Engineering and functional properties of biodegradable pellets developed from various agro-industrial wastes using extrusion technology

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Abstract Different agro-industrial wastes were mixed with different plasticizers and extruded to form the pellets to be used further for development of biodegradable molded pots. Bulk density and macro-porosity are the important engineering properties used to determine the functional characteristics of the biodegradable pellets viz., expansion volume, water solubility, product colour, flowability and compactness. Significant differences in the functional properties of pellets with varying bulk densities (loose and tapped) and macro-porosities (loose, tapped) were observed. The observed mean bulk density of biodegradable pellets made from different formulations ranged between 0.213 and 0.560 g/ml for loose fill conditions and 0.248 to 0.604 g/ml for tapped fill conditions. Biodegradable pellets bear a good compaction for both loose and tapped fill methods. The mean macro-porosity of biodegradable pellets ranged between 1.19 and 54.48 % for

loose fill condition and 0.29 to 53.35 % for tapped fill condition. Hausner ratio (HR) for biodegradable pellets varied from 1.026 to 1.328, indicating a good flowability of biodegradable pellets. Pearson's correlation between engineering properties and functional properties of biodegradable pellets revealed that from engineering properties functional properties can be predicted.

Keywords Bulk density · Biodegradable pellets · Hausner ratio · Macro-porosity · Plasticizer · Loose · Tapped fill

Introduction

Biodegradable polymers from renewable resources have attracted a lot of interest in recent years because they have positive impacts on the economy and environment. As the world energy demand soars while petroleum resources shrink, the energy sector focuses more on the conversion of biomass into useful products, and ethanol is often mentioned as an energy alternative. However, the advantage of green biomass will be fulfilled only if the “waste” products can be efficiently utilized (Keshi and Huang 2009). Nursery owners or growers use pots and cell trays of different materials, sizes, shapes, and colours to suit crop species, growing methods, and marketing strategies (Evans and Hensley 2004). At nurseries and greenhouses, seeds, bulbs, and young plants are planted in cell trays or pots containing growing substrate to start the growth under uniform cultivation conditions. However, one of the limitations of impermeable and rigid containers is that roots tend to encircle the outer perimeter of the pot which results in reduced plant growth, health, and survival once transplanted (Evans and Karcher 2004). Most of the containers used for transplanting are made of non-renewable oil-based raw materials such as polystyrene, polyethylene and polypropylene with suitable mechanical properties, chemical and microbial

Highlights 1. As these pellets are actually extruded forms of biological materials like deoiled rice bran, paddy husk, potato peel and banana peel along with glycerol and Cashew Nut Shell Liquid (CNSL) as plasticizer, microscopic pores and channels within them cannot be avoided which makes the pellets hygroscopic in nature which not only affects the volume but also influences its mass.
2. Determining the effect of fill volume and fill method (loose and tapped) on the macro-porosity, bulk density and specific weight of biodegradable pellets helps to understand the handling, storage and packaging of these pellets.
3. Correlation of engineering and Functional properties of the biodegradable pellets made from different formulations determines the functional characteristics of the biodegradable pellets viz., expansion volume, water solubility, product colour, flowability and compactness
4. Component analysis helps in the grouping of different samples.

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degradation resistance as well as durability. But at the same time disposal and recycling process of post-use plastic pots determine their uncontrolled combustion with the subsequent emission of toxic substances both into the atmosphere and into the soil. Hence, a valid alternative to the employment of petroleum based thermoplastic pots may be represented by the use of biodegradable pots (Evans and Hensley 2004; Evans and Karcher 2004; Kowalska et al. 2002; Wu et al. 2003; Yamauchi et al. 2006). Once buried, the biodegradable pots are subjected to biodegradation process, being transformed in biomass and inorganic products. Now, the attention is being focussed on the development of novel biodegradable and cost-competitive pots or multiple nurseries. These pots are biocomposites whose continuous phase is characterized by biopolymers coming from renewable and available origin, such as polysaccharides (Malinconico et al. 2002; Avella et al. 2007) and whose solid phase, dispersed within the polymeric matrix, is represented by natural fillers and fibres coming from wastes of agro-food and textile processing industries (Mohanty et al. 2000). Biodegradable plastics are an emerging generation of polymers. Protein has attracted much attention in the field of packaging due to its biodegradability (Cuq et al. 1998). Biodegradable plastics have an expanding range of potential applications, and driven by the growing use of plastics in packaging and the perception that biodegradable plastics are ‘environmentally friendly’, their use is predicted to increase. Green materials (or plant-based) have become increasingly more popular. There is a substantial interest in replacing some or all of the synthetic plastics by biodegradable polymers in several applications. Industrial production of pellets by extrusion process for manufacture of biodegradable products is relatively new and therefore the effectiveness of the availability of pellets rely upon understanding the storage stability of these pellets. A novel kind of low cost, biodegradable and medium water-resistant composite from corn protein was developed by Wu et al. (2003) wherein glycerol-plasticized pots showed good biodegradability and promising water resistance.

The thermal energy generated by viscous dissipation during extrusion combines with the shearing effect. Several studies have reported chemical changes during extrusion cooking and related them to product functional qualities such as expansion volume, water solubility and product colour (Wu et al. 2003; Manisha et al. 2012). The engineering properties of biodegradable pellets determine their overall quality. Hardness, bulk density, and porosity are the engineering properties that are used to evaluate the compactness of the pellets. The porosity of biodegradable pellets, expressed in percentage, can be defined as the ratio of volume of voids to the total filled volume. The voids volume can be divided into (i) a major volume of voids existing among the pellets (outside) when they are held in bulk, and (ii) a minor volume corresponding to the collection of minute pores, cracks, and channels present

inside pellet solids. Porosity is further classified by pellet solid's pore volume as total-porosity and apparent porosity which is also known as macro-porosity (Igathinathane et al. 2010). Bulk density is one of engineering characteristics that depend on particle size distribution, moisture content, and product processing (Barletta et al. 1993). Functional properties like compressibility and flowability are affected by engineering properties like bulk density. Bulk density and porosity affect the structures and is important in designing of hoppers, storage and transporting structures.

Knowledge of pellet's porosity helps in designing of storage silos and transportation containers as well. There are number of factors on which porosity of packed biodegradable pellets depends such as particle shape, shaking or pressing of the bed, particle size distribution and particle shrinking due to chemical conversion or agglomeration (Hamel and Krumm 2008). Although, several works on porosity of wood pellets and agricultural grains has been reported though the reported literature on biodegradable pellets made from deoiled rice bran and food industry waste is limited.

As these pellets are actually extruded forms of biological materials like deoiled rice bran, paddy husk, potato peel and banana peel along with glycerol and Cashew Nut Shell Liquid (CNSL) as plasticizer, microscopic pores and channels within them cannot be avoided. Rabier et al. (2006) reported that availability of the micro-pores and particle interaction with liquid makes the pellets hygroscopic in nature which not only affects the volume but also influences its mass.

Previous research is available on the determination of the engineering parameters like durability, bulk density and macro-porosity of wood pellets (Igathinathane et al. 2010), particle density of pellets and briquettes used for fuel (Rabier et al. 2006), the bulk densities and compaction behaviour of knife mill chopped switchgrass, wheat straw, and corn stover (Nehru et al. 2010).

In the current research, through geometrical approach, a method of stereo-metric measurements of biodegradable pellets with slight modifications was adopted to determine the macro-porosity (apparent porosity) Igathinathane et al. (2010). The geometrical approach intends to determine the whole pellet volume (including internal micro-pores) by forming the pellets to conform a geometrical shape and then applying the analytical mensuration formula to determine their volume directly.

Since literature is scanty on the macro-porosity, bulk density and specific weight of the pellets used for the development of biodegradable products made from deoiled rice bran, paddy husk, banana peels and CNSL as well as glycerol, the present study was undertaken to describe the influence of these engineering parameters on the overall quality (compactness and flowability) of pellets developed from different industrial by-products, wastes and plasticizers by extrusion technology. The main aim of the study is as follows:

1. To determine the effect of fill volume and fill method (loose and tapped) on the macro-porosity, bulk density and specific weight of biodegradable pellets using stereometric method.
2. To establish the correlation between engineering parameters.
3. To evaluate the functional properties of the biodegradable pellets made from different formulations prepared using agro-industrial wastes.
4. To statistically analyse the structural data of biodegradable pellets for natural grouping of samples.

V_p pellets volume, Wp mass of cylindrical pellets, (ϕ_p) macro-porosity, Wsp specific weight, V_f fill volume, ρ_l and ρ_t bulk densities of loose and tapped fill methods, HR hausner ratio

Materials and methods

Procurement of raw material

Paddy husk and Deoiled rice bran used for the present study were donated by M/s. AP Solvex Ltd., Dhuri (Punjab, India). Banana peels and potato peels were procured locally from local juice vendors and Hostel Mess (Canteen), respectively.

Glycerol was used as a reference plasticizer in this study and was of analytical grade (M/s. Merck Specialities Pvt. Ltd., Mumbai, India). Other plasticizer used in this study was CNSL which was kindly donated by M/s. Allen Petrochemicals Pvt Lt., Meerut (India).

Sample preparation

Preparation of paddy husk, banana and potato peel powder

Paddy husk was ground in a Laboratory grinding mill (M/s. Philips India Limited, Kolkata, India) and passed through 60 mesh screen to obtain fine powder. Banana peels and potato peels were washed with water impregnated with citric acid (1 %) and KMnSO_4 (0.5 %) to check the enzymatic browning. The peels were cut into transverse slices of about 2 mm thickness and dried in tray drier at 65 °C overnight. The dried peels were ground in a Laboratory grinding mill as mentioned above and passed through 60 mesh screen to obtain powders. All powders were stored in airtight HDPE plastic packs for further use.

Proximate composition of raw material

The raw materials were analysed for moisture, protein, fat, fibre and ash content according to AOAC (1995).

Particle size analysis

Particle size analysis was done by sophisticated instrument Shimadzu Laser Diffraction Particle Size Analyzer (SLAD-2300), M/s. SHIMADZU Corporation (Kyoto, Japan). Measuring method used was laser diffraction and laser scattering intensity pattern, with a measuring range of 0.017 to 2500 μm . Light source used was semi-conductor laser with a wavelength of 680 nm and an output of 3 mW. Operating temperatures were 10 to 30 °C, with an operating humidity of 20 to 80 % (with no condensation). A suspension was prepared with 0.5 g powder and 1 ml double de-ionized water (refractive index: 1.35) by continuous stirring on a magnetic stirrer for 3–5 min. The prepared sample was filled into the cuvette and readings were taken. The median granule diameter determines the diameter for 50 % of the normalized particle amount present.

Extrusion process for the preparation of pellets

In the blend preparation, ingredients like deoiled rice bran (DOB), paddy husk powder (PDH), potato peel powder (PPP), banana peel powder (BPP), cashew nut shell liquid (CNSL) and glycerol (GL) were used. The purpose of using different blends was to study the changes in final pellet characteristics in relation to change in concentration of one or more ingredients present in it and to finalize the formulation having desired pellet characteristics. Different formulations used for different samples were A1 (DOB (75 g), PDH (75 g) and CNSL (9 ml)), A2 ((75 g), PDH (75 g) and CNSL (15 ml)), A3 (DOB (75 g), PDH (75 g) and CNSL (21 ml)), B1 (DOB (75 g), PPP (75 g) and CNSL (9 ml)), B2 (DOB (75 g), PPP (75 g) and CNSL (15 ml)), B3 (DOB (75 g), PPP (75 g) and CNSL (21 ml)), C1 (DOB (75 g), BPP (75 g) and CNSL (9 ml)), C2 (DOB (75 g), BPP (75 g) and CNSL (15 ml)), C3 (DOB (75 g), BPP (75 g) and CNSL (21 ml)), D1 (DOB (75 g), PDH (75 g) and GL (9 ml)), D2 ((75 g), PDH (75 g) and GL (15 ml)), D3 (DOB (75 g), PDH (75 g) and GL (21 ml)).

The moisture was adjusted by sprinkling the distilled water on all the dry ingredients. All the ingredients were weighed and then mixed in a Hobart mixer (M/s. Continental Equipment India Pvt. Ltd, Delhi, India) for optimum mixing time at suitable rpm using sigmoid shape blade. This mixture was then passed through a sieve to prevent the lumps formation due to addition of moisture. After mixing, samples were stored in polyethylene bags at room temperature for 24 h (Process filed for patent, No: 111/DEL/2015).

Extrusion was performed using a co-rotating twin-screw extruder (M/s. Basic Technology Pvt. Ltd., Kolkata, India). The twin screw extruder was kept running for suitable period of time to stabilize the set temperatures and samples were then fed in to feed hopper at the feed rate of 4 kg/h for easy and

non-choking operation. Screw speed was set at 125 rpm and the suitable quantity of water was added to makeup the moisture content to 10 %. The horizontal split design of the extruder allowed the collection of samples inside the extruder channel with a 'dead-stop' run. Samples were taken along the extruder channel and made into pellets using suitable die of 4 mm diameter and high speed cutter. The length of the extruded pellets was about $(10.00 \pm 1 \text{ mm})$ and the same samples were used for further analysis. The pellets had on average about 7 % moisture content. The preliminary experiments carried out to study the moisture isotherms (unpublished) showed that the pellets being hygroscopic absorbed moisture to about 30 % in different storage conditions. This required drying of pellets to suitable moisture content before using injection moulding to prepare pots.

Measurement of macro-porosity, bulk density, and related engineering parameters of pellets

Three Graduated bulk density cylinders of Vankle's design (100 ml), (M/s. Standard Instrument Corporation, Patiala, India) were taken. Cylindrical pellets were filled to various volumes such as 30, 50 and 70 ml. The superscript with the coded sample as shown in further text and tables like in B1³⁰ denotes the measurement done for 30 ml fill and tapped volume. Filling was randomly done by selecting bulk of pellets which were poured from a height of approximately 100 mm from the brim of the cylinder. Care was taken during filling so that the top filled surface was flat. This procedure was for the loose filled condition of pellets. However, tapping the pellet filled cylinder on hard wooden surface determined the settling behaviour of the pellets in shaking or vibration during transport. Fifty taps with a tap height of about 20 mm produced a stable settled state of pellets (Chevanan et al. 2008). Digital weighing balance (0.01 g readability, 300 g capacity, M/s. Sartorius Mechatronics Pvt Lt., Bangalore, India) was used to determine the mass of the pellets directly after nullifying the cylinder mass. All the measurements were done in triplicates.

Macro-porosity can be defined as the percentage of volume of voids within the pellets when they are kept in bulk to the overall filled volume. The measurement of fill volume was easily obtained from the graduations or dimensions of the container. The voids volume was obtained indirectly by deducting the pellets volume from the fill volume (Igathinathan et al. 2010). The volume of biodegradable pellets using geometrical mensuration formula can be expressed as:

$$V_p = \frac{\pi}{4} d^2 \sum_{i=1}^n l_i \quad (1)$$

Where V_p is the pellets volume (mm^3), d is the mean diameter (mm), l_i is the variable length (mm) of the i^{th} pellet, and n is the number of pellets used in the fill.

The macro-porosity (ϕ_p) was calculated from the fill volume (V_f) and pellet volumes in percentage as:

$$\phi_p = \frac{V_f - V_p}{V_f} \times 100 \quad (2)$$

Specific weight of pellets is calculated from:

$$W_{sp} = \frac{W_p}{V_p} \quad (3)$$

Where W_{sp} is the specific weight of cylindrical pellets (g/ml), and W_p is the mass of cylindrical pellets (g).

The loose and tapped densities of biodegradable pellets are expressed as:

$$\rho_l = \frac{W_l}{V_l} \quad (4)$$

$$\rho_t = \frac{W_t}{V_t} \quad (5)$$

Where ρ_l and ρ_t are the loose and tapped bulk densities (g/ml) respectively, w_l and w_t are the masses (g), v_l and v_t are the volumes (ml) of pellets in loose and tapped fill conditions, respectively.

Hausner ratio (HR) was expressed in decimals; it is a value correlated to material flowability and is defined as the ratio of a material tapped bulk density to its loose bulk density (Grey and Beddow 1969; Garcia et al. 2007) and is given as:

$$\text{HR} = \frac{\rho_t}{\rho_l} \quad (6)$$

Where ρ_t and ρ_l are bulk density values for tapped and loose fill conditions.

Functional properties

The following functional properties of the extrudates were determined.

Water binding capacity

Water binding capacity of pellets was determined using the method described by Yamazaki (1953). A suspension of 5 g ground sample (dry weight) in 75 ml distilled water was agitated for 30 min at 25 °C and centrifuged ($3000 \times g$) for 10 min. The supernatant was discarded, drained for 5 min and the gain in weight was expressed as percentage of water binding capacity.

Expansion ratio

The ratio of diameter of extruded pellet and the diameter of die was used to express the expansion of extruded pellet (Fan et al. 1996). The diameter of extruded pellets was determined as the mean of 10 random measurements made with a Vernier caliper. The expansion ratio was calculated using below formulae:

$$\text{Expansion ratio} = \text{Extrudates' diameter} / \text{die diameter} \quad (7)$$

Colour

Colour measurements of all the extruded pellets were carried out in triplicate using a hand-held Chroma Meter (CR-400, Konica Minolta, UK) and L^* , a^* , and b^* readings were recorded. L^* , a^* and b^* represents the white–black, red–green and yellow–blue ratio of the sample, respectively. The instrument was calibrated against a standard white reference tile (L^* 48.32, a^* 48.8, b^* 27.92). All the experiments were conducted in triplicate. The values of ΔE were calculated by below equation

$$\Delta E = \sqrt{(L^* - L)^2 + (a^* - a)^2 + (b^* - b)^2} \quad (8)$$

Where L^* is the standard value and L is the obtained value. Similarly, a^* and b^* are the standard values and a , b are the obtained values.

Hardness

Hardness of the extruded pellets was determined using a crushing method using a TA – XT2i Texture Analyzer (M/s. Stable Micro Systems Ltd., Godalming, UK). Five pellets from each sample were mounted on the platform of the analyzer with a probe SMS–P/75–75 mm diameter at a crosshead speed of 2 mm/s with a target mode of 2 mm distance. The compression generates a curve with the force over distance. The highest first peak value was recorded as this value indicated the first rupture of pellet at one point and this value of force was taken as a measurement for hardness (Stojceska et al. 2008).

Pellet durability index

Durability of pellets was determined using a Two-compartment pellet-durability tester (M/s Continental Agra Equipment Inc, India). A sample of pellets to be tested was sieved using 4.0 mm screen to remove the fines. Approximately 500 g of sieved pellets were placed in each compartment of the tumbler. After tumbling for 10 min, the sample was removed and sieved using a 4.0 mm screen to remove the

fines. Pellet durability was calculated by the following equation ASABE (2002):

$$\text{Durability \%} = \frac{\text{Mass of pellets after tumbling}}{\text{Mass of pellets before tumbling}} \times 100 \quad (9)$$

Data analysis

The results obtained were statistically evaluated using Excel worksheet of MS office version 2010 (MS Office), Principal component analysis was performed to classify the samples using Statistical XLSTAT package version 2014. Statistical log software package version 7 (StatSoft Inc., OK, USA) Addinsoft (1995–2014) was used to establish a correlation among the different investigated properties and to develop regression equations from the most influencing variables.

Results and discussion

Proximate composition

The proximate analysis of raw materials revealed that DOB contained moisture content 8.08 %, ash 8.42 %, protein 14.6 %, fat 0.67 %, crude fibre 8.75 % and carbohydrates 59.48 %. BPP contained moisture content 7.62 %, ash 13.32 %, protein 1.92 %, fat 1.34 %, crude fibre 31.72 % and carbohydrates 44.08 %. PPP contained moisture content 8.93 %, ash 7.04 %, protein 10.24 %, fat 1.52 %, crude fibre 14.24 % and carbohydrates 58.03 %. PDH contained moisture content 9.41 %, ash 12.82 %, protein 0 %, fat 0 %, crude fibre 25.33 % and carbohydrates 52.44 %.

Engineering properties of biodegradable pellets

Particle size analysis

Particle size distributions of raw materials are shown in Fig. 1, wherein the granules showed a unimodal distribution profile for granule size. The result showed that granule diameter of all the samples showed a widest size distribution. In case of DOB, granule size ranged between 94.423 and 643.058 μm and the median granule diameter was 325.760 μm . The granule size of paddy husk powder ranged from 8.082 to 565.573 μm with median diameter of 180.918 μm . For potato and banana peel powders, the granule size ranged from 37.148 to 681.582 and 8.887 to 654.989 μm respectively, while median diameter was 330.353 and 321.483 μm , respectively. The variation in particle size plays an important role in handling, packing, product development, product characteristics, macro- porosity and Hausner ratio (Igathinathane et al.

Fig. 1 Particle size analysis of raw materials

2010). The median particle sizes of the blends as shown in Table 1 varied from 328.424 to 395.833 μm .

Mass

The obtained mean mass of biodegradable pellets with different formulations was in the range of 6.45 to 38.96 g for loose fill (Table 2). Here, an obvious increase in mass of samples was observed with increase in fill volume. Similar trend was also observed in case of tapped fill (Table 3). On comparing the mass of both tapped fill pellets and loose fill pellets it was

Table 1 Particle size analysis of blend of raw materials

S.No.	Raw material	Median D(μm)	Std dev
1	A1 (DOB, PDH, 9 ml CNSL)	337.552	0.195
2	A2 (DOB, PDH, 15 ml CNSL)	342.201	0.177
3	A3 (DOB, PDH, 21 ml CNSL)	362.107	0.184
4	B1 (DOB, PPP, 9 ml CNSL)	357.069	0.195
5	B2 (DOB, PPP, 15 ml CNSL)	392.420	0.121
6	B3 (DOB, PPP, 21 ml CNSL)	395.833	0.177
7	C1 (DOB, BPP, 9 ml CNSL)	328.424	0.173
8	C2 (DOB, BPP, 15 ml CNSL)	362.239	0.122
9	C3 (DOB, BPP, 21 ml CNSL)	383.178	0.130
10	D1 (DOB, PDH, 9 ml GL)	342.017	0.328
11	D2 (DOB, PDH, 15 ml GL)	351.775	0.312
12	D3 (DOB, PDH, 21 ml GL)	386.801	0.361

DOB Deoiled rice bran, PDH Paddy husk powder, PPP Potato peel powder, BPP Banana peel powder, CNSL Cashew nut shell liquid, GL Glycerol

observed that the mass in tapped fill was significantly higher than that of loose fill. As evident, increasing the amount of plasticizer increased the mass of the pellets in both the cases. The highest value of mass (38.96 g) was observed in sample D3⁷⁰ containing GL as plasticizer in case of loose fill. Also, in tapped fill, D3⁷⁰ containing GL as plasticizer showed the highest value viz., 40.37 g. Lowest values were observed in B1³⁰ (6.45, 7.52 g), respectively, for loose and tapped fill conditions (Tables 2 and 3). The results are evident as increasing the quantity of plasticizer obviously increased the mass and the same is true for the tapped fill volume as the number of pellets had to be increased to fill up the cylinder upto the designated level. The mass of pellets prepared with glycerol as plasticizer (series D) was higher as compared to those prepared with CNSL as plasticizer (series A) which may be due to higher density of glycerol (1.26 g/ml) (Christoph et al. 2000) as compared to CNSL (0.957 g/ml) (Sengar et al. 2012).

Pellet volume

The mean pellet volumes (Eq. 1) for biodegradable pellets ranged from 593.4 to 2414 and 611.34 to 2751.9 mm^3 for loose and tapped fill methods, respectively as shown in Tables 2 and 3. It is evident from the Tables that the pellet volume increased with increase in fill volume for both the cases, because the number of pellets had to increase to fill the given cylinder volume. The lowest pellet volume was observed in sample B1³⁰ for both the cases as the pellets prepared from potato peels have slightly higher expansion ratio means lesser

Table 2 Effect of different formulations and fill volume (loose fill) on biodegradable pellets by mean separation analysis

S.No.	Fill vol. (ml)	Code	Mass (g)	Pellet volume (mm ³)	Bulk density (g/ml)	Macro-porosity (%)
1	30	A1 ³⁰	11.42±0.05 ^h	771.48±0.15 ^g	0.381±0.002 ^a	3.80±0.25 ^h
2	50	A1 ⁵⁰	19.00±0.14 ^f	1184.0±1.23 ^e	0.380±0.003 ^{a A}	26.32±1.14 ^d
3	70	A1 ⁷⁰	28.00±0.02 ^c	1186.0±1.25 ^e	0.400±0.001 ^b	53.08±1.29 ^a
4	30	A2 ³⁰	15.40±0.10 ^g	810.71±2.05 ^f	0.513±0.003 ^b	2.97±0.25 ⁱ
5	50	A2 ⁵⁰	23.66±0.50 ^e	1418.7±2.12 ^c	0.473±0.010 ^{a B,C}	21.62±1.25 ^f
6	70	A2 ⁷⁰	32.90±0.20 ^b	1915.4±1.25 ^a	0.470±0.003 ^a	42.63±1.54 ^c
7	30	A3 ³⁰	15.80±0.10 ^g	797.32±1.52 ^{fg}	0.527±0.003 ^b	6.98±0.54 ^g
8	50	A3 ⁵⁰	25.00±0.06 ^d	1343.7±1.58 ^d	0.500±0.003 ^{a C}	23.12±1.10 ^e
9	70	A3 ⁷⁰	35.00±0.23 ^a	1754.3±1.02 ^b	0.500±0.003 ^b	44.93±1.52 ^b
1	30	B1 ³⁰	6.45±0.06 ^h	593.4±0.08 ^g	0.215±0.002 ^a	12.02±0.85 ^g
2	50	B1 ⁵⁰	10.66±0.15 ^g	1025.1±2.55 ^e	0.213±0.003 ^{a A}	29.49±1.15 ^d
3	70	B1 ⁷⁰	16.80±0.30 ^c	1065.3±1.65 ^e	0.240±0.004 ^b	54.48±1.24 ^a
4	30	B2 ³⁰	10.80±0.16 ^g	658.1±0.56 ^f	0.360±0.005 ^a	8.06±0.08 ⁱ
5	50	B2 ⁵⁰	20.66±0.32 ^d	1318.4±2.54 ^c	0.413±0.006 ^{b B}	23.63±1.24 ^f
6	70	B2 ⁷⁰	29.16±0.10 ^b	1888.7±3.23 ^a	0.417±0.001 ^b	43.01±1.57 ^c
7	30	B3 ³⁰	15.00±0.23 ^f	594.9±0.21 ^f	0.500±0.008 ^a	10.16±1.15 ^h
8	50	B3 ⁵⁰	25.00±0.04 ^c	1185.5±2.55 ^d	0.501±0.001 ^{a C}	26.29±1.83 ^e
9	70	B3 ⁷⁰	34.53±0.12 ^a	1705.2±1.44 ^b	0.493±0.002 ^a	45.64±1.54 ^b
1	30	C1 ³⁰	9.60±0.40 ^h	610.7±2.14 ^h	0.320±0.013 ^a	9.64±0.57 ^f
2	50	C1 ⁵⁰	18.16±0.05 ^c	1312.5±1.44 ^d	0.363±0.001 ^{b A}	23.75±1.53 ^e
3	70	C1 ⁷⁰	25.43±0.62 ^c	1863.0±4.46 ^c	0.363±0.009 ^b	43.38±2.54 ^a
4	30	C2 ³⁰	10.50±0.21 ^g	675.0±1.26 ^g	0.350±0.007 ^a	7.49±0.07 ^g
5	50	C2 ⁵⁰	19.16±0.22 ^d	939.9±0.24 ^e	0.383±0.004 ^{b A}	31.20±1.23 ^d
6	70	C2 ⁷⁰	26.60±0.11 ^b	1896.3±3.05 ^b	0.380±0.002 ^b	42.91±1.59 ^b
7	30	C3 ³⁰	16.50±0.41 ^f	867.9±1.16 ^f	0.550±0.014 ^b	7.49±0.04 ^h
8	50	C3 ⁵⁰	26.00±0.27 ^{bc}	939.9±0.06 ^e	0.520±0.005 ^{a B}	31.20±0.52 ^d
9	70	C3 ⁷⁰	37.10±0.41 ^a	2068.8±4.23 ^a	0.530±0.006 ^a	40.44±0.89 ^c
1	30	D1 ³⁰	15.60±0.35 ^g	709.6±0.36 ^f	0.520±0.012 ^a	6.34±0.72 ^e
2	50	D1 ⁵⁰	26.00±0.28 ^c	1324.5±3.44 ^d	0.520±0.006 ^{a A}	23.51±1.24 ^c
3	70	D1 ⁷⁰	35.70±0.26 ^c	2090.0±5.02 ^b	0.510±0.004 ^a	40.14±1.05 ^a
4	30	D2 ³⁰	15.90±0.11 ^{fg}	709.6±1.10 ^f	0.530±0.004 ^a	6.34±0.24 ^e
5	50	D2 ⁵⁰	26.50±0.33 ^c	1324.5±1.16 ^d	0.530±0.007 ^{a A}	23.51±1.02 ^c
6	70	D2 ⁷⁰	37.80±0.45 ^b	2090.0±4.44 ^b	0.540±0.006 ^a	40.14±1.36 ^a
7	30	D3 ³⁰	16.00±0.52 ^f	864.1±1.53 ^e	0.533±0.017 ^a	1.19±0.65 ^f
8	50	D3 ⁵⁰	28.00±0.05 ^d	1623.3±1.62 ^c	0.560±0.001 ^{b B}	17.53±1.25 ^d
9	70	D3 ⁷⁰	38.96±0.07 ^a	2414.8±3.05 ^a	0.557±0.001 ^b	35.50±1.89 ^b

The values are means ± standard deviation of three replicates

Means with different small letters in a column differ significantly ($p < 0.05$)

Means with different capital letters in the column differ significantly ($p < 0.05$) among samples with different level of plasticizers

number of pellets in a given volume. The highest pellet volume was observed in sample D3⁷⁰ containing (GL) as plasticizer for loose fill condition. However, in case of tapped fill the highest volume was observed in C3⁷⁰ containing CNSL as plasticizer. This may be due to greater compactness of pellets prepared from banana peel powder. Hence, during tapping the heavier material

settled to bottom readily which in turn required more material (pellets) to fill up the marked volume.

Bulk density

The mean bulk density of loose fill (Eq.4) of biodegradable pellets made from different formulations ranged between

Table 3 Effect of different formulations and fill volume (tapped fill) on biodegradable pellets by mean separation analysis

S.No.	Fill vol. (ml)	Code	Mass (g)	Pellet volume (mm ³)	Bulk density (g/ml)	Macro-porosity (%)
1	30	A1 ³⁰	13.16±0.06 ^h	865.76±1.04 ^e	0.439±0.002 ^a	1.41±0.12 ^e
2	50	A1 ⁵⁰	22.52±0.12 ^e	1360.9±2.11 ^d	0.450±0.002 ^{b A}	22.78±0.25 ^d
3	70	A1 ⁷⁰	32.97±0.25 ^c	1902.4±2.26 ^b	0.471±0.004 ^c	42.8±0.45 ^c
4	30	A2 ³⁰	16.87±0.33 ^g	865.80±1.25 ^e	0.562±0.011 ^c	1.13±0.89 ^a
5	50	A2 ⁵⁰	25.82±0.06 ^d	1768.9±4.21 ^c	0.516±0.012 ^{b B,C}	14.62±0.65 ^d
6	70	A2 ⁷⁰	34.80±0.12 ^b	2273.0±5.05 ^a	0.497±0.002 ^a	37.52±1.84 ^b
7	30	A3 ³⁰	17.85±0.55 ^f	879.90±1.01 ^e	0.595±0.018 ^c	0.670±0.05 ^a
8	50	A3 ⁵⁰	26.04±0.50 ^d	1334.5±2.11 ^d	0.521±0.010 ^{a C}	23.31±0.15 ^c
9	70	A3 ⁷⁰	37.46±0.28 ^a	1922.5±3.25 ^b	0.535±0.004 ^b	42.53±2.22 ^c
1	30	B1 ³⁰	7.52±0.09 ⁱ	611.34±0.50 ^h	0.248±0.007 ^a	9.62±0.43 ^g
2	50	B1 ⁵⁰	12.45±0.29 ^h	1155.5±2.02 ^e	0.249±0.006 ^{a A}	26.89±0.25 ^d
3	70	B1 ⁷⁰	18.59±0.07 ^e	1165.1±1.89 ^e	0.266±0.001 ^b	53.35±2.65 ^a
4	30	B2 ³⁰	12.98±0.60 ^g	781.55±0.89 ^f	0.433±0.020 ^a	3.94±0.52 ⁱ
5	50	B2 ⁵⁰	22.16±0.02 ^d	1416.6±1.41 ^c	0.443±0.001 ^{b B}	21.66±0.26 ^f
6	70	B2 ⁷⁰	30.86±0.22 ^b	1973.3±2.72 ^a	0.441±0.003 ^b	41.81±1.45 ^c
7	30	B3 ³⁰	17.64±0.45 ^f	708.24±0.73 ^g	0.598±0.009 ^c	6.39±0.26 ^h
8	50	B3 ⁵⁰	28.90±0.22 ^c	1270.1±1.51 ^d	0.578±0.004 ^{b C}	24.59±0.64 ^e
9	70	B3 ⁷⁰	39.05±0.23 ^a	1847.3±1.94 ^b	0.558±0.003 ^a	43.61±0.75 ^b
1	30	C1 ³⁰	12.75±0.40 ^h	753.8±0.69 ⁱ	0.425±0.013 ^c	4.87±0.24 ^g
2	50	C1 ⁵⁰	19.89±0.02 ^f	1412.0±1.54 ^f	0.398±0.001 ^{b A}	21.76±0.61 ^d
3	70	C1 ⁷⁰	26.80±0.07 ^d	1944.0±3.58 ^c	0.383±0.001 ^a	42.22±1.36 ^a
4	30	C2 ³⁰	11.20±0.23 ⁱ	867.9±1.15 ^h	0.373±0.008 ^a	1.07±0.22 ^h
5	50	C2 ⁵⁰	20.49±0.22 ^e	1439.9±3.35 ^e	0.410±0.004 ^{c A}	21.20±0.21 ^e
6	70	C2 ⁷⁰	27.30±0.14 ^b	1998.8±5.31 ^b	0.390±0.002 ^b	41.44±1.26 ^b
7	30	C3 ³⁰	18.11±0.15 ^g	897.1±0.46 ^g	0.604±0.005 ^c	0.94±0.10 ⁱ
8	50	C3 ⁵⁰	27.00±0.16 ^c	1659.9±1.79 ^d	0.540±0.003 ^{a B}	16.80±0.54 ^f
9	70	C3 ⁷⁰	39.33±0.56 ^a	2751.9±4.08 ^a	0.562±0.008 ^b	30.68±1.01 ^c
1	30	D1 ³⁰	17.01±0.58 ⁱ	891.2±0.11 ^g	0.567±0.019 ^c	0.29±0.01 ^h
2	50	D1 ⁵⁰	27.54±0.22 ^f	1743.5±2.04 ^d	0.551±0.004 ^{b A}	15.13±0.83 ^f
3	70	D1 ⁷⁰	37.32±0.75 ^c	2515.0±5.43 ^a	0.533±0.011 ^a	34.07±1.15 ^c
4	30	D2 ³⁰	17.60±0.27 ^h	827.9±1.15 ^h	0.587±0.009 ^b	2.40±0.04 ^g
5	50	D2 ⁵⁰	28.84±0.55 ^e	1424.8±1.63 ^f	0.577±0.011 ^{b B, C}	21.50±0.24 ^d
6	70	D2 ⁷⁰	39.70±0.55 ^b	2191.0±5.36 ^c	0.567±0.008 ^a	38.70±0.65 ^a
7	30	D3 ³⁰	17.90±0.29 ^g	887.0±1.08 ^g	0.597±0.010 ^b	0.431±0.54 ^h
8	50	D3 ⁵⁰	29.66±0.36 ^d	1705.8±2.07 ^c	0.593±0.007 ^{b C}	15.88±0.45 ^e
9	70	D3 ⁷⁰	40.37±0.35 ^a	2423.2±4.33 ^b	0.577±0.005 ^a	35.38±0.24 ^b

The values are means±standard deviation of three replicates

Means with different small letters in a column differ significantly ($p<0.05$)

Means with different capital letters in the column differ significantly ($p<0.05$) among samples with different level of plasticizers

0.213 and 0.560 g/ml. However, in case of tapped fill (Eq. 5) mean bulk densities ranged between 0.248 and 0.604 g/ml. As shown in Tables 2 and 3 there is an obvious increase of bulk densities in tapped fill than that of loose fill.

There was a significant change in the bulk density of pellets of “A” series in both the cases (loose and tapped) with increase in level of CNSL from 9 to 15 ml. However,

further increase in CNSL (21 ml) showed a non significant change. In case of pellets of “B” series, a significant increase in bulk density with increase in CNSL level was observed for both loose and tapped fill volumes, which may be due to increase in binding of CNSL with proteins present in higher amounts in PPP. The bulk density of pellets prepared from banana peel powder did not change

significantly with the increased amount of CNSL from 9 to 15 ml. However, it increased significantly at the higher level of CNSL (21 ml) due to greater binding of fibres by the action of plasticizer (CNSL) (Wypych 2012). Upon increasing the plasticizers, the mean bulk density increased in general in both the cases (loose and tapped).

Macro-porosity

The mean macro-porosity of biodegradable pellets with different formulations (Eq. 2) varied from 1.19 to 54.48 % for loose filled biodegradable pellets (30–70 ml) as shown in Table 2. However, in case of tapped fill biodegradable pellets the values were in the range of 0.29 to 53.35 % (Table 3). This wide range in macro-porosity may be due to the varied structural changes as well as morphological characteristics of different raw materials in the formulation like paddy husk (A), potato peel powder (B) and banana peel powder (C). The results were in agreement with the work of Igathinathane et al. (2010), who have determined the values of macro porosity of the wood pellets in the range of 39.4 to 44.9 and 36.9 to 40.4 % for loose and tapped fill, respectively. Another classification was done on dry solids by Anonymous (1979), where porosity values were categorized under different classes. Hence, porosity values if ranged in between 35 and 45 % was termed as “Medium” and grouped under “Class-3”, for 30 to 35 % as “Low” and “Class-4”, and for <30 % as “Very low” and “Class-5.” Hence, considering this classification, the porosity of biodegradable pellets made from different formulations fall under a varied range of classes for different formulation under loose fill and tapped fill conditions. However, on an average sample with codes A2⁷⁰, A3⁷⁰, B2⁷⁰, B3⁷⁰, C1⁷⁰, C2⁷⁰, C3⁷⁰, D1⁷⁰, D2⁷⁰, D3⁷⁰ belong to “Class-3” and described as “Medium” under loose fill conditions (A1⁷⁰ and B1⁷⁰ may also be considered in the same class having values in higher range). It is evident that all the macro-porosity values in **Class-3** were observed in the highest fill volume (70 ml). In case of tapped fill conditions sample with code A1⁷⁰, A2⁷⁰, A3⁷⁰, B1⁷⁰, B2⁷⁰, B3⁷⁰, C1⁷⁰, C2⁷⁰, D2⁷⁰, D3⁷⁰ belong to the same class. C2⁵⁰,

C3⁵⁰ (loose filled) and C3⁷⁰, D1⁷⁰ (tapped filled) belong to “Class-4”. Rest of the samples come under “Class-5” groups. It was observed that macro-porosity increased with increase in fill volume and similar results were also obtained by Igathinathane et al. (2010). In case of tapped fill, macro-porosity decreases due to tapping than that of loose fill method. This can be attributed to the release of air during tapping and further new pellets are added to make up the fill volume.

Correlation of engineering parameters

Coefficients of Pearson correlation were used to identify the correlation of engineering parameters of biodegradable pellets made from different formulations for both loose fill and tapped fill (Table 4). In both cases, the pellet volume showed a significant positive correlation with fill volume ($r=0.88$ for loose fill and $r=0.89$ for tapped fill) which indicated that the pellet volume increased with increase in fill volume. The similar effect was observed for mass of pellets with fill volume and pellet volume ($r=0.83$ for both loose and tapped fill and $r=0.91$ for both loose and tapped fill). The macro-porosity showed a higher positive correlation with fill volume in loose as well as in tapped fill ($r=0.96$). There was little influence of specific weight on any of the engineering properties except bulk density which showed non-significant correlation. Simple regression equations would be useful for derived engineering parameters such as macro porosity utilizing the significant pellet properties.

For Loose fill

$$\begin{aligned}\text{Macro-porosity (\%)} &= -20.948 + 0.925 \times \text{Fill volume(ml)} \quad (R^2 = 0.965) \\ \text{Macro-porosity (\%)} &= -0.367 + 0.121 \times \text{Mass (g)} \quad (R^2 = 0.815)\end{aligned}$$

For Tapped fill

$$\begin{aligned}\text{Macro-porosity(\%)} &= 29.102 + 0.987 \times \text{Fill volume(ml)} \quad (R^2 = 0.962) \\ \text{Macro-porosity (\%)} &= -8.873 + 1.234 \times \text{Mass (g)} \quad (R^2 = 0.805)\end{aligned}$$

In case of loose fill and tapped fill, similar models were adopted for pellet volume from fill volume and mass data.

Table 4 Pearson correlation for loose and tapped fill parameters with biodegradable pellets of different formulations

Loose fill Loose fill →							Tapped fill Tapped fill →					
Variables	Fill volume	Bulk density	Pellet volume	Mass	Specific weight	Macro-porosity	Fill volume	Bulk density	Pellet volume	Mass	Specific weight	Macro-porosity
Fill volume	1.00	-	-	-	-	-	1.00	-	-	-	-	-
Bulk density	0.035	1.00	-	-	-	-	0.084	1.00	-	-	-	-
Pellet volume	0.88	0.30	1.00	-	-	-	0.89	0.17	1.00	-	-	-
Mass	0.83	0.54	0.91	1.00	-	-	0.83	0.44	0.91	1.00	-	-
Specific weight	-0.09	0.64	-0.12	0.21	1.00	-	-0.25	0.74	-0.26	0.12	1.00	-
Macro-porosity	0.96	-0.11	0.73	0.71	-0.04	1.00	0.96	-0.23	0.75	0.70	-0.22	1.00

For Loose fill

$$\text{Pellet volume}(\%) = -117.117 + 27.556 \times \text{Fill volume}(\text{ml}) \quad (R^2 = 0.869)$$

$$\text{Pellet volume}(\%) = 79.186 + 52.850 \times \text{Mass}(\text{g}) \quad (R^2 = 0.910)$$

Pellet volume for mass showed highly significant values ($R^2=0.910$) and ($R^2=0.916$) for loose and tapped fill, respectively.

For Tapped fill

$$\text{Pellet volume}(\%) = -113.187 + 31.396 \times \text{Fill volume}(\text{ml}) \quad (R^2 = 0.896)$$

$$\text{Pellet volume}(\%) = 55.938 + 57.524 \times \text{Mass}(\text{g}) \quad (R^2 = 0.916)$$

Hausner ratio (HR)

Hausner ratio (HR) for biodegradable pellets varied from 1.026 to 1.328 within the different fill volumes (Table 5).

Table 5 Functional properties, Hausner ratio and pellet durability index of biodegradable pellets

S.No.	Code	Hardness (Newton)	Colour (ΔE)	Expansion ratio	Water binding capacity (g/g)	Pellet durability index (%)	Hausner ratio
1	A1 ³⁰	392.454±0.14 ^a	49.219±0.05 ^a	1.058±0.05 ^{c,f,g}	3.395±0.02 ^h	98.25±0.01 ^a	1.152±0.001 ^a
2	A1 ⁵⁰						1.185±0.002 ^b
3	A1 ⁷⁰						1.178±0.008 ^b
4	A2 ³⁰	448.592±0.05 ^b	50.383±0.12 ^a	1.020±0.02 ^{f,i,j,k}	3.734±0.10 ⁱ	98.91±0.01 ^b	1.095±0.014 ^b
5	A2 ⁵⁰						1.101±0.002 ^a
6	A2 ⁷⁰						1.058±0.003 ^a
7	A3 ³⁰	487.044±0.15 ^c	50.533±0.01 ^a	1.018±0.53 ^{h,k}	4.795±0.42 ^f	99.51±0.16 ^c	1.129±0.028 ^c
8	A3 ⁵⁰						1.033±0.001 ^a
9	A3 ⁷⁰						1.070±0.001 ^b
1	B1 ³⁰	432.119±0.16 ^d	50.525±0.16 ^a	1.814±0.42 ^a	6.195±0.24 ^{d,e}	98.42±0.01 ^a	1.155±0.021 ^b
2	B1 ⁵⁰						1.168±0.011 ^b
3	B1 ⁷⁰						1.107±0.016 ^a
4	B2 ³⁰	466.606±0.13 ^e	51.619±0.07 ^b	1.354±0.15 ^b	7.73±0.35 ^b	98.96±0.02 ^b	1.202±0.038 ^b
5	B2 ⁵⁰						1.073±0.016 ^a
6	B2 ⁷⁰						1.058±0.004 ^a
7	B3 ³⁰	475.719±0.14 ^f	50.668±0.12 ^{a,b}	1.074±0.24 ^{c,e}	7.975±0.50 ^a	99.02±0.02 ^c	1.196±0.025 ^c
8	B3 ⁵⁰						1.155±0.009 ^{b,c}
9	B3 ⁷⁰						1.131±0.003 ^a
1	C1 ³⁰	480.167±0.12 ^g	54.385±0.15 ^c	1.050±0.66 ^{d,e,f,h}	4.265±0.09Z ^g	99.15±0.04 ^a	1.328±0.014 ^c
2	C1 ⁵⁰						1.095±0.002 ^b
3	C1 ⁷⁰						1.054±0.023 ^a
4	C2 ³⁰	482.219±0.05 ^h	53.012±0.12 ^c	0.982±0.56 ^l	6.885±0.12 ^c	99.21±0.58 ^a	1.067±0.025 ^b
5	C2 ⁵⁰						1.069±0.001 ^b
6	C2 ⁷⁰						1.026±0.001 ^a
7	C3 ³⁰	495.622±0.13 ⁱ	54.811±0.05 ^c	0.053±0.25 ^m	7.475±0.37 ^b	99.62±0.57 ^b	1.098±0.018 ^b
8	C3 ⁵⁰						1.038±0.005 ^a
9	C3 ⁷⁰						1.060±0.003 ^a
1	D1 ³⁰	439.805±0.02 ^j	50.459±0.04 ^a	1.087±0.54 ^{c,j}	4.785±0.11 ^f	98.56±0.01 ^a	1.090±0.013 ^c
2	D1 ⁵⁰						1.059±0.003 ^{b,c}
3	D1 ⁷⁰						1.045±0.013 ^a
4	D2 ³⁰	448.583±0.16 ^k	49.316±0.14 ^a	1.075±0.65 ^{c,d,j}	6.14±0.16 ^e	98.69±0.01 ^b	1.107±0.009 ^c
5	D2 ⁵⁰						1.088±0.007 ^b
6	D2 ⁷⁰						1.050±0.002 ^a
7	D3 ³⁰	460.897±0.08 ^l	50.094±0.12 ^a	1.025±0.08 ^{g,h,i}	6.355±0.16 ^d	98.93±0.01 ^c	1.119±0.018 ^b
8	D3 ⁵⁰						1.059±0.011 ^a
9	D3 ⁷⁰						1.036±0.007 ^a

The values are means ± standard deviation of three replicates

Means with different letters in a column differ significantly ($p<0.05$)

On the basis of the Hausner ratio, Hayes (1987) described the groups for flow, as free flowing (1.0–1.1), medium flowing (1.1–1.25), difficult flowing (1.25–1.4), and very difficult flowing (>1.4). Hausner ratio above 1.25 is considered to represent the threshold between “free flow” and “no flow” conditions (Carr 1965; Podzeck 1998; Schussele and Bauer-Brandl 2003). Based on these classifications the nature of pellets prepared from different formulation has been described. In case of pellets of “A” series, the free flowing nature of the pellets increased with the amount of plasticizer (CNSL) in the formulation. Almost all the pellets prepared with formulation “B” showed medium flow behaviour. The results are in agreement with the microstructure of pellets formed with these formulations (result unpublished). Pellets with formulation C and D series showed free flowing behaviour as compared to “B” series. The pellets of “A” series showed medium flowability in contrast with “D” series containing glycerol as plasticizer. It showed the different behaviour of paddy husk powder with CNSL and GL. Hence, the prepared biodegradable pellets can be classified as “freely flowing” and “medium flowing”. The extruded biodegradable pellets had similar nature of flowability (1.05–1.14) as in the case of wood pellets described by Igathinathane et al. (2010).

Functional Properties

Water binding capacity

It was observed that the water binding capacity increased with the increase in the amount of the plasticizer (CNSL and glycerol) ($A1 < A2 < A3$, $B1 < B2 < B3$, $C1 < C2 < C3$, $D1 < D2 < D3$) as summarized in Table 5. This may be due to the hydrophilic and hygroscopic nature of the plasticizers and forming the large hydrodynamic plasticizer-water complex (Krochta 2002). The pellets of “B” series containing increased amount of protein present in potato peel powder had highest water binding capacity which may be due to polar groups on the protein surface forming hydrogen bonds with water (Petukhov et al.

1999; Tanya 2006; Zayas 1997). Following “B” series, the “C” series showed higher water binding capacity which may be due to the characteristic feature of fibre supplemented banana peel powder (Dreese and Hosney 1982; Haridas and Hemamalini 1991; Chaplin 2003). Fibre may interact with water by means of polar and hydrophobic interactions, hydrogen bonding and enclosure, thus contributes to higher water binding capacity (Gamlath and Ravindran 2009).

Expansion ratio

As evident from Table 5, the expansion ratio decreases with the increases in plasticizer level. This decrease can be attributed to the binding property of the plasticizer as less void spaces may be present in the residue with higher quantity of binder (Oladeji 2012). Hence, more resistance to compression as the binder level increases. Highest values for expansion ratio were observed in “B” series (Table 5) which may be due to the larger particle size of the potato peel powder. Desrumaux et al. (1998) reported that with increased particle size in a given biochemical composition, extrudates could be produced with increased expansion ratio. Extrudates produced with corn meal of higher particle sizes expanded more than the extrudates produced with smaller particle sizes (Carvalho et al. 2010).

Colour

From the observed results, it was seen that samples C (C1, C2 and C3) were different in colour than that of other samples. The obtained values of C1 (54.385), C2 (53.012), C3 (54.811) (Table 5) clearly predicts the difference in total colour change (ΔE). The intensity of the brown to black colour was greater in C samples containing banana peel powder as an ingredient. The differences in total colour change (ΔE) are probably due to the presence of poly phenols compounds in banana peel powder. It was also observed that total colour change (ΔE) in “D” series pellets containing glycerol as plasticizer was

Table 6 Correlative studies (Pearson correlation) between engineering and functional properties

	Hardness	Expansion ratio	WBC	Mass	Bulk density	Macro porosity	Hausner ratio	Pellet durability
Hardness	1.000	–	–	–	–	–	–	–
Expansion Ratio	–0.347	1.000	–	–	–	–	–	–
WBC	0.517	–0.022	1.000	–	–	–	–	–
Pellet durability	0.845	–0.389	0.339	1.000	–	–	–	–
Mass	0.149	–0.678	–0.033	0.054	1.000	–	–	–
Bulk Density	0.283	–0.488	0.082	0.312	0.547	1.000	–	–
Macro porosity	–0.033	–0.455	–0.065	–0.104	0.716	–0.120	1.000	–
Hausner ratio	–0.274	0.529	–0.075	–0.240	–0.590	–0.416	–0.356	1.000

WBC Water binding capacity

slightly lower (mean; 49.956) than that of other formulations containing CNSL as plasticizer “A” series (mean; 50.045) this decrease may be due to the inherent dark colour of the cashew nut shell liquid.

Hardness

Based on the results, it was observed that the hardness increases with the increase in plasticizer in case of CNSL ($A1 < A2 < A3$, $B1 < B2 < B3$, $C1 < C2 < C3$) as shown in Table 5. The hardness varied from 392.454 to 495.622N, wherein the lowest value was observed for A1 sample 392.454N and the highest value was observed for C3 (495.622N). The increase in the hardness in “C” series may be due to the high amount of fibre in banana peel powder. The fibre changes the cell wall thickness and makes it less porous and thicker which in turn makes the pellets harder (Charunuch et al. 2014). Similar findings were reported by Ainsworth et al. (2007); Mendonca et al. (2000) and Yanniotis et al. (2007). In case of samples containing glycerol as plasticizer (D1, D2, D3), a similar trend was observed, i.e., D1 (439.805N) < D2 (448.583N) < D3 (460.897N). The hardness of the samples was found to be inversely correlated with the expansion ratio of the pellets.

Pellet durability index

As shown in Table 5, pellets prepared from different formulations ranged between 98.25 and 99.62 % which exceeds the standard durability (97.5 %) set for handling and transportation of pellets (BSI 2010). As the amount of the plasticizer increases the pellet durability index increases. It was observed that the highest value of pellet durability index was found in C3 formulation which can be correlated with the increased hardness (495.622 N) of pellets (C3 formulation), as compared to pellets prepared from other formulations (Table 5). The increased hardness and durability can be attributed to the strong binding effect of the plasticizer causing strong binding interactions between particles that are very similar to those of solid bridges (Nalladurai and Morey 2009). Increased hardness of the pellets describes the increased durability index of the pellets as the pellets have not disintegrated during tumbling.

Correlation among engineering properties and functional properties

Pearson’s correlation between engineering properties and functional properties of biodegradable pellets has been shown in Table 6. Hardness is positively correlated ($r=0.845$) with pellet durability index indicating that the pellets with increased hardness did not disintegrate during the pellet durability test. But hardness had a negative correlation with expansion ratio ($r=-0.347$) which means that hardness increases as

expansion ratio decreases. Expansion ratio had a negative correlation with pellet durability, mass, bulk density and macro-porosity and had a positive correlation with Hausner ratio. The mass showed the positive correlation with both bulk density and macro-porosity which established that both the properties increase with increased mass. However, the mass showed negative correlation with Hausner ratio indicating heavier mass reduces the free-flowing behaviour of pellets. Correlation between engineering properties and functional properties of biodegradable pellets reveal that functional properties can be predicted from engineering properties.

Structural data analysis for biodegradable pellets

The factor loading obtained for the first two components (PCs) and the percentage of variance along with cumulative variance in case of loose fill is shown in Table 7. The first two principal components accounted for 75.087 % of the variance. The first and second principal components (F1 and F2) explained 48.386 and 26.700 % of the variance, respectively. It was observed that 48.386 % variability explained by F1 was positively correlated with mass, pellet volume and bulk density while 26.700 % variance showed by F2 was highly correlated positively with colour. It was observed from Fig. 2 that all “A” blends were positioned on left side of principal component 1 (F1) which were linked to expansion ratio values while all “D” blends were located on its right which were linked to density, mass and pellet volume. Such parameters could be used to distinguish “A” blends from “D” blends containing same raw material with different plasticizers. Similarly “B” and “C” blends containing different raw materials (PPP and BPP) with same plasticizer could be differentiated from each other on the basis of macro porosity, Hausner ratio,

Table 7 Principal component analysis: loading of first two components for loose fill

	F1	F2
Total variance explained		
Initial Eigen value	4.839	2.670
Variability (%)	48.386	26.700
Cumulative (%)	48.386	75.087
Factor loadings:	F1	F2
Mass	0.873	−0.371
Pellet volume	0.841	−0.401
Bulk density	0.869	−0.371
Macro porosity	−0.732	0.555
Hausner ratio	−0.719	0.048
Hardness	0.644	0.683
Colour	0.263	0.850
Water binding capacity	0.247	0.509
Pellet durability index	0.694	0.638
Expansion ratio	−0.732	−0.263

Fig. 2 Principal component score plot for loose filled biodegradable pellets made from different formulations. *WBC* water binding capacity, *HR* Hausner ratio, *PDI* Pellet durability index

colour, hardness, pellet durability index and water binding capacity. In case of tapped fill, first two components (F1 and F2) accounted for 74.566 % of the variance (Table 8). The first and second principle components (F1 and F2) showed 48.429 and 26.137 % of the variance, respectively. The F1 was positively correlated with expansion ratio and macro porosity and negatively correlated to mass and bulk density while F2 had a positive correlation with colour. As shown in Fig. 3 all “D” blends were positioned on left side of principal component 1 (F1) which were linked to bulk density, mass and pellet volume while all “A” blends were located on its right which were linked to expansion ratio. Such parameters could be used to differentiate “D” blends from “A” blends. On the basis of macro-porosity, Hausner ratio, pellet durability index, hardness, colour and water binding capacity “C” blends and “B” blends could be differentiated from each other.

Conclusion

The geometrical approach was adopted to determine the engineering properties of the biodegradable pellets made from different formulations. Based on the observed bulk densities

it was seen that extruded biodegradable pellets bear a good compaction for both loose and tapped fill methods. Moreover, compaction characteristics increased by tapping. These

Table 8 Principal component analysis: loading of first two components for tapped fill

Total variance explained	F1	F2
Initial Eigen value	4.843	2.614
Variability (%)	48.429	26.137
Cumulative (%)	48.429	74.566
Factor loadings:	F1	F2
Mass	−0.802	−0.424
Pellet volume	−0.799	−0.465
Bulk density	−0.805	−0.409
Macro porosity	0.850	0.335
Hausner ratio	0.724	0.056
Hardness	−0.586	0.745
Colour	−0.304	0.829
Water binding capacity	−0.248	0.497
Pellet durability index	−0.677	0.655
Expansion ratio	0.843	−0.131

Fig. 3 Principal component score plot for tapped fill biodegradable pellets made from different formulations. *WBC*: water binding capacity, *HR* Hausner ratio, *PDI* Pellet durability index

prepared pellets belong to “Class-3: Medium” and “Class-4: medium to low” for both loose and fill methods. Hausner ratio (HR) classified the extruded pellets as free flowing to medium flowing. Pellet volume and mass showed a significant positive correlation with fill volume for both loose and tapped fill volumes. The macro-porosity had a high positive correlation with fill volume in loose as well as in tapped fill. Hardness was observed to be positively correlated with pellet durability index but a negative correlation was seen with expansion ratio. These properties which affected the compactness and flowability of the pellets would help in improving the effective storage of pellets which in turn improves the product quality. On the basis of engineering and functional properties (flowability, compactness, pellet durability index, hardness, and water binding capacity) it can be predicted that the pellets developed from deoiled rice bran, banana peel powder and CNSL as plasticizer would be most shelf stable. The pellets containing glycerol as plasticizer (15 and 21 ml level) showed better results with respect to flowability, compactness, pellet durability index, hardness, and water binding capacity. Hence, further research is required to narrow down the range of plasticizer levels. Engineering and functional properties of biodegradable pellets analysed in this study could be helpful in further studies for design aspects related to the production, storage, packaging, handling and overall acceptability of these pellets.

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References

- Ainsworth P, Ibanoglu S, Plunkett A, Ibanoglu E, Stojceska V (2007) Effect of brewers spent grain addition and screw speed on the selected physical and nutritional properties of an extruded snack. *J Food Eng* 81(4):702–709
- Anonymous (1979) Classification of rocks and soils for engineering geological mapping. Part I: rock and soil materials. *Bull Int Assoc Eng Geol* 4(19):364–371
- AOAC (1995) Official methods of analysis. U.S.A Assoc of Official Anal Chem, Washington
- ASABE S269.4 (2002) Cubes, pellets, and crumbles-definitions and methods for determining density, durability, and moisture content. *Am Soc Agric Biol Eng St. Joseph US*
- Avela MD, Pace E, Immirzi B, Impallomeni G, Malinconico M, Santagata G (2007) Addition of glycerol plasticizer to seaweeds derived alginates: influence of microstructure on chemical-physical properties. *Carbohydr Polym* 69(3):503–511
- Barletta BJ, Knight KM, Barbosa-Canovas GV (1993) Compaction characteristics of agglomerated coffee during tapping. *J Texture Stud* 24(3):253–268
- BSI (2010) EN 15210–15211:2009-solid biofuels-determination of mechanical durability of pellets and briquettes. BSI, London
- Carr E (1965) Evaluating flow properties of solids. *Chem Eng* 72(2):163–168
- Carvalho CWP, Takeiti CY, Onwulata CI, Pordesimo LO (2010) Relative effect of particle size on the physical properties of corn meal extrudates: effect of particle size on the extrusion of corn meal. *J Food Eng* 98(1):103–109
- Chaplin MF (2003) Fiber and water binding. *Proc Nutr Soc* 62:223–227
- Charunuch C, Limsangouan N, Prasert W, Wongkrajang K (2014) Optimization of extrusion conditions for ready-to-eat breakfast cereal enhanced with defatted rice bran. *Int Food Res J* 21(2):713–722
- Chevanan N, Womac AR, Bitra VS (2008) Loose-Filled and Tapped Densities of Chopped Switchgrass, Corn Stover and Wheat Straw. ASABE Paper No. 084085. St. Joseph, MI

- Christoph R, Schmidt B, Steinberner U, Dilla W (2000) Glycerol. Ullmann's encyclopedia of industrial chemistry. Wiley, Weinheim, p 2
- Cuq B, Gontard N, Guilbert S (1998) Proteins as agricultural polymers for packaging production. *Cereal Chem* 75(1):1–9
- Desrumaux A, Bouvier JM, Burri J (1998) Corn grits particle size and distribution effects on the characteristics of expanded extrudates. *J Food Sci* 63(5):857–863
- Dreese PC, Hoseney RC (1982) Baking properties of bran fractions from brewer's spent grains. *Cereal Chem* 59(2):89–91
- Evans MR, Hensley DL (2004) Plant growth in plastic, peat, and processed poultry feather fiber growing containers. *Hortic Sci* 39(5):1012–1014
- Evans MR, Karcher D (2004) Properties of plastic, peat, and processed poultry feather fiber growing containers. *Hortic Sci* 39(5):1008–1011
- Fan J, Mitchell JR, Blanchard JMV (1996) The effect of sugars on the extrusion of maize grits: I. The role of the glass transition in determining product density and shape. *Int J Food Sci Technol* 31(1):55–65
- Gamlath S, Ravindran G (2009) Extruded products with fenugreek (*trigonella foenum-graecium*) chickpea and rice: physical properties, sensory acceptability and glycaemic index. *J Food Eng* 90(1):44–52
- Garcia RA, Flores RA, Mazenko CE (2007) Factors contributing to the poor bulk behavior of meat and bone meal and methods for improving these behaviours. *Bioresour Technol* 98(15):2852–2858
- Grey RO, Beddow JK (1969) On the Hausner ratio and its relationship to some properties of metal powders. *Powder Technol* 2(6):323–326
- Hamel S, Krumm W (2008) Near-wall porosity characteristics of fixed beds packed with wood chips. *Powder Technol* 188(1):55–63
- Haridas RP, Hemamalini R (1991) Effect of incorporating wheat bran on rheological characteristics and bread making quality of flour. *J Food Sci Technol* 28(2):92–97
- Hayes GD (1987) Food engineering data handbook. Longman Scientific and Technical, New York
- Igathinathane C, Jaya ST, Sokhansanj S, Bi X, Lim CJ, Melin S, Mohammad E (2010) Simple and inexpensive method of wood pellets macro-porosity measurement. *Bioresour Technol* 101(16):6528–6537
- Keshi KJ, Huang Q (2009) Engineering Zein films with controlled surface morphology and hydrophilicity. *J Agric Food Chem* 57:2186–2192
- Kowalska E, Wielgosz Z, Pelka J (2002) Use of post-life waste and production waste in thermoplastic polymer compositions. *Polym Polym Compos* 10(1):83–91
- Krochta JM (2002) Proteins as raw materials for films and coatings: definitions, current status, and opportunities. Protein-based films and coatings 1–41
- Malinconico M, Immirzi B, Massenti S, Mantia LFP, Mormile P, Petti (2002) L Blends of polyvinylalcohol and functionalized polycaprolactone. A study of the melt extrusion and post-cure of films suitable for protected cultivation. *J Mater Sci* 37(23):4973–4978
- Manisha J, Srivastava T, Saxena DC (2012) Extrusion processing of deoiled rice bran in the development of biodegradable molded sheets. *School J Agric Sci* 2(8):163–178
- Mendonca S, Grossmann MVE, Verhe R (2000) Corn bran as a fiber source in expanded snacks. *LWT Food Sci Technol* 33(1):2–8
- Mohanty AK, Misra M, Hinrichsen (2000) Biofibres, biodegradable polymers and biocomposites: an overview. *Macromol Mat Eng* 276–277(1):1–24
- Nalladurai K, Morey VR (2009) Factors affecting strength and durability of densified biomass products. *Biomass Bioenerg* 33(3):337–359
- Nehru C, Alvin R, Venkata W, Bitra SP, Igathinathane C, Yuechuan T, Petre MIY, Shahab S (2010) Bulk density and compaction behavior of knife mill chopped switchgrass, wheat straw, and corn stover. *Bioresour Technol* 101(1):207–214
- Oladeji JT (2012) A comparative study of effects of some processing parameters on densification characteristics of briquettes produced from two species of corncob. *Pac J Sci Technol* 13(1):182–192
- Petukhov M, Cregut D, Soares CM, Serrano L (1999) Local water bridges and protein conformational stability. *Protein Sci* 8(10):1982–1989
- Podzeck F (1998) Particle-particle adhesion in pharmaceutical powder handling. Imperial College Press, London, pp 95–114
- Rabier F, Temmerman M, Bohm T, Hartmann H, Jensen PD, Rathbauer J, Carrasco J, Fernandez M (2006) Particle density determination of pellets and briquettes. *Biomass Bioenergy* 30(11):954–963
- Schussele A, Bauer-Brandl A (2003) Note on the measurement of flowability according to the European pharmacopoeia. *Int J Pharm* 257(1–2):301–304
- Sengar SH, Mohod AG, Khandetod YP (2012) Performance evaluation of kiln for cashew nut shell carbonization and liquid. *Int J Energ Eng* 2(3):78–85
- Stojceska V, Ainsworth P, Plunkett A, Ibanoglu E, Ibanoglu S (2008) Cauliflower by-products as a new source of dietary fibre, antioxidants and proteins in cereal based ready to-eat expanded snacks. *J Food Eng* 87(4):554–563
- Tanya MR (2006) Water structure and interactions with protein surfaces. *Curr Opin Struct Biol* 16(2):152–159
- Wu Q, Sakabe H, Isobe S (2003) Processing and properties of low cost corn gluten meal/wood fiber composite. *Ind Eng Chem Res* 42(26):6765–6773
- Wypych G (2012) Handbook of Plasticizers (2nd edition). William Andrew Publishing, Boston. SN 978-1-895198-50-8, 11, 307–419
- Yamauchi M, Masuda S, Kihara M (2006) Recycled pots using sweet potato distillation lees. *Res Conser Recycl* 47(2):183–194
- Yamazaki WT (1953) An alkaline water retention capacity test for the evaluation of cooking baking potentialities of soft winter wheat flours. *Cereal Chem* 30(3):242–246
- Yanniotis S, Petraki A, Soumpasi E (2007) Effect of pectin and wheat fibers on quality attributes of extruded cornstarch. *J Food Eng* 80(2):594–599
- Zayas JF (1997) Solubility of proteins. In *Functionality of proteins in food* (pp. 6–75). Springer Berlin Heidelberg