

Effect of ultrasound treatment on dehulling efficiency of blackgram

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Abstract Present study was conducted to evaluate the effect of Power ultrasound, on dehulling efficiency, dhal yield, dehulling loss and total colour difference of black gram using response surface methodology. Nine treatments were performed with variation in ultrasound power 343–525 W and treatment time 1–3.5 h. It was observed that ultrasound treatment significantly improved the dehulling efficiency and dhal yield of the black gram and reduced the dehulling loss. The optimized treatment condition obtained for optimum dehulling yield (75.71%), dhal yield (74.63%) dehulling loss (12.72%), and total colour difference (5.08) was ultrasound power of 513.39 W and exposure time of 2.12 h. Moreover the blackgram pre-treated with ultrasound required lesser cooking time when compared to soaked alone sample. The SEM analysis revealed the significant effect of ultrasound on the blackgram kernel which led to uniform cavitation of the surface of the kernel compared to the soaked sample without ultrasound treatment. In food industry blackgram is pre-processed i.e. soaked and cooked to produce various soups, canned products, batter, snack foods etc. Hence ultrasonic treatment can be applied to improve and facilitate a faster dehulling efficiency, with added advantage of increased soaking rate and a decrease in the cooking time for blackgram.

Keywords Ultrasound · Dehulling · Blackgram · Soaking · Cooking

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Introduction

Pulse grains are an excellent source of primary and secondary metabolites and are consumed by large number of people across the globe as staple food in combination with cereals and depend on them for meeting their protein requirement (Singh 2017; Chen et al. 2017). Black gram (*Vigna mungo*. L) is one of the important pulse crop of India and is one of the most important cultivated pulses of ‘Vigna’ group. Globally the black gram is grown widely in countries like Iran, South East Asia and South Africa (Singh et al. 2004, 2017). The black gram is rich in nutrients as it contains higher percent of protein (24%), carbohydrate (59.6%) and fat (1.4%) (Gopalan et al. 1989). Black gram is consumed as whole grain or as dehulled splits or as flour and is extensively used as flour in various Indian fermented food products like dosa, idli, and other products like making dhal, curries, sweets and snacks. However the presence of vitreous layer of gums and mucilages, causes problems in processing hence the need for dehulling of black gram.

Dehulling can be defined as the process of removal of hull from the cotyledons of the pulses (Tiwari et al. 2007). The husk has to be separated because of anti nutrient properties of the husk. Dehulling of pulses is reported to be done by two process, wet pre-treatment dehulling and dry processing to achieve high efficiency of dehulling (Kurien 1977). These methods results in loosening of the bond between hull and the cotyledon, hull can be removed without damage to the cotyledon, ease of dehulling and improve the quality. The time of dehulling i.e., ease of milling can be achieved by different pre-treatments. In the dry method the grains are pitted, soaked in oil (1%), dried in sun (2–3 days), water spray (2.5%), tempered and dehulled (Tiwari et al. 2011). The application of oil and

water is followed in dry method. The pulses can be dehulled by wet method, which involve soaking for 6–12 h followed by drying to a moisture content of 10% (Kurien and Parpia 1968). Other method of wet processing involves soaking (12 h), heaping (16 h), mixing with red paste earth, drying (2–4 days), removal of red earth and dehulling (Tiwari et al. 2007). Different pre-milling treatments such as chemical treatments (acetic acid, sodium bicarbonate and alcohol etc.), urea solution, vegetable oil treatments, heat treatment and microwave application have been followed and tested by different researchers (Phirke and Bhole 2000; Narasimha et al. 2003; Tiwari et al. 2007, 2008; Joyner and Yadav 2013; Pal et al. 2017).

Ultrasound application in food industry is growing rapidly for both the analysis and modification of foods. Ultrasound is also applied as a pretreatment in many of the processing methods such as extraction, drying, freezing etc. to increase the efficiency of the process (Rawson et al. 2011a, b; Krishnan et al. 2015; Sunil et al. 2017; Janghu et al. 2017). The sound range employed is divided into high frequency, low energy diagnostic ultrasound and low frequency, high energy power ultrasound (Dolatowski et al. 2007). The ultrasound application as a pretreatment may have effect on the soaking time, dehulling time, dehulling yield and cooking time. Cunningham et al. (2008) observed reduction in soaking time due to reduction in the internal resistance compared to the external resistance with ultrasound application, which may be attributed to the change in microstructure by cavitation or by sponge effect causing inertial flow. A similar observation of reduction in soaking time was observed in chickpeas (Yildirim et al. 2010; Ranjbari et al. 2013), navy beans (Ghafoor et al. 2014), sorghum (Patero and Augusto 2015) and common beans (Ulloa et al. 2015). Ulloa et al. (2015) observed reduction in cooking time up to 43% in the ultrasound treated common beans. The ultrasound a novel pre-treatment can be used for increasing the dehulling yield by reducing the soaking time and can loosen the hull of the pulses during the treatment which may be due to the change in microstructure by cavitation, which also helps in faster hydration. Ultrasound treated pulses have shown reduction in the cooking time, thereby resulting in energy savings and reduction in nutrient loss due to long exposure time to heat. The literature for process and application of ultrasound as pretreatment to study the dehulling process for cereals and pulses is not available.

Since ultrasound is an emerging technology with varied applications, the present work was undertaken with an overall objective to study the application of Ultrasound as a pre-treatment on dehulling of black gram as well as its effect on the quality and optimization of the dehulling process for black gram.

Materials and method

Materials and equipments used

Black gram (ADT5 variety) was purchased from the local market and cleaned to remove the foreign matter and infested grains. The initial moisture content of the sample was 12.5% (wb). The moisture content of the black gram was determined by using oven drying method (Horwitz et al. 2000).

The water bath type of ultrasound system (Model: EN500 life-care.co.in, Mumbai, India) was used for the treatment. The ultrasound bath was of 47 L tank capacity and operated at room temperature. The maximum power input of the equipment was 600 W.

A domestic type of microwave (Model: IFB 30SC2) was used for the study. The operating frequency of the oven was 2450 MHz and had a maximum input power of 1400 W. The rated power output of the oven was 900 W at 100% power level and the power level can be varied from 10 to 100% at an interval of 10%.

Methodology

Proximate analysis

The carbohydrates, proteins, fat, fibre and ash content were determined for the grain, bran and husk using the standard AOAC methods (Horwitz et al. 2000).

Soaking

The grains (100 g) were soaked in distilled water in a glass beaker for different times ranging from 1 to 3.5 h. The soaked samples were used as control samples.

Ultrasound treatment

The sample (100 g) was taken in a glass beaker and filled with water and kept in the ultrasound water bath. The samples were treated at different power levels ranging from 343 to 525 W at different exposure times ranging from 1 to 3.5 h.

Microwave treatment

The sample of 100 g was taken in a glass beaker and kept on the turn table of the microwave oven. The sample was treated at 810 W power level for 120 s, which was selected from the earlier research findings (Joyner and Yadav 2013).

Dehulling procedure

The laboratory model emery roll polisher (Model: TM05, Satake corporation, Japan) was used for dehulling the grains. Dehulling was done for a fixed time period of 60 s for all the experiments based on the pre-trials conducted using emery roll polisher. After dehulling, different fractions were graded to whole grain, split grain, broken and bran.

Analysis of dehulling properties of black gram

Dehulling yield The dehulling yield is defined as the quantity of total whole dehulled kernels and broken kernels (except powder and husk) that are produced while dehulling of black gram. Head kernels are those which are retained over the sieve no. 10 (BSS mesh). Broken are those which are retained over sieve no. 30 and which are being passed through sieve no. 10 (BSS mesh). Again the broken are differentiated into fine broken which is being passed through sieve no. 30 (BSS mesh). So the dehulling yield is calculated by using the formula;

$$\begin{aligned} \text{Dehulling yield (\%)} &= \frac{\text{weight of dehulled kernels (g)} + \text{weight of broken (g)}}{\text{initial weight of black gram (g)}} \\ &\times 100 \end{aligned} \quad (1)$$

Dhal yield Dhal yield is defined as the yield of whole kernels and split kernels as a percentage of seed weight. Dhal yield is calculated by the given relationship;

$$\begin{aligned} \text{Dhal yield (\%)} &= \frac{\text{Mass of dehulled whole and split grains (g)}}{\text{initial weight of black gram (g)}} \times 100 \end{aligned} \quad (2)$$

Dehulling loss Dehulling loss is calculated by the weight fractions of the powder and fine broken relative to the initial weight of black gram taken for dehulling. It is calculated by using the relationship;

$$\begin{aligned} \text{Loss (\%)} &= \frac{\text{weight of powder obtained (g)} + \text{weight of fine broken (g)}}{\text{initial weight of black gram (g)}} \\ &\times 100 \end{aligned} \quad (3)$$

Colour Hunter colour lab colorimeter (Model: Colour Quest XE, USA) was used for determination of colour. The

calibration for the machine was done using black and white standard templates. The samples are placed over the eye of the CIELAB and the L^* , a^* , b^* values were monitored for each sample and readings were noted. The difference is calculated by using equation in terms of ΔE^* (Francis and Clydesdale, 1975).

$$\Delta E = \left[(L_o^* - L^*)^2 + (a_o^* - a^*)^2 + (b_o^* - b^*)^2 \right]^{1/2} \quad (4)$$

where L_o^* , a_o^* and b_o^* are the colour parameters of the control sample.

Cooking time

The cooking time of the optimized ultrasound treated, microwave treated and distilled water soaked were analyzed according to the method as suggested by Wani et al. (2015) and Joyner and Yadav (2015). A 20 g sample was added to a 250 ml beaker with distilled water in it after the water reached its boiling point. Boiling was continued and at an interval of one minute the samples were drawn from beaker and the firmness was test by the texture profile analyzer and it was also done by placing the sample between two glass plates. Then the plates were pressed against each other. The blackgram is said to be cooked when the kernel no longer had opaque or uncooked centers and the time required for cooking was noted.

Experimental design and statistical analysis

A central composite design (CCD) with axial points was used for designing the experimental data. The significant terms ($p < 0.05$) in the model were found by analysis of variance (ANOVA) based on p value. The experimental design matrix, data analysis, regression coefficient, generation of 3D graph and numerical optimization procedure were created using Design Expert Version 6.0.8 (Stat-Ease Inc., 2021 East Hennepin Ave., Suite 480 Minneapolis, MN 55413, USA).

Results and discussion

Proximate analysis

The carbohydrate, protein, fat, and ash content were determined for the grain as the composition of the seeds effect the cooking quality (Singh 1999). Proximate composition of the black gram contained 66.3% carbohydrates, 26.49% protein, 0.95% fat and 2.93% ash. Similar results were observed and reported for the composition (Bravo et al. 1999; Girish et al. 2012; Wani et al. 2013). Results

revealed that the black gram contained high protein content and thus a good source of nutrient.

Effect of ultrasound treatment on dehulling properties of black gram

Dehulling yield

The dehulling yield of nine treatments of ultrasound and exposure times is shown in Table 1. The dehulling time, increased with increase in the ultrasound power and also increase in the exposure time. The dehulling yield for the ultrasound treatment at different exposure times was ranging from 72.40 to 76.46%. The highest dehulling yield of 76.46% was recorded for 450 W ultrasound power at an exposure time of 3.5 h.

A study was also conducted to compare the dehulling yield in the water soaked sample only for same exposure time with ultrasound pretreated samples. The dehulling yield achieved using the ultrasound treatment at different exposure times was higher than the water soaked treatment for the same time periods (Table 2). The dehulling yield in water soaking pretreatment ranged from 64.35 (1.01 h) to 71.11% (3.5 h) however in case of ultrasound pretreatment

it was ranging from 72.40 (1.4 h) to 76.46% (3.5 h). The dehulling yield increased with increase in soaking time similar to ultrasound pretreatments. The increase in soaking time can result in loosening of the husk, thereby resulting increase in the dehulling yield which can be attributed to the change in microstructure by cavitation resulting in loosening of the hull. Moreover ultrasound treatment may also denature the protein and gums between the hull and cotyledon leading to an improved dehulling yield. Application of heat results in breaking the bond between hull and the cotyledon of pulses by denaturing the protein and gums present between them (Sokhansanj and Patil 2003; Joyner and Yadav 2015). The oil pretreatment which is one of the methods used for pulses dehulling shows a maximum dehulling yield of 74.1% (Sreerama et al. 2009) which is similar to the results obtained in the present study.

It is evident by the analysis of variance, the effect of ultrasound power and exposure time was significant ($p < 0.05$) on the dehulling yield of black gram (Table 3) Fig. 1a. The regression equation obtained for the linear model in terms of coded factors is as below;

$$Dehulling\ yield = 74.51 + 1.6A + 1.13B \tag{5}$$

Table 1 Effect of ultrasound treatment and exposure time on the quality parameters of black gram

Ultrasound power (W)	Exposure time (h)	De-hulling yield (%)	Dhal yield (%)	De-hulling loss (%)	Total colour difference
343	2.25	73.21	71.27	14.64	5.59
375	1.4	72.40	70.10	14.99	0.10
375	3.13	73.80	72.02	14.03	3.52
450	1.01	72.50	70.33	14.87	0.20
450	2.25	74.80	73.11	13.13	5.81
450	2.25	75.00	73.68	13.26	5.81
450	3.5	76.46	75.76	12.13	7.48
525	1.4	75.93	74.85	12.76	4.11
525	3.13	76.34	75.64	12.27	6.84

Table 2 Comparison of effect of ultrasound treatment and water soaking treatment on the de-hulling yield (same soaking times)

S. no.	Soaking time (h)	Water soaking	Ultrasound treated	
		De-hulling yield (%)	Power (W)	De-hulling yield (%)
1	1.01	64.35	450	72.50
2	1.4	65.16	375	72.40
3	1.4	65.18	525	75.93
4	2.25	68.54	343	73.21
5	2.25	68.55	450	75.00
6	2.25	68.11	450	74.8
7	3.13	70.32	375	73.80
8	3.13	70.54	525	76.34
9	3.5	71.11	450	76.46

Table 3 Analysis of the variance of the models and predictive second order polynomial equations for different ultrasound treatment of black gram

Parameters	R ²	CV	p	Lack of fit	F value	Equation
Dehulling yield	0.875	0.87	0.002	0.15	21.0	$Dehulling\ yield = 74.51 + 1.6A + 1.13B$
Dhal yield	0.886	1.16	0.0015	0.32	23.4	$Dhal\ yield = 73.01 + 2.19A + 1.58B$
Dehulling loss	0.902	2.94	0.0009	0.15	27.65	$Dehulling\ loss = 13.55 - 1.12A - 0.81B$
Total color difference	0.67	40.31	0.034		6.26	$\Delta E = 4.79 + 1.13A + 2.49B$

A ultrasound power (W); B treatment time (h)

where A ultrasound power and B exposure time.

By using the above Eq. (5) the actual and estimated values were compared, and it was evident that both the values were close to unity slope (1.156) and high R² value (0.8750) of the straight line fit between actual and estimated values.

Dhal yield

The dhal yield of all the treatments of ultrasound power level and exposure times is shown in Table 1. The dhal yield significantly increased with increase in the ultrasound power and exposure time ($p < 0.05$) given in Table 3 and Fig. 1b. The dhal yield for the ultrasound treatment at different exposure times ranged from 70.10 to 75.76%. The maximum dhal yield of 75.76% was recorded in 450 W ultrasound power and exposure time of 3.5 h. The lowest dhal yield was recorded in treatment of 375 W and 1.4 h.

The trend in the dhal yield was almost similar to the dehulling yield and it was expressed by a linear equation as expressed below;

$$Dhal\ yield = 73.01 + 2.19A + 1.58B \quad (6)$$

where A ultrasound power and B exposure time.

By using the above Eq. (6) the actual and estimated values were compared, and it was evident that both the values were close to unity slope (1.112) and high R² value (0.8864) of the straight line fit between actual and estimated values.

Dehulling loss

The dehulling loss of the treatments of ultrasound and exposure times is shown in Table 1. There was a significant decrease in dehulling loss ($p < 0.05$) with increase in the ultrasound power and exposure time (Table 3, Fig. 1c). The dehulling loss for the ultrasound treatment at different exposure times ranged from 12.13 to 14.99%. The minimum dehulling loss of 12.13% was recorded in 450 W ultrasound power and exposure time of 3.5 h. The maximum dehulling loss was observed in treatment of 375 W and 1.4 h. With the generation of cavitations during

ultrasound treatment the denaturation of the protein and gums between the hull and cotyledon and there by its loosening, reduces the force required to separate the hull from the cotyledon thereby the wear and tear of the grain is also low and lower dehulling losses.

The trend in the dehulling loss was expressed by a linear equation as expressed below;

$$Dehulling\ loss = 13.55 - 1.12A - 0.81B \quad (7)$$

where A ultrasound power and B exposure time.

By using the above Eq. (7) the actual and estimated values were compared, and it was evident that both the values were close to unity slope (1.126) and high R² value (0.9021) of the straight line fit between actual and estimated values.

Colour

The total colour difference (ΔE^*) values for different treatments ranged from 0.197 to 7.481 (Fig. 1d). The ΔE^* values increased with increase in the ultrasound treatment time and power (Table 3). The change in colour may be due to the long exposure time (soaking time) leading to rapid hydration action hence leading to change in colour. Furthermore during ultrasound treatment there may be release of some water soluble colored components from the black gram grain which may as well lead to total colour difference. Ghazali and Cheng (1991) reported that during soaking, soluble pigments such as anthocynins leach out from the seed coats into the soaking medium, leading to change in colour. Similarly in the present study the analysis of variance showed that the effect of ultrasound power and exposure time were significant ($p < 0.05$) on the colour change (Fig. 2d).

$$\Delta E = 4.79 + 1.13 * A + 2.49 * B \quad (8)$$

where A ultrasound power and B exposure time.

Cooking time

The best dhal yield samples were chosen for the cooking studies. The dehulled dhal were cooked to know the time

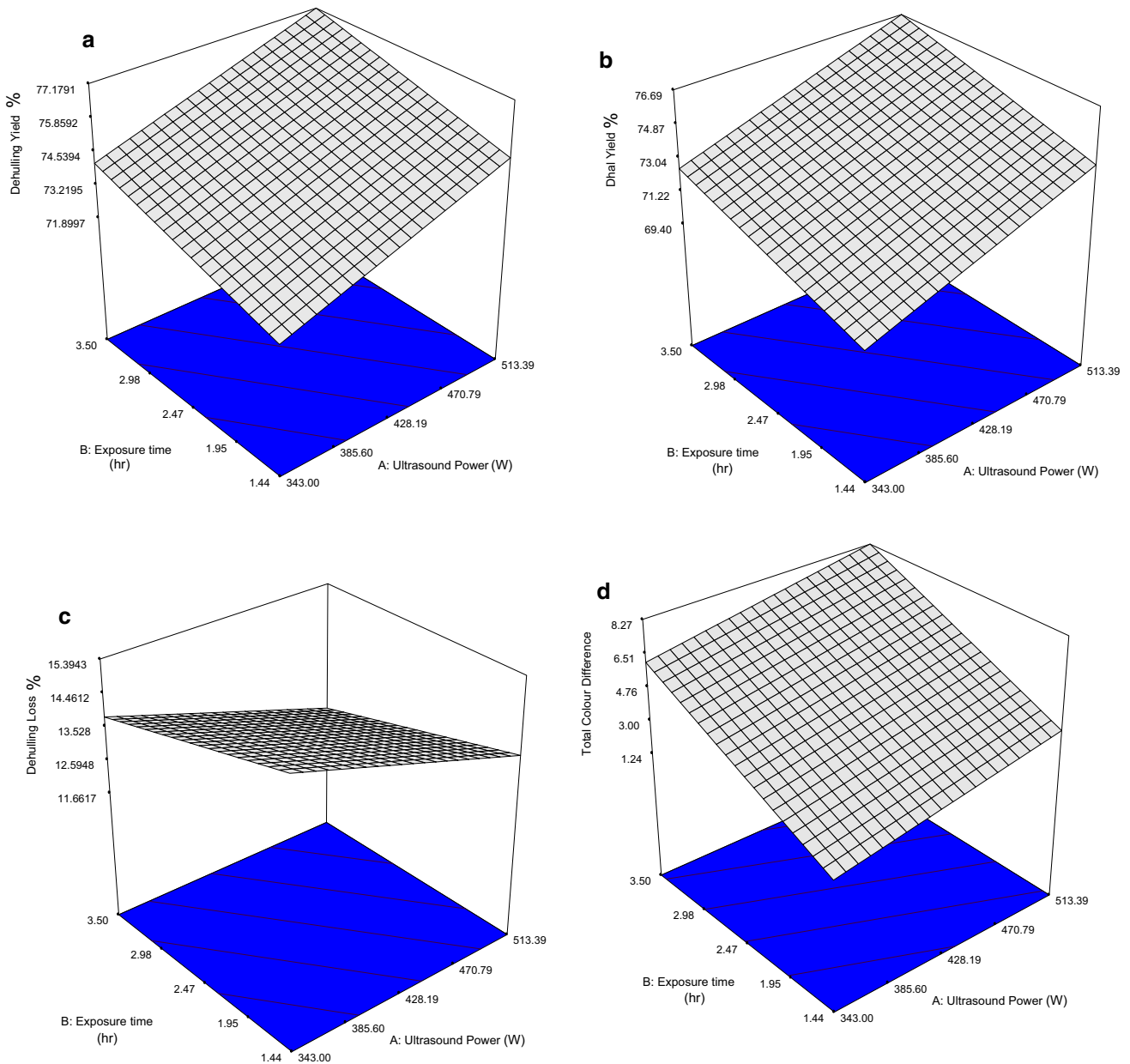


Fig. 1 Effect of ultrasound treatment on **a** dehulling yield, **b** dhal yield, **c** dehulling loss, **d** total colour difference of black gram

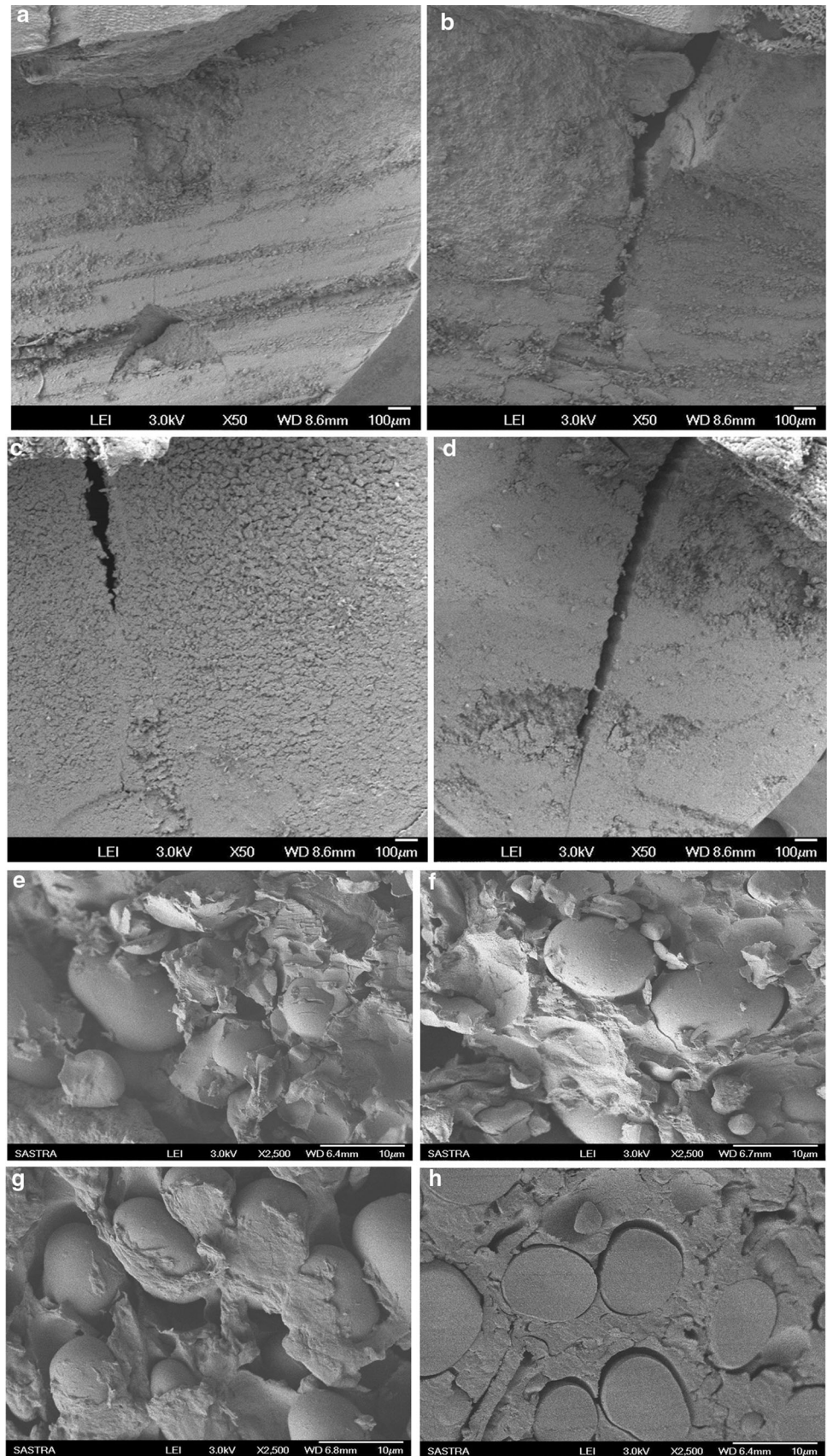
and the total energy required for cooking and to compare it with the untreated samples. It was observed that the microwave treated sample required the least time of 11 min to cook, followed by the ultrasound treated samples with 13.5 min and for the control sample (soaked initially) it was found to be 20 min. This observation was as expected; since the microwave treatment is severe which leads to precooking of the dhal as evident from the SEM analysis, discussed later in the manuscript, which on further thermal treatment requires lesser time to fully cook. Ultrasound pretreated samples showed a faster rate of cooking compared to soaking alone. Similar results were observed for

chickpeas by Yildirim et al. (2013), which was explained by the fact that ultrasonic treatment may cause a rapid series of alternative compressions and expansions, in a similar way to a sponge when it is squeezed and released repeatedly. Moreover the production of cavitations may be further helpful in improved rate of moisture uptake and significant reduce in the cooking time of chickpeas.

Optimisation of the experimental conditions

The optimum conditions for dehulling of black gram using ultrasound treatment at different exposure times was

Fig. 2 SEM micrographs of black gram **a, e** control (untreated), **b, f** soaked, **c, g** ultrasound treated, **d, h** microwave treated at different magnification



arrived using Design-Expert version 6.0.8. The conditions were optimized for maximum dehulling yield and dhal yield, minimum dehulling loss and minimum total colour difference. The software predicted that ultrasound power of 513.39 W and exposure time of 2.14 h treatment gave an optimum dehulling yield of 75.73%, dhal yield of 74.67%, dehulling loss of 12.7% and total colour difference of 5.08. The average experimental results for the same condition of ultrasound power and exposure times for dehulling yield, dhal yield, dehulling loss and total colour difference were observed to be 74.49, 72.97, 13.56 and 4.92% respectively, with low percentage error which confirms to the optimum conditions.

Scanning electron microscope analysis

SEM analysis morphological characterization of the samples was done using FE-SEM (JEOL JSM 6701-F). The dried sample was coated on a carbon tape. It was again coated with platinum in an auto fine coater and then the material was subjected to analysis.

It was observed that the sample pretreated with ultrasound was having a more uniform texture compared to normal soaked and un-soaked control sample (Fig. 2).- When the ultrasound pretreated sample was compared with microwave pretreated sample the major difference observed was that the overall morphology of the microwave treated sample looked uniform like ultrasound pretreated sample however, cavitation effect was clearly visible in the ultrasound pretreated sample. One more major difference observed was that the microwave pretreated sample seemed to be cooked which was apparent from the cutting of the sample for the SEM analysis the grains were observed to be in halves. Whereas in case of normal soaked only and ultrasound treated and soaked, it was observed that the ultrasound treatment significantly improved the dehulling yield (Table 2).

Discussion

Ultrasonic treatment leads to the formation of acoustic cavitations which includes the formation, growth, and implosive collapse of bubbles in a liquid (Suslick 1990). In the present study the formation of bubble may be due to the pre-existing bubbles in water and also due to the gas trapped in the pores of the blackgram which was immersed in water for the ultrasound treatment. Moreover a larger bubble can be created as trapped gas exits from the crevices of a blackgram under the influence of ultrasonic driving force and coalesces with pre-existing bubbles. The bubble formed may continue to grow until it reaches a critical size, known as its resonance size, which

depends on the applied frequency of the ultrasound field (Leong et al. 2011; Bang and Suslick 2010; Chowdhury and Viraraghavan 2009; Brennen 1995). Ultrasonic cavitation also produces a hydrodynamic shear force in the aqueous phase due to the rapid collapse of microbubbles, which helps in the disintegration of coarser lignocellulosic materials. Cavitation, bubble implosion and hydrodynamic shear force may lead to cell membrane permeability which can alter the surface potential resulting in activation of calcium channels, increased selectivity, accelerated nutrition transport, and enhanced mass transfer inside and outside of the cell. Substrates such as hemicellulose and lignin may be destructively partitioned from cellulose and entrapped in the interbundles of cellulose microfibril leading to dislodging. The localized increase in temperature during ultrasound treatment may also lead to denaturation of the protein and gums between the hull and cotyledon of the blackgram leading to an improved dehulling yield. Furthermore the formation of free radicals which occurs during ultrasound treatment can lead to damage of macromolecules, including cellulose, proteins and lipids (Fig. 3). Overall, the controlling mechanism of ultrasound-assisted dehulling can be attributed to mechanical, thermal and chemical effects, which results into a reduction in particle size, enhanced mass transfer across the cell membranes, and denaturation of protein and gums.

However in case of microwave assisted dehulling, though the dehulling efficiency is improved, as it causes fragmentation and swelling, leading to degradation of lignin and hemicellulose in biomass. However it is also responsible for pre-cooking of the blackgram and hence the blackgram cannot be used as seed following microwave pretreatment. Moreover the reduced cooking time in microwave treatment can be attributed to the thermal effect on denaturation of the intermolecular bonds and thereby increase in intermolecular spaces. With increased intermolecular spaces, the water penetration will be easier during cooking which in turn helps faster cooking. The microwave treatment which is a thermal treatment has led to pre-cooking of the grains as evident by the SEM analysis.

Conclusion

From the present study it can be concluded that ultrasound as a pretreatment has a potential to improve the dehulling efficiency of black gram, moreover since it aids in rapid hydration of the grain it can also decrease the concentration of anti-nutritional factors from the grains, reduce the hardness of grain and aid in cooking and if the grain is to be used as seed, its viability following the ultrasound

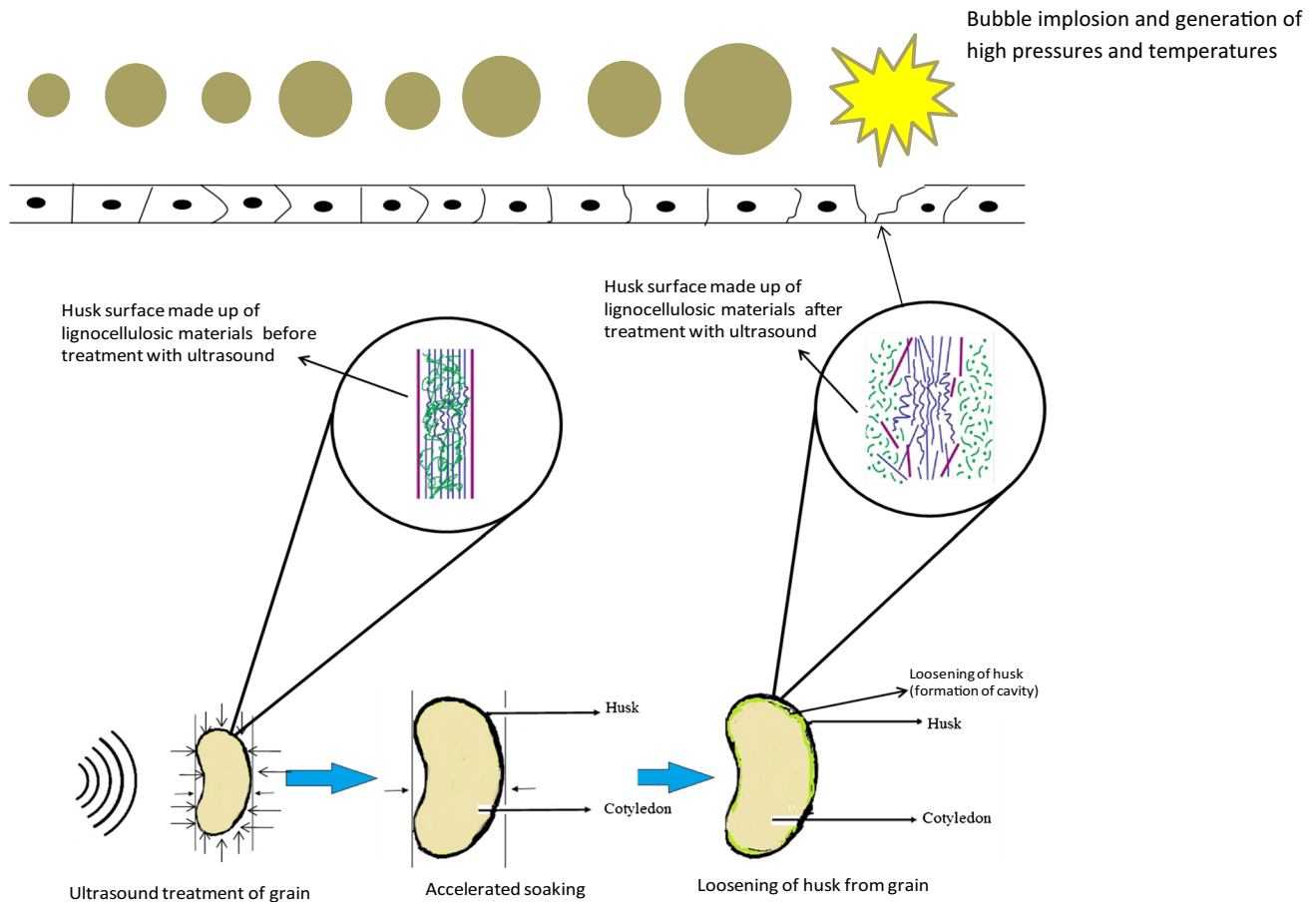


Fig. 3 The mechanism of loosening of husk from the black gram cotyledon following ultrasound treatment for improved dehulling efficiency

treatment is still valid and it can also increase the rate of germination.

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