Generation of Optimal Flowsheet for Downstream Processing in Biochemical Production of Butanol: Inclusion of Adsorption

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Adsorption is ubiquitous in the laboratory-scale as well as industrial-scale separation or purification of liquid and gaseous mixtures for the manufacture of a wide variety of chemicals, biochemicals and materials, e.g., fuel-grade ethanol by a biochemical route (*1*). Naturally, the potential of adopting adsorption for the same purpose in the biochemical production of butanol (B), ethanol (E), and acetone (A) has been well recognized (*2-7*). Apparently, however, no attempt has yet been made to incorporate adsorption into an industrial plant for the downstream processing in the biochemical production of B, E, and A. The current work explores the possibility of incorporating both adsorption and the conventional separation methods, including various types of distillation and extraction, into such a plant through synthesizing the potentially optimal and near-optimal flowsheets for it. A comparative analysis is carried out among the resultant flowsheets as well as with those consisting only of the conventional separation methods (*8*) including the ones generated in our prior work to which the current work can be regarded as a sequel. The flowsheets' generation is implemented with a graph-theoretic method for process-network synthesis based on process graphs (P-graphs), whose efficacy has been repeatedly demonstrated (*8*).

The system for the downstream processing is described in our previous paper (*8*). Two adsorbing units and one concomitant centrifuging unit are included in the current work. The fermentation broth contains 2 wt% of insoluble solids whose densities are far higher than the aqueous portion of the broth comprising B, E, A, and water (W). Hence, the insoluble solids can be readily removed from the fermentation broth by centrifugation prior to adsorption. The clear supernatant from the centrifugal unit is passed through the first of the two adsorbing units that comprises two adsorption columns. Bonopore (a divinylbenzene-styrene copolymer; Nobelkemi AB, Sweden) is adopted to adsorb butanol in the current work. The biochemical production of fuel ethanol is very similar to that of B, E, and A. Thus, the adsorption of W from the vapor phase, as practiced in the former, is also considered for inclusion in the current process (*1*); this constitutes the second of the two adsorbing units. To implement it, the fermentation broth is first concentrated in an existing gas-stripping unit, identified as Gas Stripper 1 (G1) in our previous paper (*8*), to yield the vapor phase comprising B, E, A, and W and the aqueous phase containing solid suspensions. Subsequently, W in the vapor phase is removed in two adsorption columns that are operated cyclically between the adsorption and desorption phases similar to those of the other adsorption unit.

The methodology adopted in the current work is detailed in the prior paper and literatures (*8-18*). The centrifuging unit is designated as Centrifuge C1. This supernatant yielded contains 1.5 wt% B, 0.2 wt% E, 0.6 wt% acetone (A), and 97.7 wt% water (W). The concentrated suspension containing the solids is recycled to the fermenter. The first of the

two adsorbing units, designated as Unit 24, comprises Column B1 and Column B2 both packed with Bonopore adsorbents. The clear supernatant from Unit 23, comprising Centrifuge C1, is fed to one of the columns where B, E, and A are adsorbed onto the adsorbents. The adsorption capacities of the adsorbents are approximately 50-80 mg g^{-1} adsorbent for the products. The equilibrium adsorption isotherms indicate that butanol can be concentrated from 0.5% (w/v) to 98% (w/v) (25). When the products saturate the adsorbents, they are thermally desorbed and recovered subsequently; meanwhile, the adsorbents are reactivated in the other column. The second adsorbing unit, designated as Unit 25, comprises Adsorption Column B3 and Column B4, both packed with multiple beds (trays) of thinly-layered molecular sieves. The vapor stream from Unit 3, comprising Gas Stripper G1, is fed to this unit where water is essentially completely adsorbed onto the adsorbents.

Based on the specifications of materials (*8*) the comprehensive flowsheet corresponding to the maximal structure is constructed (Figure 1). The total computing time consumed is less than 2 seconds on a PC (266MHz and 65 MB Pentium II; Windows 95). The optimal and near-optimal flowsheets are identified by resorting to algorithm ABB (*8)*. The objective function is minimized based on the costs of operating units in the flowsheet. The present values of C1, B1& B2, and B3 and B4's costs are estimated based on heuristics (*8*). Their values are 4248, 9240, and 1401×10^3US$/year. The total computing time consumed for generating optimal and near-optimal flowsheets is less than 4 seconds on the same PC used in executing algorithm MSG (266 MHz and 65 MB Pentium II; Windows 95). The optimal flowsheet is exhibited in Figure 2.

Our results show that the addition of the three operating units, increases the computing time only very slightly for generating the comprehensive flowsheet and the optimal and near-optimal flowsheets. This attests to the efficacy of the methodology adopted. The top 10 flowsheets are compared graphically in Figure 3. The optimal flowsheet is noticeably less expensive than the four flowsheets ranked second through fifth, which, in turn, are substantially less expensive than the five ranked sixth through tenth. It is also revealed that the differences in the costs among the top 10 flowsheets can be attributed to the differences in the configurations of distillation columns downstream beyond G1, B3 and B4.

None of the top 10 flowsheets contains the extracting, centrifuging, or azeotropic-distillation units. The total cost of the optimal flowsheet, $5{,}286\times10^3$US$, is 756×10^3US$ (12.5%) and $2{,}653\times10^3$US$ (33%) less than those of the second and tenth best flowsheets, respectively. There are, however, only slight differences between the cost of the second best flowsheet and those of the third best (20×10^3US$ and 0.3%), fourth best (39×10^3US$ and 0.6%) and fifth best (215×10^3US$ and 3%) flowsheets. The difference between the costs of fifth best and sixth best flowsheets is appreciable; it is $1{,}183\times10^3$US$ or 16%. The cost differences among the sixth best through tenth best flowsheets are indeed small.

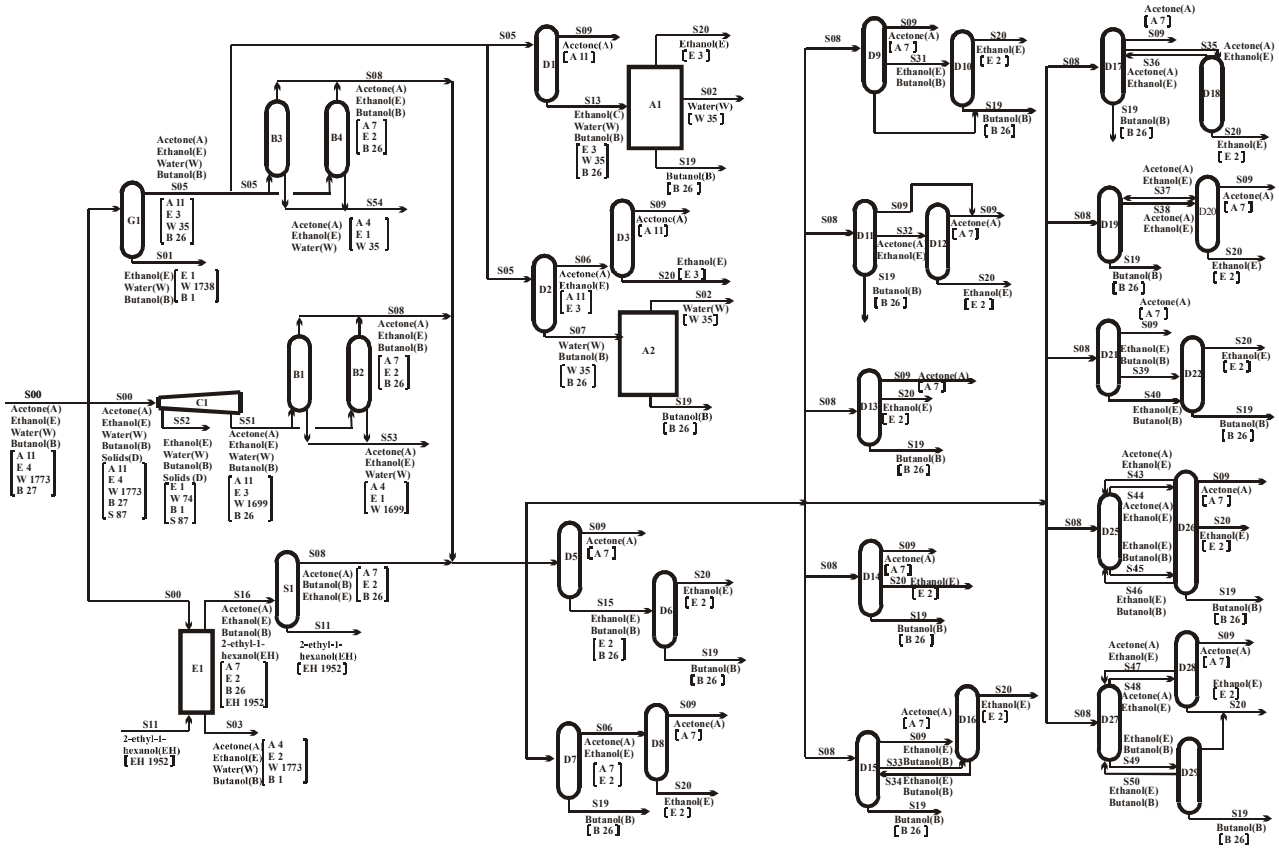

Figure 1. Comprehensive flowsheet corresponding to the maximal structure for the production of butanol, ethanol, and acetone with the inclusion of adsorption: conventional representation.

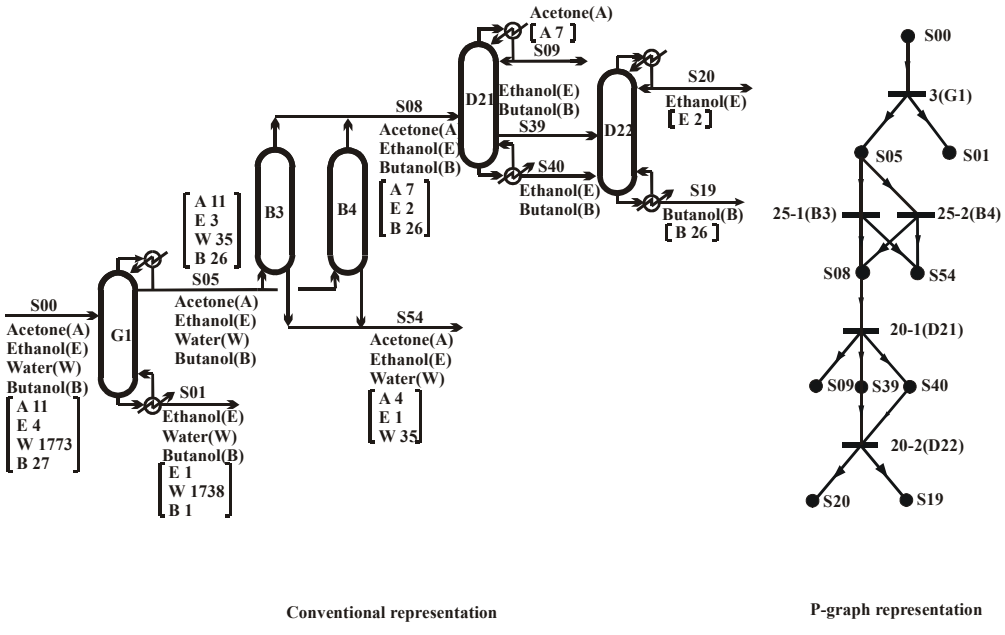

Figure 2. Optimal flowsheet with the consideration of adsorption.

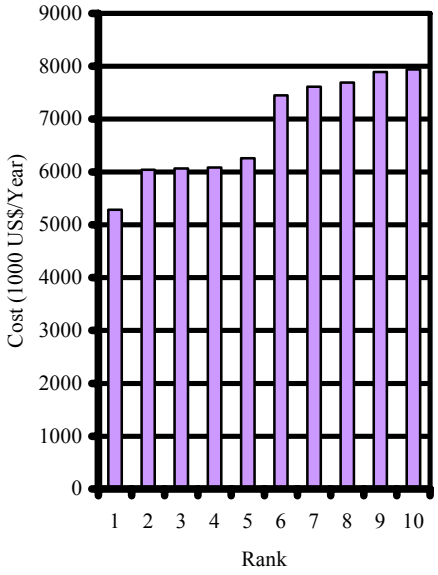

Figure 3. Comparison of the total costs of the top 10 flowsheets with the inclusion of adsorption considered.

The optimal flowsheet consists of Gas Stripper G1, Adsorption Columns B3 and B4, and Distillation Columns D21 and D22. The configuration of these two distillation columns for separating butanol (B), ethanol (E), and acetone (A) from each other is referred to in our previous paper (*8*) as the complex-direct (*26*). The configurations of the two distillation columns in the second and third best flowsheets are referred to in our previous paper (*8*) as complex-Petlyuk type IIIb and simple-indirect, respectively (*26*). Naturally, the cost of the complex-Petlyuk type IIIb is lower than that of the simple-indirect; moreover, both costs are higher than that of the complex-direct configuration of distillation columns in the optimal flowsheet. The cost differences among the top 10 flowsheets can mainly be attributed to the different configurations of the distillation columns separating B, E, and A from each other.

The top 10 flowsheets listed in Table 3 are markedly different from those generated in our previous paper with only conventional operating units (*8*), i.e., without adsorbing units. The marked cost reduction of the top 10 flowsheets is attributable to the replacement of Extractor E1 and Solvent Stripper S1 with Gas Stripper G1, and Adsorption Column B3 and Adsorption Column B4. The total cost of the optimal flowsheet, $5{,}286\times10^3$US$, is $4{,}130\times10^3$US$ (44%) less than that of the optimal flowsheet generated in our previous paper (8). In fact, even the total cost of tenth best flowsheet, $7{,}979\times10^3$US$, is $1{,}477\times10^3$US$ (16%) lower than that of the optimal flowsheet generated in our previous paper (*8*).

The newly generated top 10 flowsheets indicate that the incorporation of the adsorbing unit (Unit 25), comprising Adsorption Column B3 and Adsorption Column B4, reduces the cost immeasurably. None of them, however, contains the adsorbing unit (Unit 24), comprising Adsorption Column B1 and Adsorption Column B2.

Water and suspended solids constitute the major fraction of the fermentation broth. Gas Stripper G1 preceding Adsorption Column B3 and Adsorption Column B4 removes the

massive amount of W and almost all solids as the bottom liquid stream from the fermentation broth. The feed to Adsorption Column B3 and Adsorption Column B4 is the vapor stream from Gas Stripper G1, which is only a small fraction of the original fermentation broth, thus substantially reducing the equipment size.

In contrast to Adsorption Column B3 and Adsorption Column B4, the inclusion of Adsorption Column B1 and Adsorption Column B2 are not advantageous from the cost standpoint. Adsorption Column B1 and Adsorption Column B2 receive feed at a rate of 789×10^3 kg/hr, which is about 22 times larger than Adsorption Column B3 and Adsorption Column B4 (34×10^3 kg/hr). Moreover, the inclusion of Adsorption Column B1 and Adsorption Column B2 necessitates the additional Centrifuge C1 to remove insoluble solids from the high-volume feed, thereby incurring additional cost. Naturally, the incorporation of Adsorption Column B1 and Adsorption Column B2 dramatically magnifies the cost of the flowsheets; as a result, these two adsorption columns are totally excluded from the list of the optimal and near-optimal flowheets.

Our previous work has explored in depth the structure of the downstream processing system composed of mature conventional separating units for the biochemical production of butanol, ethanol, and acetone (*8*). This has been accomplished by expeditiously and systematically generating the optimal flowsheet as well as a set of near-optimal flowsheets in the ranked order with the aid of the highly efficient graph-theoretical algorithmic method based on process graphs (P-graphs). As a sequel, the current work extends the previous work by adding a class of non-conventional separating units based on adsorption in the mix of separating units. The results reveal that the judicious inclusion of adsorption replaces some conventional operating units, thereby substantially reducing the cost of flowsheet. The present approach represents a novel and highly robust paradigm for planning the optimal retrofitting of a downstream processing system. It is highly likely that such a paradigm can be adapted without much difficulty to take into account other criteria, such as sustainability, in the objective function to satisfy the ever-increasing environmental, societal and regulatory requirements, which naturally gives rise to a multi-objective optimization problem.

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