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Product-driven process synthesis: Extraction of polyphenols from tea

Aleksandra Zderic [a], Edwin Zondervan [b, *]

[a] Eindhoven University of Technology, De Zaale, 5612 AJ Eindhoven, The Netherlands
[b] Bremen University, Leobener Straße, D 28359 Bremen, Germany

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ABSTRACT

In this contribution, the Product-Driven Process Synthesis methodology is used as a well-defined structured approach for the conceptual design of an extraction process for polyphenols from fresh tea leaves. A detailed specification of the starting material (fresh tea leaves) and product (polyphenols) leads to a subsequent definition of fundamental tasks to convert our raw material into the desired product. Among the different mechanisms and techniques that could be used to perform the tasks under mild conditions, pulsed electric field has been selected as non-invasive and non-thermal method for cell wall disruption. To define the operating window for pulsed electric field method an experimental design has been defined and executed (varying several settings of the pulsed electric field). From the collected data, the analysis of variance has been used to determine which variables are significant i.e. electric field strength, pulse duration and number of pulses. Box-Behnken design has been used as part of statistical analysis to find optimal pulsed electric field settings to maximize the extraction yield of polyphenols (extraction yield). With the outcome of optimization of pulsed electric field settings maximum value of 32% of extraction yield was achieved. This is better as compared to a conventional process where extraction is done by hot (boiling) water or solvents while not destroying valuable components in raw material itself.

1. Introduction

Technological and scientific progress in the food industry has been pushed towards the extraction of intracellular compounds and liquids from cellular plant tissue. The shift from conventional processing of food plant material towards design of smart operation techniques is made to explore a novel product range. Therefore, in the last ten years the link between process synthesis and the development of novel consumer products became increasingly important, especially regarding the design of processes for structured products. These structured products have high added value and they are often complex multi-phase materials (*e.g.* cosmetics creams and lotions, margarine, ice cream, etc.) see e.g. Edwards (Edwards, 2006). Process synthesis is a well developed discipline in conventional process engineering (Biegler et al., 1997) Process synthesis of food processes is much more challenging due to the fact that food systems have complex microstructure and stability is not only a thermodynamic function but also microbiological factors play a role.

The Product-Driven Process Synthesis (PDPS) methodology proposed by Bongers and Almeida (Bongers and Almeida-Rivera, 2012) connects product design with process synthesis. The PDPS method comprises a multi-level decision hierarchy with increasing level of complexity that helps the user in the development of new products and processes.

In Table 1 the hierarchy is shown that starts at the framing level and ultimately leads to a complete conceptual process design (including equipment design, process control and multi-product equipment integration). Bongers and Almeida (Bongers and Almeida-Rivera, 2009) explained the complete hierarchy in more detail. Recently, the PDPS method was used for the isolation of valuable components from different feedstock, for example the isolation of isoflavones from okara (Jankowiak et al., 2015), and in the design of a sugar beet leaves biorefinery (Kiskini et al., 2016). This contribution will focus on the opportunities for product design, process synthesis and the strategies towards novel molecular and functional products. In particular, we will use the isolation of polyphenols from fresh tea leaves to illustrate the applicability and scope of the PDPS methodology.

In the following sections the first six levels of PDPS will be presented; starting from the framing level until the mechanism-

* Corresponding author.
E-mail address: edwin.zondervan@uni-bremen.de (E. Zondervan).

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Table 1
General description of each level of PDPS approach (Bongers and Almeida-Rivera, 2009).

Level	General description of each level
(0) Framing level	Description of the project background (BOSCARD) and business context as well as overall supply chain
(1) Consumer wants	Consumer wants are obtained in qualitative descriptions consumer likings, focus groups, interviews and translate them into quantifiable product attributes
(2) Product ideas	Potential products that meet the consumer wants are identified and mapped the quantifiable product attributes onto the product properties, which are measurable
(3) Input output level	Complete specification of the output is done. Inputs (ingredients or raw materials) and the descriptors of the output (*e.g.* microstructure) are specified.
(4) Task network	Definition of the fundamental tasks needed to convert input into output, taken from a cluster of tasks and its subgroup is performed. Then, a network is made from the selected tasks and clusters.
(5) Mechanism and operating window	Mechanisms[a] that can perform tasks are defined. And for every selected task, operating window needs to be defined.
(6) Multiproduct integration	The outcomes of steps 3–5 for the different products are compared to look for overlap and possibilities to combine the production.
(7) Equipment selection and design	The operating units are selected taking into consideration integration possibilities and controllability.
(8) Multi product-equipment integration	Multi-stage scheduling of the multiple products is applied, including plant-wide control.

[a] "Mechanisms" is the nomenclature used in the paper of Bongers and Almeida-Rivera (2012).

and operating window. The multiproduct integration level, equipment selection and design as well as multi product-equipment integration are not discussed in this paper. Multiproduct integration is important when a production line is used for different products. The last level of the PDPS methodology (level 8) is the translation of the governing mechanisms in the selection of the appropriate unit operations. Since in this particular case, there is only one product – polyphenols, levels 6 and 8 are not addressed.

2. Framing level, consumer wants and product ideas

At the framing level, the background of the project, the business context and the potential of polyphenols (PPs) as food additives are identified. Polyphenols present in fresh tea leaves may reduce the risk of a variety of illnesses, including cancer and coronary heart diseases. The usefulness of these polyphenols may be extended by combining them with other consumer products such as food items and vitamin supplements. The presence of PPs in fresh tea leaves is responsible for health benefits associated with green tea consumption. PPs are now gaining significant attention from both technical and consumer point of view due to potential health benefits. It has been reported that PPs may reduce risk of cancer, cardiovascular diseases, dental decay, obesity, diabetes, and improvement in the immune system (Zaveri, 2006).

From a consumer point of view, idea is to isolate PPs from fresh tea leaves and to bring and mix them back to dried spent leaves. By doing this, two objectives can be achieved: first, isolated PPs are natural products extracted directly from the plant and second, by dosing different amount of PPs to dried spent leaves, different tea products can be made. Because different amount of PPs leads to different chemical compositions and at the end a different taste and appearance of the final cup of tea. For an industry such as Unilever that has been in the tea business for a long time, these two objectives are very important: holding the "natural product" label and different tea products can be introduced to different markets. Moreover, processing fresh tea leaves in this way by isolation of PPs and drying spent leaves could subsequently lead to a sustainable process without producing waste.

Industrial tea extraction is mainly based on the maceration method (rolling) combined with stirring, circulation, ultrasonic, microwave or enzymatic treatment (Perva-Uzunalić et al., 2006); (Lee and Lee, 2008); (Zhu et al., 2012). These methods require large volumes of solvent (mostly ethanol ratio water to ethanol = 1:1 on v/v), high temperatures and significant amounts of energy. Extraction efficiencies are generally low. However, during conventional tea processing that includes cutting, rolling, and drying, the extraction step is missing. Process conditions are harsh, i.e. to destroy the remaining enzymes and to reduce the moisture content in a leaf, high temperatures have to be applied (above 80 °C). There is a need for an alternative process for the extraction of polyphenols under mild conditions. Conventional processes are applying harsh conditions and constituents in raw material are destroyed. An alternative process under harsh conditions would bring additional cost, heat, water and/or chemical reduction (e.g. use of organic, non-food-grade solvents, energy intensive recovery of the solvents, etc.). Such a novel alternative process has to be an effective and green method that will allow extraction of polyphenols from fresh tea leaves.

3. Input/output level

At the input output level a complete specification of all exchange streams to the process inputs (raw material(s)) and target output (product(s)) are identified. Fresh tea leaves (input) contain approximately 75 wt % moisture and 25 wt % of total solids. PPs account for approximately 30% of dry weight of fresh tea leaves (Zhu et al., 2012). Fresh tea leaves contain caffeine, polyphenols, polysaccharides, and necessary nutrients, such as proteins, amino acids, lipids, and vitamins. Generally, some chemical components – free amino acids, total tea polyphenols, and soluble sugars – are considered important indicators of tea quality (Ruan et al., 2009).

The output of the process is a fraction of polyphenols. Green tea leaves contain low-molecular-weight polyphenols consisting mainly of flavanol (flavan-3-ol) monomers, which are referred to as catechins. There are several isomers of this compound: catechin, catechin gallate (CG), gallocatechin, gallocatechin gallate (GCG), epicatechin, epicatechin gallate (ECG), epigallocatechin, and epigallocatechin gallate (EGCG). Normally, 10–20% of the catechins in green tea leaves are epigallocatechin and EGCG. Black tea polyphenols are formed during the enzymatic oxidation of green tea leaves. In green tea (non-oxidized leaves) mostly catechins are present, while in black tea several types of polyphenols formed by enzymatic polymerization of catechins, including theaflavins can be found. The performance of the input-output level is assessed by simple economic analysis, which basically estimates the economic potential of the process. The difference between product revenue and raw material costs is computed on a year basis for 1 ton/hr. To estimate the economic potential (EP) (Moggridge and Cussler, 2011) of the proposed process the following equation was applied:

EP = Product revenues-Raw material cost

$$EP = \sum_{i-product} C_i \cdot F_i - \sum_{j-raw\ mat.} C_j \cdot F_j \quad (1)$$