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Product Driven Process Synthesis for the Recovery of Vitality Ingredients from Plant Materials

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In this paper, the last levels of the product-driven process synthesis methodology are used to formulate a conceptual process design for the production of a purified catechins powder. First a two stage adsorption process for the separation of the catechins was designed. Secondly a spray drying process is modeled to determine the equipment and the operational conditions, as well as the economic potential. The conceptual study shows that it is possible to obtain a powder of catechins with 83.5 % purity and a particle sauter mean diameter of 7.48 μm.

1. Introduction

Tea is the second most consumed beverage in the world and it is a very rich source of polyphenols. In the case of green tea the polyphenols are mainly catechins, which represent 30 % on a dry weight basis. Tea catechins are known for their strong scavenging and antioxidant activity and are considered to have antimutagenic and anticarcinogenic properties. Therefore, tea consumption and the addition of catechins to foods are considered a way of providing health benefits. Furthermore, the use of catechins in foods can also increase the shelf life, improve the quality and prevent lipid peroxidation (see for example Zderic et al. 2013).
Caffeine is also present in green tea, although in a much smaller amount. It plays however a very important role in the precipitation of green tea catechins.
Usually, in the separation of tea catechins, organic solvents are used that can affect human health in case any residues remain in the final product. By avoiding the use of such solvents, a higher acceptance by the consumers can be achieved.
This work follows the Product Driven Process Synthesis (PDPS) methodology (Bongers & Almeida-Rivera, 2006). The PDPS combines product and process synthesis principles with an engineering overview, in a structured approach. It includes a hierarchy of 9 decision levels of increasing detail. In this particular case PDPS is applied to the separation and recovery of the targeted tea polyphenols, instead of the more common application to structured food products.
In this paper we will focus on the conceptual process design level of the PDPS. From the definition of inputs (tea leaves) and outputs (polyphenols) several task networks can be defined. The Task networks describe the tasks required to convert the inputs into the desired output at a mechanistic (driving force) level. The task network description is then use for the definition of the operational windows and the selection of appropriate equipment to execute the tasks. Ultimately all acquired data and models are used in an overall conceptual process design for the separation of catechins from green tea.

2. Adsorption

Separation of catechins from tea can be achieved by connecting two adsorption columns via a buffer tank and operating them in a batch mode, where the green tea feed is set at a flow rate of 0.84 m^3/h. Tea extract is fed to the first column via the bottom and the output stream at the top of the column is afterwards collected in the buffer tank. The output of the buffer tank is the input of the second column. The output of the second column is the final stream of the adsorption process.

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Based on the resin screening results from (Sevillano et al. 2014), the first column will be packed with HP20 resin (favorable for caffeine adsorption) and the second will be packed with the resin XAD7HP (preferred for the adsorption of catechins).

In total four operating modes are considered, based on the possibility of using the output from the adsorption or from the desorption in each column. The feed for the buffer tank, can be taken from the output of either the adsorption or desorption of the HP20 column. The output of the whole process can either be the adsorption or the desorption of the second column (XAD7 column).

The objective is to maximize the yield of catechins (Ycat) and minimize the yield of caffeine. We found that the operational mode which achieves the best result is the one that uses the adsorption output of column 1 (HP20 resin), as the input for the buffer, followed by the desorption (elution) output of column 2 (XAD7HP resin). In this mode, column 1 is used to preferably adsorb the caffeine, while column 2 mostly adsorbs the catechins. The cycle time for this operational mode is 40-45 min.

In the green tea feed (input for the first column) the relative purity of catechins was 78 %. After one cycle the catechins relative purity increased to 91 %. In figures 1 and 2 the column design and the operational modes are schematically shown.

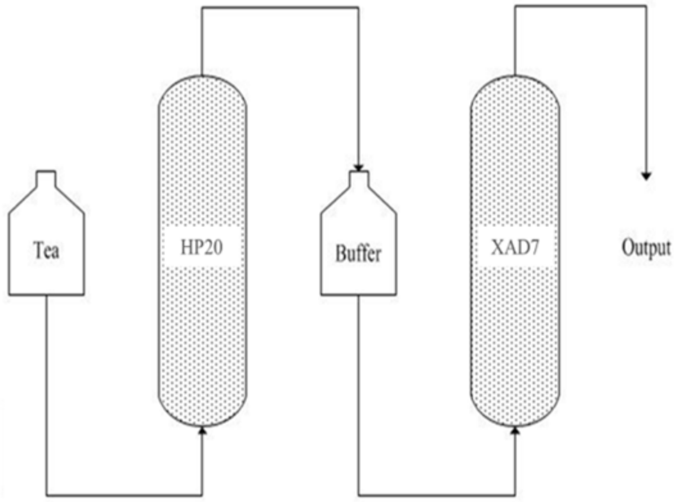

Figure 1: Two columns process design diagram

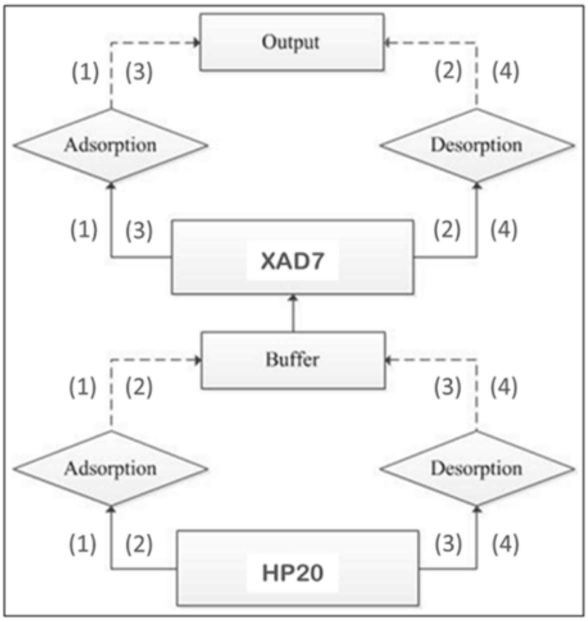

Figure 2: Four possible operational modes (right).

3. Spray Drying

Following the green tea column sorption process, a spray dry process is proposed to achieve a high purity catechins dry powder.

For the drying of tea polyphenols after the adsorption step a closed cycle spray dry process model is used. The design of the drying process is done in Aspen Plus. The spray drying unit is used to evaporate the solvent and dry the solids. Spray drying is commonly used in the food industry because it can handle temperature sensitive materials and it produces solid rounded particles with a uniform size. Another advantage is the potential combination of several operations (evaporation, crystallization, filtration, size reduction, classification and drying) in a single operation.

The input stream for the adsorption process is an aqueous green tea stream with a flow rate of 1 t/h. The input stream for the spray dyer is based on the modeled output stream from the two column adsorption process. Since the solvent is a solution of ethanol-water, nitrogen (inert gas) is used as heat carrier medium, to prevent ignition. This also has the advantage of avoiding the oxidation of the polyphenols. In addition a closed cycle drying process is used, to recirculate the nitrogen.

The heated gas is introduced through a roof disperser around the atomizer, creating a co-current flow of product and gas. The contact between the gas and the droplets as soon as they are formed causes rapid surface evaporation, and keeps the solids at relatively low temperatures. The particle formation in the dryer starts after the critical moisture content of the droplet is reached. When evaporation becomes limited by the liquid diffusion from the center of the droplet to the surface, the particles are in a less hot zone of the dryer. This is the reason why heat-sensitive products can be spray-dried even at elevated temperatures. The output from the drying model should be a solid product with less than 0.2 % moisture content, at a maximum temperature of 70 °C. The drying temperature is set at 180 ºC based on the optimized spray drying conditions reported by(Tang et al. 2011) and on the desired output settings. The nitrogen flow rate is set at 18500 kg/h.

The most important part of the spray dyer is the atomizer. In the atomizer the droplets that are formed closed to the atomizing device have a direction and velocity well defined by the atomizer. The evaporation begins when the hot gas is mixed with the droplets. The gas flow sets the droplets movement inside the dryer chamber, to allow the drying process and avoid contact with the dryer walls. In a closed cycle preferably rotary or liquid nozzle atomizers are used, with the advantage that no gas is needed for the spraying (Mujumdar 2014). The selected liquid atomizer for the dryer is a hollow cone nozzle. The spray dryer is operated in co-current mode from the top and the diameter of the dryer is proportional to the desired particle size desired in the final powder. The main characteristics of the modeled spray dryer are shown in Table 1.

Table 1. Spray dryer description.

Spray dryer	Values
Tower height (m)	6
Tower diameter (m)	2
Spray angle (deg)	45
Number of atomizers	9
Nozzle orifice diameter (mm)	1
Number of droplets intervals	4

The closed spray drying process flowsheet is depicted in Figure 3. In the dryer, the solvent evaporates into the stream of nitrogen, resulting in a dry solid material. The evaporation rate in the dryer is directly proportional to the product of the temperature variation along the dryer and the mass flow of the gas. Part of the particles are formed and collected in the dryer. The remaining particles entrained in the spent drying gas are recovered in a high efficiency cyclone or in the fabric filter with a baghouse design. All the particles formed are recovered into the solids stream. Since the formed powder is very fine, most of it is collected in the cyclone (Sauter mean diameter of 22.5 micron) and in the bag collector (Sauter mean diameter of 5.24 micron), which is the main collection point.

The humid gas afterwards passes through the filter and goes to a condenser-scrubber, where the solvent vapors are condensed. The gas without the solvent is then recirculated to the dryer. The recovered solvent is recirculated to the column adsorption process, to be used in the desorption step.

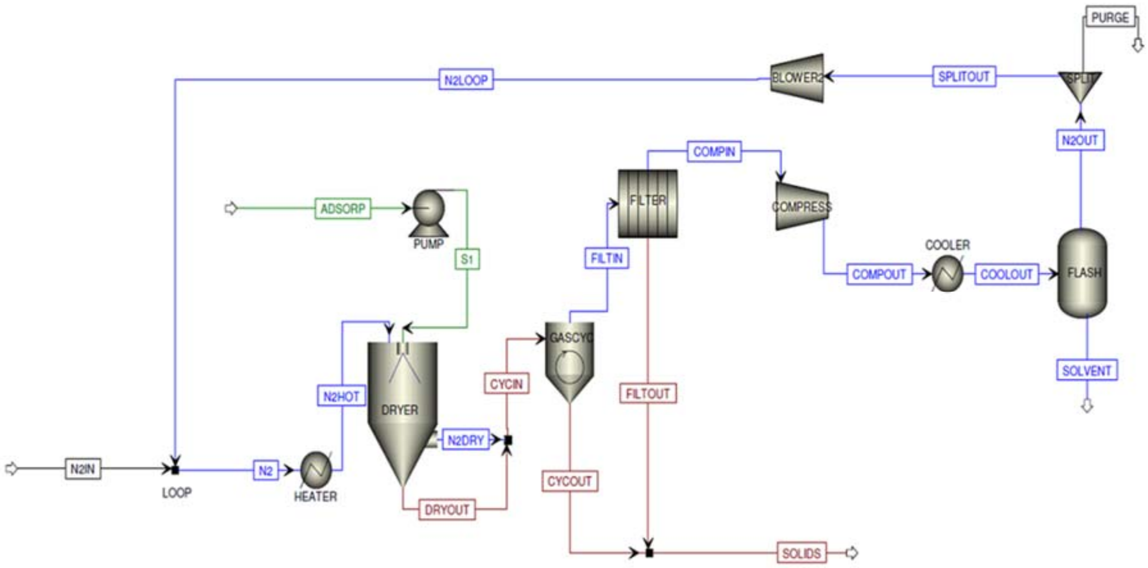

Figure 3. Spray drying process flowsheet. The gas streams are shown in blue, the solids streams in red and the input stream from the adsorption process in green.

For the droplet size distribution in the atomizers, the formation of four droplet classes is assumed (one for each droplet interval). The modeled results from the droplets and gas temperature variation and the droplet moisture content for each class along the height of the dryer are shown in Figure 4. It is observed that the classes 1 and 2 are heated to the gas temperature already in the beginning of the drying chamber. Between 1 and 2 meters the class 3 particles are dried. The last class of particles only reaches the gas temperature near the of the chamber.

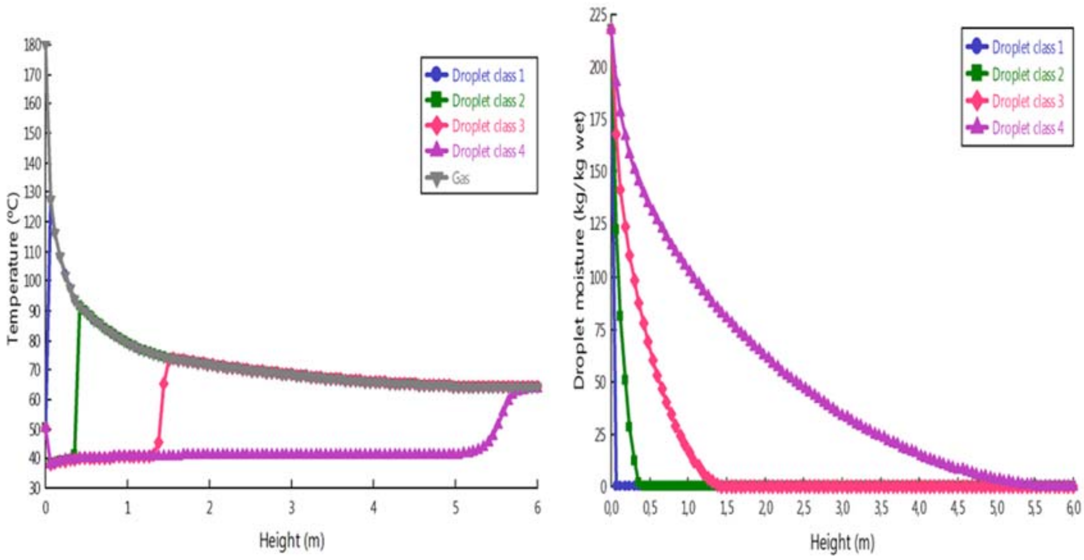

Figure 4. Temperature (left) and droplet moister content (right) curves along the height of the dryer.

A very high drying rate for classes 1 and 2, may be observed while class 4 has a slower drying rate and only reaches the a low moisture content at the bottom of the drying chamber. At the end of the drying process the solid particles have a final moisture content of 1.21 g/kg. The dryer model results are showed in table 2.
The results for the solids stream show that 4.47 kg/h of catechins are produced with 83.5 % purity, at a temperature of 64 ºC. The Sauter mean diameter of the dry particles is 7.48 micron. The advantage of obtaining such a fine powder is that it can be easily dissolved when added to other products. This specification tends to increase the commercial value of the catechins powder.

Table 2: Spray drying results

Spray dryer	Values
Inlet temperature (ºC)	180
Exhaust temperature (ºC)	64.06
Inlet solids moisture content- dry basis (kg/kg)	217.4
Outlet solids moisture content- dry basis (g/kg)	1.21
Overall evaporation rate (kg/h)	954.5
Outlet gas moisture content- dry basis (kg/kg)	0.050

5. Economic evaluation

Although cost data is scarce, it is expected that the costs of the spray drying process (including the solvent recovery) will be much higher than the adsorption costs. Both the spray dryer and the condenser for the solvent recovery are high energy consumption operations. Nevertheless, the make-up ethanol solution costs for the adsorption process are included in the overall costs of the economic analysis.
The summary with all the considered process operating costs is included in Table 3. The price for the green tea is assumed to be 50 €/t and the price for the purified catechins is assumed to be 95 €/kg. The economic calculations performed by Aspen Plus are based on a processing time of 4,800 h/y (DACE Price Booklet 2011).

Table 3: Summary of the operating cost

Utility	Values
Total heating cost flow:	0.75 M€/yr
Total cooling cost flow:	0.048 M€/yr
Net cost (Total heating cost + Total cooling cost):	0.80 M€/yr
Stream cost	Values
Net cost flow of feeds:	0.49 M€/y
Net cost flow of products:	3.00 M€/y
Overall net cost flow:	2.50 M€/y

The net cost flow of feeds is the cost of the raw materials in M€/year and the net cost flow of products is the value of the product stream, also in M€/year.

The process operating profit is calculated as:

[Operating profit] = [Products sales] - [Raw material cost] - [Utility cost] (1)

For this process the operating profit is 1.71 M€/y. For this simplified evaluation mass and energy balances are considered, but no real equipment design constraints.
To estimate the full capital and operating costs, the Aspen Process Economic Analyzer® tool is used to perform the economic evaluation within the Aspen Plus model simulation. The capital expenditures (CAPEX) and the operational expenditures (OPEX) are calculated, together with the total annual costs (TAC). The total OPEX are estimated to be 2.10 M€/year, for which the main contribution is from the cooler and the compressor. The CAPEX value is estimated to be 7.11 M€, with the major contribution coming from the compressor. The calculation of the TAC is done by summing up the OPEX with 20 % of the CAPEX (Seider et al. 2004). The TAC is estimated to be 3.52 M€/y.

6. Conclusions

In this paper a conceptual design for the separation and purification of catechins from green tea is presented. In the first part of the process a packed bed column adsorption is used, followed by a spray drying process. The two stage adsorption processes can be used to achieve (after one cycle) a 52 % yield of catechins and the yield of caffeine was 19 %, for an operation time per column of 95 minutes. The relative purity of catechins to caffeine increased from 78 % to 91 %, when compared to the feed green tea solution. The proposed design proves to be effective for separating the caffeine and still allows the recovery of more than half of the catechins. Following the green tea column adsorption process, a spray dryer is proposed to produce a high purity catechins dry powder. The spray drying process is able to produce a fine catechins powder with 83.5 % purity. The fact that the powder has a low particle Sauter mean diameter means that it can easily be dissolved in future applications. Based on the estimated economic evaluation it can be concluded that the process has the potential to be profitable.

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