Seed coats of pulses as a food ingredient: characterization, processing, and applications

Liezhou Zhong[a], Zhongxiang Fang[b], Mark L. Wahlqvist[c, d], Gangcheng Wu[a], Jonathan M. Hodgson[e, f], Stuart K. Johnson[*, a]

[a] School of Public Health, Curtin University, Perth, Western Australia, 6102

[b] Faculty of Veterinary and Agricultural Sciences, The University of Melbourne, Parkville, Victoria, 3010, Australia

[c] Fuli Institute of Food Science, Zhejiang University, Hangzhou, 310058, China

[d] Monash Asia Institute, Monash University, Melbourne, Victoria 3800, Australia

[e] School of Medical and Health Sciences, Edith Cowan University, Joondalup, Western Australia 6027, Australia

[f] Medical School, Royal Perth Hospital Unit, The University of Western Australia, Perth, Western Australia 6000, Australia

[*] Corresponding author. Tel.: +61 410 781 873. E-mail address: S.Johnson@curtin.edu.au (Stuart K. Johnson).

Abstract

Background: In recognition of their multiple benefits on environment, food security, and human health, pulses are attracting worldwide attention. The seed coat is a major by-product of pulse processing, and its only markets are as low value ruminant feed and very limited use in high fibre foods. Recently, accumulating studies have suggested that this underutilised by-product has greater potential as a novel natural "nutritious dietary fibre" which can be used as a functional food ingredient.

Scope and approach: This review discusses biochemical and physicochemical functionalities of seed coats of six globally important pulses: chickpea, field pea, faba/broad bean, lentil and mung bean with a special emphasis on the emerging food pulse lupin. Food process modification and recent human food applications of the seed coats are summarized. Bio-availability of the seed coat compounds, and phomopsins contaminated lupin seed coats as a typical example of safety issue are discussed.

Key findings and conclusions: High levels of dietary fibre, minerals and potential health-promoting phytochemicals in the seed coats indicate their great potential to be used as a natural "nutritious dietary fibre". However, further in-depth studies are required to improve their desirable nutritional, physiological and techno-functional properties whilst minimizing any undesirable ones.

Keywords

Pulses; seed coat; dietary fibre; minerals; by-product; food ingredient

1. Introduction

"Pulses" refers to those low-fat content leguminous seeds which are harvested for dry grain (FAO, 1994). So, oilseeds (e.g. soybean and peanut), leguminous green vegetables (e.g. green peas and green beans) and leguminous fodder plants (e.g. clover and alfalfa) are traditionally excluded. Pulses are historically important in both the human diet and cropping systems as crop rotations, due to their rich-protein and biological nitrogen fixation ability. Although most pulses are not traditionally typical Western-style foodstuffs, international events like "International Year of Pulses 2016" and "Global Pulses Day" suggest that they are being promoted to be important human food world widely (Foyer, et al., 2016).

As shown in Table 1, six of the 11 pulses which are covered in the FAO list, chickpea (*Cicer arietinum*), lupin (*Lupinus*), field pea (*Pisum sativum*), faba/broad bean (*Vicia fabae*), lentil (*Lens culinaris*) and mung bean (*Vigna radiate*), are the most important pulses globally, totally accounting for 79.89% of the world pulse production (81.8 million tonne) in 2016 (FAOSTAT, 2018). India is the largest pulse producer globally, followed by Canada, Myanmar and China. However, Australia is the largest lupin producer in the world, contributing an average of 58.22% of the world production in

2012-2016 (ABARES, 2018). Australian sweet lupin (ASL, *L. angustifolius*), which is also named "narrow-leafed lupin", is the most important lupin specie, constituting 93% of Australian lupin production and 52% of the world production (Pulse Australia, 2016). However, chickpea has overtaken lupin as Australia's largest pulse crop since 2011-12, with a production estimated at over 2 million tonne in 2016-17 (ABARES, 2018). As a leading pulse exporter, Australia exports over 90% of its chickpeas, faba beans, lentils and mung beans, and 60% of field peas were exported, being the largest exporter of *desi* chickpea and faba bean in the world. Notably, although Australia exported only 50% of its lupin, this accounted for 90% of world lupin export in 2013.

Pulse seed has three distinctive parts, namely the seed coat, embryonic axe and cotyledon, which generally accounts for 8-16%, 1-3% and 80-90% of the whole seed respectively (Dueñas, Hernandez, & Estrella, 2006). However, the proportions of seed coat show great genetic and environmental variability both between and within species (Table 2). For example, lupin uniquely contains a much higher percentage of seed coat than others, with up to 24% in Australian sweet lupin and around 18% in white lupin (Clements, et al., 2014). Removal of pulse seed coat (dehullinig) is a primary process to produce dehulled splits, ground flours and other fractionated pulse ingredients like pulse protein and fibre. In practice, by-product generated from the dehulling process is a mixture of seed coats, embryonic axes and brokens of cotyledons (Oomah, Caspar, Malcolmson, & Bellido, 2011; Sherasia, Garg, & Bhanderi, 2017). As a consequence,

dehulling loss which is the main waste stream of pulse processing represents as much as 31% for sweet lupin in Australia (Sipsas, 2008), and up to 28% for lentil and chickpea in India (Tiwari & Singh, 2011). Currently the primary markets for pulse seed coats are low value animal feed and only very limited use in human foods such as that added to make high fibre breads and meat products (like sausage and nuggets). This by-product not only leads to a tough disposal problem for the millers, but also wastes a potential source of novel, nutritious and health-promoting food ingredient (Sherasia, et al., 2017).

Growing evidence suggests that pulse seed coats have considerable amount of dietary fibre which is associated with diverse types of minerals and phytochemicals (bioactive secondary metabolites in plants e.g., polyphenolic antioxidants). Therefore, besides the well-documented physiological benefits of dietary fibre, seed coats provide potential for various physiological benefits, such as those related to antioxidant and anti-inflammatory activities. Available studies on pulse seed coats mainly focus on proximate compositions and anatomical structures, with little attention paid to their phytochemical properties and physiological functionalities. The present review brings together the current research on the characterization, processing and applications of seed coats from six selected pulses, i.e., chickpea, lupin, field pea, faba/broad bean, lentil and mung bean. This information should encourage strategies which might enable the more extended use of pulses and their seed coats in human foods.

2. Seed coat morphology and physical properties

The pulse seed coat (often referred as hull or testa) is a protective outer layer of the pulse seed. Structures of pulse seed coats have been overviewed by Moïse, Han, Gudynaitę-Savitch, Johnson, and Miki (2005) and Smykal, Vernoud, Blair, Soukup, and Thompson (2014). Anatomical structures of seed coats of field pea (Van Dongen, 2003), faba/broad bean (Youssef & Bushuk, 1984), chickpea (Wood, Knights, & Choct, 2011), lentil (Hughes & Swanson, 1986), lupin (Clements, Dracup, Buirchell, & Smith, 2005) and mung bean (Joseph & Swanson, 1993) have been extensively examined and show great similarities. Largely, there are three specialized cross-sectional layers in typical pulse seed coats: palisade cells (macrosclereids) layer, thick-walled hourglass cells (osteosclereids) layer, and a few layers of parenchyma (Fig 1.) (Moïse, et al., 2005; Tiwari & Singh, 2011).

The seed coats significantly affect chemical exchange (e.g. water and gas), biochemistry, mechanical properties (e.g. permeability, hardness and porosity) and physiological activities (e.g. germination and metabolism) of the pulse seeds (Moïse, et al., 2005). In addition, their chemical and physical characteristics, including composition, shape, mass, smooth or rough surface, thickness, colour, density and thermal properties (e.g. thermal conductivity and thermal diffusivity) strongly affect the whole seed properties (such as density, dehulling efficiency, and cooking quality),

124 further determine post-harvest processing, end application and market price of the seeds
125 (Souza & Marcos-Jilho, 2001). In this review, the most essential physical properties of
126 the seed coats (i.e. colour, thickness and permeability) are discussed.

127

128 *2.1. Colour*

129 As a key quality indicator, colour of the pulse seeds is crucial to consumer acceptance.
130 Seed coat colour varies significantly across different varieties (Table 2). In Australia
131 for example, over 90% of field pea is *dun* coloured type, and principally light brown
132 *desi* chickpea, beige or light brown faba bean, white with/without spots lupin, red lentil
133 and green mung bean. It has been revealed that the pigmentations of seed coats are
134 mainly attributed to chlorophyll and polyphenols (mainly flavonoids) (Hossain,
135 Panozzo, Pittock, & Ford, 2011), which mainly are located in the external palisade layer
136 (Wood, et al., 2011). The colours are associated with levels of those compounds, and
137 thus their physiological properties like antioxidant capacity. For example, dark
138 coloured pulses are reported to contain higher levels of polyphenols, mainly
139 anthocyanins and condensed tannins, and correspondingly higher antioxidant activities
140 than those of pale ones (Xu, Yuan, & Chang, 2007).

141

142 Colours of pulse seed coats are unstable during post-harvest processing and strongly
143 affected by processing conditions. For example, the extremely high temperature (>40
144 ºC) may accelerate undesirable colour darkening process in faba bean seed coat. This

145 is accompanied by a significant loss, ranging up to 86% of total polyphenols, which
146 may be explained by the polymerization of polyphenols into insoluble, un-extractable
147 high molecular weight polymers (Nasar-Abbas, et al., 2009). Similar browning was
148 found in lentils (Nozzolillo & Debezada, 1984) and chickpea (Reyes-Moreno,
149 Okamura-Esparza, Armienta-Rodelo, Gomez-Garza, & Milan-Carrillo, 2000) when
150 stored at high temperature. Moreover, the unpigmented varieties are supposed to be
151 more vulnerable to seed deterioration during storage (Souza & Marcos-Jilho, 2001).
152 However, the exact principles which are responsible for the darkening of pulse seed
153 coats are still unclear.
154
155 *2.2. Thickness and permeability*
156 In general, the palisade cell layer mainly decides the thickness of the seed coat.
157 Domesticated pulse varieties have thinner, softer, more permeable seed coats than wild
158 counterparts mainly due to decreases in thickness of the palisade layer. Moreover, the
159 proportion and thickness of the seed coat are negatively correlated with seed size. The
160 seed coat characteristics should be carefully considered during food processing
161 (especially dehulling) and application. For example, *kabuli* chickpea has a larger seed
162 size and thinner seed coat than the *desi* type (Table 2). As a result, cultivars of *kabuli*
163 are normally used as whole seeds without dehulling for paste, salads, roasted or fried
164 to make snacks (Wood, Knights, Campbell, & Choct, 2014). By contrast, cultivars of
165 the *desi* type are often dehulled to *dahl* (split) which are directly cooked or milled to

166 flour. Another example to show the associations between processing properties and
167 seed coat thickness is that lentils have thinner seed coats and thus shorter cooking times
168 than do other pulse seeds. Additionally, thicker seed coats result in longer cooking-
169 times in field peas (Wang, Daun, & Malcolmson, 2003) and faba beans than those
170 which have thin seed coats (Youssef & Bushuk, 1984).
171
172 The permeability of pulse seed coats change as the seed matures and are related to their
173 structure and chemistry (Ma, Cholewa, Mohamed, Peterson, & Gijzen, 2004). Although
174 impermeability of pulse seed coats is important to seed vitality, it is undesirable during
175 food processing. The impermeability will contribute to lower whole-seed cookability
176 ("hard-to-cook" phenomenon) and customer acceptability. For example, during
177 soaking, the thick and impermeable seed coat will slow water imbibition by the seed,
178 restrict its expansion and thus decrease the wet dehulling efficiency. The hydrophobic
179 waxy cuticle and condensed palisade cells layer of the seed coat are major contributors
180 to seed impermeability (Ma, et al., 2004).
181
182 **3. Pulse seed coat composition**
183 The nutritional composition of whole pulse seeds have been reviewed in the
184 FAO/INFOODS global food composition database for pulses (uPulses 1.0) (FAO,
185 2017). The composition of seed coats of the selected six pulses are summarized (Table
186 3). Generally, pulse seed coats have about 8-10% moisture, 3% ash, 1-3% lipids and

187 2-8% protein, with a major carbohydrate components (60-90%), mainly insoluble non-
188 starch polysaccharides (NSPs) (Tiwari & Singh, 2011). Of the macronutrients, we focus
189 on carbohydrates and minerals since they make up the majority and provide a basis for
190 the usage of the seed coat as a food ingredient.

191

192 In general, pulse seed coats have a neutral to slightly nutty flavour, although their
193 volatile profiles are largely unknown (Pfoertner & Fischer, 2001). Pulse seed coats are
194 the major contributors to the phytochemical content of the whole seeds (Dueñas, et al.,
195 2006; Luo, Cai, Wu, & Xu, 2016). Some of the phytochemicals are, historically,
196 referred to as "anti-nutritional factors (ANFs)", as with polyphenols, phytic acid and
197 alkaloids. However, numerous epidemiological studies now indicate their potential
198 benefits for human health (Rochfort & Panozzo, 2007). Investigation of the
199 micronutrients (vitamins and minerals) and other bioactive compounds in the six pulse
200 seed coats is embryonic.

201

202 *3.1. Carbohydrates*

203 As mentioned above, pulse seed coats have negligible amounts of starch and
204 oligosaccharides. Instead, they are predominantly composed of structural
205 polysaccharides (non-starch polysaccharides, NSPs), which are mainly cellulose,
206 hemicellulose, pectin (Table 3). As such, over 50 percent of the monosaccharides in
207 seed coat are glucose from the cellulose. The other principal sugars vary between

208 species. For example, the high concentrations of xylose (21.6%), uronic acids (10.0%)
209 and arabinose (8.4%) in lupin seed coat indicate relatively high contents of
210 arabinoxylan hemicellulose and pectin (Evans & Cheung, 1993). On the contrary,
211 uronic acids (22.3%), xylose (10.8%) and arabinose (5.2%) are the main sugars in field
212 pea seed coat cell walls (except glucose), indicating a high content of pectin (Guillon
213 & Champ, 2002). It is worthwhile to note that there are also significant differences
214 between NSPs in cotyledons and seed coats. For instance, the major constituent NSPs
215 of lupin seed coat are cellulose (from 45 to 56 g/100g dry matter (DM)), arabinoxylan
216 hemicelluloses (~13 g/100g DM) and pectins, whereas pectic substances and
217 hemicellulose are the predominant parts in cotyledon (Brillouet & Riochet, 1983).
218
219 Non-starch polysaccharides are classified as the principal components of the plant
220 dietary fibre (DF) (Lovegrove, et al., 2015). In principle, seed coat contributes a
221 significant proportion of the DF level of pulse because of their high content of NSPs,
222 ranging from 75 to 91 g/100g DM (Table 3). In addition, most of the DF in pulse seed
223 coats are insoluble dietary fibre (IDF), only 3.5% of total dietary fibre (TDF) in lupin
224 seed coat is soluble for example (Evans & Cheung, 1993). IDF levels of dehulled lentils,
225 peas and chickpeas decreased by 64%, 53% and 35% respectively compared to raw
226 seeds, but no significant reduction in SDF was found (Dalgetty & Baik, 2003). However,
227 regarding the newly proposed DF definition and analytic method (i.e. AOAC 2011.25),

contemporary information on DF (including oligosaccharides) for the six pulse seed coats is scarce.

3.2. Minerals and trace elements

Pulses provide substantial amounts of minerals. Pulse seed coats are rich in several minerals, e.g. Ca, Mg, Mn, Cu, Zn, B, Al and Na etc. (Tiwari & Singh, 2011). Notably, 67.5% of total Ca, and 41.3% of total Al of the whole lupin seed were reported to concentrate in its seed coat (Hung, Handson, Amenta, Kyle, & Yu, 1988). Likewise, over 70% of Ca and 50% of iron in mung bean (Singh, Singh, & Sikka, 1968), lentil (Tiwari, et al., 2012) and chickpea (Jambunathan & Singh, 1981) are found in their seed coats. Besides the inter-species variations, minerals in seed coats vary widely inner-species. For instance, contents of most of the minerals, especially Ca, Zn, Cu, and Mn, in *kabuli* chickpea seed coat are higher than *desi* type (Jambunathan & Singh, 1981). Consequently, differences in the seed coats (like thickness and proportion) between pulses are used to explain the variations in mineral levels of the whole seeds.

3.3. Phytochemicals

The major phytochemicals in different pulses vary significantly. For instance, chickpea was found to be one of the major sources of dietary saponins (Oakenfull, 1981), but alkaloids are characteristically present in lupin. Although, carotenoids (a group of lipid-

248 soluble natural plant pigments) contents of field pea (Marles, Warkentin, & Bett, 2013)
249 and chickpea (Ashokkumar, Tar'an, Diapari, Arganosa, & Warkentin, 2014) are
250 suggested to be associated with seed coat colours, pulse seed coats are generally known
251 as a poor source of carotenoids since they have low level of fat. In some cases,
252 phytochemicals may cause toxic effects (e.g. favism caused by vicine and convicine in
253 faba beans) (Klupšaitė & Juodeikienė, 2015). However, this review will discuss
254 polyphenols and phytic acid in the six pulse seed coats, and alkaloids in lupins since
255 they are more relevant to the potential positive physiological properties of the seed
256 coats.
257
258 *3.3.1. Polyphenols*
259 Polyphenols are a wide range of secondary plant metabolites, which typically have one
260 or more aromatic rings bearing several hydroxyl groups. The major polyphenols in
261 whole pulse seeds are phenolic acids (e.g., benzoic/cinnamic acids and their
262 derivatives), flavonoids (e.g., flavone and flavonol glycosides) and condensed tannins
263 (Oomah, Patras, Rawson, Singh, & Compos-Vega, 2011). Recently, a few studies have
264 investigated polyphenols in pulse seed coats, including chickpea (Sreerama, Neelam,
265 Sashikala, & Pratape, 2010), faba bean (Boudjou, Oomah, Zaidi, & Hosseinian, 2013),
266 field pea (Marles, et al., 2013), lentil (Dueñas, et al., 2006; Oomah, Caspar, et al., 2011),
267 and mung bean (Luo, et al., 2016; Muhammed, Manohar, & Junna, 2010).
268

In general, these studies confirmed that polyphenols of whole pulses seeds are essentially concentrated in the seed coats, and hence they are the predominant *in vitro* antioxidant capacity contributors. For example, 80.3-84.2% of the total polyphenol and over 83.9% of total flavonoid content of whole mung bean seed were reported to be present in the seed coat (Luo, et al., 2016; Muhammed, et al., 2010). The proportions of total polyphenol and total flavonoid content in faba bean seed coat are up to 80.0% and 89.3% of the whole seed respectively (Boudjou, et al., 2013). Similarly, total polyphenol content of chickpea seed coat (75.94 mg GAE /g DM) was relatively higher than that of cotyledon (15.24 mg GAE /g DM) (Sreerama, et al., 2010). Condensed tannins in faba bean (Boudjou, et al., 2013), mung bean (Xu, et al., 2007), and lentil (Dueñas, Hernández, & Estrella, 2002) seed coats were report to represent over 75%, 50% and 54% respectively of the total tannins in the whole seeds. Notably, Xu, et al. (2007) found that polyphenols levels and *in vitro* antioxidant activities of dark coloured (like red, bronze, and black) lentil and chickpea seeds were significantly higher than those of light coloured (like white, yellow, and green) varieties. Total free phenolic acids and condensed tannins in coloured pea seed coat reached to 78.53 g/g DM and 1560 mg CE/g DM comparing to 17.17 g/g DM and not detected for those in white seed coat (Troszyńska & Ciska, 2002).

In the case of lupins, total polyphenol content in seed coats of *L. mutabilis*, *L. albus*, and *L. angustifolius* which grown in Brazilia were reported to be 1.15-4.49 mg catechin

290 equivalents (CE)/g DM which were much lower than cotyledons (7.38-12.42 mg CE/g
291 DM) (Ranilla, Genovese, & Lajolo, 2009). The results accord with findings from
292 Lampart-Szczapa, et al. (2003), who found that polyphenols in seed coats of *L. luteus*,
293 *L. albus* and *L. angustifolius* grown in Poland (ranging from 0.16 to 0.42 mg caffeic
294 acid equivalents/g DM), were 1.30-6.52 times lower than those in cotyledons (0.32 to
295 1.88 mg caffeic acid equivalents/g DM). Additionally, these authors revealed that free
296 phenolic acids, primarily procatechuic acid and *p*-hydroxybenzoic acid, were mainly
297 present in the seed coats. Likewise, they found that concentrations of tannins in the
298 cotyledons were 4.33-31.00 times higher than that in the seed coat. On contrast,
299 Petterson (1998) reported that most tannins (include proanthocyanidins) of lupin
300 occurred in the seed coat, however, the initial data are unavailable.
301
302 These different and sometimes conflicting results of studies on polyphenols in pulse
303 seed coat are difficult to interpret and compare since the lack of consensus on extraction
304 methods and express ways (i.e., equivalents). Nonetheless, most of previous published
305 studies have only extracted polyphenols with organic solvents in which case
306 appreciable amounts of "bound" polyphenols in the seed coat matrix may remain un-
307 extracted and thus the total polyphenol levels and antioxidant capacity may be
308 underestimated (Saura-Calixto, 2012).
309

310 *3.3.2. Phytic acid*

311 Phytic acid (PA), its lower substituted homologues and its salts are referred as phytates
312 which are commonly present in pulse seeds. Phytic acid have been implicated in the
313 "hard-to-cook" phenomenon in pulse seeds. In addition, they are considered as the main
314 anti-nutritional factor due to their capacity to chelate cations (in particular calcium, iron
315 and zinc) to form insoluble complexes and therefore reduce their bio-availability
316 (Sanchez-Chino, Jimenez-Martinez, Davila-Ortiz, Alvarez-Gonzalez, & Madrigal-
317 Bujaidar, 2015). The content of phytic acid can be affected significantly by genetic and
318 environmental factors, alone and in combination. However, phytic acid in mung bean
319 (1.8-5.8 mg/g DM), pea (3.1-7.1 mg/g DM), lentil (2.5-12.2 mg/g DM), chickpea (2.8-
320 13.6 mg/g DM), lupin (6.0-8.9 mg/g) and faba bean (5.9-15.0 mg/g DM) are generally
321 lower than soybean (4.8-20.1 mg/g DM) (Campos-Vega, Loarca-Piña, & Oomah, 2010).
322 Moreover, the majority of phytic acid is demonstrated to present in "the proteins bodies"
323 in cotyledon (Campos-Vega, et al., 2010). Phytic acid of chickpea is presented in a low
324 level (0.79 mg/g DM) in seed coat but high in cotyledon (9.82 mg/g DM) (Sreerama,
325 et al., 2010). Beal and Mehta (1985) indicated that little (1 mg/g) or no phytic acid was
326 found in pea seed coats.

327

328 *3.3.3. Alkaloids*

329 Alkaloids are mainly present in lupins. Quinolizidine alkaloids, mainly lupanine, 13-
330 hydroxylupanine and angustifoline, are major contributors to the bitter taste of some

varieties of lupin seeds and are potentially toxic (Petterson, 1998). Bitter lupin varieties have alkaloid contents ranging between 0.5-6%, in contrast the sweet varieties have less than 0.02% (Resta, Boschin, D'Agostina, & Arnoldi, 2008). Moreover, alkaloids can be removed by washing with water. A maximum legal limit on alkaloid concentration in lupin flours and lupin products has been set at 0.02% by authorities of France, UK, Australia and New Zealand (Resta, et al., 2008). Little is known on distributions of alkaloids in lupin seeds, though Sipsas (2008) reported that no alkaloids were found in Australian sweet lupin seed coat, but no detailed data was found.

4. Mycotoxins contamination

Pulses are vulnerable to be contaminated by fungus and the resulting mycotoxins (e.g., alfatoxins, ochratoxins and phomopsins) during pre- or post-harvest (CAST, 2003). A further increase in human exposure of them by consuming products containing contaminated pulses may occur. However, recent systematic surveys on mycotoxins in pulses based human food are lacking. Here, phomopsins in contaminated lupin seeds, a highly representative example of mycotoxins contamination of pulses, will be discussed as a detailed case study.

Phomopsins are toxins produced by the fungus *Diaporthe toxic* (EFSA, 2012). The fungus mainly infects lupin stems but also the seeds under high humidity storage conditions (>13%). Lupin seed coat, being the outermost layer of the seed, is the most

352 vulnerable part of seed to be invaded by the fungus and thus may content the highest
353 level of phomopsins (EFSA, 2012). Phomopsins are suspected as the cause of lupinosis
354 in grazing animals. A maximum legal limit (5 μg/kg) of phomopsins in lupin seeds and
355 lupin foods has been established in Australian, New Zealand, UK and FAO (Schloss,
356 Koch, Rohn, & Maul, 2015). As other mycotoxins, phomopsins are stable to food
357 processing including soaking, cooking, and fermentation. However, seeds
358 contaminated by phomopsins can be easily removed by seeds grading and screening
359 since phomopsins is "almost entirely limited to dis-coloured seeds" (EFSA, 2012). In
360 addition, resistant varieties have been developed. Extrusion which combines high
361 pressure, high temperature and severe shear has showed the capacity to reduce other
362 mycotoxins (e.g. alfatoxins and zearalenone) (Bullerman & Bianchini, 2007), but no
363 studies on detoxifying phomopsins in lupin by extrusion cooking have been reported.
364

365 **5. Bio-availability of nutrients in pulse seed coats**

366 Bio-availability refers to the extent that nutrients can be released from food matrix into
367 digestive fluid, and thereby available for intestinal transport, biotransformation,
368 absorption and metabolism (Versantvoort, Van de Kamp, & Rompelberg, 2004). There
369 is strong evidence that structure and composition of a food matrix will govern the bio-
370 availability of many nutrients in the gastrointestinal tract (Wahlqvist, 2016).
371

A few published clinical studies have suggested that pea seed coat consumption may benefit cardiovascular and gastrointestinal biomarkers in humans, that may be due to multiple mechanisms caused by the high dietary fibre in the seed coat (Dahl, Whiting, Healey, Zello, & Hildebrandt, 2003; Flogan & Dahl, 2010; Mollard, Luhovyy, Smith, & Anderson, 2014). However, dietary fibre has been shown to significantly reduce bio-availability of several nutrients. For example, lupin seed coat in the diet decreased protein digestibility in rats (Bailey, Mills, & Hove, 1974). In contrast, removal of lentil seed coat significantly improved lentil iron bio-availability (DellaValle, Vandenberg, & Glahn, 2013). The compact inert insoluble fibre matrix of the seed coat may be a physical barrier to block the release of nutrients, give increased viscosity of digesta and therefore impair absorption. Besides, dietary fibre, polyphenols and alkaloids can also inhibit enzymes, and chemically bind some nutrients thus lowering their bio-availability (Khattab & Arntfield, 2009).

The fermentability of cellulose and hemicellulose in the colon was surprisingly reported to be high, 70% and 72% respectively, mainly degraded by some specialized series of gut bacteria (Flint, Scott, Duncan, Louis, & Forano, 2012). It suggests that "trapped" compounds (e.g., minerals and polyphenols) in pulse seed coats could be released in colon. In this context, dietary fibre could also modulate pH of human gastrointestinal tract, especially lower pH level in colon, to enhance release and absorption of minerals (Baye, Guyot, & Mouquet-Rivier, 2017). A large proportion of polyphenols are

393 reported to be not bioavailable in the upper part of the human gastrointestinal tract.
394 Instead, they will reach colon and be metabolized at a large extent by gut microbiota
395 (Saura-Calixto, 2012). However, more studies are needed to investigate digestibility of
396 pulse seed coats in human, as well as their physiological effects on human health
397 (including effects on colon and gut bacteria).
398

399 **6. Effect of processing on pulse seed coats**

400 Generally, pulses are dried in the field to achieve the target moisture of 9-20% for
401 threshing (i.e. removal of pods), then cleaned, graded and further dried to approximate
402 13% for storage. Storage conditions (e.g. seed moisture, relative humidity, duration and
403 temperature) significantly affect the seed coat characteristics. For example, the seed
404 coat colour of faba bean has been observed to darken from beige to dark brown
405 depending upon the storage conditions (Nasar-Abbas, et al., 2009). Although pulses
406 can be consumed either whole or dehulled splits, they require processing before
407 consumption to (1) reduce or eliminate anti-nutritional factors, (2) improve consumer
408 acceptability (e.g. texture, flavor), and (3) enhance nutritional properties like nutrient
409 bio-availability. There are several conventional whole seeds processing methods,
410 including soaking, dehulling, milling, cooking, puffing, germination (or sprouting) and
411 fermentation (Patterson, Curran, & Der, 2017). But only few studies are found to treat
412 isolated pulse seed coat using milling, boiling, and more recently extrusion cooking.

413 All have shown to affect composition, and physicochemical and nutritional properties
414 of the seed coats.
415
416 *6.1. Conventional processing*
417 Seed coat bulk density (weight of seed coat per unit volume) is low such that further
418 processing (like grinding) is required to increase their density to reduce its storage and
419 transport fees after dehulling (Table 4). Grinding was reported to increase solubility of
420 pea seed coat, from 4.1% to 8.6%, accompanying by reduction in water binding
421 capacity (WBC) and swelling capacity by 35.2% and 21.7% respectively (Ralet, Valle,
422 & Thibauit, 1993a). Similarly, water solubility of mung bean seed coat was 0.97% with
423 particle size of <50 mesh (<300 μm), whereas a much lower water solubility (0.79%)
424 was found with particle size of >35 mesh (>500 μm) (Huang, 2009). The authors also
425 found that mung bean seed coat with smaller particle size had a significantly higher
426 swelling capacity, WBC, and oil binding capacity but lower bulk density compared
427 with those with bigger particle size.
428
429 Soaking followed by cooking of whole pulse seeds is the traditional domestic operation
430 to produce edible pulse products. During soaking, pulses imbibe water to expand the
431 seed coats, and activate endogenous enzymes (cell wall polysaccharidases which can
432 disrupt the cell wall, and phytase which can reduce phytic acid content, for example)
433 (Wang, et al., 2003). Moreover, water soluble compounds like minerals, soluble tannins,

434 phytic acid, alkaloids and polyphenols may leach into soaking, cooking and canning
435 water (Tajoddin, Manohar, & Lalitha, 2013). As the outer layer, the seed coat plays a
436 crucial role in controlling these exchanges during soaking and cooking. Additionally,
437 Güzel (2012) found that atmospheric pressure cooking (APC) and high-pressure
438 cooking (HPC) caused darkening of chickpea and faba bean seed coats, with greater
439 effect for HPC. The colour changes may be the results of pigment degradation. Hashemi,
440 et al. (2015) found that starch in pea seed coat increased from 0.16% to 0.59% in dry
441 basis after boiling for 30 min, what may be due to the increase of starch bioavailability
442 and losses of soluble compounds during boiling.
443
444 Mung bean (Tajoddin, et al., 2013), lentil and field pea (López-Amorós, Hernández, &
445 Estrella, 2006), chickpea (Ghavidel & Prakash, 2007), and lupins (Dueñas, Hernandez,
446 Estrella, & Fernandez, 2009) have been used to germinate sprouts. Most of these studies
447 confirmed that germination will increase polyphenols (prominently flavonoids) and
448 vitamins, whereas decrease anti-nutritional factors (e.g. α-galactosides, trypsin
449 inhibitors and phytic acid). As a result, germination can improve antioxidant capacity
450 and bio-availability of the nutrients. Seed coat impermeability is the main regulator for
451 pulse germination. Moreover, the structure and composition of the seed coat will
452 change significantly just before and during germination, possibly by enzymes (Finch-
453 Savage & Leubner-Metzger, 2006). Although, so far, no study on the effect of

454 germination on pulse separated seed coat has been found, it can be hypothesised that
455 changes in composition and physico-chemical properties of pulse seed coat may occur.
456
457 *6.2. Extrusion*
458 Extrusion cooking is a high temperature short time unit operation in which food will be
459 melted in a sealed cylinder by high pressure, high temperature and high mechanical
460 shear, then passed through a die (Alam, Kaur, Khaira, & Gupta, 2016). Depending on
461 extrusion conditions (such as material particle size, feed rate, moisture, screw speed
462 and configuration, barrel temperature and die geometry), the process results in
463 disruption of cell wall structures, chemical reactions (such as polysaccharides
464 depolymerization, Maillard reaction and starch gelatinization), and physical changes
465 (e.g. solubility, morphological and rheological properties) (Singh, Gamlath, &
466 Wakeling, 2007; Wolf, 2010). Moreover, extrusion has been used to incorporate seed
467 coats of field pea (Schmidt, 1987), lupin (Tucek, 2009) into breakfast, pasta and snacks
468 to increase their dietary fibre levels. But they are beyond the scope of this review.
469
470 Extrusion cooking, mainly twin-screw extrusion, is the most used technology to modify
471 the functional properties of high fibre materials (Rashid, Rakha, Anjum, Ahmed, &
472 Sohail, 2015; Wolf, 2010; Yan, Ye, & Chen, 2015). Water solubility of pea seed coat
473 was reported to increase by 3.6-15.3% after extruded using twin screw extruder,
474 accompanied by a dramatic increase (up to 220%) in soluble dietary fibre (Ralet, Valle,

475 & Thibauit, 1993b). Similarly, single screw extruder increased soluble dietary fibre in
476 pea seed coat from 5.3% to 6.7% (Arrigoni, Caprez, Amadò, & Neukom, 1986).
477 Correspondingly, technical properties of pea seed coat, like water binding capacity and
478 swelling capacity, were increased by extrusion cooking. On the contrary, extrusion has
479 shown no or slightly increased effects on the technical properties of yellow pea seed
480 coats (Arrigoni, et al., 1986). Except the conflicting results mentioned above, data from
481 extruded wheat bran (Rashid, et al., 2015; Yan, et al., 2015), sugar beet pulp (Rouilly,
482 Jorda, & Rigal, 2006), onion waste (Ng, Lecain, Parker, Smith, & Waldron, 1999)
483 support the increase in the solubility of dietary fiber, as well as the improvements on
484 their physicochemical characteristics.
485
486 Extrusion cooking has been revealed to reduce the levels of heat sensitive extractable
487 polyphenols, which can be extracted by aqueous/organic solvents (Singh, et al., 2007).
488 However, it can release non-extractable polyphenols, which remain in the resulting
489 residues of the aqueous/organic extraction, from food matrix. Depolymerization of high
490 molecular weight polyphenols (such as condensed tannins) was also reported (Awika,
491 Dykes, Gu, Rooney, & Prior, 2003). Additionally, extrusion cooking can increase bio-
492 availability of minerals, mainly by reducing the chelating properties of dietary fibre and
493 the contents of other chelating compounds such as phytic acid and condensed tannins
494 (Singh, et al., 2007). Taken together, extrusion cooking could be an applicable
495 technology to improve the properties of pulse seed coats. However, more

496 comprehensive studies are required to investigate its effects on compositional and
497 physicochemical properties of the pulse seed coats.
498
499 **7. Application of pulse seed coats in human food**
500 Pulses have been historically important sources of energy, protein and dietary fibre in
501 human diet. Currently, pulse seed coats have only limited use in human food such as in
502 high fibre breads and meat products. However, the high content of dietary fibre in pulse
503 seed coats, along with considerable amounts of minerals, phytochemicals (e.g.
504 polyphenols) suggests they could be more widely utilized as novel functional dietary
505 fibre ingredients (Macagnan, da Silva, & Hecktheuer, 2016). There are several
506 commercial dietary fibre ingredients manufactured from pea seed coat and lupin seed
507 coat, both of them have been classified as GRAS (Generally Recognized as Safe).
508 However, lupin has been listed as a food allergen what requires mandatory labelling in
509 Europe since 2007, and most recently in Australia and New Zealand (March 2017)
510 (FSANZ, 2017). Moreover, there are several specific regulations on contaminants and
511 natural toxins levels of the six pulses and their derived food products, phomopsins and
512 lupin alkaloids in lupin seed coat for example.
513
514 Like other dietary fibre ingredients, pulse seed coats have been incorporated into baked
515 goods, in which they have shown to change physical, nutritional, and sensory properties
516 of the products. Dalgetty and Baik (2006) found that incorporations of pea, lentil, and

517 chickpea seed coats significantly increased dough mixing time, water absorption, and
518 loaf weight but decreased loaf volume. The observations are in accordance with the
519 results of Sosulski and Wu (1988) who added up to 7.7% of pea seed coat into dough.
520 The authors of these studies concluded that breads with 5% pulse seed coat addition
521 was comparable to whole wheat breads in sensory quality but had desirable higher
522 dietary fibre content.

523

524 In terms of adding pulse seed coats into meat products, Verma, Banerjee, and Sharma
525 (2012, 2015) used pea hull flour (PHF) and chickpea hull flour (CHF) as dietary fibre
526 sources to improve qualities of chicken nuggets. The studies found that incorporation
527 of the two hull flours significantly increased product yield and dietary fibre content.
528 However, both reduced emulsion stability of the product, and lowered its hardness,
529 gumminess and chewiness dramatically. Product colour was also affected by initial
530 colours of the two hull flours and formulation differences. Sensory evaluation
531 suggested that an 8% PHF addition in low salt (40% reduction) chicken nuggets were
532 acceptable to consumers.

533

534 **8. Conclusions**

535 To date, pulse seed coats are little utilised in human food. However, there is potential
536 for the seed coat to be used as a natural "nutritious dietary fibre" which could (1) fill
537 the "fibre intake gap", (2) provide considerable levels of minerals and antioxidants, and

(3) achieve greater safe and sustainable utilization of pulses by exploiting value-added applications of their by-products (Saura-Calixto, 2012; Sharma, et al., 2016). However, in-depth studies on biochemical, and nutritional properties of pulse seed coats are still lacking. In addition, physicochemical properties (e.g. solubility, swelling capacity, water and oil binding capacities, and viscosity) of pulse seed coats will significantly associate with physiological functionalities (Wahlqvist, 2016), but the impacts of processing on physicochemical properties are also still unclear. Moreover, to minimize the negative impacts of dietary fibre and other anti-nutritional factors, while improving their desirable physiological properties, further work is needed to optimize the processing. Finally, parallel to the study of pulse seed coats incorporation into food products, more nutritional and safety studies on the products are needed. These will add to what are likely to be favourable cost and sustainability profiles.

Acknowledgements

The author Liezhou Zhong wishes to thank the support from Curtin International Postgraduate Research Scholarship (CIPRS)/Health Sciences Faculty International Research Scholarship (HSFIRS). The salary of Jonathan Hodgson was supported by a National Health and Medical Research Council of Australia Senior Research Fellowship, and a Royal Perth Hospital Medical Research Foundation Fellowship. This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

559

560 **References**

561 ABARES. Agricultural commodities and trade data (2018).
562 http://www.agriculture.gov.au/abares/research-topics/agricultural-
563 commodities/agricultural-commodities-trade-data#2017 Accessed 27 April
564 2018.

565 Alam, M. S., Kaur, J., Khaira, H., & Gupta, K. (2016). Extrusion and extruded products:
566 Changes in quality attributes as affected by extrusion process parameters: A
567 review. *Critical Reviews in Food Science and Nutrition, 56*, 445-473.

568 Arrigoni, E., Caprez, A., Amadò, R., & Neukom, H. (1986). Chemical composition and
569 physical properties of modified dietary fibre sources. *Food Hydrocolloids, 1*,
570 57-64.

571 Ashokkumar, K., Tar'an, B., Diapari, M., Arganosa, G., & Warkentin, T. D. (2014).
572 Effect of cultivar and environment on carotenoid profile of pea and chickpea.
573 *Crop Science, 54*, 2225.

574 Awika, J. M., Dykes, L., Gu, L. W., Rooney, L. W., & Prior, R. L. (2003). Processing
575 of sorghum (*Sorghum bicolor*) and sorghum products alters procyanidin
576 oligomer and polymer distribution and content. *Journal of Agricultural and*
577 *Food Chemistry, 51*, 5516-5521.

578 Bailey, R. W., Mills, S. E., & Hove, E. L. (1974). Composition of sweet and bitter lupin
579 seed hulls with observations on the apparent digestibility of sweet lupin seed

580 hulls by young rats. *Journal of the Science of Food and Agriculture, 25*, 955-
581 961.
582 Baye, K., Guyot, J. P., & Mouquet-Rivier, C. (2017). The unresolved role of dietary
583 fibers on mineral absorption. *Critical Reviews in Food Science and Nutrition,*
584 *57*, 949-957.
585 Beal, L., & Mehta, T. (1985). Zinc and phytate distribution in peas-Influence of heat
586 treatment, germination, pH, substrate, and phosphorus on pea phytate and
587 phytase. *Journal of Food Science, 50*, 96-115.
588 Boudjou, S., Oomah, B. D., Zaidi, F., & Hosseinian, F. (2013). Phenolics content and
589 antioxidant and anti-inflammatory activities of legume fractions. *Food*
590 *Chemistry, 138*, 1543-1550.
591 Brillouet, J. M., & Riochet, D. (1983). Cell wall polysaccharides and lignin in
592 cotyledons and hulls of seeds from various lupin (*Lupinus* L.) species. *Journal*
593 *of the Science of Food and Agriculture, 34*, 861-868.
594 Bullerman, L. B., & Bianchini, A. (2007). Stability of mycotoxins during food
595 processing. *International Journal of Food Microbiology, 119*, 140-146.
596 Campos-Vega, R., Loarca-Piña, G., & Oomah, B. D. (2010). Minor components of
597 pulses and their potential impact on human health. *Food Research International,*
598 *43*, 461-482.
599 CAST. (2003). *Mycotoxins: Risks in Plant, Animal. and Human Systems*. USA: CAST:
600 Council for Agricultural Science and Technologyy, Ames, Iowa.

601 Clements, J. C., Dracup, M., Buirchell, B. J., & Smith, C. G. (2005). Variation for seed
602 coat and pod wall percentage and other traits in a germplasm collection and
603 historical cultivars of lupins. *Australian Journal of Agricultural Research, 56*,
604 75.
605 Clements, J. C., Galek, R., Kozak, B., Michalczyk, D. J., Piotrowicz-Cieslak, A. I.,
606 Sawicka-Sienkiewicz, E., Stawinski, S., & Zalewski, D. (2014). Diversity of
607 selected *Lupinus angustifolius* L. genotypes at the phenotypic and DNA level
608 with respect to microscopic seed coat structure and thickness. *PLoS One, 9*,
609 e102874. https://doi.org/102810.101371/journal.pone.0102874.
610 Dahl, W. J., Whiting, S. J., Healey, A., Zello, G. A., & Hildebrandt, S. L. (2003).
611 Increased stool frequency occurs when finely processed pea hull fiber is added
612 to usual foods consumed by elderly residents in long-term care. *J Am Diet Assoc,*
613 *103*, 1199-1202.
614 Dalgetty, D. D., & Baik, B.-K. (2006). Fortification of bread with hulls and cotyledon
615 fibers isolated from peas, lentils, and chickpeas. *Cereal Chemistry, 83*, 269-274.
616 Dalgetty, D. D., & Baik, B. K. (2003). Isolation and characterization of cotyledon fibers
617 from peas, lentils, and chickpeas. *Cereal Chemistry, 80*, 310-315.
618 DellaValle, D. M., Vandenberg, A., & Glahn, R. P. (2013). Seed coat removal improves
619 iron bioavailability in cooked lentils: studies using an *in vitro* digestion/Caco-2
620 cell culture model. *Journal of Agricultural and Food Chemistry, 61*, 8084-8089.
621 Dueñas, M., Hernandez, T., & Estrella, I. (2006). Assessment of i*n vitro* antioxidant

622 capacity of the seed coat and the cotyledon of legumes in relation to their
623 phenolic contents. *Food Chemistry, 98*, 95-103.
624 Dueñas, M., Hernández, T., & Estrella, I. (2002). Phenolic composition of the
625 cotyledon and the seed coat of lentils (*Lens culinaris* L.). *European Food*
626 *Research and Technology, 215*, 478-483.
627 Dueñas, M., Hernandez, T., Estrella, I., & Fernandez, D. (2009). Germination as a
628 process to increase the polyphenol content and antioxidant activity of lupin
629 seeds (*Lupinus angustifolius* L.). *Food Chemistry, 117*, 599-607.
630 EFSA. (2012). Scientific opinion on the risks for animal and public health related to
631 the presence of phomopsins in feed and food. *EFSA Journal, 10*, 2567.
632 Evans, A. J., & Cheung, P. C.-K. (1993). The carbohydrate composition of cotyledons
633 and hulls of cultivars of *Lupinus angustfolius* from Western Australia. *Journal*
634 *of the Science of Food and Agriculture, 61*, 189-194.
635 FAO. Pulses and derived products (1994). http://www.fao.org/es/faodef/fdef04e.htm
636 Accessed 23 May 2017.
637 FAO. FAO/INFOODS global food composition database for pulses Version 1.0 -
638 uPulses 1.0 (2017). http://www.fao.org/infoods/infoods/tables-and-
639 databases/faoinfoods-databases/en/ Accessed 1 May 2018.
640 FAOSTAT. FAO statistical databases (2018). http://faostat3.fao.org/home/E Accessed
641 26 April 2018.
642 Finch-Savage, W. E., & Leubner-Metzger, G. (2006). Seed dormancy and the control

643 of germination. *New Phytologist, 171*, 501-523.

644 Flint, H. J., Scott, K. P., Duncan, S. H., Louis, P., & Forano, E. (2012). Microbial
645 degradation of complex carbohydrates in the gut. *Gut Microbes, 3*, 289-306.

646 Flogan, C., & Dahl, W. (2010). Fiber fortification improves gastrointestinal function
647 and decreases energy intake in children with a history of constipation. *Infant,*
648 *Child, & Adolescent Nutrition, 2*, 312-317.

649 Foyer, C. H., Lam, H.-M., Nguyen, H. T., Siddique, K. H. M., Varshney, R. K., Colmer,
650 T. D., Cowling, W., Bramley, H., Mori, T. A., Hodgson, J. M., Cooper, J. W.,
651 Miller, A. J., Kunert, K., Vorster, J., Cullis, C., Ozga, J. A., Wahlqvist, M. L.,
652 Liang, Y., Shou, H., Shi, K., Yu, J., Fodor, N., Kaiser, B. N., Wong, F.-L.,
653 Valliyodan, B., & Considine, M. J. (2016). Neglecting legumes has
654 compromised human health and sustainable food production. *Nature Plants, 2*,
655 16112.

656 FSANZ. Approval report-Proposal P1026: Lupin as an allergen (2017).
657 http://www.foodstandards.gov.au/code/proposals/Pages/proposalp1026lupinas
658 5830.aspx Accessed 23 April 2018.

659 Ghavidel, R. A., & Prakash, J. (2007). The impact of germination and dehulling on
660 nutrients, antinutrients, *in vitro* iron and calcium bioavailability and *in vitro*
661 starch and protein digestibility of some legume seeds. *LWT - Food Science and*
662 *Technology, 40*, 1292-1299.

663 Guillon, F., & Champ, M. M.-J. (2002). Carbohydrate fractions of legumes: uses in

human nutrition and potential for health. *British Journal of Nutrition, 88*, S293-S306.

Güzel, D. S., Sedat. (2012). Effect of cooking methods on selected physicochemical and nutritional properties of barlotto bean, chickpea, faba bean, and white kidney bean. *Journal of Food Science and Technology, 49*, 89-95.

Hashemi, Z., Yang, K., Yang, H., Jin, A., Ozga, J., & Chan, C. B. (2015). Cooking enhances beneficial effects of pea seed coat consumption on glucose tolerance, incretin, and pancreatic hormones in high-fat-diet-fed rats. *Applied Physiology, Nutrition, and Metabolism, 40*, 323-333.

Hossain, S., Panozzo, J. F., Pittock, C., & Ford, R. (2011). Quantitative trait loci analysis of seed coat color components for selective breeding in chickpea (*Cicer arietinum* L.). *Canadian Journal of Plant Science, 91*, 49-55.

Huang, S.-C., Lia, T.-S., Cheng, T.-C., Chan, H.Y., Hwang, S.-M., Hwang, D.-F.,. (2009). *In vitro* interactions on glucose by different fiber materials prepared from mung bean hulls, rice bran and lemon pomace. *Journal of Food and Drug Analysis, 17*, 307-314.

Hughes, J. S., & Swanson, B. G. (1986). Microstructure of lentil seeds (*Lens culinaris*). *Food Structure, 5*, 241-246.

Hung, T. V., Handson, P. D., Amenta, V. C., Kyle, W. S. A., & Yu, R. S. T. (1988). Mineral-composition and distribution in lupin seeds and in flour, spray-dried powder and protein isolate produced from the seeds. *Journal of the Science of*

685 *Food and Agriculture, 45*, 145-154.
686 Jambunathan, R., & Singh, U. (1981). Studies on desi and kabuli chickpea (*Cicer*
687 *arietinum* L.) cultivars. 3. Mineral and trace elements composition. *Journal of*
688 *Agricultural and Food Chemistry, 29*, 1091-1093.
689 Joseph, E., & Swanson, S. G. C. B. G. (1993). Microstructure of black green and red
690 gram. *Food Structure, 12*, 155-162.
691 Khattab, R. Y., & Arntfield, S. D. (2009). Nutritional quality of legume seeds as
692 affected by some physical treatments 2. Antinutritional factors. *LWT - Food*
693 *Science and Technology, 42*, 1113-1118.
694 Klupšaitė, D., & Juodeikienė, G. (2015). Legume: composition, protein extraction and
695 functional properties. A review. *Chemical Technology, 66*.
696 Lampart-Szczapa, E., Siger, A., Trojanowska, K., Nogala-Kalucka, M., Malecka, M.,
697 & Pacholek, B. (2003). Chemical composition and antibacterial activities of
698 lupin seeds extracts. *Nahrung-Food, 47*, 286-290.
699 López-Amorós, M. L., Hernández, T., & Estrella, I. (2006). Effect of germination on
700 legume phenolic compounds and their antioxidant activity. *Journal of Food*
701 *Composition and Analysis, 19*, 277-283.
702 Lovegrove, A., Edwards, C. H., Noni, I. D., Patel, H., El, S. N., Grassby, T., Zielke, C.,
703 M. Ulmius, L. N., Butterworth, P. J., Ellis, P. R., & Shewry, P. R. (2015). Role
704 of polysaccharides in food, digestion and health. *Critical Reviews in Food*
705 *Science and Nutrition*.

706 Luo, J., Cai, W., Wu, T., & Xu, B. (2016). Phytochemical distribution in hull and
707 cotyledon of adzuki bean (*Vigna angularis* L.) and mung bean (*Vigna radiate*
708 L.), and their contribution to antioxidant, anti-inflammatory and anti-diabetic
709 activities. *Food Chemistry, 201*, 350-360.
710 Ma, F. S., Cholewa, E., Mohamed, T., Peterson, C. A., & Gijzen, M. (2004). Cracks in
711 the palisade cuticle of soybean seed coats correlate with their permeability to
712 water. *Annals of Botany, 94*, 213-228.
713 Macagnan, F. T., da Silva, L. P., & Hecktheuer, L. H. (2016). Dietary fibre: The
714 scientific search for an ideal definition and methodology of analysis, and its
715 physiological importance as a carrier of bioactive compounds. *Food Research*
716 *International, 85*, 144-154.
717 Marles, M. A., Warkentin, T. D., & Bett, K. E. (2013). Genotypic abundance of
718 carotenoids and polyphenolics in the hull of field pea (*Pisum sativum* L.).
719 *Journal of the Science of Food and Agriculture, 93*, 463-470.
720 Moïse, J. A., Han, S., Gudynaitę-Savitch, L., Johnson, D. A., & Miki, B. L. A. (2005).
721 Seed coats: structure, development, composition, and biotechnology. *In Vitro*
722 *Cellular & Developmental Biology - Plant, 41*, 620-644.
723 Mollard, R. C., Luhovyy, B. L., Smith, C., & Anderson, G. H. (2014). Acute effects of
724 pea protein and hull fibre alone and combined on blood glucose, appetite, and
725 food intake in healthy young men - a randomized crossover trial. *Applied*
726 *Physiology Nutrition and Metabolism, 39*, 1360-1365.

727 Muhammed, T., Manohar, S., & Junna, L. (2010). Polyphenols of mung bean
728 (*Phaseolus aureus* L.) cultivars differing in seed coat color: effect of dehulling.
729 *Journal of New Seeds, 11*, 369-379.
730 Nasar-Abbas, S. M., Siddique, K. H. M., Plummer, J. A., White, P. F., Harris, D., Dods,
731 K., & D'Antuono, M. (2009). Faba bean (*Vicia faba* L.) seeds darken rapidly
732 and phenolic content falls when stored at higher temperature, moisture and light
733 intensity. *LWT - Food Science and Technology, 42*, 1703-1711.
734 Ng, A., Lecain, S., Parker, M. L., Smith, A. C., & Waldron, K. W. (1999). Modification
735 of cell-wall polymers of onion waste III. Effect of extrusion-cooking on cell-
736 wall material of outer fleshy tissues. *Carbohydrate Polymers, 39*, 341-349.
737 Nozzolillo, C., & Debezada, M. (1984). Browning of lentil seeds, concomitant loss of
738 viability, and the possible role of soluble tannins in both phenomena. *Canadian*
739 *Journal of Plant Science, 64*, 815-824.
740 Oakenfull, D. (1981). Saponins in food - a review. *Food Chemistry, 7*, 19-40.
741 Oomah, B. D., Caspar, F., Malcolmson, L., & Bellido, A.-S. (2011). Phenolics and
742 antioxidant activity of lentil and pea hulls. *Food Research International, 44*,
743 436-441.
744 Oomah, B. D., Patras, A., Rawson, A., Singh, N., & Compos-Vega, R. (2011).
745 Chemistry of pulses. In B. K. Tiwari, A. Gowen & B. McKenna (Eds.), *Pulse*
746 *Foods: Processing, Quality and Nutraceutical Applications* (1st ed., pp. 9-40).
747 USA: Academic Press.

748 Patterson, C. A., Curran, J., & Der, T. (2017). Effect of processing on antinutrient
749 compounds in pulses. *Cereal Chemistry Journal, 94*, 2-10.
750 Petterson, D. S. (1998). Composition and food uses of lupins. In J. S. Gladstones, C.
751 Atkins & J. Hamblin (Eds.), *Lupins as crop plants: Biology, production and*
752 *utilization* (pp. 353-384). Oxon: CAB International.
753 Pfoertner, H. N., & Fischer, J. (2001). Dietary fibres of lupins and other grain legumes.
754 In B. V. McCleary & L. Prosky (Eds.), *Advanced Dietary Fibre Technology*
755 (pp. 361-366). Oxford: Blackwell Science Ltd.
756 Pulse Australia. Australian sweet lupin production (2010/11 to 2015/16) (2016).
757 http://www.pulseaus.com.au/storage/app/media/industry/AU-ASlupin-
758 production.pdf Accessed 24 May 2017.
759 Ralet, M. C., Valle, G. D., & Thibauit, J. F. (1993a). Raw and extruded fibre from pea
760 hulls. Part I: Composition and physico-chemical properties. *Carbohydrate*
761 *Polymers, 20*, 17-23.
762 Ralet, M. C., Valle, G. D., & Thibauit, J. F. (1993b). Raw and extruded fibre from pea
763 hulls. Part II: Structural study of the water-soluble polysaccharides.
764 *Carbohydrate Polymers, 20*, 25-34.
765 Ranilla, L. G., Genovese, M. I., & Lajolo, F. M. (2009). Isoflavones and antioxidant
766 capacity of Peruvian and Brazilian lupin cultivars. *Journal of Food Composition*
767 *and Analysis, 22*, 397-404.
768 Rashid, S., Rakha, A., Anjum, F. M., Ahmed, W., & Sohail, M. (2015). Effects of

769 extrusion cooking on the dietary fibre content and water solubility index of
770 wheat bran extrudates. *International Journal of Food Science & Technology,*
771 *50*, 1533-1537.
772 Resta, D., Boschin, G., D'Agostina, A., & Arnoldi, A. (2008). Evaluation of total
773 quinolizidine alkaloids content in lupin flours, lupin-based ingredients, and
774 foods. *Molecular Nutrition & Food Research, 52*, 490-495.
775 Reyes-Moreno, C., Okamura-Esparza, J., Armienta-Rodelo, E., Gomez-Garza, R. M.,
776 & Milan-Carrillo, J. (2000). Hard-to-cook phenomenon in chickpeas (*Cicer*
777 *arietinum* L): Effect of accelerated storage on quality. *Plant Foods for Human*
778 *Nutrition, 55*, 229-241.
779 Rochfort, S., & Panozzo, J. (2007). Phytochemicals for health, the role of pulses.
780 *Journal of Agricultural and Food Chemistry, 55*, 7981-7994.
781 Rouilly, A., Jorda, J., & Rigal, L. (2006). Thermo-mechanical processing of sugar beet
782 pulp. II. Thermal and rheological properties of thermoplastic SBP.
783 *Carbohydrate Polymers, 66*, 117-125.
784 Sanchez-Chino, X., Jimenez-Martinez, C., Davila-Ortiz, G., Alvarez-Gonzalez, I., &
785 Madrigal-Bujaidar, E. (2015). Nutrient and nonnutrient components of legumes,
786 and its chemopreventive activity: a review. *Nutrition and Cancer 67*, 401-410.
787 Saura-Calixto, F. (2012). Concept and health-related properties of nonextractable
788 polyphenols: the missing dietary polyphenols. *Journal of Agricultural and Food*
789 *Chemistry, 60*, 11195-11200.

790 Schloss, S., Koch, M., Rohn, S., & Maul, R. (2015). Development of a SIDA-LC-
791 MS/MS method for the determination of phomopsin A in legumes. *Journal of*
792 *Agricultural and Food Chemistry, 63*, 10543-10549.
793 Schmidt, E. D. (1987). High fiber bread and extruded products. In: Google Patents.
794 Sharma, S. K., Bansal, S., Mangal, M., Dixit, A. K., Gupta, R. K., & Mangal, A. K.
795 (2016). Utilization of food processing by-products as dietary, functional and
796 novel fibre: a review. *Critical Reviews in Food Science and Nutrition, 56*, 1647-
797 1661.
798 Sherasia, P. L., Garg, M. R., & Bhanderi, B. M. Pulses and their by-products as animal
799 feed (2017). http://www.fao.org/3/a-i7779e.pdf Accessed 27 April 2018.
800 Singh, S., Gamlath, S., & Wakeling, L. (2007). Nutritional aspects of food extrusion: a
801 review. *International Journal of Food Science & Technology, 42*, 916-929.
802 Singh, S., Singh, H. D., & Sikka, K. C. (1968). Distribution of nutrients in the
803 anatomical parts of common Indian pulses. *Cereal Chemistry, 45*, 13-18.
804 Sipsas, S. (2008). Lupin products - concepts and reality. In J. A. Palta & J. B. Berger
805 (Eds.), *2008 'Lupins for Health and Wealth' Proceedings of the 12th*
806 *International Lupin Conference*. Fremantle, Western Australia.
807 Smykal, P., Vernoud, V., Blair, M. W., Soukup, A., & Thompson, R. D. (2014). The
808 role of the testa during development and in establishment of dormancy of the
809 legume seed. *Frontiers in Plant Science, 5*, 351.
810 Sosulski, F. W., & Wu, K. K. (1988). High-fiber breads containing field pea hulls,

811 wheat, corn, and wild oat brans. *Cereal Chemistry, 65*, 186-191.
812 Souza, F. h. D. d., & Marcos-Jilho, J. (2001). The seed coat as a modulator of seed-
813 environment relationships in Fabaceae. *Brazilian Journal of Botany, 24*, 365-
814 375.
815 Sreerama, Y. N., Neelam, D. A., Sashikala, V. B., & Pratape, V. M. (2010). Distribution
816 of nutrients and antinutrients in milled fractions of chickpea and horse gram:
817 seed coat phenolics and their distinct modes of enzyme inhibition. *Journal of*
818 *Agricultural and Food Chemistry, 58*, 4322-4330.
819 Tajoddin, M., Manohar, S., & Lalitha, J. (2013). Effect of soaking and germination on
820 polyphenol content and polyphenol oxidase activity of mung bean (*Phaseolus*
821 *aureus* L.) cultivars differing in seed color. *International Journal of Food*
822 *Properties, 17*, 782-790.
823 Tiwari, B. K., & Singh, N. (2011). *Pulse chemistry and technology* (1 st ed.).
824 Cambridge, UK: Royal Society of Chemistry.
825 Troszyńska, A., & Ciska, E. (2002). Phenolic compounds of seed coats of white and
826 coloured varieties of pea (*Pisum sativum* L.) and their total antioxidant activity.
827 *Czech Journal of Food Sciences, 20*, 15-22.
828 Tucek, M. (2009). Use of lupin bran in high-fibre food products. In: Google Patents.
829 Turnbull, C. M., Baxter, A. L., & Johnson, S. K. (2005). Water-binding capacity and
830 viscosity of Australian sweet lupin kernel fibre under *in vitro* conditions
831 simulating the human upper gastrointestinal tract. *International Journal of Food*

Sciences and Nutrition, 56, 87-94.

Van Dongen, J. T. (2003). Structure of the developing pea seed coat and the post-phloem transport pathway of nutrients. *Annals of Botany, 91*, 729-737.

Verma, A. K., Banerjee, R., & Sharma, B. D. (2012). Quality of low fat chicken nuggets: effect of sodium chloride replacement and added chickpea (*Cicer arietinum* L.) hull flour. *Asian-Australasian Journal of Animal Sciences, 25*, 291-298.

Verma, A. K., Banerjee, R., & Sharma, B. D. (2015). Quality characteristics of low fat chicken nuggets: effect of salt substitute blend and pea hull flour. *Journal of Food Science and Technology-Mysore, 52*, 2288-2295.

Versantvoort, C. H. M., Van de Kamp, E., & Rompelberg, C. J. M. (2004). Development and applicability of an *in vitro* digestion model in assessing the bioaccessibility of contaminants from food. In *RIVM report 320102002*.

Wahlqvist, M. L. (2016). Food structure is critical for optimal health. *Food & Function, 7*, 1245-1250.

Wang, N., Daun, J. K., & Malcolmson, L. J. (2003). Relationship between physicochemical and cooking properties, and effects of cooking on antinutrients, of yellow field peas (*Pisum sativum*). *Journal of the Science of Food and Agriculture, 83*, 1228-1237.

Wolf, B. (2010). Polysaccharide functionality through extrusion processing. *Current Opinion in Colloid & Interface Science, 15*, 50-54.

Wood, J. A., Knights, E. J., Campbell, G. M., & Choct, M. (2014). Differences between

853 easy- and difficult-to-mill chickpea (*Cicer arietinum* L.) genotypes. Part I:
854 broad chemical composition. *Journal of the Science of Food and Agriculture,*
855 *94*, 1437-1445.
856 Wood, J. A., Knights, E. J., & Choct, M. (2011). Morphology of chickpea seeds (*Cicer*
857 *arietinum* L.): comparison of desi and kabuli types. *International Journal of*
858 *Plant Sciences, 172*, 632-643.
859 Xu, B. J., Yuan, S. H., & Chang, S. K. C. (2007). Comparative analyses of phenolic
860 composition, antioxidant capacity, and color of cool season legumes and other
861 selected food legumes. *Journal of Food Science, 72*, S167-S177.
862 Yan, X., Ye, R., & Chen, Y. (2015). Blasting extrusion processing: the increase of
863 soluble dietary fiber content and extraction of soluble-fiber polysaccharides
864 from wheat bran. *Food Chemistry, 180*, 106-115.
865 Youssef, M. M., & Bushuk, W. (1984). Microstructure of the seed coat of faba bean
866 (*Vicia faba* L.) seeds of different cookability. *Cereal Chemistry, 61*, 381-383.
867

868 **Tables**

869

870 **Table 1** Names and major types of the selected six pulses

871 **Table 2.** Morphological and physical properties of the selected six pulse seed coats

872 **Table 3.** Main carbohydrates and dietary fibre of three selected pulses seed coats

873 **Table 4.** Selected physico-chemical properties of pulse seed coats

874

875 **Table 1.** Names and major types of the selected six pulses

876

Botanical name	FAO commodity class	Common/Alternative names	2016 World production (million tonne)	Main market types in Australia (• most common)
Pisum sativum	Peas, dry	Field pea; Protein pea; Austrian winter peas (black peas); Canadian field peas (spring peas)	14.36	• Dun (including Kaspa type) White/blue Green
Vicia fabae	Broad beans, dry	Faba bean /Tickbean Broad bean	4.46	• *V. faba* var. minor (faba bean) *V. faba* var. major (broad bean)
Cicer arietinum	Chickpeas	Garbanzo beans (US); Bengal gram (India)	12.09	• Desi Kabuli
Lens culinaris	Lentils	Masurdahl, adas	6.32	• Red Green
Lupinus spp.	Lupins	Blue lupin; narrow-leaved lupin European white lupin	1.28	• *L. angustifolius* Albus (*L. albus*)
Vigna radiate	Beans, dry	Mung bean (Australia) Green/golden gram (India)	~3.0	• *Vigna radiata* (green) *Vigna mungo* (black)

877 Ref: FAO (1994); FAOSTAT (2018); Sherasia, et al. (2017); Tiwari, et al. (2011); Pulse

878 Australia (2016).

879

880 **Table 2.** Morphological and physical properties of the selected six pulse seed coats

881

Pulses	Colour	Seed weight (g/100 seeds)	Hull percentage (%)*	Hull thickness (µm)
Lupin				
L. angustifoliu	speckled	3.1-23.8	19.4-38.8	257.0-335.0
L. albus	white	12.0-86.9	12.2-27.5	nd
Field pea	green/yellow	18.7-25.6	7.2-14.0	55.9-72.0
Vicia fabae				
Faba bean	beige	40.0-95.0	11.0-15.4	141.0-248.0
Broad bean	beige	110.0-145.0	nd	
Chickpea				
desi	dark/brown	12.0-27.0	10.1-22.0	343.0-423.0
kabuli	beige/yellow	20.0-65.0	4.5-9.5	251.0
Lentil	red/green	4.5-7.5	7.0-11.0	25.0-65.0
Mung bean	green	2.5-4.7	8.6-23.5	30.0-330.0

882 * Data are dry basis; nd: no data were found;

883 Ref.: Huang, et al. (1988); Miao, et al. (2001); Clements, et al. (2005, 2014); Van

884 Dongen, et al. (2003); Youssef, et al., 1984; Wood, et al., 2011; Tajoddin, et al., 2010;

885 Hasan, 2010.

886

887 **Table 3.** Main carbohydrates and dietary fibre of three selected pulses seed coats

888

Pulses	*L. angustifoliu*	Field pea	Chickpea
Starch (g/100g)	0.4-0.9	0.16-1.8	0.2-0.5
Oligosaccharides (g/100g)	0.4	nd	nd
NSP (g/100g)			
Total	79.8-89.1	68.0	45.9-72.4*
Soluble	5.0	3.0-4.0	1.9-2.5
Insoluble	80.6-84.1	64.0-65.0	49.1-52.9*
Cellulose (g/100g)	44.5-51.7	62.3	18.2-29.0
Hemicellulose (g/100g)	12.7-14.4	8.2	30.4
Pectins (g/100g)	15.6-27.7	nd	0.1
Lignin (g/100g)	0.3-2.1	3.5	1.4-4.1
Dietary fibre (g/100g)			
Total	88-90.5	81.0-91.5	74.9-84.2
Soluble	3.1-3.8	4.1-11.0	nd
Insoluble	84.2-87.4	70.0-87.4	nd

889 Data are in dry basis.

890 NSP: Non-starch polysaccharides; nd: no data were found.

891 *: A remaining 15% was not hydrolysed by the NSP analyses which was supposed to

892 be “highly bound ligno-cellulosic compounds” (Wood, et al. 2014).

893 Ref.: Miao, et al. (2001); Evans, et al. (1993); Evans (1994); Brillouet, et al. (1983);

894 Guillon & Champ (2002); Hashemi, et al. (2015); Bailey, et al. (1974); Sosulski, et

895 al.(1988); Dalgetty & Baik (2003); Ralet, et al. (1993a); Wood, et al. (2014).

Table 4. Selected physico-chemical properties of pulse seed coats

Physical properties	***L. angustifoliu***	**Field pea**	**Chickpea**	**Lentil**	**Mung bean**
Direct Density (g/mL)	nd	0.6	0.4	0.7	nd
Bulk Density (g/mL)	nd	0.8	0.7	0.8	0.45-0.64
Swelling capacity(mL/g)	nd	1.9-6.0	3.6	2.4	5.51-9.20
Water binding (mL/g)	7.0-8.0	4.0-7.1	6.2	3.6	3.13-4.44
Oil binding (mL/g)	1.6-1.7	1.5-2.0	1.8	1.6	1.49-1.83

nd: no data were found.

Ref.: Guillon & Champ (2002); Dalgetty, et al. (2003); Pfoertner (2001); Turnbull, Baxter, and Johnson (2005); Ralet, et al. (1993b); Huang, et al. (2009).