluation of whole wheat flour for 11 weeks demonstrated that hydrolytic rancidity was inversely related to the acceptability of bread made from these flours (Hansen and Rose, 1996). Whole wheat flour with a high content of non-esterified fatty acids has been described as musty, bitter, and rancid (Heinio et al., 2002).

Products of hydrolytic rancidity have an effect on baking quality (Bell et al., 1979; Tait and Galliard, 1988). At low concentrations, non-esterified polyunsaturated fatty acids have a positive effect on loaf volume through co-oxidation of gluten protein sulfhydryl groups during mixing. However, at high concentrations non-esterified polyunsaturated fatty acids affect dough mixing by reducing lipid binding capacity of gluten. This reduces gas holding capacity and elasticity of gluten (Carr et al., 1992; Miller and Kummerow, 1948). Interestingly, saturated fatty acids seem to have no effect on dough and baking properties (Bell et al., 1979). In addition to direct effects on bread quality, non-esterified polyunsaturated fatty acids are substrates for lipoxygenase, an enzyme that generates oxidation products that decrease the quality and acceptability of whole wheat flour (Loiseau et al., 2001).

3.1.2. *Oxidative rancidity*

Lipids can be oxidized in whole wheat flour enzymically (Brash, 1999; Galliard, 1986a, 1986b) or through autoxidation (Robards and Kerr, 1988). Enzymic lipid oxidation occurs through the action of lipoxygenase (EC 1.13.11.12). Lipoxygenase in wheat is located in the germ and bran of the grain (Galliard, 1994; Loiseau et al., 2001). It consists of a group of isozymes with a molecular mass of ~110 kDa and optimal activity at pH between 4.5 and 6.0 (Loiseau et al.,

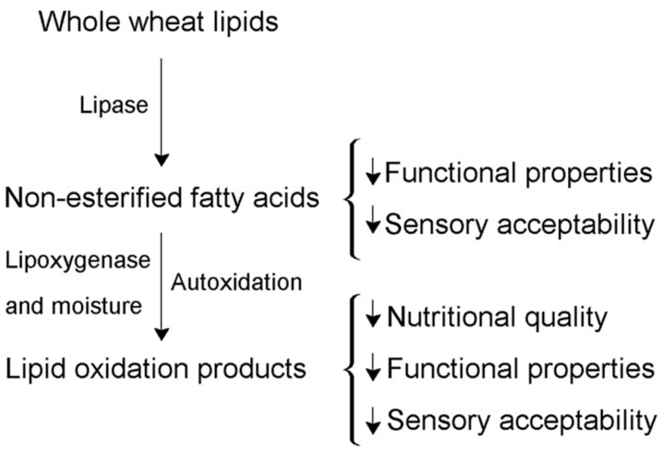

Fig. 2. Causes and consequences of lipid degradation in whole wheat flour during storage.

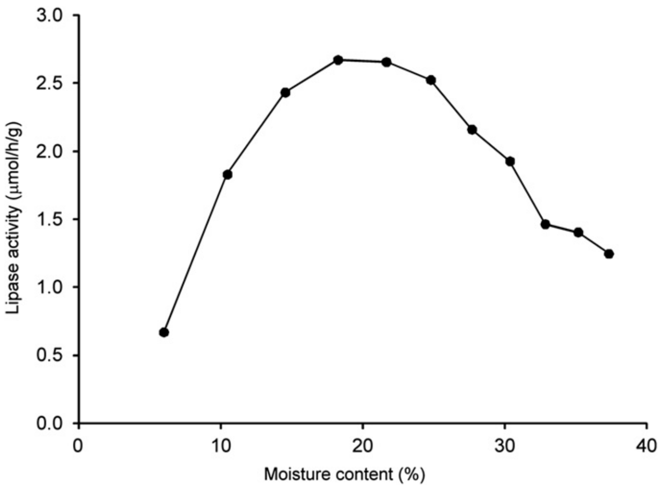

Fig. 3. Lipase activity as a function of wheat bran moisture content; reprinted with permission from Rose and Pike (2006); copyright 2006 Springer.

2001). Lipoxygenase attacks the methylene group between two double bonds in polyunsaturated fatty acids, preferentially non-esterified polyunsaturated fatty acids (Galliard, 1986a; Morrison and Panpaprai, 1975). Autoxidation can occur by non-enzymic reaction of grain lipids with atmospheric oxygen. Under both mechanisms, lipid oxidation involves addition of oxygen to polyunsaturated fatty acids, forming hydroperoxides (Loiseau et al., 2001), followed by fissure of the carbon chain into smaller, volatile compounds (e.g., epoxyaldehydes, ketones, lactones, furans; McWilliams, 2005; Robards and Kerr, 1988).

Lipid oxidation during storage of whole wheat flour is a much slower process than lipid hydrolysis (Galliard, 1994). This is because, unlike lipase, lipoxygenase exhibits very little activity at moisture contents typically found during storage (Wang and Toledo, 1987), and because whole wheat flour contains high levels of protective antioxidants (Adom et al., 2005).

Despite being a slower process than lipid hydrolysis during storage, lipid oxidation can contribute substantially to loss of product quality. While minimally active in dry flour, lipoxygenase becomes active when stored flour is mixed with water and rapidly oxidizes non-esterified fatty acids present in the flour from the action of lipase (Galliard, 1986b). This is evident in Fig. 4, which shows that whole wheat flour containing high non-esterified fatty acid content exhibits high oxygen consumption from lipid oxidation when the flour is mixed with water.

Oxidation of lipids can lead to a decrease in nutritional quality and consumer acceptability of whole wheat flour and whole wheat flour-based products. Lipid oxidation reduces nutritional quality through loss of essential fatty acids (Pokorny and Velisek, 1995), although, more significantly, reduced nutritional quality is affected through co-oxidation of other flour components. Free radicals that are generated can denature proteins (Warwick et al., 1979) and convert essential amino acids into unavailable derivatives (Pokorny and Velisek, 1995). Lipoxygenase activity also causes significant losses of carotenoids (Leenhardt et al., 2006) and vitamin E (Lehtinen et al., 2003). Consumer acceptability of whole wheat flour declines as a result of lipid oxidation (Galliard and Gallagher, 1988; Tait and Galliard, 1988), which can generate undesirable odor components that affect sensory acceptability of whole wheat flour-based products (Galliard and Gallagher, 1988; Heinio et al., 2002).

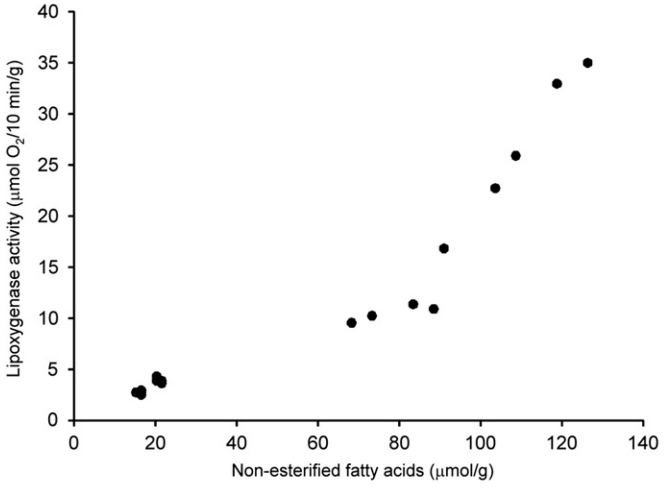

Fig. 4. Lipoxygenase activity (LOX) in aqueous suspensions of whole wheat flour as a function of non-esterified fatty acid (NEFA) content; for linear relationship: LOX = 0.238*NEFA − 2.15 ($R^2 = 0.87$; $p < 0.0001$); for quadratic relationship: LOX = 0.00365*NEFA^2 − 0.223*NEFA + 6.13 ($R^2 = 0.98$; $p < 0.0001$); figure produced from data in Galliard (1986b).

3.2. Protein degradation during whole wheat flour storage

Wheat proteins are unique due to their ability to form a viscoelastic dough. Wheat storage proteins (i.e., gliadin and glutenin) contain intra- and inter-molecular disulfide bonds that are important contributors to their functionality: gliadin is responsible for the cohesiveness of the dough and contains intra-molecular disulfide bonds, while glutenin provides elasticity to the dough and contains inter- and intra-molecular disulfide bonds (Veraverbeke and Delcour, 2002). Right after milling, flour proteins contain a high proportion of sulfhydryl groups and exhibit poor quality for bread making. Short-term (months) aging or chemical bleaching improves flour functionality through sulfhydryl/disulfide interchange among gluten proteins (mainly glutenin; Goesaert et al., 2005; Veraverbeke and Delcour, 2002).

During long-term storage, however, protein functionality is reduced. Wilkes and Copeland (2008) found an increase in wheat flour protein solubility over 270 d of storage at 30 °C. The most substantial increase was in the high molecular weight glutenin fraction. This may be a result of sulfhydryl/disulfide interchange with low molecular weight sulfhydryl compounds such as glutathione, which would decrease elasticity of dough. These effects may be more pronounced in whole wheat flour due to a higher glutathione content compared with wheat flour (Every et al., 2006b).

Changes in gluten functionality could also be a result of co-oxidation with lipids, due to close interactions between protein and lipid radicals (Dean et al., 1997). The properties of gluten are dependent on binding of lipid components from flour (Goesaert et al., 2005); thus degradation of the lipids in whole wheat flour may result in poor gluten development. In addition, lipid oxidation can convert lysine, cysteine, methionine, and tryptophan into unavailable derivatives (Pokorny and Velisek, 1995; Rehman and Shah, 1999).

3.3. Carbohydrate degradation during whole wheat flour storage

In wheat flour, resistance to stretching decreased in doughs made from three different flours stored for 24 months (Bell et al., 1979). This phenomenon could indicate modifications in protein, but also could be attributed to changes in the starch as a result of endogenous amylolytic activity (Rehman and Shah, 1999). Indeed, an increase in low molecular weight carbohydrates has been reported in whole wheat flour during storage (Marathe et al., 2002). Low molecular weight carbohydrates in dough increase bread crust coloration due to the Maillard reaction (Pomeranz, 1988). However, whole wheat bread made from flour or wheat stored for extremely long periods of time may exhibit more pale crust rather than a darker crust (unpublished observations while preparing bread for Rose et al., 2011). This may be due to reduction in amino acids available for the Maillard reaction. While this has not been shown conclusively, Rehman and Shah (1999) reported a reduction in total available lysine by 18% after storage of wheat at 25 °C for 6 months.

Non-esterified fatty acids may also modify starch characteristics during storage. Salman and Copeland (2007) reported an increase in final viscosity of wheat flours heated in a Rapid Visco Analyzer over 12 months of storage at 20 and 30 °C. Iodine binding and non-esterified fatty acid analyses suggested the formation of amylose-fatty acid complexes during storage.

3.4. Degradation of other components during whole wheat flour storage

Wennermark and Jägerstad (1992) demonstrated a decrease in vitamin E activity by 40% during the storage of whole wheat flour at 20 °C for 12 months. Nielsen and Hansen (2008) showed similar results: 32% decrease in vitamin E over 297 d of storage at room temperature. Decrease of vitamin E content is associated with lipid oxidation (Lehtinen et al., 2003; Nielsen and Hansen, 2008). Carotenoids also oxidize during flour storage as a result of lipid oxidation (Arya and Parihar, 1981; Farrington et al., 1981). There was a 7.2–11.5% reduction in thiamin during 12 months of storage of whole wheat flour under varying conditions (8–12% moisture content; 10–32 °C; 25–55% relative humidity; Franz, 1968).

3.5. Strategies to improve whole wheat flour storage stability

As the above discussion illustrates, lipid degradation in whole wheat flour during storage is the major contributor to loss of product quality. The rate of lipid degradation can be reduced by cold storage. Indeed, compared with 20 °C, storage of whole wheat flour at −20 °C for 20 weeks has been shown to prevent the deleterious changes in lipids that accompany loss in whole wheat flour functionality (Tait and Galliard, 1988). Unfortunately, refrigerated transport and storage is probably cost prohibitive for the flour industry.

Other strategies to stabilize lipids in foods have included modified atmosphere and addition of antioxidants. These strategies may be more cost effective, but would likely be minimally effective. The former would be less effective because degradation of whole wheat flour lipids begins with hydrolytic rancidity, which is enzymatic and does not require oxygen. For instance, over 6 months of storage of whole wheat flour, no differences in non-esterified fatty acid development were observed at 25 °C and 35 °C (Fig. 5). When the flour was stored under abusive conditions (45 °C) a slight reduction in non-esterified fatty acids was observed after 6 months of storage without oxygen, although most likely a result of oxidation of non-esterified fatty acids in the sample with atmospheric oxygen in the headspace, rather than an actual decrease in release of non-esterified fatty acids (Rose, 2005).

Addition of antioxidants to whole wheat flour would also likely be minimally effective at prolonging shelf-life. This is because non-esterified fatty acids produced during storage are rapidly oxidized by lipoxygenase when the flour is mixed with water (Fig. 4), as would occur in any baking situation, and antioxidants are only marginally effective against lipoxygenase-mediated oxidation (Galliard, 1994; Reddanna et al., 1985).

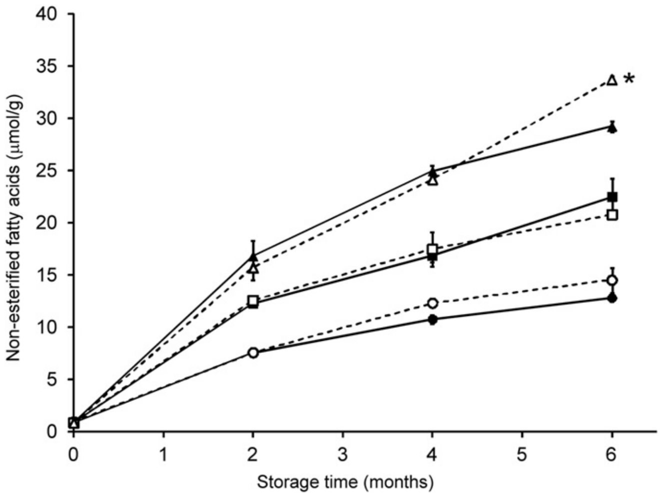

Fig. 5. Non-esterified fatty acids in whole wheat flour (10.6% moisture) stored at 25 °C (circles), 35 °C (squares), and 45 °C (triangles) in foil-lined laminate pouches sealed with atmospheric oxygen (filled shapes) or with an oxygen absorber packet (Ageless, Mitsubishi Gas Chemical America, New York, NY USA) that reduced oxygen to $<0.1\%$ (as measured by a 3500-series oxygen analyzer, Illinois Instruments, Johnsburg, IL USA; open shapes); error bars show standard deviation; $n = 2$; some error bars were too small to plot; *indicates significantly different (Student's *t*-test; $p < 0.05$) from the sample stored at that temperature under atmospheric oxygen at each time point (Rose, 2005).

Therefore, other strategies to stabilize lipids in whole wheat flour must be employed. The obvious strategy to control rancidity of whole wheat flour would be inhibition of lipase activity, the first step of lipid degradation (Fig. 2). This would inhibit the generation of substrates for lipoxygenase during oxidative rancidity (Galliard, 1994).

A number of heat processing approaches have been explored to inhibit lipolytic activity in whole wheat flour. Since the lipase activity is concentrated in the bran, this fraction can be heated separately and then added to wheat flour in the proper proportions to make whole wheat flour (Rose et al., 2008). This allows for the inhibition of lipase without risking influencing the flour functional properties. Furthermore, in wheat, rice, and oats, lipase is more stable than lipoxygenase (O'Connor et al., 1992). Therefore, if lipase is denatured, lipoxygenase is also denatured. Vetrimani and Haridas Rao (1990) reduced lipase activity in wheat bran by 40% by heating at 175 °C for 40 min Rose et al. (2008) reported 74, 93 and 96% reduction in lipase activity when wheat bran was dry heated at 175 °C for 25 min, 60 s of microwave (1000 W), or 60 s of steam, respectively.

The challenge with heat treatments to inactivate lipase is that they can easily promote autoxidation; that is, heat treatments that totally inactive lipase have resulted in flours that oxidize more rapidly than control flour (Cuendet et al., 1954; Molteberg et al., 1995). Lehtinen et al. (2003) demonstrated that the majority of oxidation in heat treated oats was from polar, membrane-bound lipids, rather than storage triacylglycerols; thus, they suggested that premature lipid oxidation in heat treated flours was due to disintegration of membrane structures and inactivation of heat labile antioxidants.

As a result, other strategies that do not involve heat have been employed to inhibit lipase activity. Because the activity of lipase can be influenced by the presence of metal ions (Barros et al., 2010), Munshi et al. (1993) treated rice bran with $ZnCl_2$, $NiCl_2$, $FeCl_3$, or $CuCl_2$, by dissolving each salt in HCl or methanol and spraying the solution in a fine mist over the bran. The salts were applied at 25–200 μg of the metal ion/g bran. They found that the effectiveness of these salts against lipase activity during 10 d of storage was $NiCl_2 > FeCl_3 > ZnCl_2 > CuCl_2$. The practical applicability of this approach may be limited, however, since $NiCl_2$ addition to whole wheat flour for food use would be unacceptable, and $FeCl_3$, while perhaps more nutritionally acceptable, would probably promote lipid oxidation (Huma et al., 2007). In a similar approach, Prabhakar and Venkatesh (1986) treated rice bran with HCl to reduce the pH. This decreased the non-esterified fatty acid concentration by about 80% comparing with an untreated bran after 30 d of storage at 25–30 °C and 50–65% relative humidity. However, this treatment would likely affect flour functionality. Champagne and Hron (1994) treated rice bran with boiling ethanol vapors to denature lipase. Rice bran was placed in a vessel affixed with a condenser above and boiling ethanol beneath. They found that the vapors were effective in denaturing lipase, but the vapor extracted antioxidants as it passed through the bran, thus increasing susceptibility to lipid oxidation. Addition of citric acid or butylated hydroxytoluene to the ethanol helped stave off premature lipid oxidation.

Besides lipase inactivation, other strategies involving new processing technologies for food applications could be used as solutions to increase shelf-life of whole wheat flour. Marathe et al. (2002) tested gamma irradiation to extend shelf-life of whole wheat flour. After 6 months of storage, chapatis, an Indian unleavened bread, made with whole wheat flour that had been treated with 0.25 kGy irradiation were significantly preferred over those made with untreated whole wheat flour.

4. Conclusions

Selection of wheat and milling technique may be different when producing whole wheat flour compared to wheat flour. Chemical components and physical properties of the outer portions of the wheat kernel influence baking properties. We are only beginning to understand these effects; more research is necessary to identify components with the greatest influence that can be manipulated to create whole wheat flours with optimum functionality.

During whole wheat flour storage, the products of lipase and lipoxygenase activity are the major culprits in the loss of sensory acceptability, nutritional value and functional quality. The strategy to control rancidity of whole wheat flour has been inhibition of lipase activity, thus halting or slowing the early steps of lipids degradation. Unfortunately, these approaches have been met with only marginal success.

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