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Review

Integrated sustainable process design framework for cassava biobased packaging materials: Critical review of current challenges, emerging trends and prospects

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ABSTRACT

Background: Cassava represents a reasonable share in biobased material development globally. The production of its biopolymer derivatives using conventional techniques/methods is accompanied by significant wastes with potential negative environmental impact. Among the biopolymer derivatives, starch dominates as lone additive in cast matrices with packaging limitations, requiring other biopolymer derivatives, and/or external-source modifiers for matrix improvement. Exploiting integrated sustainable engineering process design of all biopolymer derivatives, is a novel approach in designing efficient system of cassava biobased materials for food and non-food applications.

Scope and approach: A critical review on the current and emerging techniques and methodologies to address cassava wastes and challenges of cassava research for application on biobased packaging are provided. The potential of integrated sustainable engineering process design framework for packaging system is discussed, and prospects for improvement suggested.

Key findings and conclusions: Challenges of significant waste generated during conventional processing and on the application process aiming at tailoring materials to industrial needs are reported. These materials should be improved using a holistic approach reflecting the target products, variable environment, minimising production costs and energy. Use of novel material resources, eliminating waste, and employing a standardised methodology via desirability optimisation, present a promising process integration tool for development of sustainable cassava biobased systems.

1. Introduction

The substantial global dependence on petrochemical based materials has given rise to packaging security concerns. These concerns, together with negative environmental impacts (Emmambux et al., 2004), increased population pressure on finite and dwindling natural resources and competition for food supply, have drawn the extensive research and development of sustainable alternatives. The sustainable alternatives that are green, clean, post-use biodegradable, compostable, efficient and sustainable are desired (Coombs & Hall, 2000). The based materials, which have emerged as main alternatives to address the concerns, are obtained from renewable resources which is a component of a sustainable biobased industry. Cassava (*Manihot esculenta* Crantz) represents a sustainable resource of biobased products (Hood, Teoh, Devaiah, & Requesens, 2013).

Thus, this critical review reports: (i) the current technologies and methodologies used to address cassava wastes, to apply cassava biobased materials in food industry; (ii) challenges with biobased material development using conventional processes; (iii) potential of integrated green engineering process design framework for sustainable packaging system development; and (iv) prospects for further improvement of the integrated process design.

2. Cassava as a versatile crop resource of biomaterials

Cassava is categorised into sweet or bitter, with sweet cassava being edible and safe for immediate use in fresh and processed forms, while the bitter ones are unsafe for immediate consumption.

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Cassava is consumed widely and highly valued as food security anchorage for tropical and sub-tropical countries. Mainly in Africa whereby more than half of the world's cassava or about 162.5 million t from over 15 million hectares, compared to more than 33 t (3.0 ha) and 92 t (5.0 ha) millions in Latin America and Asia, are cultivated (FAOSTAT, 2015).

Advancements in the biopolymer research triggered, in the last decade, the paradigm shift towards a fully industrial-applied sweet cassava (Adetunji, Isadare, Akinluwade, & Adewoye, 2015). Increasing awareness of the association between cassava biopolymer derivatives and cheap industrial biobased products might account for this trend. This popularity is due to its easily processed low cost biopolymer derivatives (Starch, cellulosic fibres, lignin, and hemicellulose) (Table 1). Of the biopolymer derivatives, starch has been extensively studied, perhaps due to its high root proportionality, chemical and functionality (Blazek & Copeland, 2009), and received a higher attention for biobased materials production (Paunonen, 2013). Starch molecular structures, with differentiated amylose (20–30%) and amylopectin (70–80%) contents (Mufumbo et al., 2011), presents unique polymer functionality in wide range applications. The proportionality of amylose and amylopectin in extracted and applied starch can differ significantly depending on production methodology and amounts used to prepare products. Amylose is a nearly linear polymer of α-1, 4 anhydroglucose units that has excellent film-forming ability, rendering strong, isotropic, odourless, tasteless, and colourless film (Campos, Gerschenson, & Flores, 2011). Amylopectin is a highly branched polymer of short α-1, 4 chains linked by α-1, 6 glucosidic branching points occurring every 25–30 glucose units (Liu, 2005). Consequently, amylose and amylopectin provide materials of varying viscosities, crystalline quality and the energy required to melt the material (Mufumbo et al., 2011).

3. Cassava starch production and environmental impact

Due to sweet cassava starch ease of processing, low cost and potential high yields, conventional methods have been used for its extraction, purification and drying. Wet milling is the most common and simple conventional method, using at industrial level, simple equipment and heavy investment, depending on the desired final product (Lundy, Ostertag, & Best, 2002). Cassava starch can be obtained from fresh roots or its non-edible parts, stems, peels and leaves, primarily by wet milling and starch has also been produced from dry cassava chips. The complete step-wise process (using simple or large scale extraction) can be divided into four main stages: (i) preparation (peeling and washing); (ii) rasping/pulping/grating; (iii) recovery (starch sedimentation, washing, dewatering, drying); and (iv) finishing (milling and packaging).

Beyond starch extraction, cassava processing also generates large amounts of wastes as waste solids and wastewaters (Adeola, 2011). The United Nations Statistics Division, *Glossary of Environment Statistics* defines wastes as materials that are not prime products for which the initial user has no further use in terms of his/her own purposes of production, transformation or consumption, and of which he/she wants to dispose. Wastes can be generated during the extraction of raw materials, the processing of raw materials into intermediate and final products, the consumption of final products, and other human activities (UNSD, 1997). According to Food and Agriculture Organisation of the United Nations (FAO, 2013), starch roots, mainly cassava contributes over 700 MT wastes in the global upstream food wastes, requiring conversion into valuable products and energy in an environmentally friendly manner. Besides, an active starch plant can generate up to 47% total fresh disposable cassava by-products (Heuzé, Tran, Archimède, Lebas, & Regnier, 2013). When disposed for a given period in the environment, these could be typically associated with emission of strong unpleasant smells, carbon-dioxide and total cyanogens. Cassava wastes -rich total cyanogens can contaminate surface water, groundwater, soil, and air which causes more problems for humans, other species, and ecosystems (Simonetto, de, & Borenstein, 2007). In addition, cassava wastes can also be a source for rodents and insects, which can harbour gastrointestinal parasites, yellow fever, worms, the plague and other conditions for humans. Moreover, the increasing nature of non-beneficial sweet cassava competition might exacerbate waste disposal problems arising from more use of bitter cassava. During the traditional processing, huge waste solids and wastewater are generated from bitter varieties in order to avoid total cyanogens contained in the peels (Cardoso et al., 2005; Tumwesigye, Oliveira, & Sousa-Gallagher, 2016b). With insufficient prioritization of packaging source reduction, recyclability, compostability, recycled content and recycling policies (MacKerron & Hoover, 2015), wastes are likely to increase in the years ahead.

Table 1
Composition of sweet cassava root and different components.

Component	Per 100 g (On a fresh weight (dry matter) basis)	
Cassava root (Uchechukwu-Agua, Caleb, & Opara, 2015)		
Water, g	60	
Protein, g	1.4	
Fat, g	0.28	
Carbohydrate, g	38	
Fibre, g	1.8	
Sugar, g	1.7	
Minerals, g	0.46	
Vitamins, g	0.07	
Cassava peeled & unpeeled root (Ospina & Ceballos, 2002)		
	Peeled	Unpeeled
Water, g	71.50	68.06
Carbohydrate, g	26.82	29.06
Crude fibre, g	0.12	0.99
Crude protein, g	0.74	0.87
Ash, g	0.13	0.17
Vitamins	0.69	0.85
Cassava waste solids (peel and edible fibre) polysaccharides (Salvador, Suganuma, Kitahara, Tanoue, & Ichiki, 2000)		
Others	1.8	
Pectin	17.8	
Hemicellulose	22.8	
Cellulose	48.2	

4. Emerging trends for sweet cassava biobased packaging material development and technological challenges

A number of conventional methods/techniques for the development of biobased packaging materials have been reported in literature, and they include: extrusion (sheet/film, reactive), baking, injection moulding, blow moulding, compression moulding, vacuum foaming, casting, spraying, lamination, calendaring and thermoforming (Imam et al., 2008). Casting has been the commonest technique used for producing edible and biodegradable starch films (Table 2), and was adequately described by Jiménez, Fabra, Talens, and Chiralt (2012). Regardless of the technique used, the production and characterisation of BPM is a four-stage process: (i) preheating homogenisation of additives; (ii) heating of polymeric solution; (iii) drying; and (iv) structural and functional characterisation. Heating and drying are vital steps in the production of desired BPM because they can alter the structure and affect the functional application (Tumwesigye et al., 2016b). The most common convention characterisation techniques for cassava reported are: (i) thickness measurements (Micrometer); (ii) optical (colour-chroma;

Table 2
Summary of cassava biobased material mechanical and barrier properties.

Materials	Method	Thickness, mm	Property				Reference
			Tensile strength, MPa	Elongation, %	Water vapour permeability	Oxygen permeability	
Starch*, glycerol	Casting	0.08	4.0–49.0	3.0–46.0	4.02–8.33***u***	33	Mali et al. 2006
Starch*, Chitosan flake, Acetic acid (glacial)	Casting	0.10–0.12	0.38–21.02	3.16–39.86	2.30–3.15**	–	Bangyekan et al. 2006
Starch*, amylose, glycerol	Casting	0.10	2.2–52.8	4.5–263.1	0.24–0.49***u***	–	Alves et al. 2007
Starch*, glycerol, potassium sorbate	Casting	–	0.16–2.35	1.30–29.0	6.10–16.1***u***	–	Flores et al. 2007
Modified starch		0.06–0.12	–	–	0.12–0.21**n**	–	Henrique et al. 2007
Starch*, montmorillonite, chitosan, glycerol, acetic acid	Casting	0.071	21.2–24.64	1.1–4.5	1082–2000**	–	Kampeerapappun et al. 2007
Starch*, agar, glycerol, Polyethylene glycol200	Casting	0.014–0.048	1.39–42.11	–	10.3–137.0***u***	–	The et al. 2008
Starch*, chitosan, gelatin, glycerol	Casting	0.083–0.117	13.63–49.40	4.51–110.7	5.78–10.17***u***	0.64–2.58 ***p***	Zhong & Xia, 2008
Starch*, chitosan, glycerol, oregano oil	Extruder	–	1.43–2.54	21.95–48.40	0.62–1.39***u***	–	Pelissari et al. 2009
Starch*, PBAT, tween80, polyoxyethylene sorbitan monooleate	Casting	0.15–1.21	0.5–14.5	12.5–32.5	0.0009–0.0115 ***u***	–	Brandelero et al. 2010
Starch*, xanthan gum, potassium sorbate	Extruder	–	Negligible	19.0–85.0	3.70–6.70***u***	–	Flores et al. 2010
Starch*, sucrose, inverted sugar, spinach	Casting	0.084–0.116	1.81–7.75	65.0–217.0	12.3–99.9***u***	–	Veiga-Santos et al. 2008
Starch*, carboxymethyl cellulose, glycerol	Casting	–	2.0–30.0	4.0–86.0	–	–	Tongdeesoontorn et al. 2011
Starch*, cornstarch, glycerin, stearic acid, Sugarcane bagasse	Casting	–	2.20–3.80	10.4–57.2	–	–	Vallejosa et al. 2011
Starch*, glycerol, carnauba wax type 1, stearic acid	Casting	0.128–0.132	0.25–2.14	19.30–51.06	32.75–54.74***R***	–	Chiumarelli & Hubinger 2012
Starch*, chitosan, glycerol	Extruder	0.19–0.23	085–2.71	21.95–74.04	1.00–2.22***u***	–	Pelissari et al. 2012
Starch*, natural sodium montmorillonite, ethanol, glycerol, liquid inverted sugar, sucrose	Casting	0.077–0.092	1.85–6.06	89.9–213.4	3.36–7.08**n**	36,979.2 –366,163.2***p***	Souza et al. 2012
Starch*, xanthan gum, potassium sorbate, glycerol	Casting	0. 18–0.26	0.38–2.00	71.6–280.6	0.17–0.23***u***	–	Arismendi et al. 2013
Starch*, glycerol, β-zeolite nano-crystal/Na-beidellite	Casting	0.148–0.210	1.40–2.70	25.0–110.0	2.3–3.5***u***	–	Belibi et al. 2013
Starch*, glycerol, agar, span80	Casting	0.027–0.046	–	–	0.33–0.56***u***	402.0 –496.0***p***	Maran et al. 2013
Flour, Glycerol, Sorbitol, Polyethylene glycol	Casting	0.096–0.099	5.29–28.65	4.13–28.24	31.20–36.68**	48.67 –55.33***T***	Suppakul et al. 2013
Starch*, cashew tree gum, carnauba wax, tween80, span80, glycerol	Extruder	0.15	0.76–1.48	76.5–136.3	3.70–6.70***uv***	–	Rodrigues et al. 2014

* Commercially purchased, native.
** Water vapour transmission rate, g/(m^2.day)***u***, $\times 10^{-10}$ g m^{-1} s^{-1} Pa^{-1}***uv,*** g mmkPa^{-1} h^{-1} m^{-2}***n***, g mm m^{-2}. d^{-1}.kPa^{-1}***p***, $\times 10^{-8}$ cm^3 m^{-1} s^{-1} Pa^{-1} ***R,*** water vapour resistance (scm^{-1})***T***, oxygen transmission rate (cm^3 m^{-2} d^{-1} atm).

transmission-spectrophotometry); (iii) structure and morphology (scanning electron microscopy); (iv) surface energy-sessile drop technique (optical tensiometer); (v) chemical and functional-fourier transform infrared (UV/Vis spectrophotometer); (vi) barrier-gravimetric measurement for water vapour permeability (with acrylic permeation cell), oxygen and carbon-dioxide transmission rate (Presensor/Dansensor); (vii) mechanical-Texture analyser; (viii) thermal techniques (differential scanning calorimetry, thermogravimetric analysis, Crystallinity); and (ix) x-ray diffraction.

The greatest practical challenges with cassava biobased packaging materials developments using conventional approaches are associated with starch based matrices limitations in industrial application of food packaging materials. In the last decade, there has been intensified research on cassava material physicochemical and functional properties (mechanical, barrier and thermal) (Table 2) and antimicrobial (Table 3) in order to attain functional properties closer to the traditional plastic packages. Unfortunately, the approach has been a piecemeal development, associated with uncontrolled variability in material properties (Table 2). This is due to native cassava starch inherent polar and hydrophilic nature, brittleness, resultant inferior functional properties and vulnerability to degradation. In order to improve starch properties, its matrices have to be modified to enhance process stability, performance and biocompatibility (see section 5). While improvers have shown to yield better properties, structure and general appearance of biobased packaging materials (see section 5), they have the disadvantage of imparting unnecessary colours to the starch films leading to variations in opaqueness (Tumwesigye et al., 2016b). Additionally, various starch reinforcements tend to distort film morphology and introduce surface heterogeneity (de Moraes, Muller, & Laurindo, 2012). Although it has sufficed to extrinsically modify the starches and cellulose derivatives prior to or during matrix formulations to make them more highly functional, it has rendered the process lengthier and this could perhaps explain the high production costs, perhaps due to non-cost effectiveness and energy inefficiency of feedstocks.

In any case, the trend to focus on property improvements without considerations of the holistic approach, which considers useful validation information on package-product compatibility and behaviour during realistic distribution could account for food

Table 3
Summary of cassava biobased material antimicrobial properties.

Source of antimicrobials	Active substance	Incorporation style in starch based matrix	Intended study/function	Reference
Fruit and vegetable pomace extracts	Anthocyanin, flavonoids & chlorophyll	Direct addition	Antioxidant properties (general)	Hyashi et al. 2006
Commercial	Potassium sorbate	Direct addition	Antimicrobial release model and kinetics (general)	Flores et al. 2007
Commercial	Chitosan	Direct addition	Fungistatic activity (general)	Zhong & Xia, 2008
Commercial	Chitosan	Direct addition	Antimicrobial performance on global quality of salmon muscle	Vásconez et al. 2009
Commercial	Chitosan & oregano essential oil	Direct addition	Antimicrobial against *Bacillus cereus* ATCC 25923, *E. coli* ATCC 25922, *S. aureus* FRI196e, and S. enteritidis,	Pelissari et al. 2009
Commercial	Clove powder, volatile oils, cinnamon powder, red pepper powder, honey propolis, coffee powder & orange essential oil	Direct addition	Antimicrobial against yeast & mold counts of white pan bread slices	Kechichian et al. 2010
Mango & acerola pulps	Carotenoid, polyphenol, & vitamin C	Direct addition	Antioxidants to preserve palm oil	Souza et al. 2011
Commercial	Nisin & potassium sorbate	Direct addition	Antimicrobial effectiveness against Listeria innocua & Zygosaccharomyces bailii	Basch et al. 2012
Commercial	Natamycin	Direct addition	Antimicrobial effectiveness against *Saccharomyces cerevisiae*	Ollé Resa et al., 2013
Commercial cinnamon & clove essential oils	Cinnamaldehyde & eugenol	Direct addition	Inhibitory against Penicillium commune & Eurotium amstelodami	Souza et al. 2013
Commercial	Potassium sorbate	Direct addition	Effective antimicrobial barrier against *ygosaccharomyces bailii*	Arismendi et al. 2013
Commercial	Cin-namaldehyde	Supercritical fluid technology	Antimicrobial inhibition of proliferation of Penicillium commune & Eurotium amstelodami fungi in bread products	de Souza et al. 2014
Green tea and palm oil carotenoids extracts	Peroxides, total carotenoids, and total polyphenol	Direct addition	Inhibit oxidation & as a scavenger of oxygen radicals; oxidative protection in packaged butter	Perazzo et al. 2014
Red propolis & licuri leaves	Propolis & cellulose nanocrystals extracts	Direct addition	Coagulase-positive staphylococci in cheese curds & antioxidant against butter	Costa et al. 2014
Oregano & clove essential oils	Carvacrol (2-methyl-5-[1-methylethyl]phenol) and thymol (5-methyl-2-[1-methylethyl) phenol])	Direct addition & surface coating	Antimicrobial against molds, yeasts, & Gram-positive and Gram-negative bacteria	Debiagi et al. 2014

packaging application limitations. For example, by saying a given property reduced, improved, increased, etc. without testing/validating the research outputs' performance with target products could be the major cause for low end use adoption of these materials. Most developed films did not include validation pathways in situ that represent the real conditions while considering envisaged applications. To put it right, most techniques applied in situ tend to differ from ones developed by research in vitro. Generally, wide material property variations, which are not optimised and directed, had made it difficult for food industry to compare results and make objective application decisions.

Limitations still lie in optimisations, modelling or simulations to bring out the optimum performance of films and coatings. According to Van Boekel (2008), a dynamic modelling approach permits advancement of an application scheme specific to a product and to select the most suitable regimes without the necessity for extensive testing of the product and indicator. Limited reports have focused on simulation for mechanical and barrier properties of starch-based composite matrix (Arismendi et al., 2013; Suppakul, Chalernsook, Ratisuthawat, Prapasitthi, & Munchukangwan, 2013). In spite of these advantages, few of these packaging systems are commercialized because of high cost, strict safety and hygiene regulations or limited consumer acceptance. Therefore more research is needed to develop cheaper, more easily applicable and effective packaging systems for various foods.

Careful manipulations in the perforation's density, pore localisations, dimensions, micropore structure, and the technique of microperforation can effectively permit desired in-package environment envisaged for a named product. However, the current concept of developing biobased materials to cater for different properties and functions could influence a particular product' in-package atmosphere, thereby making it difficult to match product and package properties. Besides, the constant use of perforations could make the process inefficient, damage the package or introduce contaminants particularly in dust-prone areas with differentiated pressures.

Recently, some research has been concentrated on design techniques to improve the casting technique and starch biobased materials towards prescribing a uniform and standard system. In addition, work has also been intensified in reinforcing starch matrices in order to improve the properties by which tailored materials would be developed. Industries have demanded efficient and economy biobased materials, which has resulted into new innovations, with robust and cheap production processes.

4.1. Starch reinforcement techniques

Reinforcing starch-based materials helps to overcome weaknesses inherent in starch matrices (mentioned previously) in order to improve their mechanical, water resistance, and generally functional properties. Starch reinforcements have been mainly studied for improving mechanical (Scheibe, De Moraes, & Laurindo, 2014; Versino & García, 2014; Zainuddin, Ahmad, Kargarzadeh, Abdullah, & Dufresne, 2013), improving water resistance (Rodrigues et al., 2014) or barrier to moisture and gas (Argüello-García et al., 2014; Cardoso et al., 2005; De Pauli, Quast, Demiate, & Sakanaka, 2011).

While reinforcements improve starch material properties, they have the disadvantage of exhibiting non-cost effectiveness and energy inefficiency and their processing at source since in all the starch reinforcements, fillers and reinforcers are externally sourced. This entails many individual stand-alone processes executed independently of each other. The extra energy and costs implied in stand-alone developments could be avoided if the individual processes were integrated.

4.2. *Antimicrobial biobased materials intended for active packaging*

The rapid development of cassava biobased materials, in particular the edible types, their compatibility to packaging material development, and successful research into extraction of bioactive functional natural compounds, has led to functional food packaging material development (Flores, Famá, Rojas, Goyanes, & Gerschenson, 2007; Kechichian, Ditchfield, Veiga-Santos, & Tadini, 2010; de Souza, Dias, Sousa, & Tadini, 2014). Different antimicrobial substances were added to cassava biobased materials in different ways intended to deliver various functions (Table 3). However, research information on the influence of antimicrobial packaging materials on in-package atmospheres and corresponding packed products' quality and safety is scanty, and this presents challenges to their full potential use in modified atmospheric packaging.

Hayashi, Veiga-Santos, Ditchfield, and Tadini (2006) observed a significant antioxidant effect on the packed in cassava starch plasticised materials soybean oil with grape pomace (1.69 and 8.16% total solids) while (Flores et al. (2007) found that high amorphous film matrix relaxation greatly contributes to sorbate release kinetics. The cassava-chitosan fungistatic activities inhibited growth of phytopathogen on mango fruit surfaces (Zhong & Xia, 2008) and reduced Zygosaccharomyces bailii external spoilage in a semisolid product but were not effective against Lactobacillus spp. (Vásconez, Flores, Campos, Alvarado, & Gerschenson, 2009), while those with oregano essential oil had a higher inhibition on *B. cereus* than *S. enteritidis* (Pelissari et al., 2009). Cassava films containing mango and acerola pulps with carotenoids and total polyphenols presented antioxidant effectiveness while acerola pulp vitamin C was a pro-oxidant agent (de Souza et al., 2014). Ollé Resa, Gerschenson, and Jagus (2013) reported antimicrobial effectiveness against bacterial or yeast culture with nisin and potassium sorbate in starch films, while Souza, Goto, Mainardi, Coelho, and Tadini (2013) reported fungicidal action against *Saccharomyces cerevisiae* with starch matrix containing natamycin. Similar results were observed on effective antimicrobial activity against *P. commune* and *E. amstelodami* bread product fungi by cinnamon and clove essential (Souza et al., 2013) and effective antimicrobial barrier against *Zygosaccharomyces bailii* external contamination (Arismendi et al., 2013). de Souza et al. (2014) reported success in supercritical impregnation of cinnamaldehyde (2.49 ± 0.30 mgCN/gfilm, 250 bar). Films with green tea extract and oil palm colorant exhibited oxidative protection in packaged butter, by decrease peroxide index (Perazzo et al., 2014), while those with cellulose nanocrystals (0–1%) and activated with alcoholic extracts of red propolis were effective on coagulase-positive staphylococci in cheese curds and reduced the oxidation of butter during storage (Costa, Druzian, Machado, De Souza, & Guimaraes, 2014). Although antimicrobial active packaging is important in reducing spoilage in packages, it leads to addition of more process with energy and cost consequences.

4.3. *Utilisation of valuable cassava wastes derivatives and by-products*

As mentioned earlier, most studies have evaluated potential of extrinsic fillers and reinforcements for starch modification without considering intrinsic modifiers inherent in cassava parts. Daud, Kassim, Aripin, Awang, and Hatta (2013) characterised the chemical composition of cassava waste peel (% w/w) as: holocellululose (cellulose + hemicellulose) (66.0), cellulose (37.9), hemicellulose (23.9), lignin (7.5), hot water solubility (7.6), 1% NaOH solubility (27.5), ash (4.5) and moisture (14.0).

However, within the last 5 years research into use of cassava waste as reinforcement fillers and source of bioactive extracts has been intensified. Wicaksono, Syamsu, Yuliasih, and Nasir (2013) studied cassava bagasse-based cellulose nanofiber (5–8 nm) for application on tapioca-film. The authors reported good stability of cellulose nanofiber suspensions, successful removal of hemicelluloses and lignin from the fibre structure and improved films tensile strength and decreased elongation at break by 69%. Other studies include use of: (i) cassava roots peel and bagasse as natural fillers of thermoplastic materials using cassava bagasse (1.5%) to increased elastic modulus (by 260%) and maximum tensile stress (by 128%) of thermoplastic starch composites (Versino, López, & García, 2015); (ii) fibrous residue of cassava starch extraction to achieve UV-barrier capacity and water vapour barrier properties (14.6 ± 0.7 10–11 g/m s Pa), tensile strength (18.01 ± 0.19 MPa), mechanical resistance increase (>900%) with 1.5% residue, eco-compatible heat-sealed materials (Versino & García, 2014); (iii) cassava bagasse to develop biodegradable trays with effective antimicrobial activity against moulds, yeasts, and Gram-positive and Gram-negative bacteria, less resistant and more flexible trays, with a decrease in the water absorption and adsorption capacities (Debiagi, Kobayashi, Nakazato, Panagio, & Mali, 2014).

While the above studies show good progress in waste minimisation, they are limited in use by the lack of an integrated process design which would otherwise reduce the energy and costs implied in individual processes. To overcome this scenario, an improved process SRRC, which includes components of integrated process was studied (discussed in subsection 6.1).

5. Sustainable inexpensive and green technologies and processes

Despite recent developments, in the last 5 years, to improve the biobased material production technologies, processes and functional properties, the proactive and robust holistic approach system is still insufficient. The application of integrated approaches to the development of materials hinged on cost-effectiveness, energy efficiency and zero environmental impact can be a promise for developing sustainable biobased materials tailored for broad applications. Of the inexpensive technologies, tape casting has found use in cassava biobased material production, notably during solution casting and drying. Among the green technologies, very few cassava biobased material development studies have been reported in literature, and include: (i) supercritical fluid technology (de Souza et al., 2014); and (ii) simultaneous release recovery cyanogenesis (SRRC) (Tumwesigye et al., 2016b).

5.1. *Tape casting*

Until recently solution casting has been the widely used technique for laboratory-scale production of biobased materials. However, with the shortfalls of this technique to produce uniform materials, handle thicker gels or adjust when varying production volumes, alternative techniques have been investigated. Among the inexpensive techniques, tape casting, successful in paper, plastic, ceramics and paint industries (Mistler & Twiname, 2000), has been investigated for large-scale material production that delivers a continuous process with success. Tape casting technique is a promising tool for production of multi-layered (Tanimoto, Hayakawa, Sakae, & Nemoto, 2006), thick, strong, uniform, varying size biocomposites films at industrial scale (de Moraes, Scheibe, Augusto, Carciofi, & Laurindo, 2015; de Moraes, Scheibe, Sereno, & Laurindo, 2013). Typically, tape casting technique consists of micrometric screw-adjusted blade that helps spread the cast solution on batch or continuous carrier-tapes, ensuring uniform thickness (0.2–1 mm) of films (Larotonda, 2007). The nature of cast solution in terms of rheological behaviour were previously well-described (de Moraes et al., 2013).

Even though materials produced by tape casting are reported to be homogeneous, reproducible and showing quick drying (60 °C, 2.3h) (de Moraes et al., 2015), their stability under high temperatures such roll-to-roll processing (Zucca et al., 2015) remains to be empirically proven. Besides, the use of heat could add to the energy requirements of the process. Integration of this technique with other processes would be crucial to energy reductions. Notwithstanding the energy cost implications, tape casting becomes relatively inexpensive compared to the conventional solution casting.

5.2. Supercritical fluid technology antimicrobial active package development

Among the major roadblocks in active packaging of cassava biobased material research has been to find a near best technique to help in controlled loading and unloading of antimicrobial compounds, and in particular those that are hydrophobic in nature such as organic lipids. Today, only a single study using supercritical fluid technology (Souza et al., 2013) is found in literature. The effect of impregnating antimicrobial compound cinnamaldehyde in cassava starch-nanoclay biocomposite films via supercritical carbon-dioxide ($scCO_2$) was studied by (Souza et al., 2013), reporting successful incorporation, with highest conditions, cinnamaldehyde loading (2.49 ± 0.30 mgCA/gfilm), pressure (250 bar), time (15 h) and at depressurization rate (10 bar min−1). They further reported that all impregnated cinnamaldehyde contents, irrespective of the amounts, were able to deter *P. commune* growth and increased film surface hydrophobicity. They found that cinnamaldehyde -treated films had reduced water vapour permeability (WVP) (4.09 ± 0.84) g mm $m^{-2}day^{-1}kPa^{-1}$ than WVP 10.09 ± 0.35) g mm $m^{-2}day^{-1}kPa^{-1}$) of untreated films. They concluded that: (i) the solubility of cinnamaldehyde in SC_{CO2} dictated the impregnation process; and it is possible to produce better cassava films for packaging with supercritical fluid technology using low energy.

5.3. The SRRC concept: exploiting intact (whole) cassava

5.3.1. Bitter cassava as a potential sustainable source of green biomaterials

The demand for low-cost material resources has led to emergency in research of unexploited plants-derived feedstock (Tumwesigye et al., 2016b). Bitter cassava is an example of unexploited plants-derived feedstock which has not been conventionally utilised in biobased material development. The bitter cassava has many similarities with sweet cassava, with the two differing in the amount of total cyanogen. The bitter cassava contains 900–2000 ppm total cyanogens (Cardoso et al., 2005; Tumwesigye, Morales-Oyervides, Oliveira, & Gallagher, 2016a), whereas sweet cassava has lower total cyanogens (TC) ($0 \geq TC \leq 100$ ppm (Tumwesigye, Baguma, Kyamuhangire, & Mpango, 2006). The properties and benefits that make bitter cassava a sustainable feedstock source for biobased materials have been reported (Tumwesigyeet al., 2006): (i) adaptation to diverse climatic conditions; (ii) high tolerance to drought, low soil fertility and low soil structure; (iii) high yield in energy per unit area per unit labour; (iv) planting and harvesting time allow for a greater flexibility; (v) the economically viable parts (FAO, 2007); (vi) gluten-free; and (vii) the high yield and bright colours of the biopolymers including the resistance to pests, rodents and swine (Tumwesigye, Montañez, Oliveira, & Sousa-Gallagher, 2016c). Latest findings by Tumwesigye et al. (2016a) had explored another benefit of bitter cassava as an effective biomaterial for production of food and non-food packages, and with significantly better properties than sweet cassava films.

However, during traditional processing, huge amount of waste is generated, in which 16–30% are waste solids (peels, fibres, rejects) and wastewaters with unspecified total cyanogens (Heuzé et al., 2013). To transform BC into a green biomaterial required new approaches.

5.3.2. Simultaneous release recovery cyanogenesis (SRRC)

Improved novel alternative methodologies have gained interest in most processing research, and in addition to the principle of green process, more attention is likely. Simultaneous release recovery cyanogenesis (SRRC) can be defined as an improved downstream process that is capable of exploiting intrinsic system's nature, under predetermined conditions, to cause chemical and physical transformations in the products (Fig. 1).

Recently, SRRC technique was applied to investigate the potential of using intact bitter cassava to minimise wastes and produce acceptable biopolymer derivatives capable of developing food packaging materials (Tumwesigye et al., 2016b). The authors reported significantly ($p < 0.05$) higher biopolymer derivative yields and 16% waste decrease. Furthermore, the method effectively reduced total cyanogen content from >1000 ppm to <3 ppm, which is within acceptable limits for use of these poisonous cassava derivatives in food processing. More transparent, homogeneous and strong packaging materials were developed and found to be suitable for packaging application like current commercial packages which are on the market. An added advantage of SRRC is the development of transparent films as a requirement for food packaging. Most cellulose and other starch reinforced biocomposites produce coloured films suitable for only foods that undergo oxidation. The significance of the SRRC outcome implies that peeling cassava can be avoided (Tumwesigye et al., 2016b). When this happens, then there is limited chance of wastes accumulation into the environment. This could also have a possibility of reducing amount of water, energy and costs implications in: (i) extrinsic processing and modification of starch using reinforcements; and (ii) waste management. Moreover, the wastes are converted into useful added value products, human safety ensured, environmental impact reduced, and social-economic welfare of society improved.

The SRRC concept can be an initial step for the regenerative design models for the future to create sustainable systems, which integrate materials needs, society socio-economic requirements and environmental integrity.

6. Process integration as a holistic approach for cassava biobased material development and sustainable process design framework

Until very recently, most cassava biobased materials were developed following stand-alone approaches intended for mono-functional applications. The current growth in process integration has put pressing challenges to biobased material development. Currently, little is known about integrated process designs which give insights into different cassava biobased material property interactions and their synergistic effects on the performance, economy and quality of materials and products. Process integration designs ensure an output material has multifunctional applications.

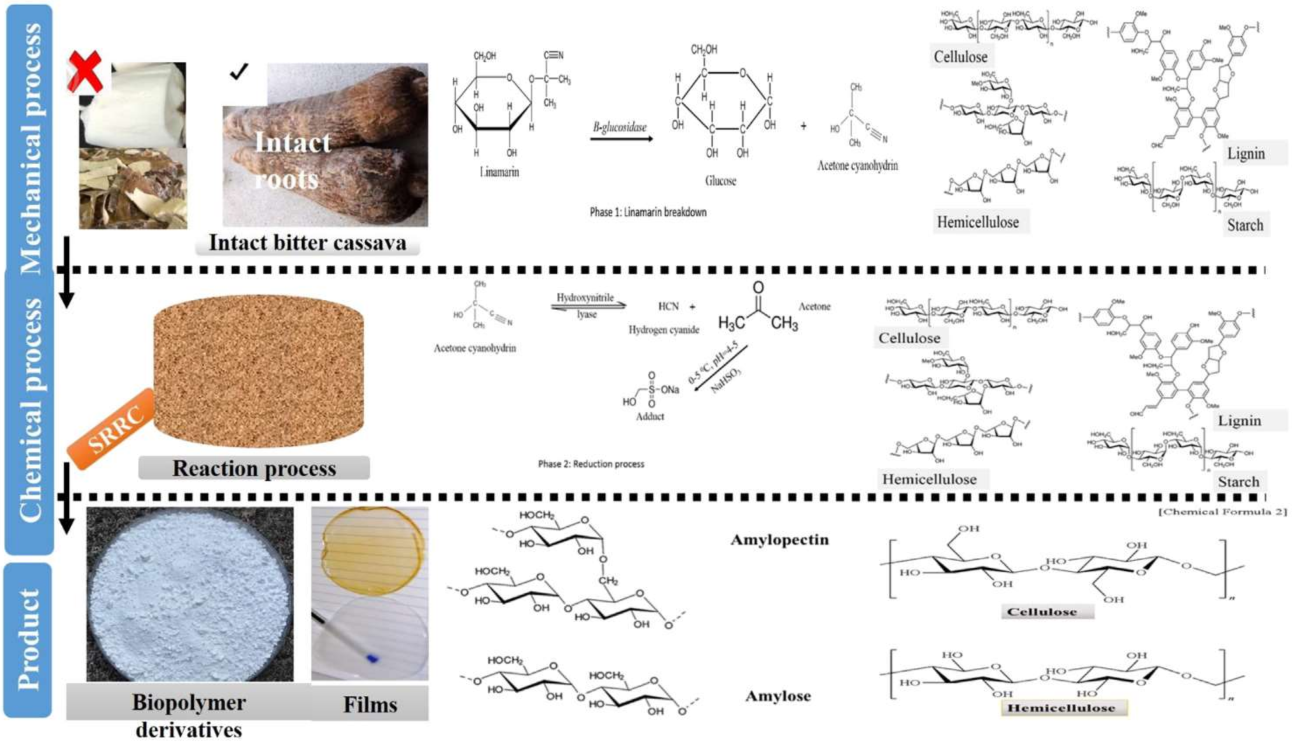

Fig. 1. Overview of simultaneous release recovery cyanogenesis (SRRC) concept using intact (whole) cassava for sustainable waste minimisation, recovery of valuable biopolymer derivatives, and food packaging development.

This is achieved by evaluating and optimising individual processes, followed by integrating their impacts and synergies/interrelationships to obtain an efficient and sustainable system.

Process integration for cassava biobased material development was achieved by integrating SRRC green processes using intact bitter cassava (Tumwesigye et al., 2016a) (discussed above), modified atmosphere packaging, desirable package optimisation, and package performance simulating real conditions (Fig. 2). Integrating SRRC-assisted waste minimisation and package production as well as desirable modified atmosphere packaging optimisation and validation can improve energy efficiency, reduce costs and lead to sustainable cassava biobased systems.

6.1. Process modelling, optimisation and integration of cassava development processes

The potential of desirability optimisation of individual SRRC to improve the efficiency of processes and reduce total cyanogens at

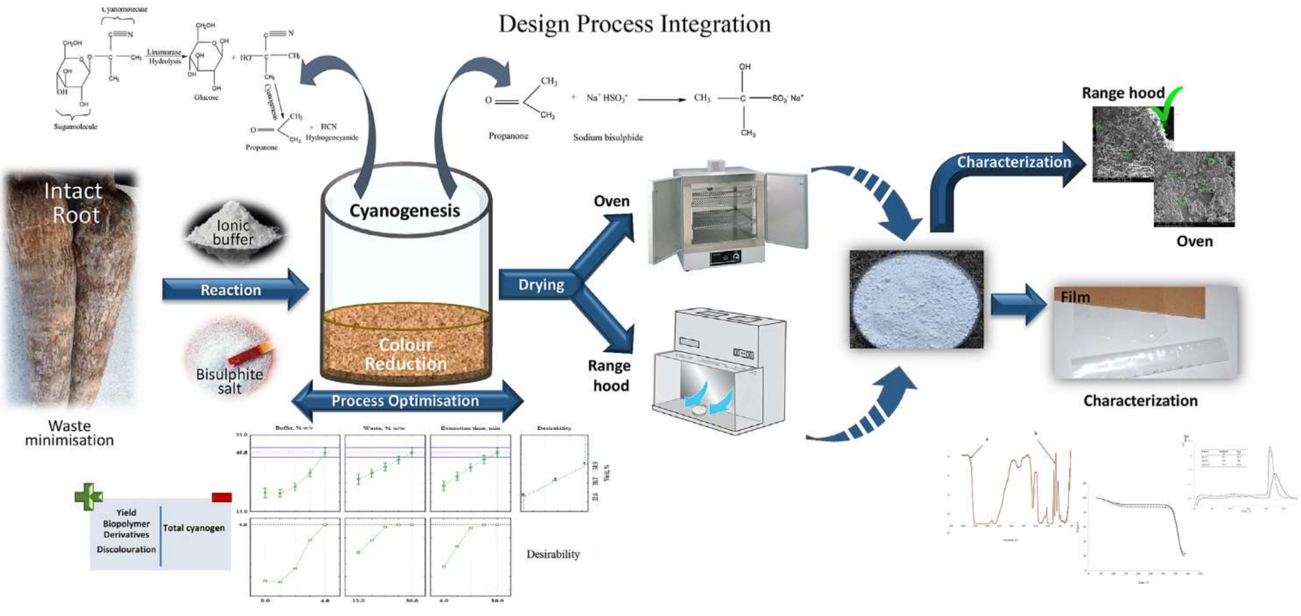

Fig. 2. Process design integration concept, showing how different processes are integrated into the development of cassava biopolymers and packaging films.

low cost and energy were fully studied (Tumwesigye et al., 2016a). It was observed that functionalisation of the reaction with buffer (4% v/v), waste biomass (30% w/v) and extraction time (10 min) led to a considerable increase in yield of cassava derivatives from the

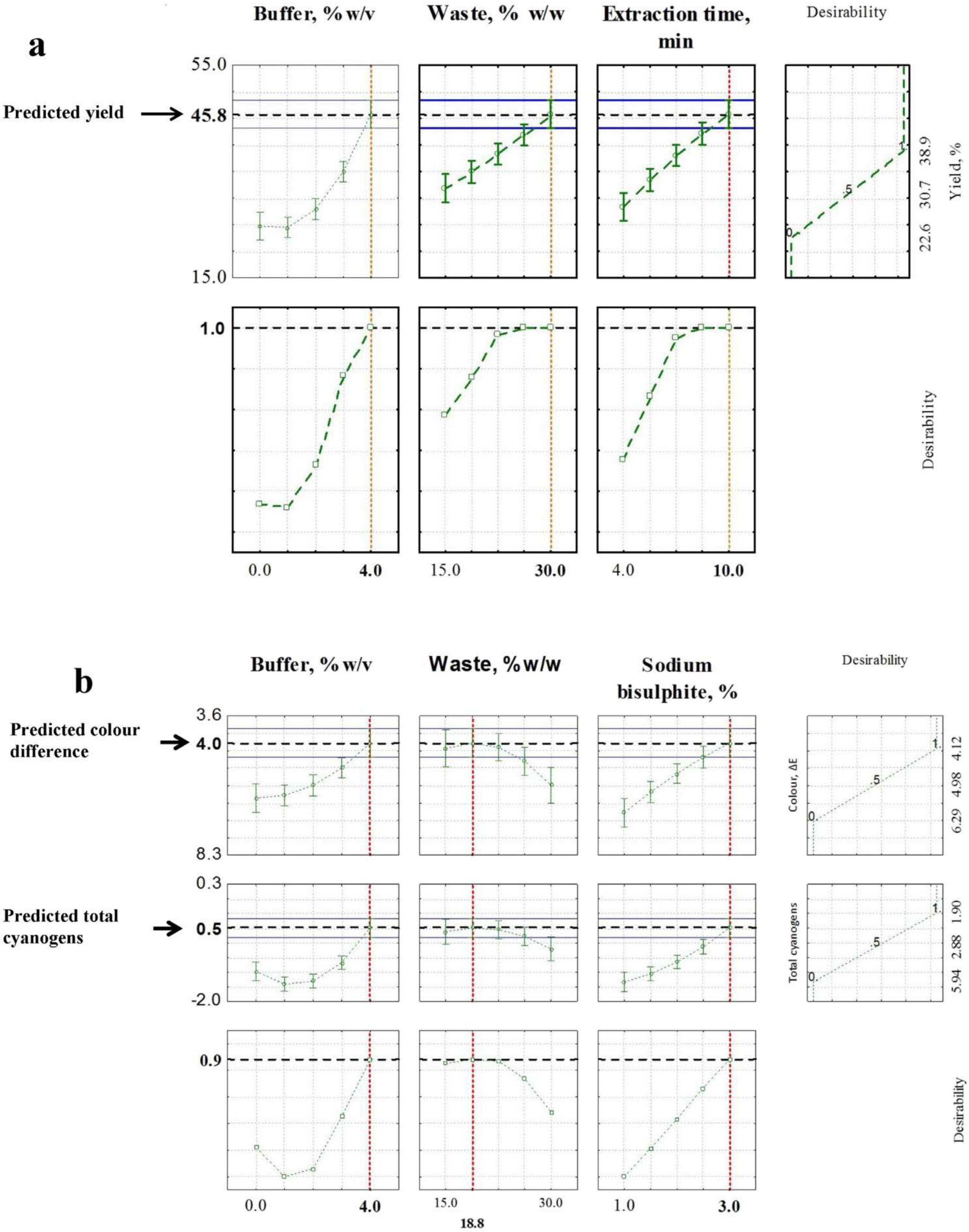

Fig. 3. Global desirability analysis of (a) yield, and (b) total cyanogens and colour difference.

conventional 30–40% to 45.8% (Fig. 3a). Essentially, it meant that all the waste biomass was utilised, and thus reducing the byproduct waste in one process. Fig. 3.

In the same desirability optimisation study, the impact of functionalisation on the reaction stage was found to significantly reduce total cyanogens to 0.5 ppm and showed a significant improvement in colour of the byproduct biomass (Fig. 3b). Conventionally, the above concurrent improved reactions were achieved in more than one process, thereby raising energy and cost requirements.

When desirability optimisation was studied in respect to the effect of recycled waste water to process more cassava waste biomass on the saving energy on deionising water and maximise resource utilisation, it was observed that the integrated reuse save about 60% of extraction solvent by the fourth cycle, in comparison to the traditional process (Tumwesigye et al., 2016a).

Similarly, Tumwesigye et al. (2016a) found better drying rates of cassava derivative powders and film prototypes when the rangehood was integrated into the process design than the conventionally used heat oven. They concluded that drying of cassava biobased materials could be an option in energy-efficient and low-cost process designs.

The same research studied integrating package development into the process design. The aim was to exploit available waste derivatives, the same solvent source and a rangehood drying process in order to minimise environmental wastes and energy for solvent purification and materials drying. It was observed that it is possible to integrate film package development using optimised processes (Tumwesigye et al., 2016a). The concept is shown in Figs. 2 and 4.

6.2. Desirability optimisation package design and package performance

Desirability optimisation package design is a non-conventional method in atmosphere packaging. Desirability optimisation package design uses multi-response optimisation based on desirability function (Derringer, 1980), and use process targets and response deviations to represent a single objective. To achieve the desired products with highest productivity, some processing inputs and outcome measures must be assigned weights so as to keep them small or large. Both inputs and outputs are transformed from multiple objectives to sole objective by transforming: (i) individual performance measures into individual desirability function; and (ii) multiple individual desirability functions into one objective function, the overall (global) desirability function.

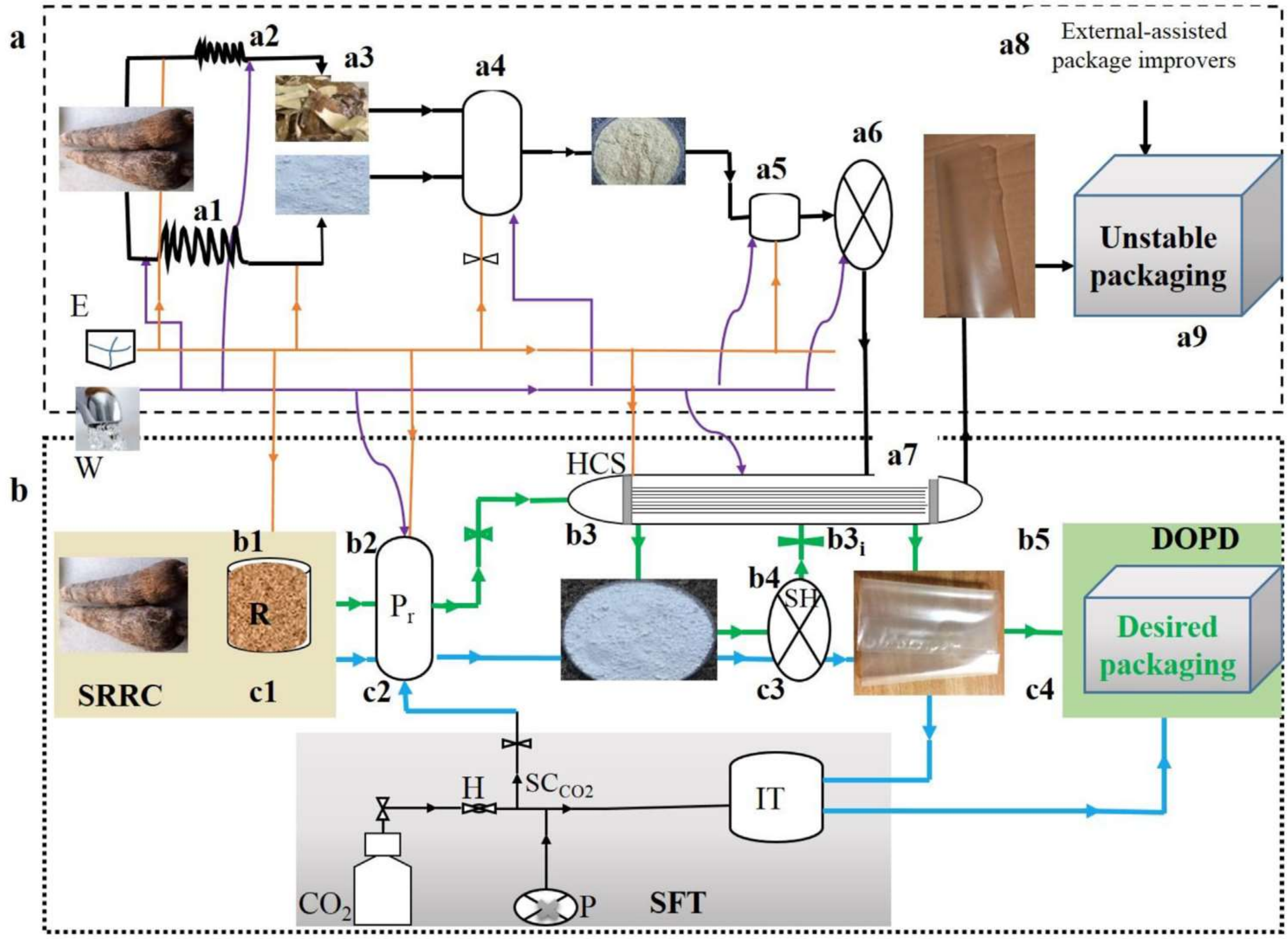

Fig. 4. Flowsheet of cassava biobased materials using conventional process (a) **(a1-a9)** and proposed process considering integrated process design (b) by combining either: simultaneous release recovery cyanogenesis (SRRC) & desirability optimisation packaging design (DOPD) **(b1-b5)** or SRRC, superfluid technology (SFT) & desirability optimisation packaging design (DOPD) **(c1-c4)** to reduce cost and energy for every process. E, energy source; W, water source; HCS, heating & cooling system; Pr, recovery; SH, solution heating; H, filter; P, pump; IT, CO_2 impregnation tank; a1 & a2, series of steps for starch production & waste peel preparations; a3, bagasse refinement; a4, reaction tank; a5, biocomposite refinement, a6, solution preparation and casting; a7, drying & cooling; b1, pulp tank; b2, reaction tank; b3 & b3i, heating & cooling; b4, solution preparation and casting; c1, pulp tank; c2, reaction tank; c3, solution preparation and casting.

Limited use of desirability optimisation package design in the development of cassava biopolymer derivatives and packaging films has been reported in literature (Tumwesigye et al., 2016b).

Unfortunately, there is no information in literature showing use of desirability optimisation package design in modified atmosphere packaging designs. Since the package environment is influenced by many factors, and only a few have been used in the modified atmosphere packaging designs and development, it is necessary that desirability optimisation package design is applied, the models developed thereof validated and package performance is done with real conditions. The scale-up of this design could help obtain an efficient modified atmosphere packaging designs with wide applications. In recent unpublished study of desirability optimisation of equilibrium modified atmosphere packaging design parameters, a desirable 5.85% O_2 concentration was obtained with optimal design parameters of temperature (10 °C), RH (95%), one perforation (0.27 mm), and desirability of 0.97 (Sousa et al., unpublished work) Consequently, desirability optimisation package design was integrated in the process design (Fig. 4).

7. Prospects for future work: integrated process design tools

Up until now, cassava biobased packaging development has concentrated on optimisation of individual processes. However, the holistic approach to integrating cassava processes is promising (Tumwesigye et al., 2016a), and with industry demand for integrated processes growing rapidly, further improvement using emerging techniques and methods is crucial. Among the optimisation techniques, pinch analysis and mathematical optimisations have been well-studied and employed in understanding many processes. Comprehensive investigation of pinch analysis and mathematical optimisations will ensure inexpensive, energy-efficient and time saving. Pinch analysis and mathematical optimisations is used synonymously with process integration, and is based on computing feasible low energy and low-cost process targets by optimising integrated process network elements, all underpinned by better process understanding (Kemp, 2007). Besides, the concept of impregnation of functional chemicals using supercritical fluids has the beneficial impact of green processing. Supercritical fluid impregnation has been demonstrated in dyeing textile materials (Liu et al., 2006)

Although reduction of polymer matrix-dye during SRRC process in bitter cassava was demonstrated using food grade sodium bisulphite to improve the colour of intact bitter biopolymer derivatives (Tumwesigye et al., 2016a), furthering this technique could also be explored to lower energy and costs of production.

Bearing in mind global interest in green processes, use of supercritical fluid drying technique would ensure development of homogeneous cassava packaging films, and avoid energy and cost implications of using drying ovens to remove water from cast solutions, which has always posed challenges of non-uniformity in materials with multiple heterogeneous surfaces and various thicknesses (Benali & Boumghar, 2015; de Moraes, et al., 2015; Tumwesigye et al., 2016a; Tumwesigye et al., 2016b; Tumwesigye et al., 2016c).

8. Conclusion

The high starch demand and its production using conventional methods is associated with environmental wastes.

Various approaches have been employed for developing biobased materials, with a growing trend in the materials structural and functional improvements using reinforcements such as fillers, bioactive compounds and chemical modification. Although, their superior properties are better than those of starch matrices, they have the potential disadvantage of increasing the costs and energy associated with material production. The materials are limited to the development stage but fall short of evaluations with intended application conditions. Their interaction with in-package environment under real conditions is non-existent, and their influence and performance is not known. Thus, the exploration of these materials in commercial use, mainly food packaging, is challenging.

Holistic studies, integrating cost-effective, energy-efficient, green processes, using standard methodology, optimising conditions and properties and validation with specific products and environments, can be a sustainable strategy for increased commercial use, primarily food packaging. Understanding the performance of cassava biobased materials in target foods and environment is crucial to their accelerated adoption.

These findings however reflect a great potential of cassava biobased materials, as packages, in wide-range applications.

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