Journal of Cleaner Production 387 (2023) 135830

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Greenhouse gas emissions of biobased diapers containing chemically modified protein superabsorbents

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ARTICLE INFO

Handling Editor: Prof. Jiri Jaromir Klemeš

Keywords:
Superabsorbent
Diapers
Protein
Sustainability
Circularity

ABSTRACT

Replacing the current mainly fossil-based, disposable, and non-biodegradable sanitary products with sustainable, functional alternatives is an industry priority. Suggested biobased alternatives require evaluation of their actual impact on greenhouse gas (GHG) emissions. We evaluated GHG emissions of biobased baby diapers as the most consumed sanitary product, using a biodegradable functionalized protein superabsorbent polymer (bioSAP) and compared them with currently used fossil-based counterparts. Assessment of the diapers also included estimated GHG emissions from the production of the biobased components, transport, and end-of-life combustion of these items. It was shown that only a few of the biobased diaper alternatives resulted in lower GHG emissions than commercial diapers containing fossil-based materials. At the same time, it was demonstrated that the production of the bioSAP via chemical modification of a protein raw material is the primary GHG contributor, with 78% of the total emissions. Reduction of the GHG contribution of the bioSAP production was achieved via a proposed recycling route of the functionalization agent, reducing the GHG emissions by 13% than if no recycling was carried out. Overall, we demonstrated that reduced and competitive GHG emissions could be achieved in sanitary articles using biobased materials, thereby contributing to a sanitary industry producing disposable products with less environmental pollution while allowing customers to keep their current consumption patterns.

1. Introduction

The global market for hygiene products has continuously increased over recent decades, and the forecast for total market value is expected to reach 650 billion USD by the end of 2022 (Essity Year, 2019; Smithers Smithers forecasts global hygiene, 2022). In this market, about 50% accounts for baby and adult single-use diapers that are also disposable (ARC, 2022; IMARC Diaper Market, 2022). The combination of convenient and effective single-use diapers with a liquid absorption capacity above 20 g/g (saline solution) and an increasing world population are reasons for the approximate 8% increase in the use of these products compared to 2018.

The major constituents of a conventional single-use diaper (and most sanitary products) are an outer layer of a polyethylene/polypropylene film, an inner layer of polyethylene/polypropylene nonwoven, and a middle layer of cellulose pulp combined with a synthetically produced superabsorbent polymer (SAP) that absorbs liquid and prevent its leakage (Buchholz and Graham, 1998; Capezza et al., 2019a). The SAP in this product engineering context represents the most important part for the functionality of the diaper with a weight fraction representing ca. 30% of the overall diaper weight (Buchholz and Graham, 1998; Buchholz, 1994). The SAP's high absorbance results from the polymeric chains that are charged and partially crosslinked, explaining also the retention of liquids (Capezza et al., 2019a; Cuadri et al., 2016, 2017; Damodaran, 2001, 2004; Pourjavadi et al., 2008; Rathna et al., 2004). However, the present industrially produced single-use diapers, with their materials mainly constructed from the polymerization of petroleum-derived monomers, has a substantial carbon footprint (Capezza et al., 2021; Cordella et al., 2015; Mendoza et al., 2019). The associated high amount of waste generated for incineration or landfilling (around 20 million tons/year globally of disposable baby diapers)

Abbreviations: FU, functional unit, one diaper.

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https://doi.org/10.1016/j.jclepro.2022.135830
Received 10 February 2022; Received in revised form 19 October 2022; Accepted 30 December 2022
Available online 31 December 2022

Abbreviation list

GHG	Greenhouse gas
bioSAP	Protein superabsorbent polymer
SAP	Superabsorbent polymer
TRL	Technology readiness level
WG	Wheat gluten protein
PPC	Potato protein concentrate
MQw	MilliQ water
EDTAD	Ethylenediaminetetraacetic dianhydride
SA	Succinic anhydride
SN	Supernatant
SC	Swelling capacity
NaPA	Sodium Polyacrylate
PP	Polypropylene
bioPP	bio Polypropylene
PE	Polyethylene
bioPE	bio Polyethylene
PLA	Polylactic acid
PU	Polyurethane
GWP	Global warming potential

(IMARC Diaper Market, 2022; Capezza et al., 2021; IMARC Personal Hygiene Market, 2021) contribute additionally to a negative environmental impact. Efforts that critically review the selection of the materials used in disposable diapers, being the most consumed hygiene product, are needed.

So far, some studies have focused on replacing the outer fossil-based plastic layers with biobased polyolefins counterparts as a more sustainable material for diapers (Siracusa and Blanco, 2020). To our knowledge, studies measuring the impact of replacing the SAP with more sustainable alternatives in single-use diapers are scarce, while the SAP has been reported to contribute significantly to the carbon footprint compared to the outer plastic layers in these products (Cordella et al., 2015; Edana Sustainability Report, 2005). Partially, this can be associated with the challenge of finding alternative high-performance absorption materials that are biobased and biodegradable, making the task of replacing the industrially produced SAP seemingly difficult (Capezza et al., 2021; Mülhaupt, 2013). Still, there is an urgent need for making more sustainable diapers and replacing all fossil-based components with biobased and biodegradable alternatives allowing society to reach a more circular lifestyle, in agreement with the United Nations 2030 Agenda. Here, a critical point regarding the sustainability of disposable sanitary items in today's market is that recycling methods for these products become difficult as they are contaminated with biological waste once used.

Plant-based protein concentrates are promising candidates for the production of sustainable SAPs with the potential to replace synthetic and fossil-based alternatives (Cuadri et al., 2016, 2017, 2018; Capezza et al., 2019b, 2019c, 2020a). Protein concentrates that are not used for food production are readily available as industrial co-products (*e.g.,* potato protein, porcine plasma), and provide an extensive range in their ability to provide fluid absorption in their processed states (Álvarez-Castillo et al., 2020; Capezza et al., 2020b; Muneer et al., 2018; Newson et al., 2015). Although proteins are heterogeneous materials, facile reactions that modify their liquid swelling performance have been demonstrated, increasing their liquid absorption capacity by 4000% (Capezza et al., 2020a). At present, no investigations have been reported evaluating the environmental impact of using such protein-based SAPs in hygiene products. Furthermore, as the protein-based SAP is currently only produced on a laboratory scale (low technology readiness level, TRL), there is a motivation to obtain more knowledge regarding protein-based SAP production sustainability and then use it as a guiding tool when developing upscaling paths/techniques.

In the present study, step-wise replacements of fossil-based components to develop and produce single-use diapers were evaluated, and the environmental impact of these replacements in terms of GHG emissions with a cradle-to-grave perspective were assessed. For a broad understanding, the 1-min swelling capacity was evaluated experimentally in a reference product (SAP from a commercial diaper) and biobased and biodegradable alternatives of such a product. The study focused on the industrial co-products wheat gluten and potato protein concentrate as protein-based sources for fabricating the biodegradable SAP alternatives. Then, a GHG emission assessment was carried out regarding replacing the other fossil-based components in diapers. The different options for achieving partial or complete renewability and biodegradability allowed us to propose a product configuration where the GHG emissions from the replacement of various parts could be compared with current commercial products. The suggestions proposed pave an avenue for designing effective and sustainable disposable sanitary items that comply with current market trends based on products with no possibility of being recyclable once used.

2. Materials and methods

This section summarizes the experimental work to produce the superabsorbent protein polymer (bioSAP) and describes the suggested production routes to decrease their greenhouse gas (GHG) emissions during production. We also describe a system study to assess the GHG emissions of the proposed product alternatives, from material use to the disposal of the used product.

2.1. Material preparation

2.1.1. Protein and SAP material

Wheat gluten protein (WG) and potato protein concentrate (PPC) were used as received (both co-products from industrial processes). The WG was provided by Lantmännen Reppe AB, Sweden, with a protein content of 86% (Dumas method, NMKL 6:2003, Nx6.25). The PPC was provided by Lyckeby Starch AB, Sweden, with a protein content of 82% (Dumas method, Nx6.25). Ethylenediaminetetraacetic dianhydride (EDTAD, >98%) and succinic anhydride (SA, >99%) were purchased from Sigma-Aldrich, Sweden. Synthetic sodium-neutralized polyacrylic acid superabsorbent (SAP) was extracted from commercial diapers for babies between 0.5 and 2 kg, purchased locally. Briefly, commercial diapers were cut in half, and the SAP particles were removed from the core. Any attached cotton fibers were removed from the SAP powder.

2.1.2. Synthesis of bioSAP

For each of the two protein sources (WG and PPC), the two most promising acylation modification routes reported in the literature were selected, (Capezza et al., 2019b, 2020c, 2021; Cuadri et al., 2018), and a total of four bioSAPs were produced and designated as WG1, WG2, PPC1, and PPC2.

The WG1 sample was produced following the methodology described in Capezza et al. (2020c) Briefly, 8 wt% protein suspension was prepared by mixing as-received WG powder with MilliQ water (MQw). The pH was adjusted to 11 using 1M sodium hydroxide (NaOH) under constant stirring. The suspension was heated at 90 °C for 30 min, cooled to room temperature, and adjusted to pH 12 for optimal acylation conditions. Then, 25 g of EDTAD/100 g of WG were gradually added to the suspension over 30 min, and the suspension was kept under stirring for another 90 min. The pH of the suspensions was constantly monitored and adjusted to pH 12 using 1 M NaOH. Thereafter, the unreacted EDTAD was removed by precipitating the protein at pH 3–3.5 using 1M HCl, centrifuging the suspension at 2000 RCF for 10 min, decanting the supernatant (SN), and replacing it with fresh MQw. The cleaning process was repeated, and thereafter the centrifuged pellet was neutralized to pH 7–7.5 using 1 M NaOH. The pellet was dried in a forced-air oven at

Table 1
Recipes for producing 1 kg of biodegradable functionalized protein superabsorbent polymer (bioSAP) as used in the life cycle assessment.

BioSAP label	Protein	Water	EDTAD	SA	NaOH, 1 M	HCl, 1 M	Total
	[g]	[g]	[g]	[g]	[g]	[g]	[g]
WG1	1300	50000	325	0	2080	816	54521
WG2	1300	50000	0	0	624	612	52536
PPC1	1300	195000	325	0	2080	816	199521
PPC2	1300	10000	0	2600	2080	0	15980

WG = wheat gluten, PPC = potato protein concentrate, EDTAD = ethylenediaminetetraacetic dianhydride, SA = succinic anhydride. The BioSAP abbreviation refers to the sample preparation method (see section 2.1.2).

50 °C for 12 h, and the dried films were ground to particles using a mortar and pestle.

The preparation of the WG2 sample was identical to the one described for WG1, except that no EDTAD was added, and the protein modification relied only on the alkaline treatment (pH 11, 1 M NaOH).

The PPC1 sample was prepared using wet acylation following previous literature (Capezza et al., 2021) and the process described for WG1 with the following exceptions: i) the protein concentration of the suspension was 2 wt%, and ii) the precipitation of the EDTAD-functionalized protein material was performed at the reaction pH (pH 12, using 1 M NaOH) instead of acidic pH.

For the preparation of the PPC2 sample, a dry acylation procedure was used as reported in previous works (Capezza et al., 2019b; Chiou et al., 2013; Robertson et al., 2014). The as-received PPC was mixed with MQw forming a 40 wt% mixture. The pH of the mixture was adjusted to pH 11 (using 1 M NaOH) and then placed in a reactor pre-heated to 70 °C. Succinic anhydride (SA) was added to the reactor to obtain a mass ratio of 0.5:1 (protein: SA). Then, the reactor's temperature was gradually increased to 140 °C for 30 min. After reaching 140 °C, the acylation process continued for 1.5 h. The mixture was poured into a beaker containing an excess of fresh MQw and vigorously mixed to remove the unreacted SA. The suspension was adjusted to pH 7, filtered with filter paper, and dried in a forced-air oven at 50 °C for 12 h.

2.1.3. Swelling characterization

The teabag method was used to obtain the materials' swelling capacity (SC), following the nonwoven standard procedure 240.0.R2 (Edana, 2015). Plastic teabags were filled with ca. 150 mg of each powder, and the bags were immersed in beakers containing MQw for 1 min. The bags were kept hanging 10 s outside the liquid and gently placed on tissue paper for 10 s to remove excess water. Identically handled teabags without any sample were immersed in MQw and used as a reference for calculating a correction factor (B). The SC of a synthetic SAP obtained from a commercial diaper was used as reference material. The results were calculated using Equation (1) and are reported as the mean and standard deviation of triplicates.

$$Swelling\left(\frac{g}{g}\right) = \frac{Wi - (Wb * B) - Wd}{Wd} \quad \text{(Equation 1)}$$

where *Wi* is the weight of the sample and teabag after the respective immersion time, *Wb* is the weight of the teabag, *B* is the correction factor for the wet teabag, and *Wd* is the weight of the dry SAP.

2.1.4. Recovering EDTA

For evaluating the possibilities for recovering EDTA (produced by the EDTAD acylation process, see section 2.1.2), the pH of the supernatant obtained after the first cleaning process (PPC/ED SN) was reduced to 6.06, 5.01, 4, 3.11, 2.03, 1.04, and 0.66, using 1 M HCl. Before each pH adjustment, aliquots of the PPC/ED SN suspension were taken and centrifuged at 5500 rpm for 15 min. The SN from each pH precipitation was decanted, and the resulting pellet and SN were frozen at −25 °C and lyophilized for 72 h.

The resulting lyophilized SN and pellets from each pH precipitation were evaluated using FTIR. A Perkin−Elmer Spectrum 400 (USA) coupled to an ATR Golden Gate unit and a triglycine sulphate (TGS) detector (Graseby Specac LTD, England) was used to obtain the FTIR data. The spectrum was obtained after 32 consecutive scans per sample with a step of 1.0 cm^{-1} and a resolution of 4.0 cm^{-1}. The lyophilized SN from each pH precipitation was also analyzed using ^{1}H NMR at room temperature for a quantitative analysis of the EDTA left in the SN. A Bruker Avance III HD 400 MHz instrument with a BBFO probe equipped with a Z-gradient coil was used for structural analysis (Bruker, UK). The data were processed with MestreNova (Mestrelab Research) using the residual solvent signal of D_2O as a reference. For the study, 50 mg of the lyophilized SN powders were individually dissolved in 1 mL of D_2O. The undissolved protein components were filtered using a 0.45 μm polypropylene membrane.

2.2. Systems study

A systems study was carried out to assess the potential environmental benefits and possible pitfalls in terms of GHG emissions of different alternative configurations of diapers for newborns.

2.2.1. Approach

Climate impact from the production and end-of-life treatment of diapers and other sanitary products can potentially be high due to the large annual production and consumption as well as the fossil origin of many of the components of such products. This study does not assume a decrease in diaper products but aims to investigate the potential GHG emission reductions when fossil-based material components are replaced with alternatives with a lower climate impact. It is worth mentioning that some of the materials suggested as replacements for the fossil-based components are still not, to our knowledge, commercial products but are reported as successful laboratory results (TRL < 3–4). Consequently, in this study, the GHG emissions from the production of alternatives of partly or wholly renewable and partially or fully biodegradable diapers for newborn babies were assessed and compared to the production of a conventional fossil-based diaper. Furthermore, different bioSAPs were compared with the current fossil-based standard SAP of sodium polyacrylate (NaPA). The focus was on GHG emissions as the first proof-of-concept step using a life cycle assessment methodology and principles (ISO, 2006). An economic assessment was not included in this study but is considered one of the next steps in developing the gluten and potato protein-based bioSAPs. To demonstrate the variation in the data used, we tested low and high GHG emission cases in all comparisons based on the range of emission data used.

2.2.1.1. BioSAP composition. For the GHG emission assessment, the same protein sources (WG and PPC) and synthesis routes were used as described in section 2.1.2, and these are summarized together with reagents used to produce 1 kg of the bioSAP in Table 1.

A recovery rate of 80% of EDTA was used in the assessment based on experimental studies on recovering EDTA described in section 2.1.4. For succinic acid, higher recovery rates have been described in previous studies (Nghiem et al., 2017; Mesch and Wittwer, 1976); however, based

Table 2
Composition of the reference and alternative diapers as investigated in the GHG emission assessment. Grey = fossil-based resources; orange = biobased, non-biodegradable resources; green = biobased, biodegradable resources.

Diaper materials	Reference fossil SAP	Alternative I bioSAP	Alternative II Biobased, non-biodegradable	Alternative III biobased and biodegradable
SAP	Sodium polyacrylate	bioSAP	bioSAP	bioSAP
Fiber pulp	cellulose	cellulose	cellulose	cellulose
PP and replacements	PP	PP	bioPP	PLA nonwovens
(LD)PE and replacements	PE	PE	bioPE	PLA nonwovens
Adhesives	Synthetic rubber	Synthetic rubber	Avoided by welding instead	
Elastic bands	Synthetic rubber & PU	Synthetic rubber & PU	bio-based PU[2]	Natural rubber
Others (*e.g.*, tape, elastic back ear, other synthetic polymers)	PP	PP	bioPP	PLA nonwovens

(LD)PE = (low density) polyethylene, PP = polypropylene, PU = polyurethane, PLA = polylactic acid.

on technical aspects related to opportunities for comparisons, we assumed the same recovery rate of 80% for SA as was used on experimental results for EDTAD.

2.2.1.2. Overall composition of the different diaper alternatives. Conventional diapers use mainly fossil-based materials, except the cellulose fluff pulp to direct the liquids toward the SAP component (Table 2). We investigated three (3) alternatives to compare conventional commercial diapers to more sustainable alternatives. The first alternative examined a diaper where a bioSAP replaces the SAP component. The second alternative was a diaper where all structural materials (including the SAP) were replaced with biobased materials that were, however, non-biodegradable. The third alternative consisted of a diaper where all structural materials were replaced with biobased and biodegradable materials. The details of the different alternatives, and motivations for the choice of these, are provided below.

In our assessment, we assumed a reference diaper for newborn babies of 0.5–2 kg weight, composed of 19.5 g SAP, 2.15 g fiber pulp, 4.5 g polypropylene (PP), 2.1 g low-density polyethylene (PE), 0.9 g adhesive, 0.3 g elastics bands, and 1.2 g other materials, adding up to a total weight of 50 g (Edana Sustainability Report, 2005). This recipe was based on that presented in EDANA (2005) (Edana Sustainability Report, 2005). It should be mentioned that the fiber pulp used was reduced by 90% from 21.5 g per diaper. The modification was done to increase the amount of bioSAP used in the three alternatives to allow the suggested product to match the 1-min swelling performance of the current commercial SAP. The modification was also considered realistic, considering the current trend toward reduced fiber content in diapers (Cordella et al., 2015). Table 2 shows the diaper configuration used for the different GHG emission assessments.

For the Alternative I diaper, the fossil-SAP was replaced with bioSAP, providing the same performance in the 1-min swelling capacity. Higher content of bioSAP than fossil-SAP was needed in the above EDANA-based recipe (Edana Sustainability Report, 2005), based on the bioSAP having lower swelling capacity than fossil-based SAP. Thus, compared to the 19.5 g fossil-SAP per diaper utilized for the assessment, the corresponding content of bioSAP was 79.6, 92.9, 47.8, and 99.5 g per diaper for WG1, WG2, PPC1, and PPC2, respectively.

For Alternative II, a diaper containing biobased but non-biodegradable materials, we replaced fossil-based PE, PP, and synthetic rubber with biobased PE, PP, and biobased PU, respectively.

For Alternative III, a biobased and biodegradable diaper was considered using PLA nonwoven materials to replace PP and PE components. Natural rubber was again used for the elastic parts. In alternatives II and III, welding was used to connect the layer materials rather than adhesives (see Table 2). Note that, in this study, we assumed that the structural materials exchange was possible from a technical perspective and that the exchange occurred on a 1:1 mass ratio.

Table 3
Emission factors for fossil SAP and the compounds used to produce the bioSAPs.

Compound	GHG emissions [g CO_2-eq/kg]		References
	Low	High	
Fossil SAP[a]	3290	3814	(Mirabella et al., 2013; Gontia and Janssen, 2016)
Wheat gluten (WG)	1072	1551	(Deng, 2014), present calculations
Potato protein concentrate (PPC)[b]	371	1000	(Tromp, 2020; Röös, 2013)
EDTAD[c]	4750	5700	Wernet et al. (2016)
Succinic anhydride (SA), fossil[d]	2370	3539	(Patel et al., 2018; Cok et al., 2014)
Succinic anhydride (SA), renewable[d]	504	870	(Moussa et al., 2016; González-García et al., 2018)
Sodium hydroxide, NaOH, 1 M	10	53	Dahlgren et al. (2015)
Hydrochloric acid, HCl, 1 M	36	160	CoW Winnipeg sewage treatment program (2011)

[a] Sodium polyacrylate.
[b] Estimated from the carbon footprint from potato starch production and production efficiency of 256 and 13.7 kg per ton potato starch and PPC, respectively.
[c] Estimated from the carbon footprint of EDTA and increased by 20% for the heating required to synthesize EDTAD.
[d] Assumed the same as succinic acid based on the assumption that the energy used for the dehydration reaction is negligible.

2.2.2. GHG assessment

2.2.2.1. Superabsorbent component. The production of bioSAP/SAP was assessed concerning GHG emission to determine if bioSAPs can fulfill the requirements of a lower environmental impact compared with the fossil reference. The emissions considered in this assessment occur in processes where energy and materials are used according to section 2.1.1. Sodium polyacrylate (NaPA) was used as a reference SAP. The reference case was based on fossil-derived acrylic acid and the current carbon intensity of the energy used in the manufacturing process.

As this is a proof-of-concept study, we have assumed that the

Table 4
GHG emission factors for materials used in the production of the investigated diaper alternatives.

Component	GHG emissions [g CO_2-eq/kg]		References
	Low	High	
Cellulose fiber pulp	554	560	(Wernet et al., 2016; Husgafvel et al., 2016)
PP	1630	1903	(Wernet et al., 2016; Europe, 2014)
bioPP[a]	723	1560	Moretti et al. (2020)
PE	1870	2936	(Europe, 2014; Benavides et al., 2020)
bioPE[a]	677	2109	(Benavides et al., 2020; Koch and Mihalyi, 2018)
Synthetic rubber, PU[b]	2403	5336	Wernet et al. (2016)
Natural rubber	535.8	2678	(Wernet et al., 2016; Jawjit et al., 2010)
biobased-PU	3358	4075	Manzardo et al. (2019)
PLA non-wovens	2352	3563	(Benavides et al., 2020; Walker and Rothman, 2020)

PP = polypropylene, PE = polyethylene, PU = polyurethane, PLA = polylactic acid.
[a] bioPP and bioPE are chemically identical to PP and PE, respectively, but are produced from renewable raw materials.
[b] The low value refers to synthetic rubber, the high value refers to PU.

Table 5
Assumptions used in assessing GHG emissions from the transport of diapers. VW = vehicle weight.

Parameter	Unit	Truck and trailer	Truck and semi-trailer
		(low case)	(high case)
Maximum total truck weight	[ton]	62	42
Transport capacity	[ton]	36	20
	[m^3]	150	135
	EUR pallets[a]	48	26
Transport distance	[km]	200	500
Diesel consumption [b]	[L/100 km]	$6.60 \bullet 10^4 \bullet VW + 13.777$	$5.83 \bullet 10^4 \bullet VW + 12.392$

[a] EUR pallets have a base area of 1.2 m × 0.8 m, and a loading height from the base of 1.8 m was assumed. Each pallet was supposed to hold 50 boxes containing 88 diapers each.
[b] Regression model based on data presented by Agioutantis et al., 2013). (Agioutantis et al., 2013).

assembly processes of the bioSAP diapers are similar to those of conventional diapers. Consequently, we have concentrated on the emissions from the raw material used in the process.

The environmental impact assessment was carried out using GHG emission factors for the production means stated in Table 1. In the "high" case, no recovery of the functionalization agents (EDTAD and succinic anhydride) was considered in the GHG assessment. A "low" case scenario was also assessed due to the significant impact of using these agents, assuming an 80% recovery of unused agents from the reactor, as described in section 2.1.4 and 2.2.1.1. For the recovery, additional amounts of HCl for pH adjustment were considered, but no additional energy costs, resulting in pH-driven precipitation of the agents, such as EDTA and succinic acid, were implemented. We also assumed that only a minor amount of energy is required to dry the precipitate and thermal conversion to EDTAD and succinic anhydride. The emission factors for production means used for the reference and alternative SAPs are presented in Table 3.

2.2.2.2. Diaper materials (non-absorbent components). The GHG emission assessment for the complete diapers was carried out using emission factors for the amounts of production means as described in section 2.2.1.2. The emission factors for production means used for the reference and alternative SAPs are presented in Table 4.

Table 6
Carbon content and proportion of fossil carbon for production means. The global warming potential (GWP) was calculated from fossil carbon, assuming complete combustion.

Compound	Carbon content	Fraction of fossil carbon	GWP
	[%]	[%]	[g CO_2-eq/kg]
Fossil SAP $(C_3H_3NaO_2)_n$	38.3	100	1405
Wheat gluten (WG)	n/d	0	0
Potato protein concentrate (PPC)	n/d	0	0
EDTAD ($C_{10}H_{12}N_2O_6$)	46.9	100	1719
Succinic anhydride ($C_4H_4O_3$), renewable	40.7	0	0
NaOH, 1 M	0	n/a	0
HCl, 1 M	0	n/a	0
Cellulose fiber pulp	n/d	0	0
PP, fossil[a]	85.7	99.1	3115
PE, fossil[a]	85.7	98.3	3090
bioPE and bioPP	85.7	0	0
Synthetic rubber[b] ($C_{15}H_{24}$)	88.2	100	3233
Natural rubber	38.1	0	0
PLA non-wovens	50.0	0	0
Elastic[c] - polyurethane ($C_{17}H_8N_2O_4$), fossil	67.1	100	2461
Elastics[c] - polyurethane ($C_{17}H_8N_2O_4$), renewable	67.1	0	0

[a] Renewable carbon content according to industry standards (Europe, 2017).
[b] Assumed as styrene-butadiene rubber (SBR).
[c] Assumed as 50% styrene-butadiene rubber (SBR) and 50% polyurethane.

2.2.3. Transport of diapers

For the transport of the diapers, two different transport options were implemented as a low and high case of GHG emissions, representing two transportation options for road transport as commonly used in product distribution. Transport with a higher transport capacity for a shorter transportation distance (60-ton truck and semi-trailer, 200 km) and transport with a lower transportation capacity for a longer distance (42-ton truck with a semi-trailer, 500 km) was assumed for the low and high case, respectively (Table 5). Despite the increase in the amount of bio-SAP in the diapers resulting in a diaper weight increase with a negligible increase in volume, the same number of diapers were assumed to be transported for both reference and alternative diapers (221200 and 114400 diapers per truckload in the low and high case, respectively). However, according to a regression model, fuel use was assumed to change due to weight changes (Table 5). GHG emissions from diesel consumption were calculated based on diesel energy content (37.4 MJ/L) and combustion emissions (83.8 g CO_2-eq./MJ) (Union, 2009).

2.2.4. End-of-life emissions of the disposed diaper

Combustion was considered for the end-of-life handling of the diaper, including the SAP component. We accounted for carbon dioxide emissions as a resulting product that affects the climate negatively. Only carbon emissions from a fossil source were accounted in the GHG assessment; biogenic carbon was regarded as not adding to the environmental effect. The resulting global warming potential emission factors were estimated based on the carbon content of the compounds (Table 6).

3. Results and discussion

3.1. Experimental evaluation of the BioSAP

The swelling capacity (SC) of the bioSAP was evaluated to allow

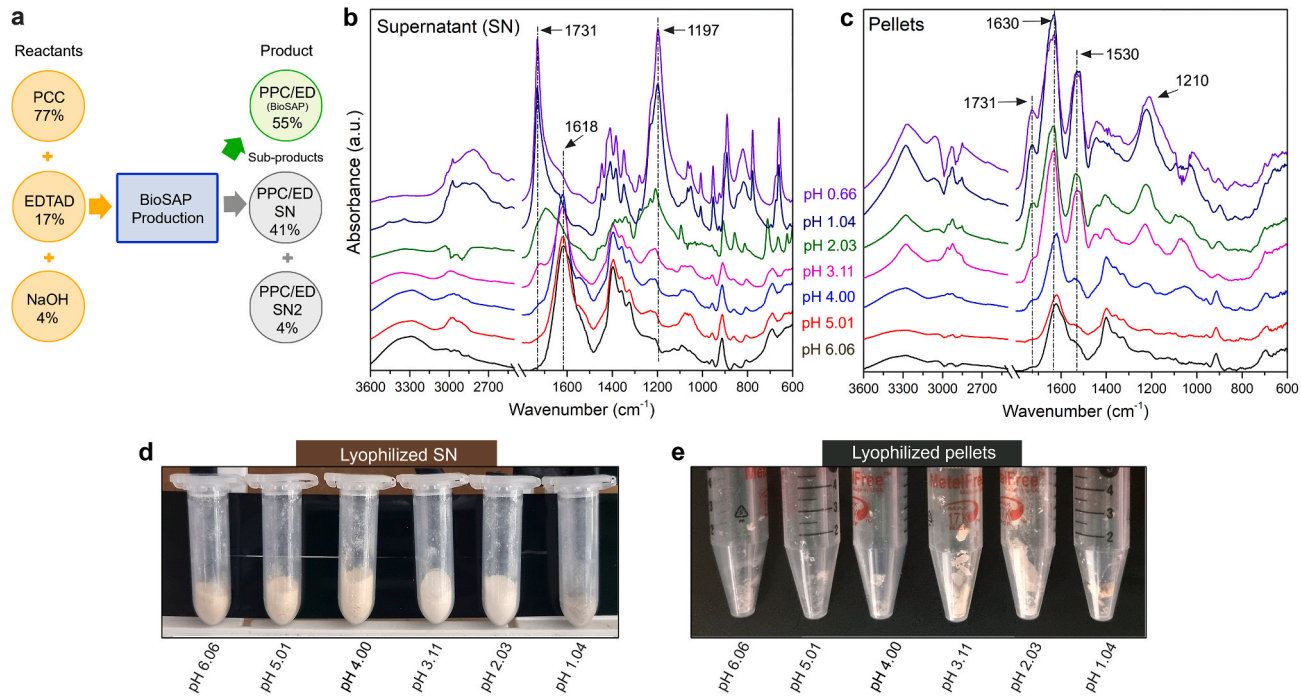

Fig. 1. Overall experimental evaluation of the recycling process of EDTA from the EDTAD protein acylation process. (a) Mass balance of the reactants and product/sub-products, (b–c) FTIR of the samples processed at different pHs and lyophilized: (b) showing the SN and (c) showing the pellets. (d–e) Visual appearance of the lyophilized samples at different pHs: (d) showing the SN fractions and (e) the pellet fractions. Arrows in the FTIR spectrum point at the main peaks used for the discussion.

environmental impact comparisons of the bioSAP alternatives with the current commercial diapers. Furthermore, to reduce the negative environmental impact of the production of the bioSAP (discussed in section 4.2), a recycling step of the reagents used was tested here. The recycling step as a parameter in the assessments was also evaluated experimentally. The outcome of these evaluations is presented and discussed below.

3.1.1. *Swelling performance*

The investigated bioSAPs showed varying swelling capacities after 1-min immersion in saline solution (0.9 wt% NaCl), *i.e.*, 5, 4, 8, and 4 g/g for WG1, WG2, PPC1, and PPC2, respectively. The use of the saline solution is motivated since this liquid is used as an industry standard to assess the liquid swelling capacity of superabsorbents in sanitary articles (Capezza et al., 2019a). The 1-min swelling capacity of the bioSAPs in saline solution corresponded to 20–41% of that of the fossil reference SAP extracted from commercial diapers. The swelling capacity for the bioSAP presented here was similar to that previously reported for absorbents produced using acylation reactions and similar protein sources (Capezza et al., 2019b, 2020a, 2020c, 2021; Cuadri et al., 2018). The increase in liquid absorbance of the proteins after acylation is ascribed to the functionalization of the amino acid groups (*e.g.*, lysine), forming charged entities that increase electrostatic repulsion of the polypeptide chains, which allow higher liquid encapsulation compared to untreated proteins (Capezza et al., 2019a; Damodaran, 2001, 2004).

The results showed a lower superabsorbent capacity of the gluten (WG) and potato protein (PPC) bioSAPs compared to the synthetic SAP. Thus, an increased bioSAP content in diapers is required to secure similar liquid uptake as commercial alternatives. We suggest a scenario where 70–90% of the pulp fiber is replaced by bioSAP to maintain the weight of future diaper alternatives. The bioSAP materials have been shown to have excellent liquid spreading properties compared to commercial SAP, as illustrated in Supplementary Video 1. Consequently, this demonstrates that the pulp fiber, with the function of spreading the liquid throughout the entire diaper, can be replaced without changing the product functionality. Moreover, the PPC and WG proteins are considered suitable as feedstock to produce bioSAP alternatives to commercial SAP due to their large availability and low cost as industrial co-streams from the global starch industry with a price below 2 EUR/kg (Capezza et al., 2019a; Damodaran, 2001, 2004). However, environmental assessment of the production process of these bioSAPs has not been performed, which is the next step to ensure that the design of the process is in-line with low environmental impacts.

Supplementary video related to this article can be found at https://doi.org/10.1016/j.jclepro.2022.135830

3.1.2. *Recycling of reagents for decreasing the environmental impact of the bioSAP production*

Recycling possibilities of unreacted reagents from the protein modification process is crucial for environmental and economic sustainability. In the case of bio-succinic acid, systems are already developed to recover it from fermentation broths, followed by conversion to anhydride (Mesch and Wittwer, 1976; Nghiem et al., 2017). The bioSAP matrix is considerably less complex than the industrial fermentation broths, indicating that the succinic acid (SA) recovery is also less complicated. Methods used on industrial fermentation broths typically entail precipitating SA as calcium succinate or forming diammonium succinate, separating the succinate from the solution. Subsequently, the semi-purified SA is treated with acid (*e.g.*, H_2SO_4), yielding calcium or ammonium sulphate with free SA, which after concentrating the SA solution and crystallization yields pure SA (Nghiem et al., 2017). The conversion of the succinic acid to the anhydride form is then accomplished through thermal dehydration, with or without a solvent (Mesch and Wittwer, 1976). Also, SA waste from the production of the bioSAP could potentially be used as raw material for the production of polyesters (Ito et al., 2002).

We evaluated EDTA recycling experimentally from EDTAD not bound to the PPC bioSAP (PPC1) after the functionalization step. As shown in Fig. 1a, the acylation of the PPC with EDTAD resulted in circa 55% of the dry initial mass leading to a PPC1 bioSAP (PPC/ED), while

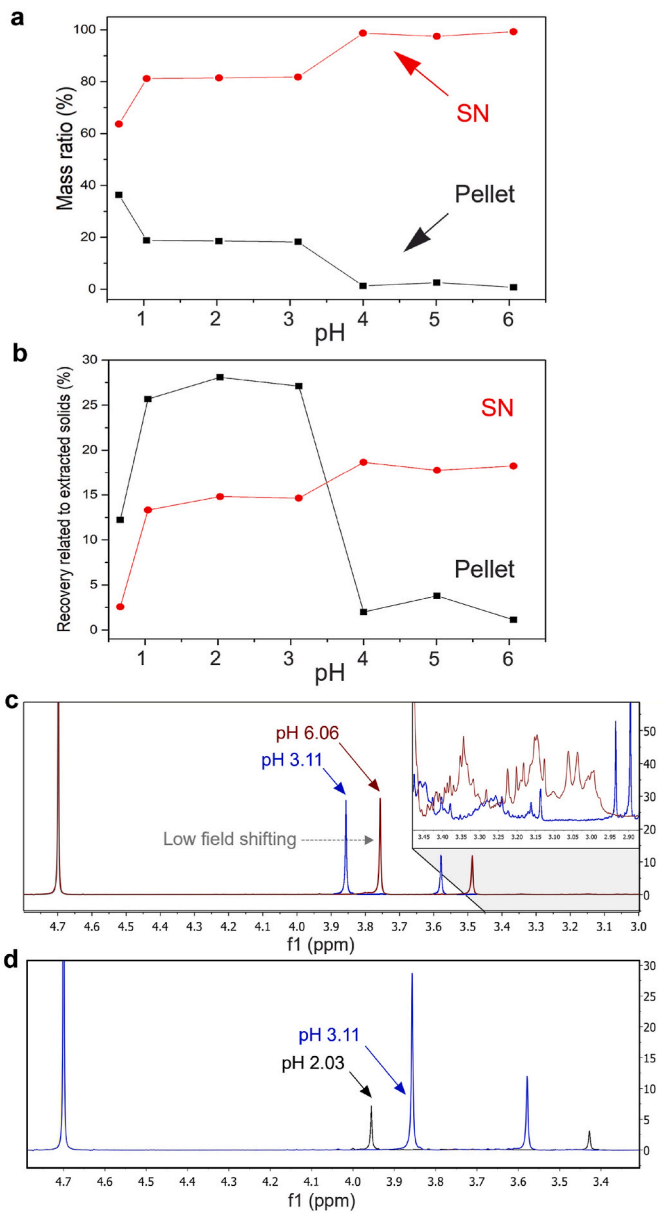

Fig. 2. Mass evaluation of the recycling process of EDTA from the EDTAD protein acylation and ^{1}H NMR. (a) Mass ratio of pellet and SN for each precipitation pH and (b) the relative mass to the total theoretical mass of pellet and SN for each precipitation pH. (c-d) ^{1}H NMR spectrum of SN for (c) pH 6.06 and 3.11 and (d) pH 3.11 and 2.03.

41% of the dry mass ended up in the supernatant SN (PPC/ED SN) after the first cleaning process. The PPC/ED SN is rich in unreacted EDTAD (in the EDTA form) and soluble protein.

The recovery of the unreacted EDTAD was evaluated by treating the PPC/ED SN at different pHs and centrifuging the suspension to separate the soluble supernatant (SN) and non-soluble fractions at each pH (pellet). Fig. 1b shows that the FTIR spectrum was similar to the SN from the centrifuged PPC/ED SN treated at pH 6.06 and 5.01. The band centered at 1618 cm^{-1} originates from the combination of the Amide I and Amide II peaks from the protein still soluble at these pHs (1630 and 1530 cm^{-1}, respectively) (Cho et al., 2011). The strong peak observed at 1575 cm^{-1} is assigned to the asymmetric stretching vibration of the carbonyl group in the ionic form (-COO^{-}) from the EDTA (Fig. 1b). (Suárez et al., 2013) A similar FTIR spectrum was also obtained for the PPC/ED SN pellets after the centrifugation at pH 6.06 and 5.01 (Fig. 1c). However, the low recovered solid content (<2%) of these pellets suggests that both protein and reagents were traces of SN that was not decanted from the pellet (hence not shown in Fig. 1d and e).

Fig. 1d and e shows the lyophilized SN and pellet, respectively, after the PPC/ED SN treatments at different pHs with further centrifugation to recover the unreacted EDTA. The color of the lyophilized SN after centrifugation at pH 6.06, 5.01, and 4 was light brown, while at pH 3.11 and 2.03, the color was light cream (Fig. 1d). The color of the pellet from the centrifugation of the PPC/ED SN was generally light brown, independently of the precipitation pH (Fig. 1e).

Fig. 2 shows the mass balances of pellets and SN obtained from the centrifugation of the treated PPC/ED SN at the different pHs. Our results suggest that most soluble proteins precipitate at pH 3.11, leaving an EDTA-rich SN after PPC/ED SN centrifugation. The relative content of EDTA in the SN after centrifugation of PPC/ED SN at pH 3.11 was, on average, 80%, while the solid content of the pellet after this centrifugation was higher than when other pHs were applied (>25%, Fig. 2b). The results agrees with the FTIR of the SN showing the peak formed at 1210-1197 cm^{-1} (assigned to C–N from tertiary amides), suggesting the presence of tri/di/monosodium EDTA complexes in the SN and the pellet precipitated at pH 2.03 (Fig. 1b).

The ^{1}H NMR confirmed a high concentration of free EDTA in the SN of the centrifuged PPC/ED SN at pH 6.06. Fig. 2c shows two single peaks at $\delta = 3.76$ and 3.49 ppm; the latter corresponds to the 8 methylene protons of the acetyl moieties, and the former to the 4 protons of the ethylenediamine moiety (Mónico et al., 2017). Many small peaks could be observed between $\delta = 3.44$ and 2.95 ppm at pH 6, probably from dissolved protein fractions in the SN of the centrifuged PPC/ED SN (see the inset image in Fig. 2c).

Fig. 1c shows that the amide I and amide II peaks at 1630 and 1530 cm^{-1} increase in intensity in the pellet of the centrifuged PPC/ED SN while pH treatment was changed from 4 to 3.11. The FTIR spectrum of the SN from the centrifuged PPC/ED SN treated at pH 3.11 showed a reduced peak intensity at 1530 cm^{-1}, but an increasingly intense peak at 1731 cm^{-1} assigned to the stretching of the –COOH vibration of saturated dimers of carboxylic acid from the mixture of tri/di/monosodium EDTA complexes (Lanigan and Pidsosny, 2007) (Fig. 1b). In addition, the ^{1}H NMR profile for the SN from the centrifuged PPC/ED SN treated at pH 3.11 showed that the small peaks with $\delta = 3.56$ and 3.10 ppm were reduced, which may originate from the protein being precipitated from the SN (inset image in Fig. 2c). The decrease of pH when treating the SN also led to a low field shifting of the EDTA signals due to the different protolysis levels, an effect that has previously been described by Hafer et al. (2020)

The ^{1}H NMR of the SN after centrifugation of the PPC/ED SN at pH 2.03 revealed that the signals of the free EDTA singlets were reduced significantly, suggesting that most EDTA commenced precipitating at this pH (Fig. 2d). Furthermore, the two singlet peaks from EDTA became much more distanced, from $\delta = 3.86$ and 3.58 ppm at pH 3.11 to $\delta = 3.96$ and 3.48 ppm at pH 2.03. Previous studies have also reported this distance broadening of the singlets arising from the low pH (Mónico et al., 2017). Thus, our result indicates that protonated EDTA precipitated in treatments of pH $\leq$ 2.03, and thereby the pellets obtained from the centrifugation after treatment at pH 2.03, 1.04, and 0.66 consisted mainly of protonated EDTA and precipitated protein. Therefore, the optimal pH to precipitate the protein from the PPC/ED SN is ca. 3, simultaneously as the EDTA complexes are left in the SN, resulting in an estimated recovery of 80%. An additional precipitation step at pH < 2 can be used on the SN of the centrifuged PPC/ED SN at pH 3 to precipitate the EDTA and reduce the amount of water that needs to be evaporated in the recovery process.

Overall, even with a potential 80% recirculation of SA and EDTA from the production of bioSAP, it should be pointed out that any wastewater from the production process should be treated in a wastewater treatment plant. This is especially important with the EDTA complexes, which are highly chelating. However, these salts are

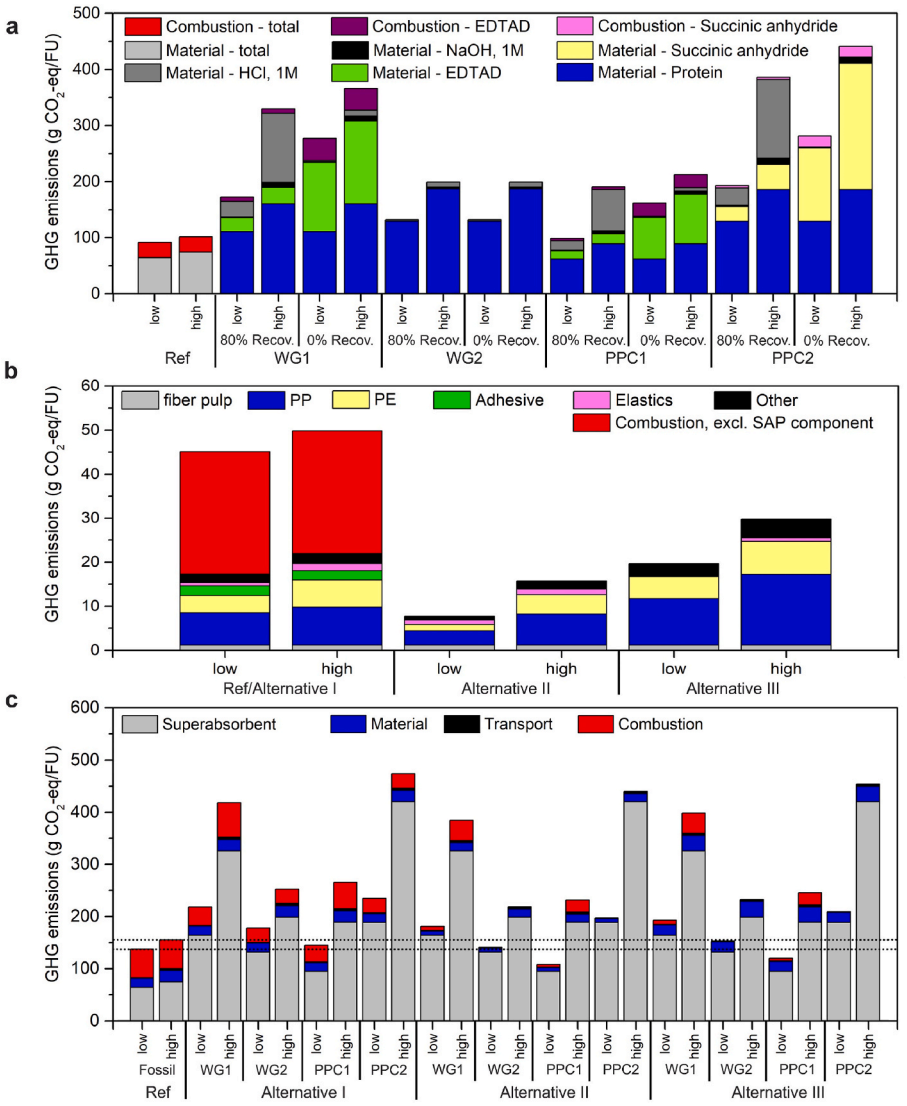

Fig. 3. Assessment study in terms of Greenhouse gas (GHG) emissions. GHG emissions for: (a) the amount of SAP used per FU based on emissions from SAP production and end-of-life combustion, (b) the materials (excluding the SAP component) used per FU in the reference diaper and the alternatives, and (c) the total GHG emissions from the whole diaper (FU). PP and PE mark both fossil-based polypropylene and polyethylene materials and their biobased replacements.

efficiently removed if treated/degraded by wastewater treatment processes (Zoe et al., 2012).

3.2. GHG emissions of suggested diaper alternatives

3.2.1. SAP component

It is considered here that bioSAPs need to match the performance of the reference SAPs to compete with these. We compared the different SAPs using the swelling capacity in saline solution (0.9 wt% NaCl) after 1 min as a performance indicator. Fig. 3a shows that GHG emissions varied considerably for the production (material) and end-of-life (combustion) of the different SAPs. Emissions from the sodium polyacrylate (NaPA) used as reference superabsorbent polymer (SAP) varied between 92 (low case) and 102 (high case) g $CO_{2\text{-eq}}$ per FU (functional unit/one diaper), using fossil-derived acrylic acid and current carbon intensity energy in the manufacturing process. The greenhouse gas (GHG) emissions from the production and end-of-life combustion of the bioSAPs ranged between 99 and 441 g $CO_{2\text{-eq}}$ per FU. GHG costs are mainly caused by the protein component and the acylation agent.

A high GHG emission from bioSAP alternatives (Fig. 3) is, to a great extent, the result of a lower swelling capacity of the bioSAPs compared to commercial counterparts, which results in a need to increase the amount of SAP material to obtain the same total liquid absorption. The functionalizing agents, EDTAD and succinic anhydride contribute significantly to the GHG emissions, which calls for recycling these agents to reduce GHG from bioSAP production. Recovery of the acylation agents resulted in a strongly reduced amount of the agents needed for production. Instead, a considerable amount of HCl is required for the pH adjustment, impacting the GHG cost (Fig. 3a).

With an 80% recovery of EDTAD, the potato protein-based PPC1 in its low case performed similarly to the fossil-based reference (Fig. 3a). The second best alternative was WG2, although with at least 30% higher GHG emissions than the reference diaper.

3.2.2. Diaper item (non-SAP diaper components)

Fig. 3b shows that the materials used in the reference diaper (similar to the alternative I) resulted in considerably higher GHG emissions than alternatives II and III (see Table 2 for their description). The high GHG emissions are primarily due to non-renewable materials that release fossil carbon during end-of-life combustion. Replacing the fossil-based PP and PE components with biobased PP and PE (alternative II) reduced GHG emissions from the diaper material mix compared to the

reference diaper. The utilization of only biobased and biodegradable materials (alternative III) increased the material-related GHG emissions compared to all other alternatives, while the total GHG emissions were still considerably lower than the reference when the combustion-related GHG emissions were also considered.

Fig. 3c shows that the SAP component was the single most significant contributor to GHG emissions in all diapers. SAP represented 47–48% of the total GHG emissions in the fossil-based reference diaper (alternative I). In the alternatives II and III herein presented, the contribution from the bioSAP component was even higher, 65–86, 81–94, and 77–89%, for alternatives I, II, and III, respectively. The non-SAP material of the diapers contributed with 3–16%, while transport contributed with 1–2%. Combustion emissions contributed 36–40%, 9–22%, 0–10%, and 0–10% from the fossil-based reference and the alternatives I, II, and III, respectively. Combustion emissions from the non-SAP materials contributed 28 g CO_2-eq/FU in the reference diaper and zero in the alternative diapers (not shown). In the bioSAP, the acylation agent EDTAD contributed to the combustion of GHG emissions, while a renewable succinic anhydride was used in the SAP synthesis.

The GHG emission estimated from the different bioSAP-based diapers differed substantially. However, only the PPC1 "low" alternatives II and III led to lower GHG emissions than the reference, while WG2 "low" alternatives II and III were on par with the reference diapers (Fig. 3c). These most favorable bioSAP diaper alternatives with similar GHG emissions as commercial SAP (Fig. 3a), even with additional material, resulted in lower GHG emissions than commercial diapers (Fig. 3b).

3.3. Degradability

Combustion is the standard end-of-life scenario for diapers in many industrial countries, where energy recovery is part of waste treatment. In other countries, diapers and other SAP-containing products end up in the environment, *e.g.*, in landfills or sewerage systems (Ntekpe et al., 2020). Public agencies have disclosed disposal of used sanitary items, such as diapers, as problematic due to reported contamination of groundwater with pathogens and chemicals such as acrylic acid from SAP degradation that can leach slowly into the environment over a long period. In addition, the long-term degradation of the PE-PP contained in current commercial diapers (Reference) and the suggested bioPE-bioPP alternatives (Table 1) may lead to the production of microplastics lixiviating when disposed of in landfills and in underground waters (Zhang et al., 2022). In this regard, alternative III with biobased, biodegradable non-sodium acrylate SAP, *i.e.,* protein-based bioSAP may reduce the abovementioned environmental loads from the polymers in these sanitary products. The advantage of using bioSAP alternatives is that it is well known that bioplastics based on proteins are degraded into safe molecules thereby not producing toxic microplastics (Capezza et al., 2021; Rosenboom et al., 2022; Jiménez-Rosado et al., 2020).

The use of cellulose to produce hydrogels and biobased residual streams from pulp production for the production of polyacrylates have been suggested as other approaches to improve the sustainability of SAPs (Gontia and Janssen, 2016; Alam et al., 2019). Gontia and Janssen (Mesch and Wittwer, 1976) (1976) showed that a de-fossilization of the sodium polyacrylate (NaPA) production process could reduce emissions from SAP materials by 40% (Gontia and Janssen, 2016). Still, the obtainable sustainability, even of biobased polyacrylates, suffers from the low degradability of the SAP. Thus, leaking degradation products may cause other environmental impacts such as higher levels of eutrophication, acidification, and ozone impacts compared to the fossil-based sodium polyacrylate (Gontia and Janssen, 2016). Advantages of using proteins as bioSAP not only relies on their fast biodegradability (being 50% degraded in 15 days) (Capezza et al., 2021) but also on their recently demonstrated biostimulation effect when incorporated into crops as absorbents (Jiménez-Rosado et al., 2021).

3.4. Potential improvements

3.4.1. Further GHG emissions reductions

There are different potential approaches to further reduce the GHG emissions of the investigated bioSAPs, which include performance and material-based approaches. Regarding performance, swelling capacity of SAPs is linked to the amount of SAP needed to match the requirements for their use in conventional diapers (Buchholz, 1994). For example, more efficient absorbents will imply less material needed in these products while new materials with less swelling capacity will require the addition of more material (Capezza et al., 2021). Therefore, performance improvements will affect the material-related emissions and the diaper's weight. Furthermore, weight changes will affect the emissions originating from the transport of the diapers, although this might be a minor effect.

Regarding the material-based approaches, reducing the amount of acylation and functionalization agents, *e.g.,* EDTAD and succinic anhydride is here seen as a way forward to decrease emissions. This is indicated by the marginal consumption of these compounds in the acylation process, resulting in a significant part of the agents remaining unreacted after manufacturing of the suggested bioSAPs, as reported here and in literature (Damodaran, 2001, 2004; Capezza et al., 2019b, 2020c, 2021). Overall, it was herein concluded that the recirculation and possible upcycling of these agents is paramount for decreasing emissions related to bioSAP production and future sustainable diapers. Alternatively, since recirculation/recycling of the acetylation agents leads to non-negligible GHG emissions, biobased acetylation agents could be used. Alternative production methods based on renewable feedstock exist for these components, *e.g.*, via biomethanol (Shamsul et al., 2014) or sugar-based succinic acid (Johansson et al., 2015), respectively, which can be used to reduce fossil carbon emission from SAP synthesis further.

3.4.2. Feasibility

The protein co-streams, WG and PPC, originate from the bioethanol and starch industry (Capezza et al., 2020b, 2021; Muneer et al., 2018; Newson et al., 2015). The agro-food industry is well established, thereby continuously producing side- and co-stream proteins as long as the processing exists to extract starch-based products. Utilizing these streams for bioSAP production will not negatively affect the food production system as a direct competition with resources needed in the food industry is avoided (*e.g.,* starch, dietary fibers, etc.). Furthermore, the valorization of these streams will add value to the agro-industrial product chain and strengthen the food industry with increased resource use efficiency and additional income (Álvarez-Castillo et al., 2021). However, competing uses for WG and PPC exist, *e.g.,* in packaging, cosmetics, detergents, adhesives, animal feed, and food ingredients (Bietz and Lookhart, 1996).

The protein components used to produce the proposed bioSAP are readily available in relatively large amounts on the market (Capezza et al., 2020c, 2021). Still, there is a need to evaluate the economic costs of replacing conventional SAPs with bioSAPs. However, a positive indication is that raw materials for bioSAP production are available at bulk prices similar to those of the fossil-based SAP counterpart (Capezza et al., 2019a). Also, the cost to adapt the production lines for producing bioSAP is considered low, as they rely on conventional reaction methods, *e.g.*, the use of reactors and other technologies already used industrially (Capezza et al., 2020b). Other methods of introducing carboxyl functionality to proteins include bulk esterification with polycarboxylic acids (Chiou et al., 2013), where biobased non-anhydride reagents should have a lower GHG potential and unreacted reagent recycling should be more efficient.

The overall technology readiness of the presented concept was assessed in a preliminary and simplified manner to indicate its technological status. Technology readiness levels (TRL) are case-sensitive and need to be evaluated closely with participating business partners, *e.g.*,

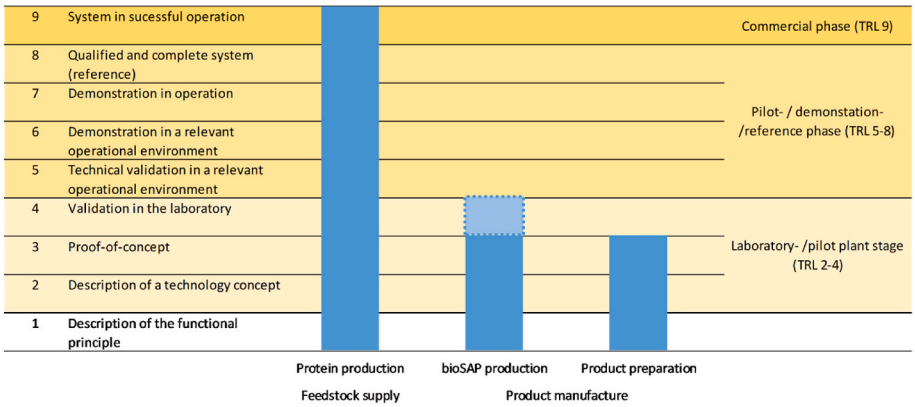

Fig. 4. Technology readiness levels (TRLs) as preliminary assessed for the production steps of the concept, adjusted based on the TRL concept of the European Commission (EC, 2012) (Commission, 2012).

when an innovation program is implemented.

The starch production industry and its operations are considered to have reached TRL9 (Fig. 4), including the protein side- and co-streams of WG and PPC, which are traded internationally as a commodity. For example, the US WG market is expected to be 2.58 billion USD in 2022 (Markets, 2026), and the world PPC market is expected to be over 88 million USD by 2022 (Markets, 2022).

Currently, no industrial processes are being implemented for the commercial production of protein-based superabsorbent polymers. Therefore, developing an industrial product and eventual commercialization of bioSAPs will have to go through a pilot phase that builds on the current laboratory validation (TRL4, as shown in Fig. 4).

Literature reports on protein-based bioSAPs are based on laboratory-scale investigations. The focus has been on using protein-based materials for diapers and agricultural uses (Damodaran, 2001; Capezza et al., 2021; Jiménez-Rosado et al., 2021). However, new applications have emerged for these types of materials, *e.g.*, in the packaging industry, medical applications, and other applications requiring high-liquid uptake and retention (Dhanapal and Subramanian, 2021; Schröfl et al., 2022). Similar to the presented application in diapers, these new utilization possibilities for bioSAP material require laboratory validation (TRL 4).

SAP products are traditionally delivered as ready-for-use powders for the final product manufacture, *e.g.*, diapers. Depending on the scale of the manufacturer involved, product packages may range from 20 kg sacks or bulk bags (ca. 700 kg) to rail car-scale deliveries. Particle sizes are typically 150–600 μm for diaper applications (Buchholz and Graham, 1998), while in agricultural and waste remediation applications, they can be as large as 4 mm (Technologies, 2022). For product preparation of the bioSAP material, laboratory validation (TRL 4) is needed.

4. Conclusions

Fully biobased diapers can contribute lower GHG emissions than currently used fossil-based diapers. The GHG emission of the diapers can be reduced by having the diaper designed with bioSAPs (biodegradable) combined with biobased materials. The superabsorbent component (either bioSAP or fossil-based SAP) was revealed to be the responsible for the highest share of the total GHG emissions, involving the manufacturing (cradle) to the end-of-life disposal of the diaper (grave). However, bioSAPs produced using raw materials as a side- or co-products from the food industry have the advantage of being biodegradable, contrary to fossil-based SAPs. The high GHG from the production of the bioSAP alternatives results from the need for a higher content of the bioSAPs in the diapers and the chemical functionalization of the proteins to obtain the bioSAP. We demonstrated the possibility of reaching >80% recovery of the chemicals used to functionalize the proteins (EDTAD, succinic anhydride), which decreased the GHG emissions by 29–39%. Using biodegradable polymers to fabricate the next generation of disposable single-use sanitary articles and their environmental validation is a significant step towards a more sustainable industry and society and a realistic path for it to comply with the Sustainable Development Agenda for 2030.

CRediT authorship contribution statement

Antonio J. Capezza: Conceptualization, Formal analysis, (Experimental), Methodology, (Experimental), Investigation, (Experimental), Data curation, Visualization, Funding acquisition, Writing – original draft, Writing – review and editing. **William R. Newson:** Data curation, Formal analysis, Investigation, Writing – original draft, Writing – review and editing. **Faraz Muneer:** Formal analysis, Writing – original draft, Investigation, Writing – review and editing. **Eva Johansson:** Validation, Investigation, Writing – review and editing. **Yuxiao Cui:** Formal analysis, Data curation, Writing – original draft. **Mikael S. Hedenqvist:** Investigation, Validation, Writing – review and editing. **Richard T. Olsson:** Investigation, Validation, Writing – review and editing. **Thomas Prade:** Conceptualization, Formal analysis, (GHG), Methodology, (GHG), Investigation, (GHG), Data curation, (GHG), Funding acquisition, Writing – original draft, Writing – review and editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Acknowledgments and funding

The Bo Rydins Stiftelse for independent research (grant F 30/19) is acknowledged for the financial support provided to Antonio Capezza. This study was also supported by South-Baltic Area Interreg project *Bioeconomy in the south Baltic area: Biomass-based Innovation and Green Growth* (BioBIGG) and the strong research area *Trees and Crops for the Future* (TC4F).

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