

SURPLUS DIAGRAM AND CASCADE ANALYSIS TECHNIQUE FOR TARGETING PROPERTY-BASED MATERIAL REUSE NETWORK

Dominic Chwan Yee **FOO**
Chemical Engineering Pilot Plant (CEPP)
Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia

Vasiliki **KAZANTZI**, Mahmoud M. **EL-HALWAGI**
Chemical Engineering Department
Texas A&M University, College Station, TX 77843, USA

Zainuddin Abdul **MANAN**
Chemical Engineering Department
Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia

ABSTRACT

Recycle of process and waste streams are among the most effective resource conservation and waste reduction strategies. In many cases, recycle/reuse is dictated by sink constraints on properties of the recycled streams. In this work, we introduce an algebraic technique to establish rigorous targets on the minimum usage of fresh resources, maximum direct reuse, and minimum waste discharge for property-based material reuse network. Two new tools have been developed. A new graphical tool called the property surplus diagram is firstly introduced to provide a basic framework for the determining rigorous targets for minimum fresh usage, maximum recycle, and minimum waste discharge. The tools also determines the property-based material recycle pinch location. The Property Cascade Analysis (PCA) technique is next established to set targets via a tabular approach. PCA eliminates the iterative steps typically associated with a graphical approach. Along with the minimum fresh and waste targets, the material allocation target is another key feature of the PCA. A network design technique is also presented in this paper to synthesise a maximum resource recovery (MRR) network that achieves the various established targets. The procedures developed in this paper constitute a generalisation to the composition-based graphical and algebraic techniques developed for water and hydrogen recovery networks. Two case studies are solved to illustrate the applicability of the developed procedures.

Keywords: Property integration, process integration, pinch analysis, resource recovery, material surplus, cascade analysis.

INTRODUCTION

The efficient use of resources is recognised as a key element of sustainable development and an effective strategy for cost reduction and environmental acceptability of process industry. Recently, significant progress has been made in the optimisation of material recycle and reuse. In particular, mass integration has emerged as an effective and holistic framework for optimising the allocation, generation, and separation of streams and

species throughout a process. Recent reviews of mass integration can be found in literature (e.g. El-Halwagi, 1997; El-Halwagi, 1998; El-Halwagi and Spriggs, 1998; Dunn and El-Halwagi, 2003). In the area of recycle/reuse, extensive work has been done to target the minimum fresh material usage (e.g., water, hydrogen, volatile organic compounds, etc.) and minimum waste discharge. Examples of these research efforts can be found in literature (e.g., Wang and Smith, 1994; Dhole *et al.*, 1996; Sorin and Bédard, 1999; Polley and Polley, 2000; Bagajewicz and Savelski, 2001; Savelski and Bagajewicz, 2000a, 2000b, 2001; Zhou *et al.*, 2001; Alves and Towler, 2002; Hallale, 2002; El-Halwagi *et al.*, 2003; Manan and Foo, 2003; Hamad and Fayed, 2004; Manan *et al.*, 2004; Foo *et al.*, 2005; Aly *et al.*, 2005; Almutlaq and El-Halwagi, 2005; Almutlaq *et al.*, 2005).

Despite the importance of mass integration techniques for material recycle/reuse, these are limited to address problems that are governed by the composition of process streams. However, composition is only one of the many chemical and physical properties that are essential in a chemical process. Other properties (or functionalities) that are commonly encountered include pH, density, viscosity, reflectivity, turbidity, colour and solubility, to name a few. Process network synthesis associated with these chemical properties clearly does not fall into the conventional mass integration problems, and hence another generic approach is needed to address this issue.

Unlike individual components, however, properties or functionalities of process streams are not conserved. Hence, a technique is needed to track these properties in a stream. A new trend of process integration techniques emerged when Shelley and El-Halwagi (2000) introduced the concept of surrogate properties called “clusters” that enable the conservative tracking of properties and functionalities of streams. This technique was originally developed for the recovery and allocation of volatile organic compounds (VOCs) to avoid the tracking of individual components in complex hydrocarbon mixtures (Shelley and El-Halwagi, 2000).

It is worth emphasising here that there are several differences between property-based problems and component-based problems. First, while components are conserved through material balances, properties are not conserved. Also, whereas mixing of components is linear, mixing of properties is not necessarily linear. Thus, a nonlinear property-mixing rule may need to be used. It is also important to note that component-free streams (pure fresh) constitute the basis for adjusting calculations in component-based cascade analysis. There is not an equivalent notion of “property-free” streams in property-based problems. Moreover, in the cases of impurities (component-based problems), lower compositions are always more preferred. In the case of properties, higher or lower properties may be preferred.

El-Halwagi *et al.* (2004) later generalised this newly developed network synthesis technique to address the so-called “*property integration*” problems which may be defined as “a functionality-based holistic approach for the allocation and manipulation of streams and processing units, which is based on functionality tracking, adjustment, and assignment throughout the process.” For a network with up to three properties in

concern, graphical tools may be used to guide the synthesis and analysis tasks (Shelley and El-Halwagi, 2000; El-Halwagi *et al.*, 2004). The concepts of clustering and property integration have been used for process modification (Kazantzi *et al.*, 2004a, b). When the number of properties is larger than three, algebraic tools have been developed by Qin *et al.*, 2004. Moreover, the clustering concept has been extended to various simultaneous process and product design problems (Eden *et al.*, 2002, 2004, 2005; Grooms *et al.*, 2005).

For systems characterized by one key property, Kazantzi and El-Halwagi (2005) introduced a pinch-based graphical targeting technique that determines rigorous targets for minimum fresh usage, maximum recycle, and minimum waste discharge. This is a generalization of the conventional material reuse/recycle pinch diagram (El-Halwagi *et al.*, 2003) which can be modified to include property operators to track properties as streams are segregated, mixed, and recycled. The graphical technique possesses the advantage of providing good visualization insight for targeting and network synthesis. Notwithstanding this advantage, there is a need for a computational tool such as an algebraic technique. There are several cases when an algebraic approach is desirable. These cases include:

- Scaling problems: when the property operators and loads of process sources or sinks are of different magnitudes, the accuracy of the graphical approach becomes questionable since the graph will be skewed by the larger loads and operators.
- Problem dimensionality: when there are numerous sources and sinks, there is a need to handle the data algebraically in favour of graphically.
- Computational effectiveness: the algebraic procedures can be easily automated and coded to enhance computational effectiveness. This serves several purposes. For instance, in sensitivity analysis, the algebraic technique can be readily used to assess the solution sensitivity to variations in input data by running what-if scenarios. Graphical procedures are cumbersome in sensitivity analysis, since they may entail the re-plotting of the composite curves for each variation.
- Interaction with process simulators: the algebraic procedure is naturally implemented on a spreadsheet. Many computer-aided process simulation tools are interactive with spreadsheets. Hence, the property information can be automatically extracted from the simulation and the targeted results from the spreadsheet are fed back to the simulator.

In this paper we focus on developing an algebraic techniques to solve the problem of identifying rigorous targets for property-based material recycle/reuse. A key element of the developed techniques is the concept of *material surplus* which is a generalization of the concept developed for the problems of hydrogen and water network synthesis (Alves and Towler, 2002; Hallale, 2002). For applications in property-based material reuse network, the property load surplus diagram is first used to provide a basic understanding of material flow throughout the network. Next, we develop an algebraic approach called the *Property Cascade Analysis* (PCA) to identify the various performance targets for maximum resource recovery (MRR) network. This newly developed cascade analysis technique is a generalisation of the cascade analysis for water and hydrogen network synthesis (Manan and Foo, 2003; Manan *et al.*, 2004). PCA offers a complimentary role

to the property surplus diagram in identifying minimum resource targets, pinch locations, as well as resource allocation targets for a MRR network via a computationally efficient algebraic approach. Network design technique for the synthesis of a MRR network, as well as a systematic procedure for identifying optimum process modification strategies are also introduced in this paper. Two case studies on resource conservation are presented here to show the versatility of these newly developed techniques.

PROBLEM STATEMENT

The problem definition of a property-based material reuse network is given as follows:

“Given a process with a number N_{SK} of process sinks (units) and a number N_{SR} of process sources (e.g. process and/or waste streams) that can be considered for possible reuse and replacement of the fresh material. It is desired to determine a network of interconnections among the property sinks and sources, so that the overall fresh resource flowrate F_F and waste discharge flowrate F_D is minimised, while the sinks receive resources of adequate quality.

Each sink, j , requires a feed with a given flowrate, F_j , and an inlet property, p_j^in , that satisfies the following constraints:

$$p_j^{\min} \leq p_j^in \leq p_j^{\max} ; \text{ where } j = 1, 2, \dots, N_{SK} \quad (1)$$

where p_j^{\min} and p_j^{\max} are the specified lower and upper bounds on admissible properties of streams to unit j . Each source i , has a given flowrate, F_i , and a given property, p_i . Also available for service is a fresh (external) resource whose property value is p_F and can be purchased to supplement the use of process sources in sinks. Each process source may be intercepted via design and/or operating changes to modify the flowrate and property of what each sink accepts and discharges”.

The problem can be schematically represented by a source-sink allocation, as shown in Figure 1. According to this representation, each source is allowed to be split and be forwarded to any sink. In particular, the objective here is to determine the optimum flowrate from each source to each sink, so as to minimise the consumption of the fresh resource within a property-based integration context.

BACKGROUND CONCEPT

When different sources (each with a flowrate value of F_i and property value of p_i) are mixed, the resulting mixture will possess a total flowrate of $\sum_i F_i$ and a mean property of \bar{p} . The targeted property could be temperature, composition, density, miscibility, etc., depending on the mixture specifications. Hence, a general mixing rule is needed to define all possible mixing patterns among these individual properties. This can be given

by the following general equation (Shelley and El-Halwagi, 2000; El-Halwagi *et al.*, 2004; Kazantzi and El-Halwagi, 2004):

$$\psi(\bar{p}) = \sum_i x_i \psi(p_i) \quad (2)$$

where $\psi(p_i)$ and $\psi(\bar{p}_i)$ are operators on property p_i and mixture property \bar{p}_i respectively; x_i is the fractional contribution of source i towards the total flowrate of the mixture, i.e.

$$x_i = \frac{F_i}{\sum_i F_i} \quad (3)$$

Eq. (2) is arranged in such a way that the weighted average summation of the operators on individual properties will yield the operator on the mean property of the mixture. Numerous properties can be expressed using this general linear mixing rule in Eq. (2). One example would be the mixing of different sources with individual density ρ_i to form a mixture with mean density $\bar{\rho}$. The mixing rule for density follows Eq. 4:

$$\frac{1}{\bar{\rho}} = \sum_i \frac{x_i}{\rho_i} \quad (4)$$

If we compare Eq. 4 with the general rule of property mixing (Eq. 2), we conclude that the operator for density is given as:

$$\psi(\rho_i) = \frac{1}{\rho_i} \quad (5)$$

Similarly, we can define the operator for some common properties as shown in Table 1. Eq. 2 can be applied to a wide range of properties having different patterns of mixing rule. Operators for other product-related properties (e.g. RON number for oil mixture, miscibility for liquids, etc.) that follow the general mixing rule in Eq. 2 can also be defined in a similar way. For simplicity, $\psi(p_i)$ will be denoted as ψ in the remainder of the text.

Having defined the general rule for deriving an operator for a given property, the sink constraints in Eq. 1 can be rewritten using the new definition of operator, as follows:

$$\psi_j^{\min} \leq \psi_j^{in} \leq \psi_j^{\max} \quad ; \text{ where } j = 1, 2, \dots, N_{SK} \quad (6)$$

Although there is an upper and a lower bound of the operator constraints in Eq. 6, only one of these bounds will be used as the *limiting data* in any network synthesis problem. Specifications of bounds and the concept of the limiting data will be further described when the example is introduced in the later sections of this paper.

Next, another important parameter needs to be defined in property integration. This is the so-called *property load* Δm , which is the product of the flowrate of a source (F_i) or sink (F_j) with its associated property operator (ψ_i and ψ_j respectively). The property loads for a source i , Δm_i and a sink j , Δm_j are given in Eq. 7a and Eq. 7b respectively:

$$\Delta m_i = F_i \psi_i \quad (7a)$$

$$\Delta m_j = F_j \psi_j \quad (7b)$$

This newly defined parameter provides information that resembles the information given by the mass load in the conventional mass integration approach. Note that from Eq 1 and Eq 6, the property of a sink is always bounded within a range of properties or its associated operators. Consequently, due to the constant flowrate required by a sink, the constraints of the sink in Eq. 6 can be rewritten in terms of property load, as in Eq. 8:

$$\Delta m_j^{\min} \leq \Delta m_j^{\text{in}} \leq \Delta m_j^{\max} \quad ; \text{ where } j = 1, 2, \dots, N_{\text{SK}} \quad (8)$$

Eq 8 implies that when feeding a process source to a process sink, its property load should not fall beyond the range of property loads, which are acceptable by the sink. This equation can now replace Eq 1 and Eq 6 for imposing constraints while feeding process source(s) to a sink.

Based on the problem statement and Eq 8, it is now clear that we need to fulfil two important criteria, in order to achieve the MRR objective in a property-based material reuse network. First, each process sink requires a certain amount of flowrate F_j to be fed from the process source(s), including the fresh resource; and second, each of the sinks also requires a certain amount of property load Δm_j , which must obey the constraints described in Eq. 8. Hence, any targeting technique to be developed should meet the two aforementioned criteria. The following metal degreasing example will be used to demonstrate this concept of surplus diagram and cascade analysis for the synthesis of property-based materials reuse network.

EXAMPLE 1 - METAL DEGREASING PROCESS

Figure 2 shows a metal degreasing process taken from Kazantzi and El-Halwagi (2005) that will be used to demonstrate how the conventional pinch-based approach can be applied to the synthesis of a property-based material reuse network. Currently, a fresh organic solvent is used in the degreaser of a reactive thermal degreasing process to decompose the grease and its organic additives. The solvent is then regenerated for reuse in the degreaser. Fresh solvent is also used in an absorber to capture light gases that escape from the solvent regeneration unit before its gaseous overhead is sent to flare. It was proposed to reuse and recycle part of the solvent that is currently discharged from the process to reduce the present high consumption of fresh solvent.

The main property of the solvent that is considered in evaluating its reuse and recycle, is the Reid Vapour Pressure (RVP), which is important in characterising the volatility, makeup and regeneration of the solvent. Although this can be viewed as a material reuse/recycle problem, the conventional mass integration techniques (e.g. El-Halwagi, 1997; Dunn and El-Halwagi, 2003) could not be applied here since the targeted property, i.e. RVP, is not adequately addressed in terms of concentration. One way to resolve this problem is by employing property integration techniques.

The first step in synthesising a property-based material reuse network is to identify the solvent sources and sinks, as well as their associated property data. From Figure 2, it is observed that the process produces two condensate streams: Condensate I (Source 1, denoted as SR1) from the solvent regeneration unit and Condensate II (denoted as SR2) from the degreaser. The two streams are currently sent to hazardous waste disposal. Since these two streams possess many desirable properties, it is advisable their in-plant reuse/recycle to be considered in order to reduce the fresh solvent consumption of the process. Both the degreaser (as Sink 1, denoted as SK1) and the absorber (denoted as SK2) can receive recycled solvent from the process, and hence can be designated as process sinks. Data for the solvent sinks and sources are given in the first four columns of Table 2. Note that the two process sinks (degreaser and absorber) impose constraints on the quality of the solvent to be recycled to these units.

Next, we transform the raw property values of all solvent sinks and sources into its associated operator values. The mixing rule for RVP is given below (Kazantzi and El-Halwagi 2004):

$$\overline{\text{RVP}}^{1.44} = \sum_i x_i \text{RVP}_i^{1.44} \quad (9)$$

From Table 1 (or from Eq 2 to Eq 5), we can express the operator for RVP, $\psi(\text{RVP})$, as follows:

$$\psi(\text{RVP}_i) = \text{RVP}_i^{1.44} \quad (10)$$

Hence, the RVP values of all the solvent sinks and sources are transformed to the corresponding operator values. These are given in the final two columns of Table 2.

In Table 2, it can be observed that the fresh solvent has a relatively low operator value compare to that of the two process sources (SR1 and SR2). In order to achieve the MRR objective, the reuse of process solvent sources is to be maximised before the utilisation of the fresh solvent. Hence, the upper bound of the operator value in Table 2 is assigned to be the limiting value for the process sink. These limiting operators multiplied by the sink flowrates will then define the *limiting property loads* of the sinks, following Eq. 7b. The limiting load indicates the maximum acceptable property load for a given sink. On the other hand, we can also define the property load contained in the solvent sources using Eq. 7a. The limiting data for all solvent sinks and sources of the metal degreasing process is shown in Table 3. After identifying the property sources and sinks, along with their relevant limiting data, the minimum resource targets will be established using the new targeting techniques described in the following sections.

THE CONCEPT OF MATERIAL SURPLUS

In this section, a new graphical tool called the *property surplus diagram* is developed to provide a fundamental understanding of material flow in a property-based material reuse network. This serves as the basis for the algebraic targeting approach that will be discussed in the next section of the paper.

The property surplus diagram is based on the concept of material surplus that was firstly introduced by Alves and Towler (2002) for the analysis of an integrated refinery hydrogen network. Prior to detailed network design, the hydrogen surplus diagram identifies the minimum fresh hydrogen feed and waste stream targets in a hydrogen network. Later, Hallale (2002) extended this targeting tool into water network, where the water surplus diagram is used to target the minimum fresh water consumption and minimum wastewater generation. Both the aforementioned surplus diagrams are built on the concept of the grand composite curves in heat integration (Linnhoff *et al.*, 1984). This material surplus concept is now extended to property-based material reuse network. Obviously, the property surplus diagram provides the same information as the surplus diagrams in hydrogen (Alves and Towler, 2002) and water (Hallale, 2002) networks, as well as the grand composite curves in heat integration (Linnhoff *et al.*, 1984).

In order to generate the property surplus diagram, we first make use of another plot called the *material sink and source composite diagrams*. These composite plots have also been utilised in hydrogen and water network analysis (Alves and Towler, 2002; Hallale, 2002). However, instead of the composition values in the hydrogen and water networks, the property operator values are now plotted versus their corresponding flowrates of each process sink and source. Utilising the limiting data in Table 3, the material sink and source composite plots for Example 1 are shown in Figure 3. Note that, the flowrate of the fresh solvent has first been set to zero. This value will be re-examined in a later targeting stage.

As has been discussed in the previous section of this paper, two objectives need to be fulfilled in synthesising a MRR network. These are the flowrate and the property load requirements of all process sinks. We need to ensure that the total flowrate fed from the process sources satisfies both these criteria for all process sinks in the network. The first criterion, i.e. flowrate feasibility, is easy to inspect from the composite curves. One has to make sure that the total flowrate of the sources is larger than or equal to the total feed flowrate that the sinks require. The composite plots can provide a clear indication if this requirement is satisfied. As long as the source composite extends to the right or touches on the sink composite, this requirement has been fulfilled (Hallale, 2002). From Figure 3, it is evidence that, without feeding fresh solvent to the network, process sources are capable of fulfilling the flowrate requirement of the process sinks. A careful observation would indicate that the total flowrate of the sources equals the total flowrate required by the sinks implying that this is a zero discharge process. However, this claim is only valid, once the second feasibility criterion of the network, i.e. the property load feasibility, is also fulfilled.

To fulfil the second feasibility criterion, we rely on the newly developed graphical tool called the property surplus diagram. To construct this plot, information from both the sink and source composite plots is needed. In Figure 3, the area below the sink composite plot, which is the product between the total sink flowrate and its associated operators, gives the total limiting property load that is acceptable by all process sinks. Similarly, the area below the source composite plot gives the total available property load possessed by the sources. Hence, the area between these composite plots indicates the area where property load is in excess (surplus) or in shortage (deficit) at its associated operator level. Hence, when a source composite lies above the sink composite, the area between the two composite plots indicates a property load surplus for this region. On the other hand, a sink composite lying above a source composite indicates a property load deficit (Figure 4a). The operator values (levels) are next plotted against their associated cumulative values of these property load surplus or deficit to form the *property surplus diagram*, as shown in Figure 4(b). Note that, since the cumulative value of the property load is plotted, a surplus load causes the surplus diagram to move to the right; while a deficit load moves it to the left.

A complete property surplus diagram with the assumed 0 kg/s fresh solvent feed is shown in Figure 4(b). Notice that part of this plot lies in the negative region of the property load surplus axis (x -axis), and this indicates that property load infeasibility occurs in this property range. This happens because the property load supplied by the sources is not sufficient to meet the load requirements of the sinks without the use of fresh solvent. Hence, the assumed zero fresh solvent flowrate does not fulfil the second feasibility criterion for the network synthesis. In other words, fresh solvent is needed to fulfil the second feasibility criterion and to restore the network feasibility. Hence, the aforementioned procedure of developing composite curves and surplus diagrams has to be repeated until the surplus diagram touches the property operator axis (y -axis), and no part of the surplus diagram lies in the negative region

Considering that the objective for network synthesis is to minimise the resource targets (minimum fresh solvent feed and waste discharge), we need to determine the minimum amount of fresh solvent flowrate that yields the minimum waste for the network. However, similar to the situation in hydrogen and water network synthesis problems (Alves & Towler, 2002; Hallale, 2002), the determination of minimum fresh solvent feed through the surplus diagram involves a tedious and time consuming trial-and-error solution. The tedious iterative procedure to construct a water surplus diagram is shown in Figure 5 (Manan *et al.*, 2004). In addition, the surplus diagram suffers from its limitation of generating highly accurate targets due to its graphical nature. In view of the above shortcoming in the property surplus diagram, we propose another numerically equivalent solution to locate the minimum resource targets. This is discussed in the following section.

ALGEBRAIC TARGETING APPROACH

The property surplus diagram presented in the previous section provides a basic understanding of the material flow in a property network. The drawbacks of the graphical approach can be resolved by introducing a numerically equivalent tool called the *Property Cascade Analysis* (PCA) technique. The PCA technique for the property surplus diagrams in property-based material reuse network is similar to the hydrogen cascade analysis technique for hydrogen network synthesis (Manan and Foo, 2003) and the water cascade analysis technique for water network synthesis (Manan *et al.*, 2004). The previously described Example 1 is used again to illustrate the PCA technique.

The first step in the PCA is to set up the *interval material balance table* (Table 4) to determine the net material source or sink at each operator level. This step is equivalent to constructing the material sink and source composite plots (Figure 3) for the property surplus diagram. The first column of Table 4 contains the operator levels (ψ) arranged in ascending order. The number of operator levels (n) equals the total number of property sinks (N_{SK}) and sources (N_{SR} ; including fresh feed) minus any duplicate operator (N_{DP}):

$$n = N_{SK} + N_{SR} - N_{DP} \quad (11)$$

Next, the property load surplus or deficit values between the sink and source composite plots are determined numerically. Note that in the previous property surplus diagram procedure, this step is done manually by calculating the area between the two composite plots (as shown in Figure 4).

Firstly, the property operator difference ($\Delta\psi$) between intervals k and $k + 1$ in column 2 of Table 4 is calculated as follows:

$$\Delta\psi = \psi_k - \psi_{k+1} \quad (12)$$

Next, columns 3 and 4 of Table 4 contain the total flowrates for the material sinks (in this case the solvent sink), $\sum_j F_{j,k}$ and the material sources, $\sum_i F_{i,k}$ at their corresponding operator levels k respectively. The *net interval material flowrate* at property level k , $F_{sum,k}$ (indicated in column 5), is given as the difference between the total flowrates of the material sources and the material sinks, i.e.

$$F_{sum,k} = \sum_i F_{i,k} - \sum_j F_{j,k} \quad (13)$$

The positive value of $F_{sum,k}$ at each individual purity level represents the *net material source*, whereas negative values represent *net material sinks* (see column 6).

The next key step in the PCA is to establish the fresh material (solvent) target for the network. In order to do so, it is important to consider both the material flowrate balance and the property load, so that the true minimum resource targets can be obtained. The material flowrate balance involves the use of the *material cascade diagram* shown in Figure 6, to obtain the *cumulative net material source/sink* for a process (F_C). The

material cascade diagram is similar to the mass exchange cascade diagram in mass integration (El-Halwagi, 1997). A conceptual illustration of how material cascading can minimise fresh material needs is given in Figure 7. A flowrate of 3 kg/s of the net solvent source is available at the second operator level of 3.741 atm^{1.44} ($k = 2$; column 5 of Table 4). By partly using this solvent source to satisfy the solvent sink (5 kg/s) at the operator level of 4.865 m^{1.44} ($k = 3$), it is possible to eliminate the effluent totally, and the consumption of fresh solvent by 60% (Figure 7b). It should also be noted that, since the solvent source has a lower operator value compared to the solvent sink, the property load sent from the source is smaller than the maximum acceptable property load of the sink.

Targeting using the material cascade diagram has been previously discussed for water cascade analysis in Manan *et al.* (2004). Hence, it will only be briefly described here. Figure 6(a) shows the material cascade diagram with zero fresh solvent flowrate ($F_F = 0$), supplied at operator level of 2.713 m^{1.44}, leads to a zero material discharge ($F_D = 0$). Next, the property load feasibility is examined using the *property load cascade* in Figure 6(b), which numerically representing the property surplus diagram in Figure 4(b). Notice that cumulative property load deficits are observed in the last three operator levels of the property load cascade, i.e. $\psi_4 - \psi_6$. These property load deficits (correspond to the negative region of the material surplus diagram in Figure 4b) indicate that the property load cascade is “infeasible” as a result of the zero fresh solvent flowrate during material cascading. To ensure that there is sufficient fresh solvent at all points in the network, one will need to feed the right amount of minimum flowrate to the network. This corresponds to the largest negative flowrate among all *interval fresh material demand* that are found in Figure 6(c). The interval fresh material demand for each operator level k ($F_{F,k}$) is obtained by dividing the cumulative property load surplus/deficit by the operator difference between the fresh solvent supply (ψ_F) and the operator level of interest (ψ_k), as follows:

$$F_{F,k} = \frac{\text{cumulative property load surplus/deficit}}{\psi_F - \psi_k} \quad (14)$$

From Figure 6(c), the largest negative $F_{F,k}$ value is found at the fifth operator level ($\psi_5 = 13.199 \text{ m}^{1.44}$), i.e. -2.38 kg/s . This same quantity of fresh solvent is added at the highest purity level of the feasible material cascade in Figure 8(a), and results in positive or zero cumulative property load surplus value in the cumulative property load cascade (Figure 8b). The feasible material cascade yields the true minimum fresh solvent target (F_F) of 2.38 kg/s and a discharge flowrate (F_D) of 2.38 kg/s, match the targets identified using the graphical pinch diagram of Kazantzi & El-Halwagi (2005). By plotting the feasible cumulative property loads at each operator level in Figure 8(b), a feasible property surplus diagram is created in Figure 9. Notice that the entire surplus diagram is now shifted to the positive region of the property load surplus axis (x -axis). The material cascade and the property load surplus diagrams can be integrated with the interval material balance table to form the *property cascade table* (PCT) shown in Table 5.

Apart from its power to quickly yield the resource targets, the PCA enables the designer to clearly identify the *pinch-causing source* and the *resource allocation targets*, i.e. the exact material allocation for the regions above and below the pinch. Hallale (2002) showed that in a water recovery network, a pinch will always occur at the composition of a water source, where the source composite switches from being below the sink composite (i.e. deficit) to being above the sink composite (i.e. surplus). In property-based material reuse network, this is the point where the property load surplus just equals the property load deficit in the network. Referring to the PCT in Table 5, a zero cumulative property load surplus at the operator level of $13.199 \text{ m}^{1.44}$ ($k = 5$) represents the pinch point of the property network. From Table 3, Process Condensate I (SR1) with a flowrate of 4.0 kg/s is identified as the pinch-causing source for this example.

Several conclusions can be drawn from this observation. The pinch location corresponds to the point in the diagram, for which any attempts to pass load through it, will result in a network design “penalty” of additional fresh usage and waste discharge. Above the pinch, the property load supplied from the sources is in balance with the load required by the sinks in this region. On the other hand, property load is in excess in the region below the pinch. Due to unmatched property load restrictions, this “excess” load cannot be reused and has to be discharged from the network. Also, no load is to be transferred across the pinch to ensure that the MRR objective is achieved. In order to achieve the minimum resource targets, 1.62 kg/s of solvent source SR1 (found at the interval between ψ_4 and ψ_5) is sent to the region above the pinch, while 2.38 kg/s (found below ψ_5) is sent to the region below the pinch, which eventually becomes the discharge stream (F_D).

A “balanced” material sink and source composite diagram constructed using the targeted fresh solvent flowrate (Figure 10) can be used to summarise the above-mentioned insight. As shown, the purest segment of the source composite curve in Figure 10 represents the fresh solvent feed of 2.38 kg/s . The pinch operator found previously divides the network into two thermodynamic regions, i.e. one above the pinch and the other below the pinch. Also, Figure 10 shows that, the pinch-causing source (at the lowest operator level among all sources) has its exact allocation flowrates for network above and below the pinch, as have been targeted earlier. These exact material allocation flowrates can also be verified with the detailed network design technique, which will be discussed in the next section of this paper.

NETWORK DESIGN

This section discusses a simple technique for synthesising a property network that achieves the resource targets previously established. Example 1 will be used to illustrate the design procedure.

The pinch operator found previously divides the network into two thermodynamic regions, i.e. one above the pinch and the other below the pinch. Observing the pinch division is essential in achieving the minimum resource targets determined during the targeting stage. This observation implies that one should not feed a source located above the pinch (including the fresh resource) to a sink below the pinch in designing a property

network. Violating this rule will incur a higher resource penalty. This guideline also holds for mixing process sources from different thermodynamic regions. However, an exception to this rule is for stream(s) found at the pinch operator (SR1 in Example 1), since these stream(s) belong to both regions.

After observing the above guidelines, a property network can be designed using the following equations, which were originally developed for mass exchange network design (El-Halwagi, 1997):

(a) For sinks
Flowrate:

$$\sum_i F_{i,j} = F_j \quad (15)$$

where $F_{i,j}$ is the flowrate fed from source i to sink j .

Property load:

$$\sum_i \Delta m_{i,j} \leq \Delta m_j^{\max} \quad (16)$$

where $\Delta m_{i,j}$ is the property load fed from source i to sink j , that is given as the product of $F_{i,j}$ and ψ_i .

(b) For sources
Flowrate:

$$\sum_j F_{i,j} \leq F_i \quad (17)$$

Eq 15 and 16 provide expressions for a fixed flowrate and the maximum acceptable property load required by a sink, respectively; whereas Eq 18 indicates that the flowrate fed from source i to sink j is bounded within its available limit. The remaining portion of the source that is not fed to a sink will leave as a waste stream. One possible network for Example 1 using the above formulation is shown in Figure 11, where SR1 (Process Condensate I) is reused in both sinks and SR2 (Process Condensate II) in sink SK1 (Degreaser) alone. For cases involving more sources and sinks, many alternative design options are possible. One may also impose other constraints into the network design, e.g. forbidden matches for safety or operability reasons.

For cases where process sinks appear in different regions network design is best conducted independently for each thermodynamic region so as to prevent the sources from different thermodynamic regions to be fed to the sinks and to avoid unnecessary mixing of sources across these regions. This is particularly important in multiple pinch problems where more thermodynamic regions exist in a network.

PROCESS MODIFICATIONS

Making appropriate changes to a process has been widely accepted as an effective measure to further reduce resource targets in mass integration (e.g., El-Halwagi, 1997). The same measure can be applied for property integration. We will again demonstrate this approach using Example 1.

Figure 12 shows the final configuration of the degreaser plant in Example 1 that was designed using the previously described network design techniques. A fresh solvent supply at 2.38 kg/s is currently fed to the process, while the same amount of waste solvent is discharged from the process. It is now desirable to further reduce the fresh solvent consumption by making changes in the process. We also observe that the total flowrate of the process sources, i.e. SR1 and SR2 is equal to the total flowrate requirements of the sinks (SK1 and SK2). To achieve zero discharge, one possible option is to modify the property of SR1 (since this is the only source that is currently discharged from the process), so that it can be reused further in both process sinks.

To fulfil the flowrate requirements of both sinks, 2.0 kg/s of solvent from SR1 is to be fed to each of these sinks. Since the degreaser has already been fed with the entire SR2 (with a flowrate of 3.0 kg/s) it is not necessary to make any changes in this solvent source. However, for any attempt to reuse more solvent from SR1, we will have to take into consideration the maximum property load constraint of the sinks as imposed by Eq. 8. Eq. 19 shows that the RVP for Condensate I, RVP_{SR1} , is a function of the operating temperature in the solvent regeneration unit (Kazantzi and El-Halwagi, 2004):

$$RVP_{SR1} = 0.56e^{\left(\frac{T-100}{175}\right)} \quad (19)$$

where T is the temperature of the thermal processing system in Kelvin (K). The acceptable range of this temperature is between 430 and 520 K. At present, the thermal processing system operates at 515 K, leading to an RVP of 6.0 atm. This operating condition can be modified to enable more solvent from SR1 to be reused in the sinks.

Analysis of the property load currently received by the process sinks in Figure 12 indicates that both sinks are currently receiving property loads at their maximum acceptable level, i.e. 24.323 kg.atm^{1.44}/s and 14.723 kg.atm^{1.44}/s respectively. In this regard, the degreaser receives 9.659 kg.atm^{1.44}/s from SR1, 11.223 kg.atm^{1.44}/s from SR2 and the remaining 3.440 kg.atm^{1.44}/s load from the fresh solvent. In addition, the absorber receives a load of 11.702 kg.atm^{1.44}/s from SR1 and 3.011 kg.atm^{1.44}/s from the fresh solvent. Hence, to entirely substitute the current fresh solvent with SR1, one should not feed more than 13.099 kg.atm^{1.44}/s of property load to the degreaser (recalling that a load of 11.223 kg.atm^{1.44}/s is currently fed from SR1) and 14.723 kg.atm^{1.44}/s of property load to the absorber. Based on these constraints, one can back calculate the maximum acceptable property operator for the degreaser and the absorber at 6.55 atm^{1.44} and 7.36 atm^{1.44} respectively. A lower value is chosen in this case in order to satisfy both sinks. From Eq. 19, this corresponds to an operating temperature of 430 K, which lies within the

acceptable range of temperatures for the solvent regeneration unit. The final flowsheet after process modification is shown in Figure 13.

EXAMPLE 2 – PAPERMAKING PROCESS

Previous sections of the paper have shown how PCA can be used to assess the various recycle/reuse and process modification strategies in a metal degreasing process. Note that the fresh solvent that is used in the degreasing process is fed at the lowest operator level compared to all other process sources (see Table 2). Unlike the cases in water and hydrogen network synthesis, fresh process feed in property integration problems do not always available at the highest (or “superior”) operator level except in some cases. Note also that this has an effect on how the limiting data is extracted for the case study. We will demonstrate the use of PCA to handle cases with fresh feed of lowest (or “inferior”) operator value using the following example.

Figure 14 shows a papermaking process taken from Kazantzi and El-Halwagi (2005) and EL-Halwagi et al (2004). Wood chips are digested and chemically treated in the Kraft digester before the produced pulp is sent to the bleaching section. The product from this section, i.e. bleached fibre is then sent to two paper machines (Paper Machine I and II), where they are converted into final paper products. Rejected products from Machine I are further treated in Hydro Pulper and Hydro Sieve before the waste and waste fibre streams (broke) are finally discharged. However, due to environmental concerns, it is proposed to recycle the broke back to the process.

The candidates that can be regarded as process sinks, where waste fibre is recycled, are the two paper machines. An external fresh fibre source is currently fed to paper machine II to supplement its fibre need. Thus, by recycling the broke, resource usage is maximised and fresh fibre consumption can be reduced.

To evaluate the quality of the broke to be used as a feed stream to the sinks, we focus on reflectivity R_∞ , which is a dimensionless property for the produced paper. It is defined as the reflectance of an infinitely thick material compared to an absolute standard, i.e. magnesium oxide. The mixing rule for reflectivity R_∞ has been given in Table 1.

Flowrate and reflectivity data for the fibre sinks and sources are given in the first four columns of Table 6. As shown there, the fresh fibre feed possesses a reflectivity value of 0.95, which is at the highest level compared to all other limiting operators. In order to minimise the usage of the fresh feed, we shall define the lower bound of the reflectivity value to be the limiting operators of the process sinks. This, in turn, leads to lower limiting loads required by the process sinks (compared to that defined by the upper bound as the limiting operator). Limiting operators and loads for all fibre sinks and sources are listed in the final two columns of Table 6.

PCA is carried out to locate the minimum resource targets for the example. Note that, since the fresh fibre feed possesses a higher operator value (or is “inferior” to the other sources), the operators are arranged in descending order, in contrast to the metal

degreasing example. This is shown in the interval material balance table (Table 7) and PCT (Table 8). The minimum fresh fibre feed (F_F) and discharged flowrate (F_D) are easily identified from the PCT as 14.95 ton/h and 24.95 ton/h respectively. These resource targets agree with those obtained by Kazantzi and El-Halwagi (2005). In addition, PCT also identifies the broke stream as the pinch-causing stream from which 35.05 ton/h is fed to the region above the pinch; whereas 25.95 ton/h goes to the region below the pinch, with the final network design shown in Figure 15.

CONCLUSIONS

This work presents the developments of a graphical technique called property surplus diagram and a new numerical targeting technique called property cascade analysis (PCA) to establish the resource targets within a property integration framework. PCA compliments the property surplus diagram that has been adapted from the targeting techniques developed for water and hydrogen networks synthesis. As an algebraic alternative to the surplus diagram, PCA quickly yields accurate resource targets, pinch locations, as well as material allocation targets for a property network, ahead of detailed network design. A procedure for synthesis of a maximum resource recovery (MRR) network has also been presented. To further reduce resource consumption in a property-based material reuse network, a systematic procedure to identify scope for process modifications has been outlined. The establishment of a comprehensive framework for the synthesis of a resource recovery network allow the procedure to be extended to a wider range of problems for which property is the main concern.

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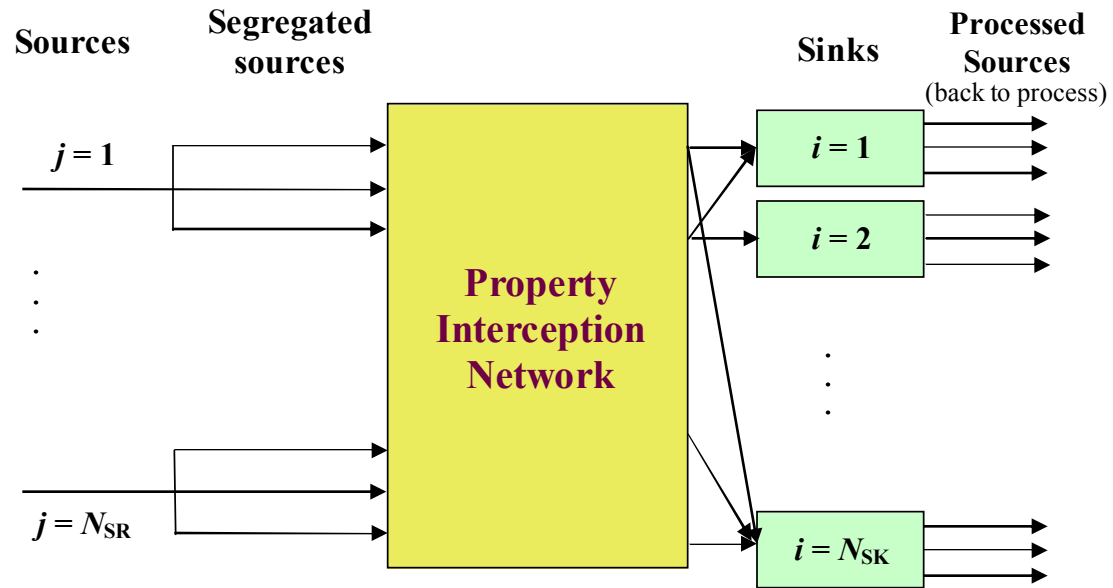


Figure 1. Source-sink representation for a property-based material reuse network

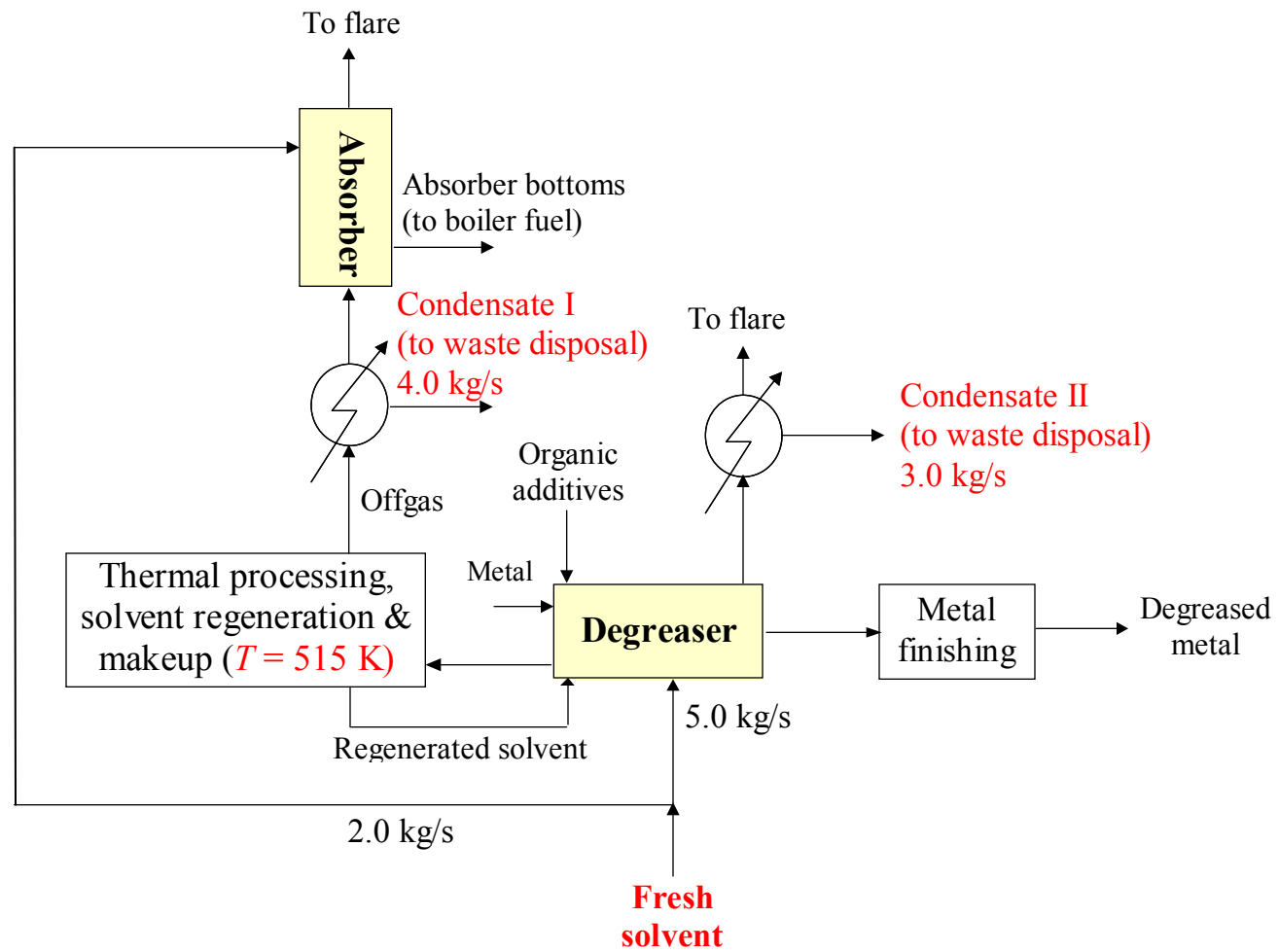


Figure 2. Example 1: A metal degreasing process (Kazantzi and El-Halwagi, 2005)

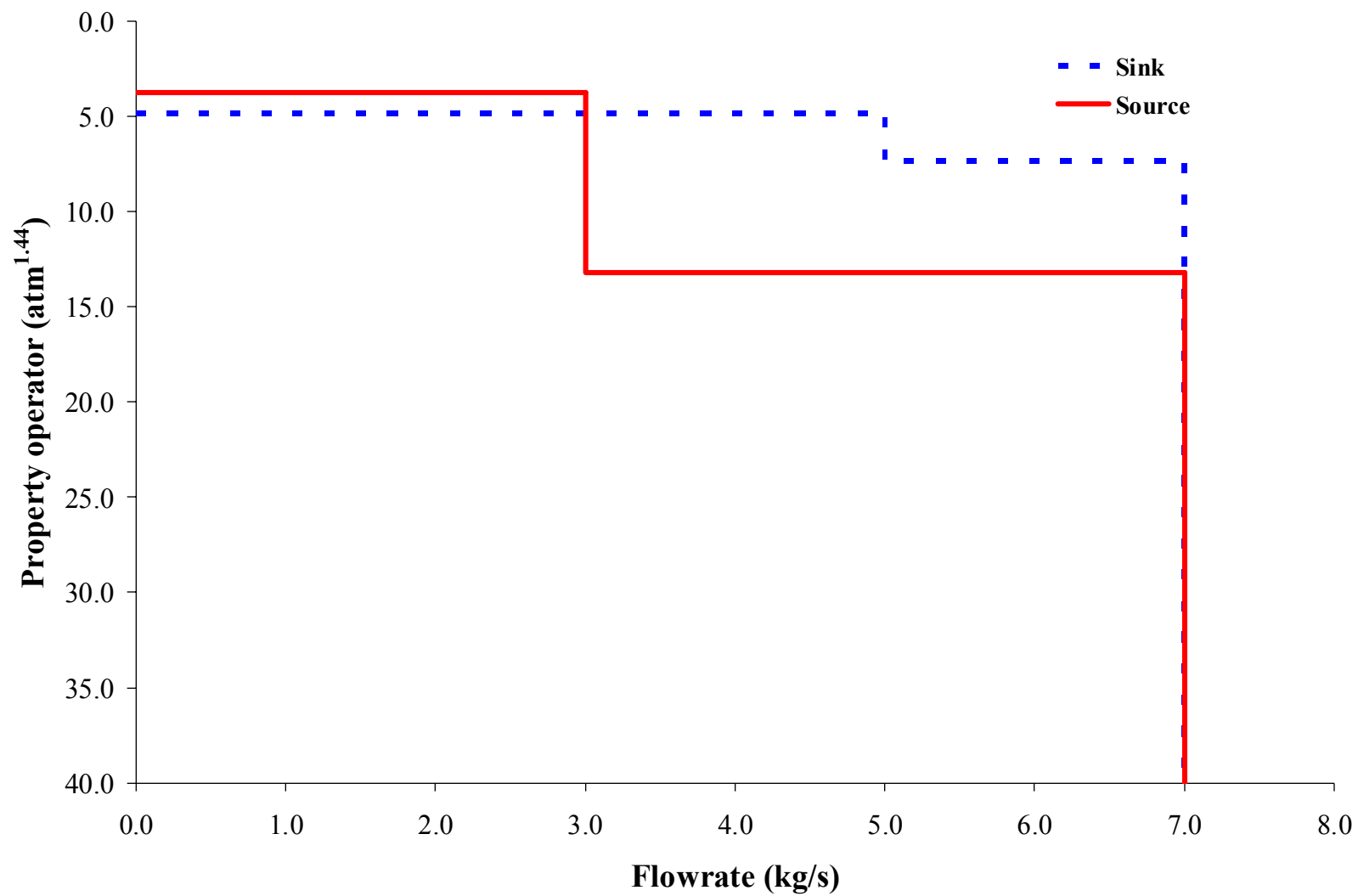
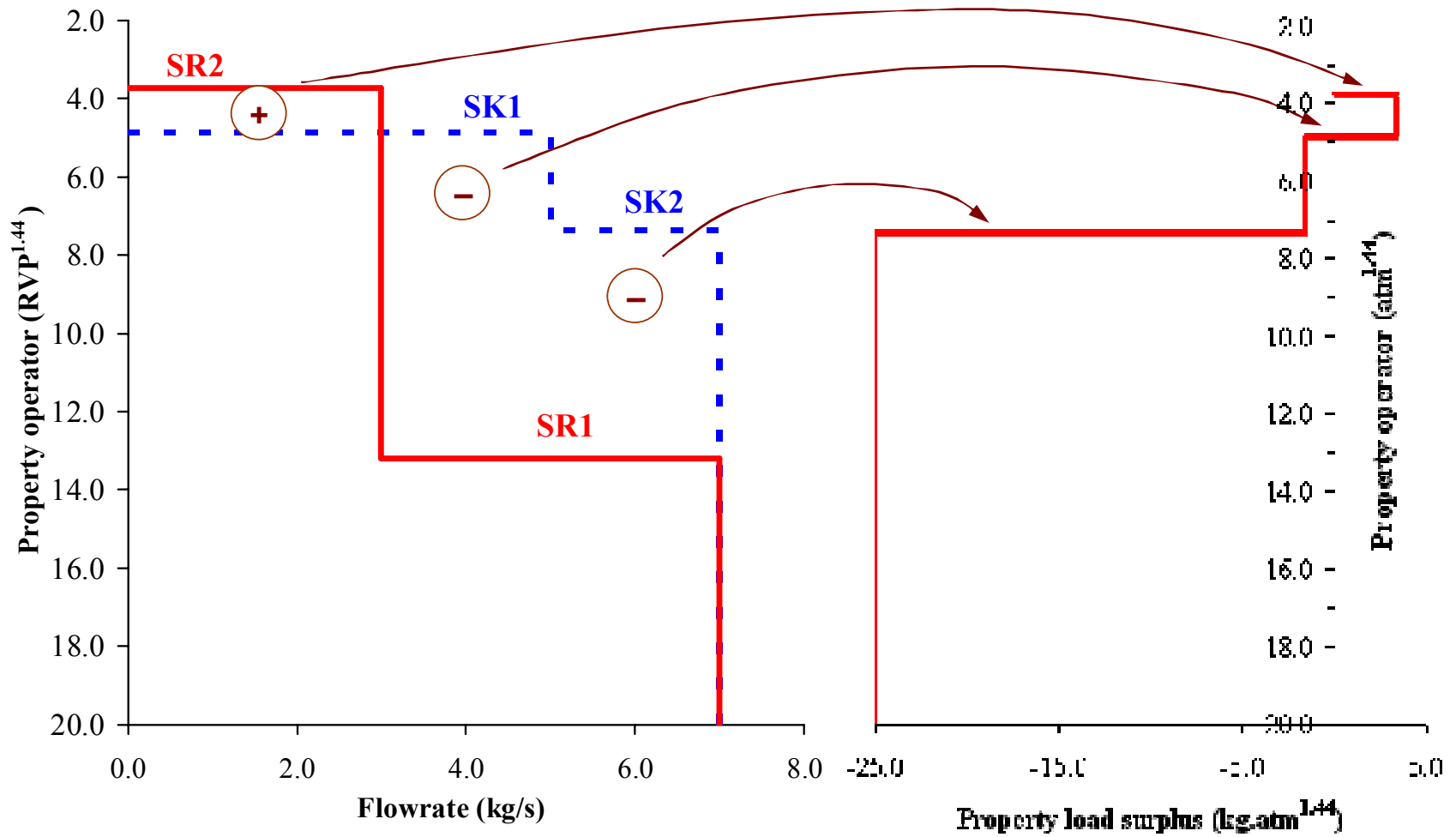


Figure 3. Sink and source composite plots for Example 1, with the solvent flowrate set to 0 kg/s



(a) Sink and source composite plots

(b) Property surplus diagram

Figure 4 Construction of a property surplus diagram

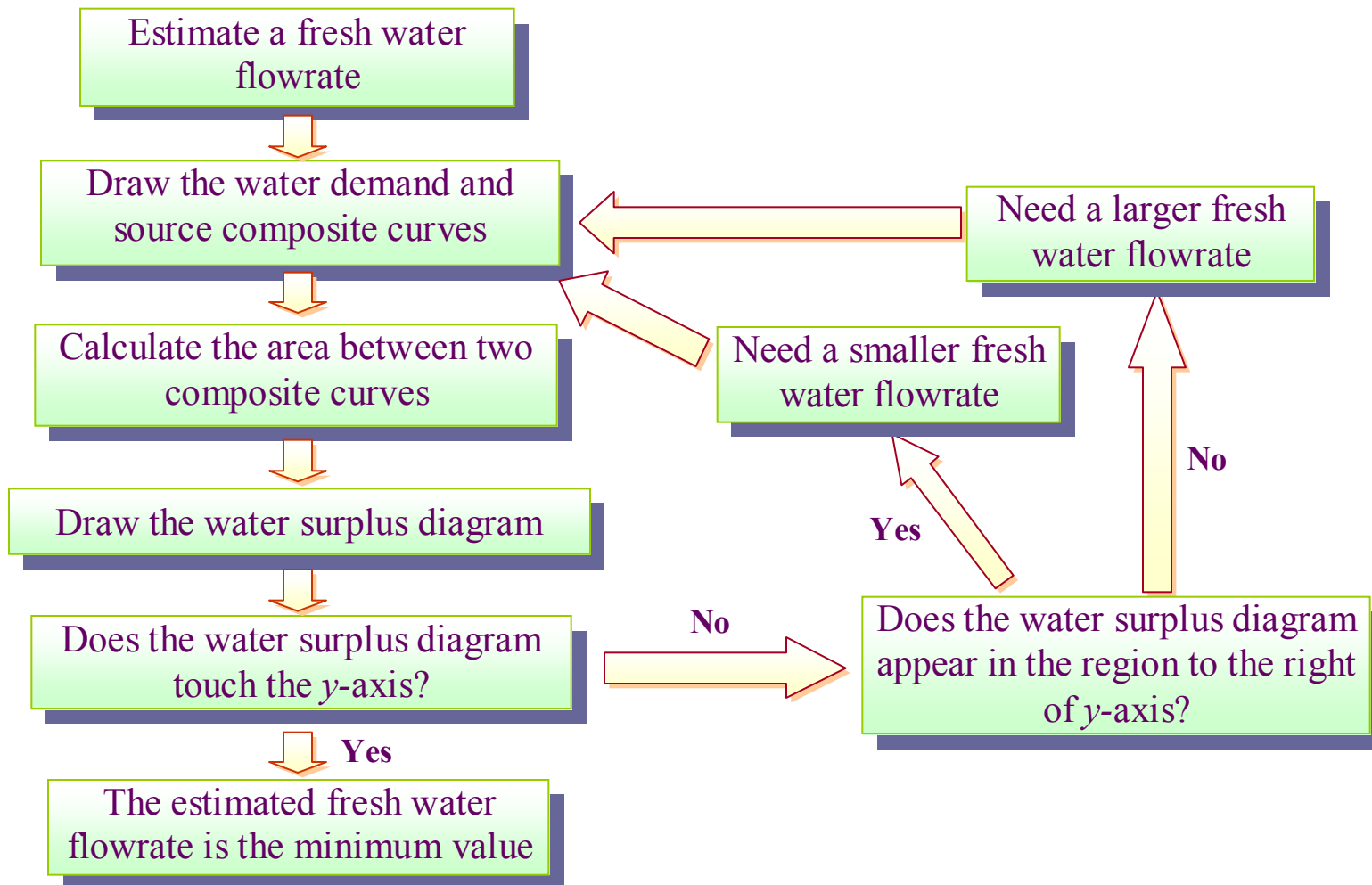


Figure 5. The tedious iterative steps for constructing a water surplus diagram (Manan *et al.*, 2004)

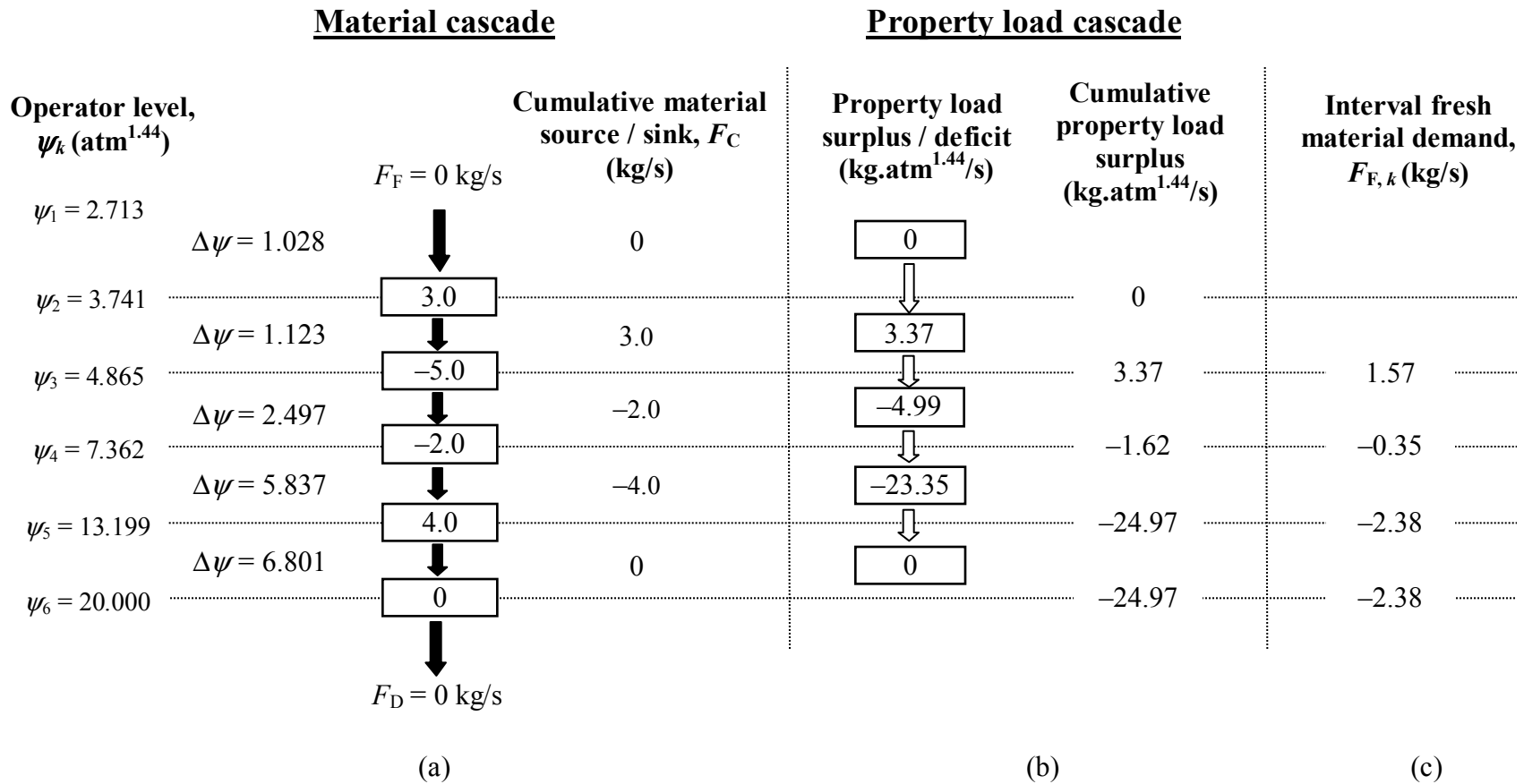


Figure 6. (a) Material cascade diagram with an assumed fresh solvent flowrate of 0 kg/s; (b) pure property load cascade is used to check the feasibility of the property network; (c) determining interval material flowrates at each purity interval k

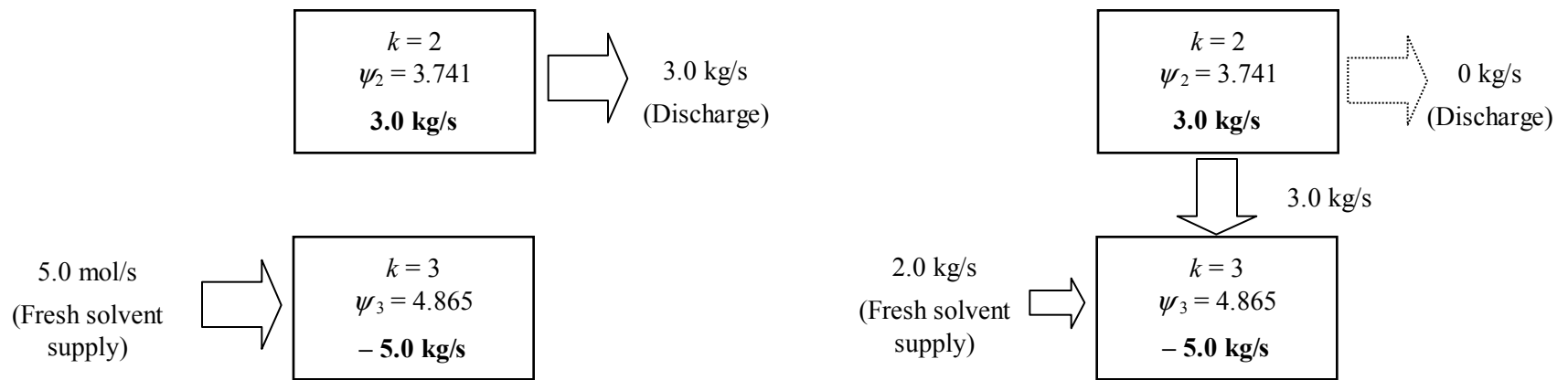


Figure 7. (a) No solvent cascading; (b) with solvent cascading, discharge is eliminated and fresh solvent intake is reduced

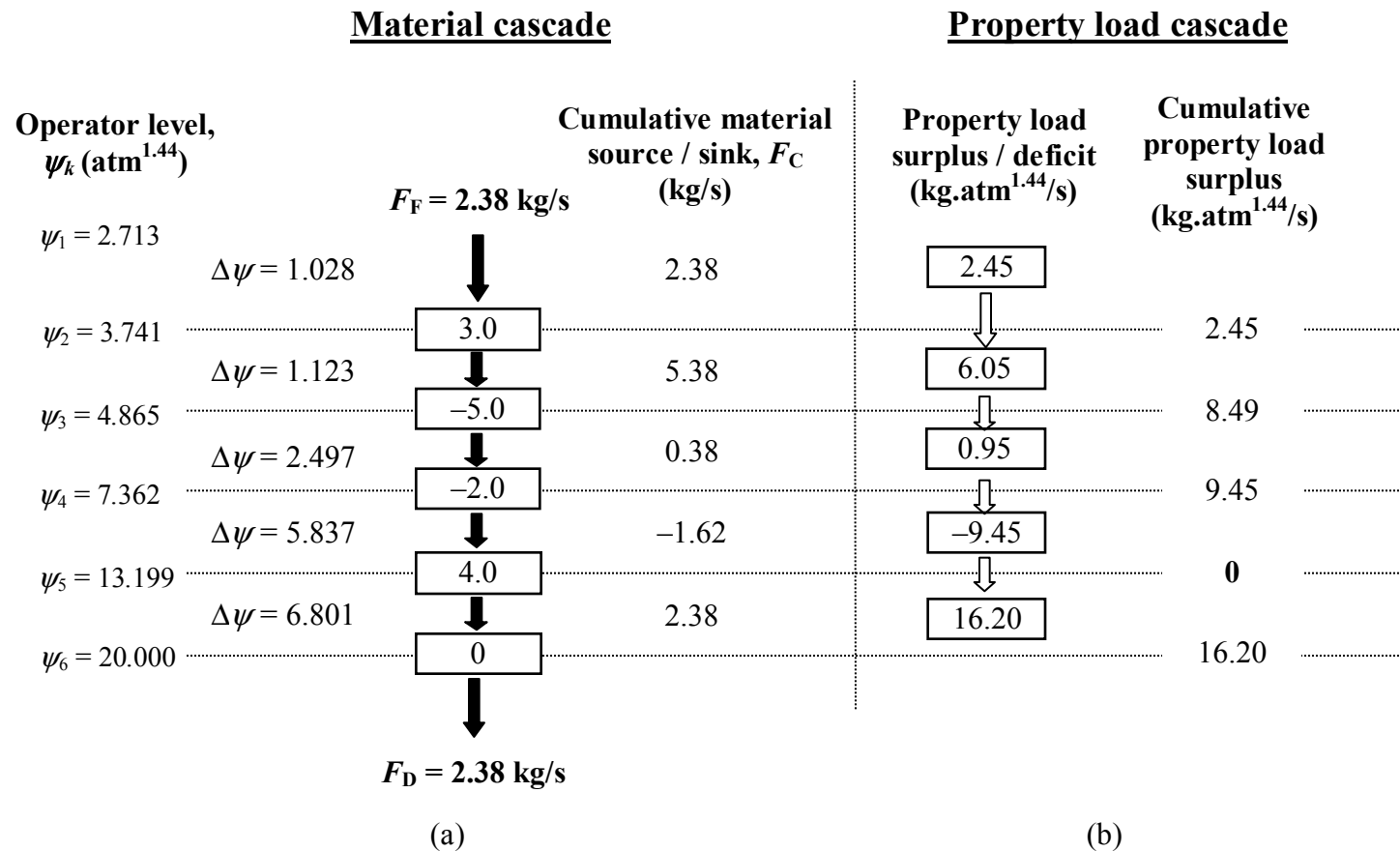


Figure 8. A feasible material cascade for Example 1

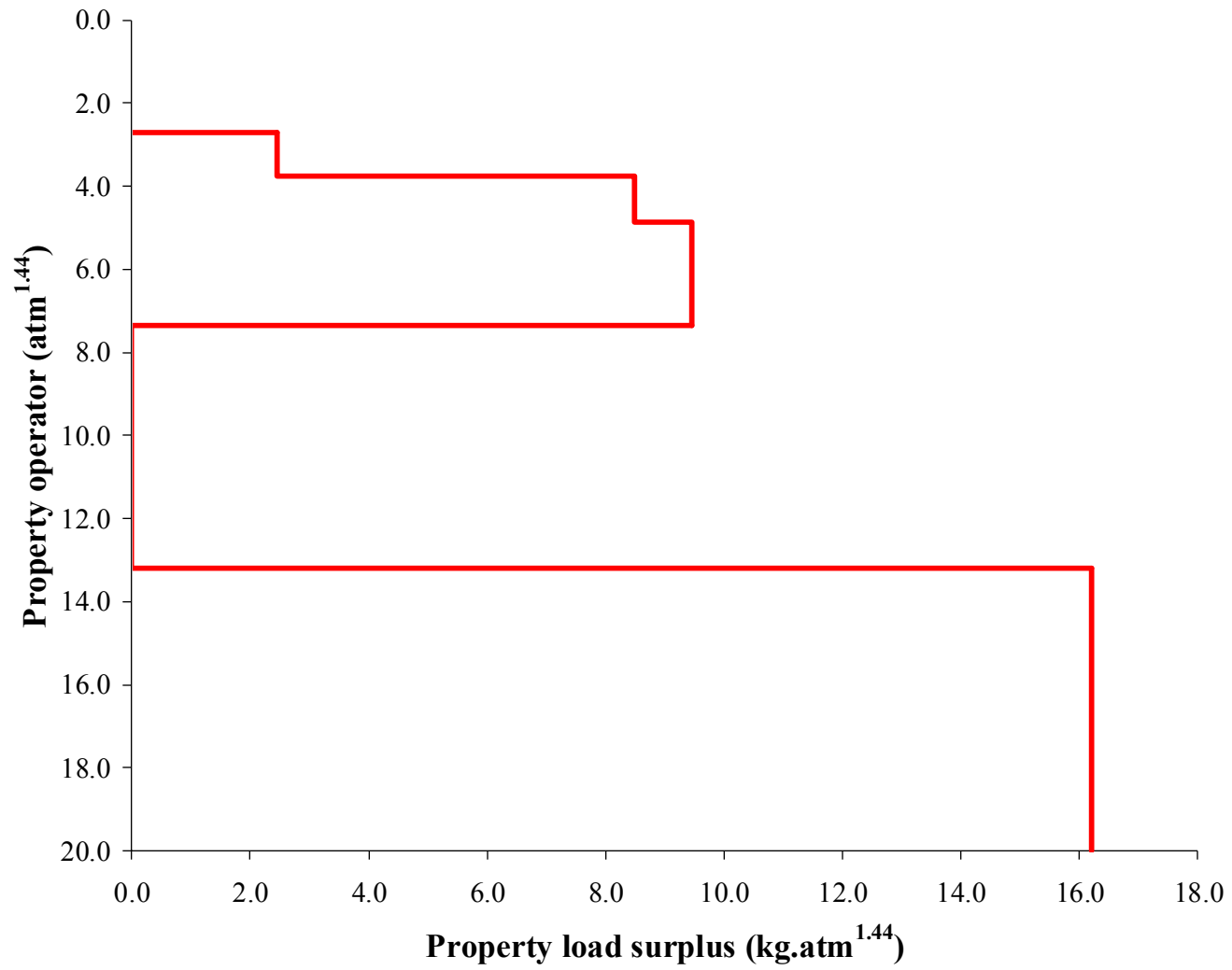


Figure 9. A feasible property surplus diagram for Example 1

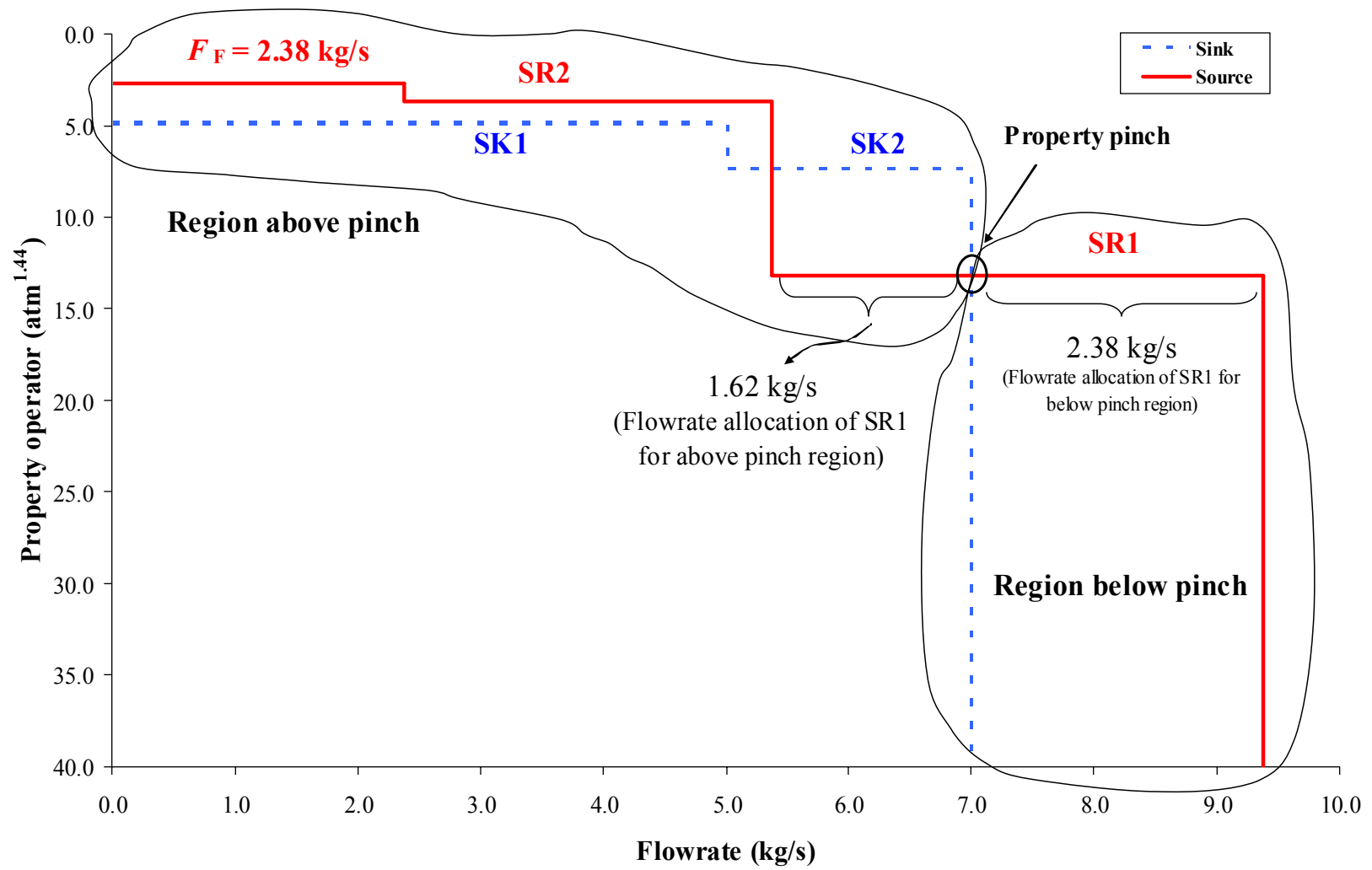
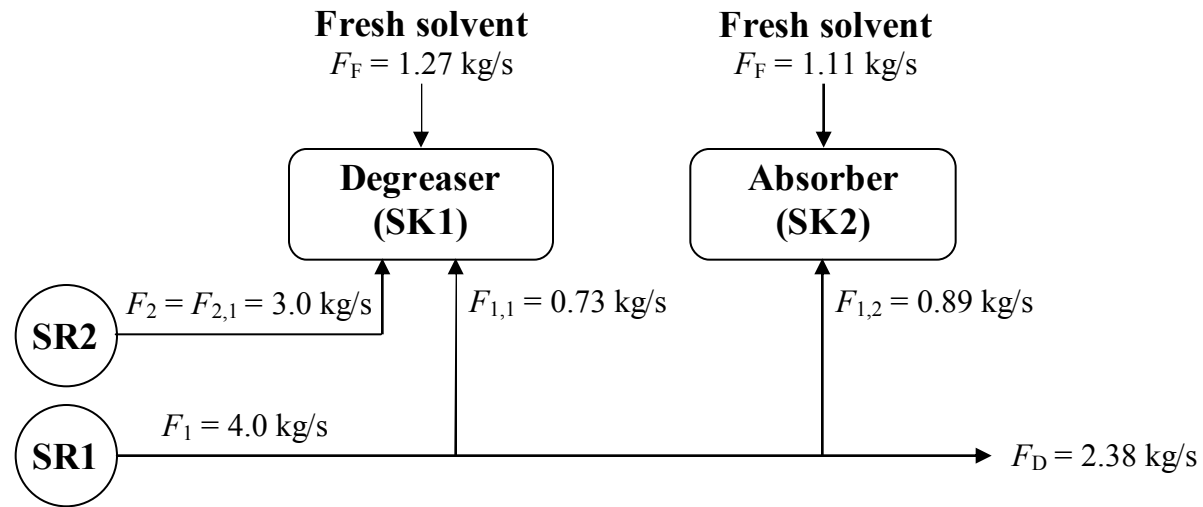


Figure 10. A feasible property surplus diagram for Example 1



Sinks	Sources	$F_{i,j}$	x_i	RVP	$\Psi_{i,j}$	$x_i \Psi_{i,j}$	Δm_{ij}
SK1	Fresh	1.27	0.254	2.0	2.713	0.689	3.446
	SR2	3.00	0.600	2.5	3.741	2.245	11.224
	SR1	0.73	0.146	6.0	13.199	1.927	9.635
	TOTAL	5.0	1.0			4.861	24.305
SK2	Fresh	1.11	0.557	2.0	2.713	1.510	3.021
	SR1	0.89	0.443	6.0	13.199	5.851	11.702
	TOTAL	2.0	1.0			7.362	14.723

Figure 11. Network design for Example 1

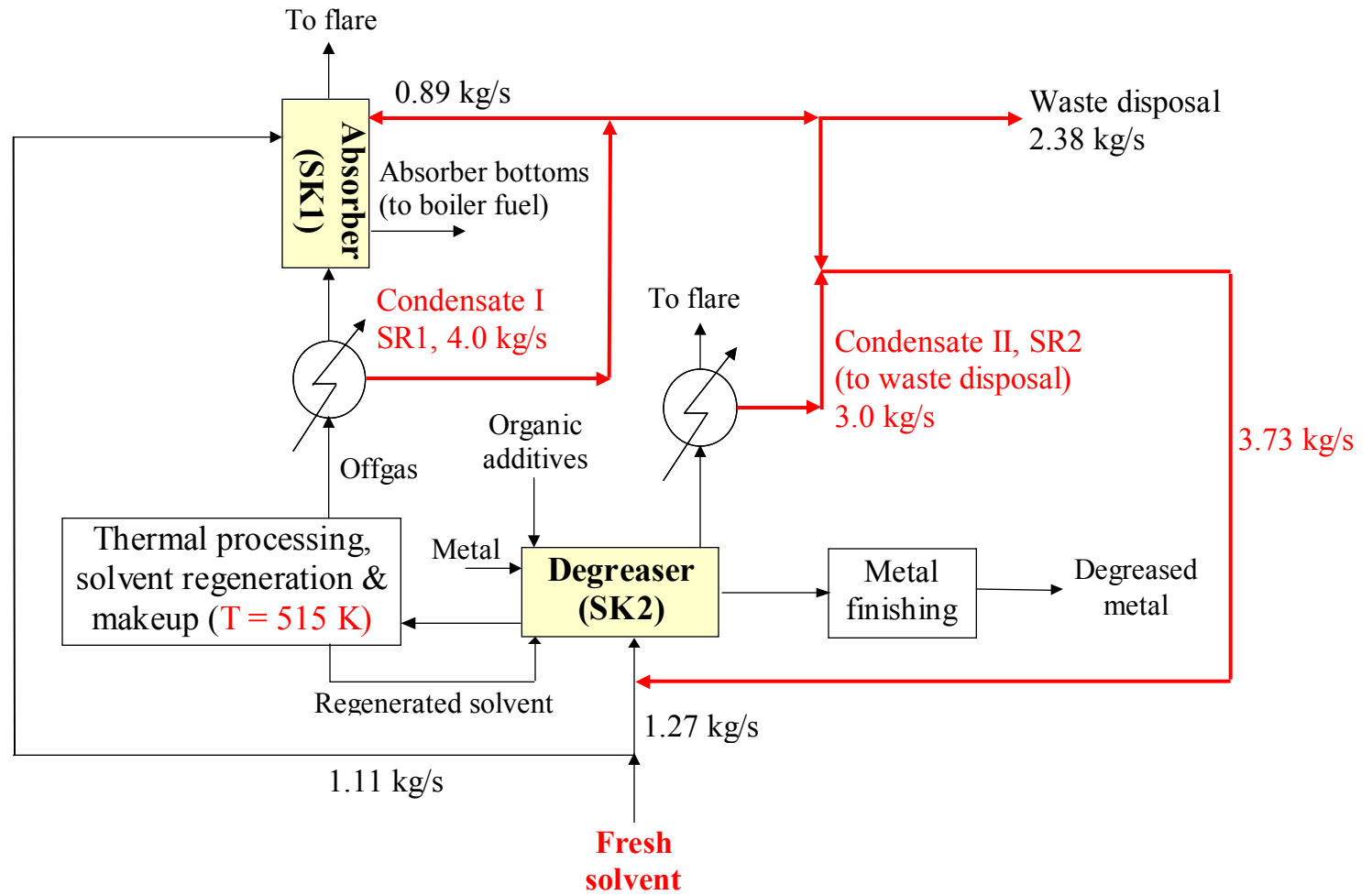


Figure 12. Network design for degreaser plant to achieve MRR objective (before process modification)

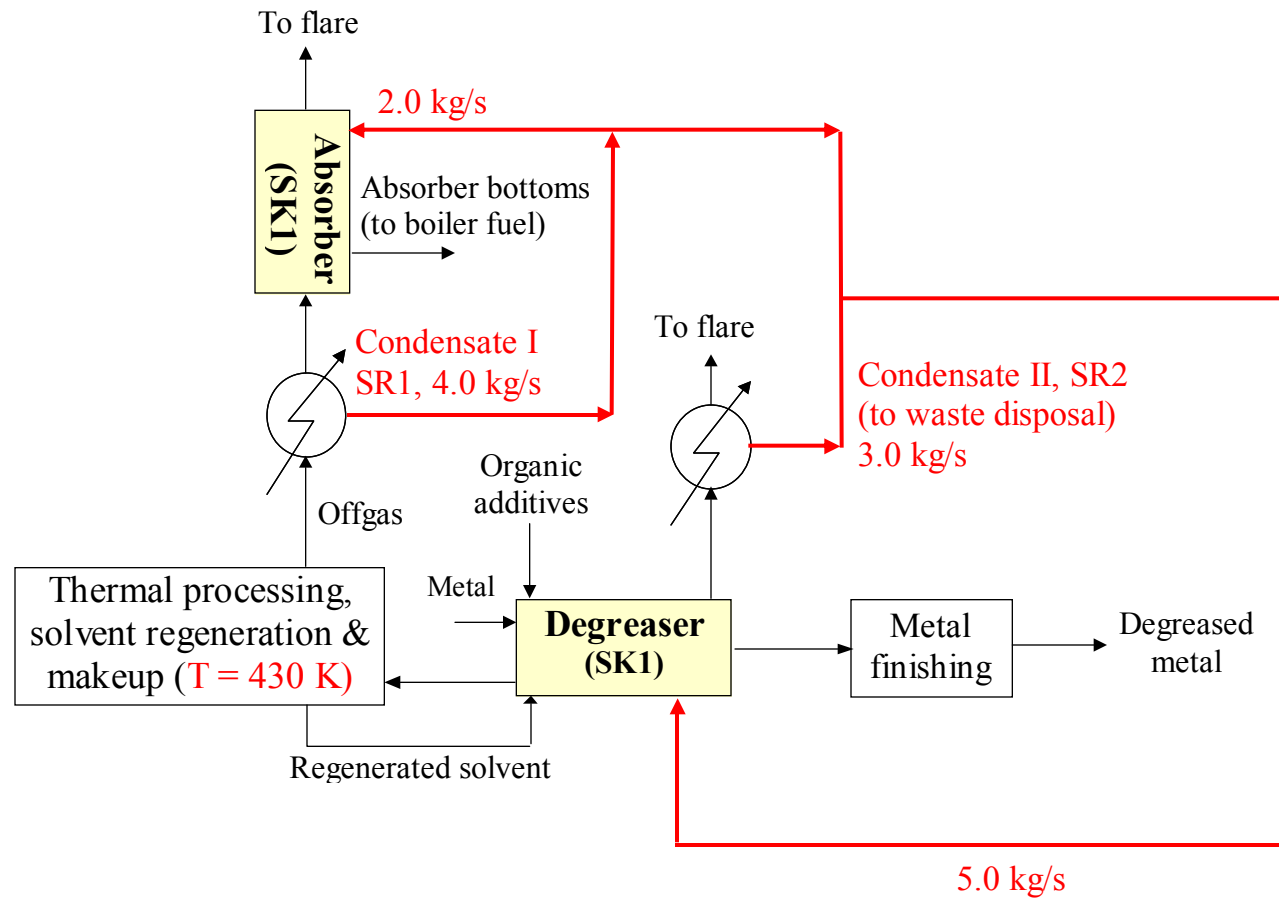


Figure 13. Network design for metal degreasing process after process modification

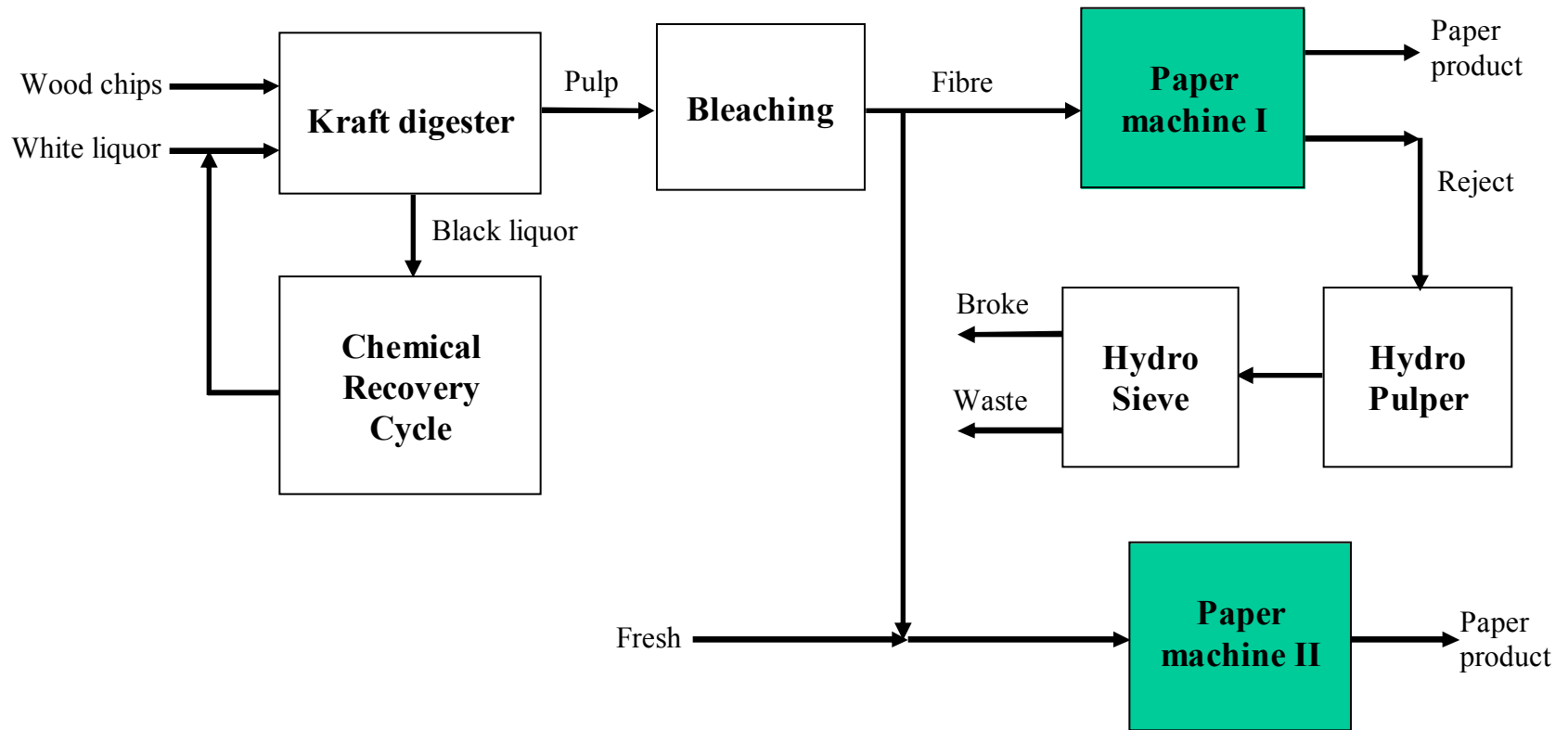


Figure 14. A papermaking process (Kazantzi and El-Halwagi, 2005)

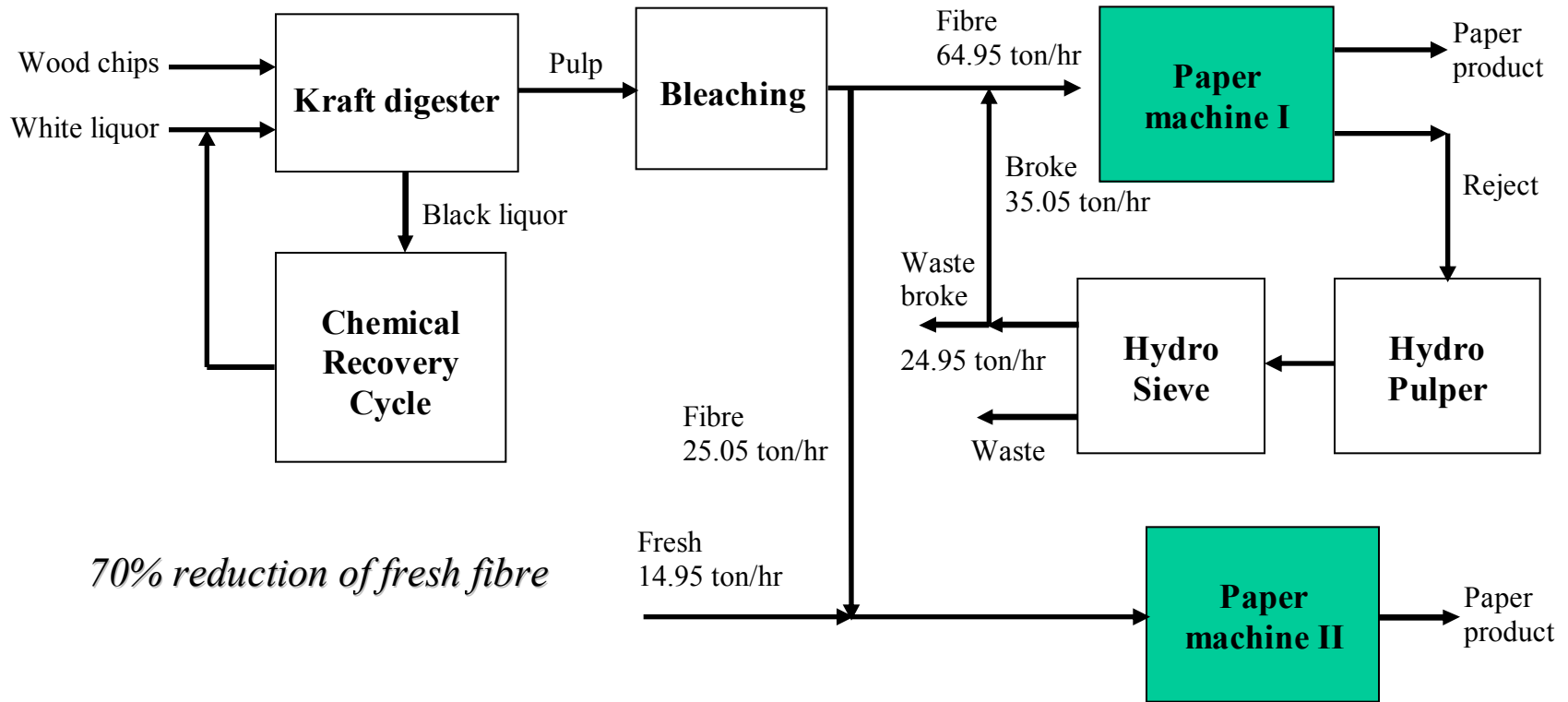


Figure 15. Final configuration of the papermaking process

Table 1. Operator expressions for common properties

Property of mixture	Mixing rule	Operator	Reference
Density, $\bar{\rho}$	$\frac{1}{\bar{\rho}} = \sum_i \frac{x_i}{\rho_i}$	$\psi(\rho_i) = \frac{1}{\rho_i}$	Shelley and El-Halwagi (2002)
Reid Vapor Pressure, \overline{RVP}	$\overline{RVP} = \sum_i x_i RVP_i^{1.44}$	$\psi(RVP_i) = RVP_i^{1.44}$	Shelley and El-Halwagi (2002)
Material content, \bar{M}	$\bar{M} = \sum_i x_i M_i$	$\psi(M_i) = M_i$	Shelley and El-Halwagi (2002); El-Halwagi <i>et al.</i> (2002)
Electric resistivity, \bar{R}	$\frac{1}{\bar{R}} = \sum_i \frac{x_i}{R_i}$	$\psi(R_i) = \frac{1}{R_i}$	Kazantzi and El-Halwagi (2004)
Viscosity, μ	$\log(\bar{\mu}) = \sum_{i=1}^{N_s} x_i \log(\mu_i)$	$\psi(\mu_i) = \log(\mu_i)$	Qin <i>et al.</i> (2004)
Paper reflectivity, \bar{R}_∞	$\bar{R}_\infty = \sum_i x_i R_{\infty,i}^{5.92}$	$\psi(R_{\infty,i}) = \sum_i x_i R_{\infty,i}^{5.92}$	El-Halwagi <i>et al.</i> (2002)

Table 2. Data for Example 1

Process	Flowrate (kg/s)	Reid Vapor Pressure, RVP (atm)		Property operator, ψ (atm ^{1.44})	
		Lower bound	Upper bound	Lower bound	Upper bound
(Sink)					
Degreaser (SK1)	5.0	2.0	3.0	2.713	4.865
Absorber (SK2)	2.0	2.0	4.0	2.713	7.362
(Source)					
Condensate I (SR1)	4.0		6.0		13.199
Condensate II (SR2)	3.0		2.5		3.741
Fresh solvent	To be determined		2.0		2.713

Table 3. Limiting data for Example 1

Process	Flowrate, F (kg/s)	Limiting operator, Ψ (atm^{1.44})	Limiting load, Δm (kg.atm^{1.44}/s)
(Sinks)			
Degreaser (SK1)	5.0	4.865	24.323
Absorber (SK2)	2.0	7.362	14.723
(Sources)			
Process Condensate I (SR1)	4.0	13.199	52.796
Process Condensate II (SR2)	3.0	3.741	11.224
Fresh Solvent	To be determined	2.713	

Table 4. Interval material balance table for Example 1

Column no.	1	2	3	4	5	6
Level, <i>k</i>	Operator ψ_k (atm ^{1.44})	$\Delta\psi$ (atm ^{1.44})	$\sum_j F_j$ (kg/s)	$\sum_i F_i$ (kg/s)	$F_{sum, k}$ (kg/s)	Net material surplus/deficit
1	2.713				0	
2	3.741	1.028		3.0	3.0	Surplus
3	4.865	1.123	-5.0		-5.0	Deficit
4	7.362	2.497	-2.0		-2.0	Deficit
5	13.199	5.837		4.0	4.0	Surplus

Table 5. The Property Cascade Table for metal degreasing process

Level, <i>k</i>	Operator ψ_k (atm ^{1.44})	$\sum_j F_j$ (kg/s)	$\sum_i F_i$ (kg/s)	$F_{sum, k}$ (kg/s)	F_c (kg/s)	Property load surplus (kg.atm ^{1.44} /s)	Cumulative property load surplus (kg.atm ^{1.44} /s)
					$F_F = 2.38$		
1	2.713			0	2.38	2.45	
2	3.741		3.0	3.0	5.38	6.05	2.45
3	4.865	-5.0		-5.0	0.38	0.95	8.49
4	7.362	-2.0		-2.0	-1.62	-9.45	9.45
5	13.199		4.0	4.0	$F_D = 2.38$	16.20	0
6	20.000						16.20

Table 6. Limiting data for Example 2 (papermaking process)

Process	Flowrate (ton/h)	Reflectivity, R_{∞} (dimensionless)		Limiting operator, Ψ (dimensionless)	Limiting load, Δm (ton/h)
		Lower bound	Upper bound		
(Sinks)					
Paper Machine I	100	0.85	0.95	0.382	38.209
Paper Machine II	40	0.90	0.95	0.536	21.438
(Sources)					
Process Fibre	90		0.88	0.469	42.226
Broke	60		0.75	0.182	10.927
Fresh fibre	To be determined		0.95	0.738	Nil

Table 7. Interval material balance table for Example 2

Column no.	1	2	3	4	5	6
Level, k	Operator ψ_k	$\Delta\psi$	$\sum_j F_j$ (ton/h)	$\sum_i F_i$ (ton/h)	$F_{\text{sum}, k}$ (ton/h)	Net material surplus/deficit
1	0.738					
2	0.536	0.202				
3	0.469	0.067				
4	0.382	0.087				
5	0.182	0.200				
6	0.000	0.182				
			-40		-40	Deficit
				90	90	Surplus
			-100		-100	Deficit
				60	60	Surplus

Table 8. The Property Cascade Table for papermaking process

Level, k	Operator ψ_k	$\sum_j F_j$ (ton/h)	$\sum_i F_i$ (ton/h)	$F_{\text{sum}, k}$ (ton/h)	F_c (ton/h)	Property load surplus (kg.atm ^{1.44} /s)	Cumulative property load surplus (kg.atm ^{1.44} /s)
					$F_F = 14.95$		
1	0.738				14.95	3.02	
2	0.536	-40		-40	-25.05	-1.67	3.02
3	0.469		90	90	64.95	5.66	1.35
4	0.382	-100		-100	-35.05	-7.01	7.01
5	0.182		60	60	$F_D = 24.95$	4.54	0
6	0.000						4.54