

PROCESS DESIGN AND CONTROL

Ecological and Economic Objective Functions for Screening in Integrated Development of Fine Chemical Processes. 1. Flexible and Expandable Framework Using Indices

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A general procedure for ecological and economic assessment during process design is presented. General objective functions to be used in the integrated development of fine chemical processes are defined. These are intended to accompany and guide process design from the very early design stages onward. The method uses three types of indices all based on simple mass balances. Mass balances are set up during the development, continually incorporating increasing complexity and knowledge. Correspondingly, mass-balance regions are extended from reaction to downstream processing and to waste treatment. Mass balances are systematically extended for multistep reactions. Mass-loss indices (MLI) are combined with ecological and economic weighing factors to yield environmental indices (EI) and economic indices (CI). Environmental indices are based on ABC-classification schemes related to environmental laws and regulations. Economic indices rely on raw material and waste treatment costs and may also include equipment and operating costs. These screening indices support condensed and lucid presentation and interpretation of mass-balance data and can be used as objective functions for the integrated development of chemical processes especially during the early design stage. Process examples are given in part 2 of this paper.

Introduction

Design Methods. Design methods in all development phases require assessment criteria and methods to characterize the performance of various process alternatives for comparison and to identify critical process steps. A variety of methods are available for quick economic and safety assessment (Peters and Timmerhaus, 1991; Keller et al., 1997).

The ecological assessment is still established the least. The most interesting but not well-enough-developed method for this purpose is life cycle analysis (LCA). Various groups have realized that the application of LCA in design of either products or processes needs special screening and streamlined methods (Schmidt et al., 1993; Schmidt-Bleek, 1993; Bretz and Fankhauser, 1996; Weidenhaupt and Hungerbühler, 1997). Particularly in the early design stages where most influence on the final process design is possible, most of the data necessary are not available for a detailed LCA. Mallick

et al. (1996) recently presented a pollution reduction methodology using indices similar to those developed in this study but particularly suited for pollution reduction in combination with process simulation. They used two methods of ranking human health and environmental effects.

Sheldon (1994) defined an environmental quotient, $EQ = E \times Q$. Sheldon called E the environmental factor which characterizes the amount of waste produced per unit amount of product. Q characterizes the environmental unfriendliness of the produced waste. Ideally, the value of E is 0 and increases with increasing formation of coupled products and byproducts as well as with increasing loss of solvents and other auxiliary materials. Typical values for the E factor range from about 0.1 in oil refining to over 100 in pharmaceuticals. E is determined from mass balances. It is almost impossible to characterize the whole waste produced by one single factor Q . Additionally, Q is a function of waste material allocation and various operational parameters. Our paper is essentially presenting a methodology to derive equivalents for E and Q in fine chemical processes during their development.

Process Development. Process development for the manufacturing of a specified fine chemical product starts with the definition of a new project, usually for the manufacturing of a new product or less frequently

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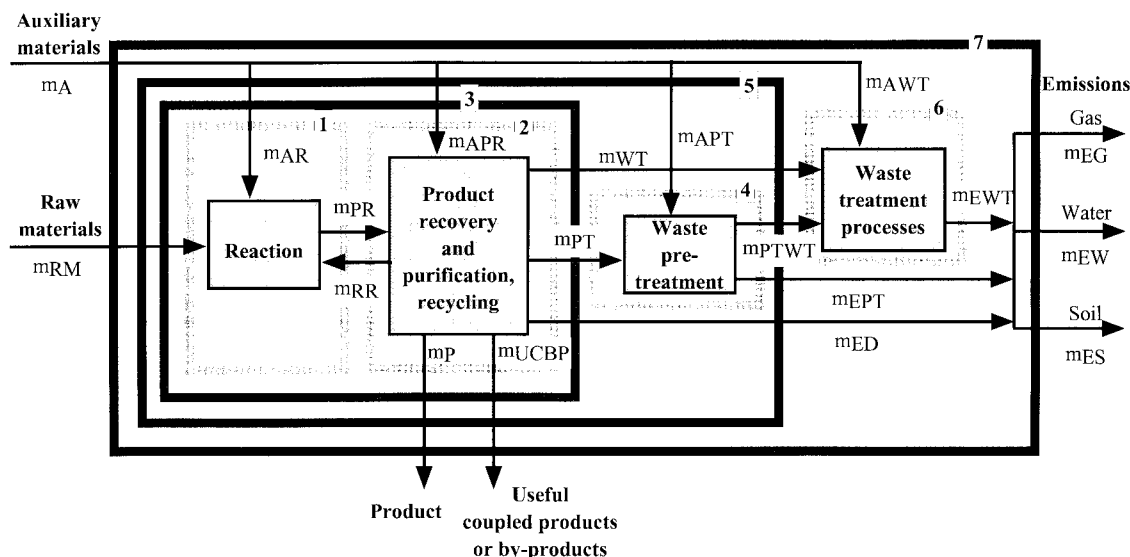


Figure 1. Simplified mass-balance regions of chemical production.

for retrofitting an existing process. In the case of a new process the available data are usually very limited and characterized by high degree of uncertainty at the beginning. After a new project is started, the chemical synthesis route is defined first. For the synthesis of modern complex chemicals for biological applications (food, pharmaceuticals, and agriculture) reactions have to be very selective with various respects (chemo-, diastereo-, enantio-, regio-, stereoselectivity). As a consequence, synthesis routes are often complex with multiple steps. For pharmaceutical products the synthesis route is usually fixed very early because of registration procedures carried out with governmental authorities (e.g., FDA). The choice of a particular synthesis route is most important for the environmental performance of a resulting technical process. Coupled products are inherently bound to a particular synthesis route. Besides required substrates, usually also solvents and other auxiliary materials are already selected at this point. Basic steps of synthesis as well as separation are defined. At this point main goals are the yield and product quality. After the definition of the chemistry, reaction conditions and the basic layout of a future process are defined. This includes the process scheme with reaction and separation steps. In this step pilot experiments may be necessary. In a next step scale-up is made, reaction and separation processes are assigned to distinct devices, and all the detailed engineering work is completed. After implementation, production starts and usually there is a learning phase during which the process is further optimized as far as what is permitted from regulatory authorities.

In the development procedure, time is usually a critical factor. Prolonged development time reduces time for production under patent protection. An ideal development procedure would, therefore, involve fast computer simulation and data generation as far as possible. Presently there are, however, only few simulation tools available which are generally suitable for batch process development. As an example, a group at MIT created the so-called Batch Design Kit which supports development and design of batch processes for the manufacturing of fine chemicals and pharmaceuticals (Linniger et al., 1995; 1996) and is now available from Hypotech. AspenTech is offering a related tool

called BATCH PLUS. These tools are, however, only of limited use for the early process design phase. Therefore, new computer methods as well as optimized laboratory techniques are needed. For the assessment of process alternatives suitable design criteria have to be defined, particularly for environmental characteristics. This type of assessment in early phases requires simplified methods.

Design Problem. Most of the costs and environmental burdens are fixed in early stages of development when the information is usually rather sparse and uncertain (Züst, 1996). Industry-producing electrical equipment found that 70% of the final costs were already determined during the development phase. The development process itself, however, contributed only 5% to the total costs (Phoenix, 1997). It seems, therefore, of utmost importance that in this phase any useful information is included in the design and assessment. As this has to be done under the pressure of time, a simple and flexible methodology, but yet scientifically based, is required to accomplish this problem. We will approach the assessment problem using a flexible and expandable framework with indices for mass intensity, for costs, and for environmental load. This framework will also suit redesign problems, especially when considering drastic changes.

As with all design problems and with any future prediction in complex systems, there is always uncertainty. It is relatively trivial to identify most important factors of an existing plant, where everything is known, but this is always a historical process. In the design process we can never be sure whether we know all important data and interactions. There are two basic ways to validate our assessment methodology, by the incorporation of experience from case studies and by careful reasoning.

System Boundaries and Mass Balances. Fine chemical processes can be simplified as shown in Figure 1. The reaction step, being the heart of any chemical process, is usually followed by a downstream section to recover and purify the desired product. Material other than the product is either recycled or treated such that the residuals can be released to the environment in an acceptable form.

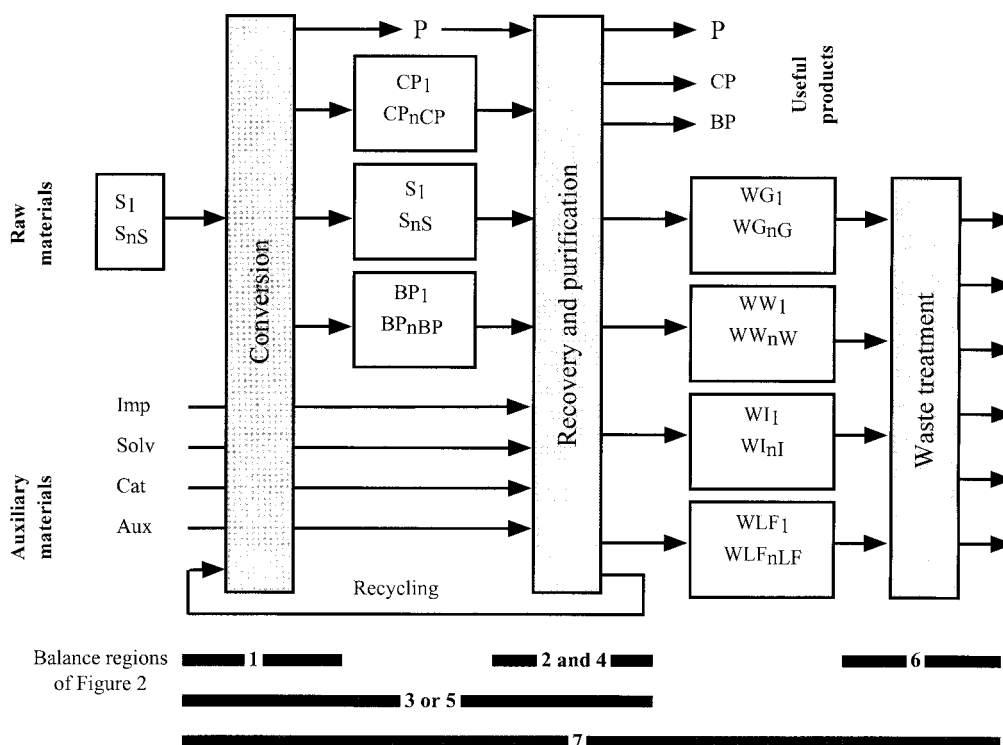
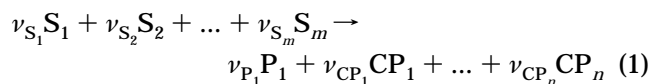


Figure 2. Material conversion and flow allocation to waste treatment systems. S: substrate. Imp: impurity. Cat: catalyst. Solv: solvent. Aux: other auxiliary material. P: product. CP: coupled product. BP: byproduct. WG: waste gas. WW: wastewater. WI: waste for incineration. WLF: waste for landfill. *n*: indicates the number of the corresponding entity.

A single reaction is characterized by



S_1 – S_m are the substrates of this reaction. If P_1 is the desired product, CP_1 – CP_n are coupled products and are unavoidably produced by this reaction. ν_i are the stoichiometric coefficients. The amount of the inherently produced coupled products cannot be changed by any means of process optimization. Only the selection of another reaction can change it.

In real systems side reactions will occur producing byproducts (Figure 2). Part of the substrate is usually not converted. Impurities contained in the substrate are usually directly transferred to the downstream recovery and purification system without reacting with any species. The same happens with catalysts, solvents, and other auxiliary materials. The recovery and purification system may consist of several units which are all lumped together in the present analysis. In this system additional materials are needed, and everything which is not recycled and which is not contained in the desired final product, recovered coupled products, or byproducts is waste. This waste can usually not directly be released to the environment. All factories now operate general waste treatment systems (e.g., biological wastewater treatment or waste incineration). Streams entering such systems often need pretreatment to be acceptable (e.g., neutralization or precipitation). After waste gas treatment (incineration, biofilter, absorber), wastewater treatment (oxidation, biological treatment), and incineration, only less harmful materials (CO_2 , mineral salts, water) are released. Some inorganic material needs landfill.

Several balance regions are defined in Figure 1: 1, reactor; 2, product recovery and purification; 3, reactor including product recovery and purification; 4, waste pretreatment; 5, reactor including product recovery and purification and waste pretreatment; 6, standard waste treatment systems; 7, whole system including reactor, product recovery and purification, waste pretreatment, and final standard waste treatment processes. Balance region 2 includes all processes for internal recycling of materials (e.g., solvents). Only waste pretreatment processes dedicated to the particular production process are in region 4 of Figure 1. For any balance region at steady state, the mass entering the region equals the mass leaving it. Assuming that neither product, nor coupled product, nor byproduct are recycled to the reactor, the mass balance around the reactor is

$$m_{1,in} = m_{RM} + m_{AR} + m_{RR} = m_{RM,S} + m_{RM,Imp} + m_{RM,Cat} + m_{RM,Solv} + m_{AR,Aux} + m_{RR,S} + m_{RR,Imp} + m_{RR,Cat} + m_{RR,Solv} + m_{RR,Aux}$$

$$m_{1,out} = m_{PR} = m_{PR,P} + m_{PR,CP} + m_{PR,BP} + m_{PR,S} + m_{PR,Imp} + m_{PR,Cat} + m_{PR,Solv} + m_{PR,Aux} \quad (2)$$

where m is the mass flow rate ($kg\ s^{-1}$). The various materials and their flows are also depicted in Figures 2 and 3. Indices are the following: 1, region 1 of Figure 1 (reactor); in, reactor inlet; out, outlet; RM, raw material for reaction; AR, auxiliary material supply to the reactor; RR, recycle to the reactor; PR, reactor outlet; S, substrate; Imp, impurity; Cat, catalyst; Solv, solvent; Aux, other auxiliary material; P, product; CP, coupled product; BP, byproduct. Usually, there is only one stream out of the reactor consisting of the various components as defined by eq 2. Each stream consists of components of classes mentioned above, for example,

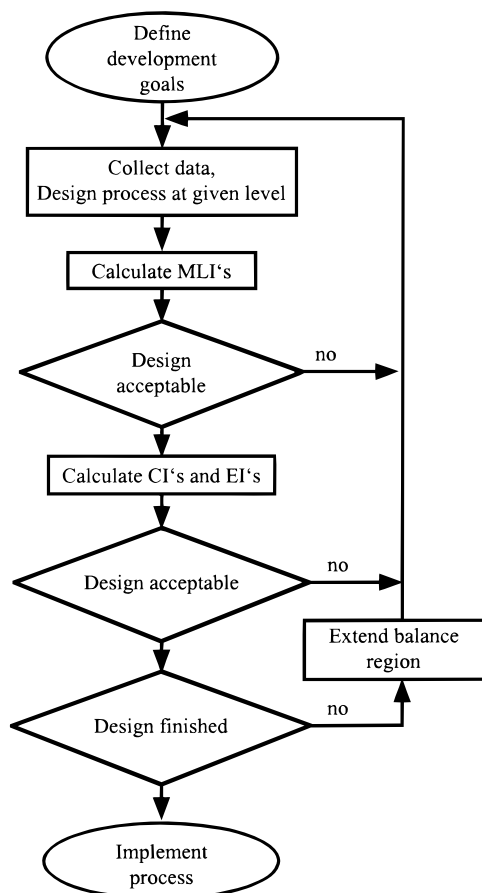


Figure 3. Proposed design and assessment procedure.

the raw material flow, m_{RM} , consists of

$$m_{RM} = m_{RM,S} + m_{RM,Imp} + m_{RM,Cat} + m_{RM,Solv} = \sum_{i=1}^{nS} m_{RM,S,i} + \sum_{i=1}^{nImp} m_{RM,Imp,i} + \sum_{i=1}^{nCat} m_{RM,Cat,i} + \sum_{i=1}^{nSolv} m_{RM,Solv,i} \quad (3)$$

Analogous to eq 2 mass balances around the reactor and product recovery and purification region (region 3 of Figure 1) and around the whole system (region 7 in Figure 1) are made. The latter would be the basis for a classical environmental assessment (e.g., life cycle analysis, LCA) considering raw material input and emissions to the environment. Such an analysis is, however, of very limited use for the design of fine chemical processes since the data basis, particularly quantitative emission data, is not available during early design stages. A complete environmental assessment needs the consideration of the fate of all compounds involved and its later impacts. This is obviously beyond the capability of an early screening assessment.

During the development process, information is first available about balance region 1 and then about regions 2 and 3. Only later are operations of region 4 defined. Therefore, indices for the design procedure are defined here for regions 1 and 3.

Definition of Mass Indices

Reaction Indices. Using eq 2 without consideration of recycle streams, m_{RR} , and relating all reactor mass

flows to the flow of product out of the reactor, $m_{PR,P}$, gives

$$\frac{m_{1,in}}{m_{PR,P}} = \frac{m_{1,out}}{m_{PR,P}} = 1 + \frac{m_{PR,CP}}{m_{PR,P}} + \frac{m_{PR,BP}}{m_{PR,P}} + \frac{m_{PR,S}}{m_{PR,P}} + \frac{m_{PR,Imp}}{m_{PR,P}} + \frac{m_{PR,Cat}}{m_{PR,P}} + \frac{m_{PR,Solv}}{m_{PR,P}} + \frac{m_{PR,Aux}}{m_{PR,P}} \quad (4)$$

Detailed definitions of the variables of eq 4 are given in Table 1. All terms in eq 4 are dimensionless relating the various mass flows to product mass flow. We define these terms as mass-loss indices, MLI_1 , where 1 indicates mass-balance region 1 in Figure 1. Rewriting eq 4, the mass index, MI_1 , for the same balance region is defined as

$$\begin{aligned} MI_1 &= \frac{m_{1,in}}{m_{PR,P}} \\ &= 1 + MLI_{1,CP} + MLI_{1,BP} + MLI_{1,S,nc} + \\ &\quad MLI_{1,Imp} + MLI_{1,Cat} + MLI_{1,Solv} + MLI_{1,Aux} \\ &= 1 + \sum_{i=1}^{nI} MLI_{1,i} \\ &= 1 + E \end{aligned} \quad (5)$$

where nI is the number of indices calculated. MI_1 can be calculated knowing the product flow out of the reactor and all reactor inputs. All indices, taking into account incomplete conversion and need for solvent and other auxiliary materials during reaction, are defined in detail in Table 1. Recycling is not taken into account in these indices. E is the environmental factor as defined by Sheldon (1994). If all component flow rates out of the reactor are known, as given in eq 5, it is also possible to allocate the origin of the various waste streams. All mass-loss indices characterize in fact reaction imperfections.

$MLI_{1,CP}$ originates from a nonideal reaction creating coupled products. $MLI_{1,CP}$ is related to the atom utilization as defined by Sheldon (1994). A simple example is the synthesis of maleic anhydride either by oxidation of benzene ($C_6H_6 + 4.5O_2 \rightarrow C_4O_3H_2 + 2CO_2 + 2H_2O$, $MLI_{1,CP} = 1.27$) or by oxidation of *cis*-2-butene ($C_4H_8 + 3O_2 \rightarrow C_4O_3H_2 + 3H_2O$, $MLI_{1,CP} = 0.55$). Side reactions produce byproducts characterized by $MLI_{1,BP}$. Incomplete conversion results in $MLI_{1,S}$. Typically, byproducts are not known in detail and are not quantified. Their mass flow can, however, also be calculated from a substrate balance since all reacting substrates which do not react in the desired way form byproducts. $MLI_{1,Imp}$ is related to the quality of raw materials used. $MLI_{1,Cat}$, $MLI_{1,Solv}$, and $MLI_{1,Aux}$ are caused by the requirement of catalysts, solvents, and other auxiliary materials. The amount of solvents has also a significant impact on space-time yield. The allocation will further help in the rational optimization of the process. In an ideal, fictive reaction with no coupled products, no byproduct formation, and 100% conversion the value of MI_1 is 1.

A more comprehensive assessment would also include environmental effects associated with the equipment used and with the energy consumed. The equipment contribution is usually negligible unless a lot of special,

Table 1. Mass Loss Indices, MLI, Defined for the Reaction/Reactor System (Region 1 of Figure 1) without Consideration of Recycling of Any Material ($m_{RR,i} = 0$)

mass index	definition	description
$MLI_{I,CP}$	$\frac{m_{PR,CP}}{m_{PR,P}} = \frac{\sum_{i=1}^{nCP} \nu_{CP,i} MM_{CP,i}}{\nu_p MM_p} = \frac{\sum_{i=1}^{nCP} m_{PR,CP,i}}{m_{PR,P}}$	formation of nCP coupled products CP_i
$MLI_{I,S}$	$\frac{m_{PR,S}}{m_{PR,P}} = \frac{\sum_{i=1}^{nS} m_{PR,S,i}}{m_{PR,P}}$	loss of nonreacted reactants S_i caused by incomplete conversion
$MLI_{I,BP}$	$\frac{m_{PR,BP}}{m_{PR,P}} = \frac{\sum_{i=1}^{nBP} m_{PR,BP,i}}{m_{PR,P}}$	formation of byproducts BP_i by other reactions
	$= \frac{\sum_{i=1}^{nS} \left(m_{RM,S,i} - m_{PR,S,i} - \frac{\nu_{S,i} MM_{S,i} m_{PR,P}}{\nu_p MM_p} \right)}{m_{PR,P}}$	calculated from substrate loss in a reaction/reactor
$MLI_{I,Imp}$	$\frac{m_{PR,Imp}}{m_{PR,P}} = \frac{\sum_{i=1}^{nImp} m_{PR,Imp,i}}{m_{PR,P}}$	impurities, Imp_i , contained in the substrates
$MLI_{I,Solv}$	$\frac{m_{PR,Solv}}{m_{PR,P}} = \frac{\sum_{i=1}^{nSolv} m_{PR,Solv,i}}{m_{PR,P}}$	solvent consumption
$MLI_{I,Cat}$	$\frac{m_{PR,Cat}}{m_{PR,P}} = \frac{\sum_{i=1}^{nCat} m_{PR,Cat,i}}{m_{PR,P}}$	catalyst consumption
$MLI_{I,Aux}$	$\frac{m_{PR,Aux}}{m_{PR,P}} = \frac{\sum_{i=1}^{nAux} m_{PR,Aux,i}}{m_{PR,P}}$	consumption of other auxiliary materials (e.g., neutralization agents)
$MLI_{I,Equip}$	$\frac{\sum_{i=1}^{nEquip} M_{I,Equip,i} \frac{t_{1,i}}{t_{Life,i}}}{M_{PR,P}}$	equipment quota for balance region 1. $M_{I,Equip,i}$: mass of equipment i . $M_{PR,P}$: mass of product produced in the reactor in a considered period. $t_{1,i}$: time of utilization of equipment i in this period. $t_{Life,i}$: total lifetime of equipment i .
$MLI_{I,Ind}$	$\frac{\sum_{i=1}^{nInd} m_{R,Ind,i}}{m_{PR,P}}$	quota of mass indirectly used for energy supply

dedicated equipment is required which is not used most of the time. The energy consumption is usually not available in early stages. Table 1 specifies an equipment index, $MLI_{I,Equip}$, and energy index, $MLI_{I,Ind}$. For the calculation of the equipment index the equipment occupation time, $t_{1,i}$, equipment mass, $M_{I,Equip,i}$, and equipment lifetime, $t_{Life,i}$, are related to the mass of product made during this period. Mass equivalents used for energy supply may be derived from data collections (e.g., ESU, 1996), provided a first guess of

energy consumption is available (e.g., 1 TJ electrical energy of the European mix corresponds to 5400 kg of material). The other indices indirectly take into account equipment use and energy consumption because higher values of mass-loss indices will generally require more complex equipment and will also consume more energy.

Indices for Reaction Including Product Recovery and Purification, Recycling, and Product Losses during These Processes. It is assumed that there is no reaction in the product recovery and puri-

Table 2. Modified Mass-Loss Indices Including Product Recovery and Purification Section as Well as Recycling of Substrate, Solvents, and Catalysts, MLI₃ (Balance Region 3 of Figure 1); Modified Mass-Loss Indices Also Including Waste Pretreatment (Balance Region 4), MLI₄; Other Indices Are Unchanged from Table 1

mass index	definition	description
MLI _{3,S}	$\frac{\sum_{i=1}^{nS} m_{PR,S,i} - \sum_{i=1}^{nS} m_{RR,S,i}}{m_{PR,P}}$	loss of nonreacted reactants S _i caused by incomplete conversion
MLI _{3,Solv}	$\frac{\sum_{i=1}^{nSolv} m_{PR,Solv,i} - \sum_{i=1}^{nSolv} m_{RR,Solv,i}}{m_{PR,P}}$	solvent consumption
MLI _{3,Cat}	$\frac{\sum_{i=1}^{nCat} m_{PR,Cat,i} - \sum_{i=1}^{nCat} m_{RR,Cat,i}}{m_{PR,P}}$	catalyst consumption
MLI _{3,Aux}	$\frac{\sum_{i=1}^{nAux} m_{AR,Aux,i} + \sum_{i=1}^{nAux} m_{APR,Aux,i}}{m_{PR,P}}$	other auxiliary materials (e.g., neutralization agents)
MLI _{3,Equip}	$\frac{\sum_{i=1}^{nEquip} M_{3,Equip,i} \frac{t_{3,i}}{t_{Life,i}}}{m_{PR,P}}$	equipment quota for balance region 3. $M_{3,Equip,i}$: mass of equipment <i>i</i> in balance region 3. $m_{PR,P}$: mass of product produced in reactor in considered time period. $t_{3,i}$: time of utilization of equipment <i>i</i> in this period.
MLI _{3,Ind}	$\frac{\sum_{i=1}^{nInd} m_{3,Ind,i}}{m_{PR,P}}$	quota of mass indirectly used (e.g., for energy supply)
MLI _{4,Aux}	$\frac{\sum_{i=1}^{nAux} m_{APT,Aux,i}}{m_P}$	other auxiliary materials used for waste pretreatment in balance region 4
MLI _{4,Equip}	$\frac{\sum_{i=1}^{nEquip} M_{4,Equip,i} \frac{t_{4,i}}{t_{Life,i}}}{M_P}$	equipment quota for balance region 4. $M_{4,Equip,i}$: mass of equipment <i>i</i> in balance region 4. M_P : mass of product produced in whole system in considered time period. $t_{4,i}$: time of utilization of equipment <i>i</i> in this period.
MLI _{4,Ind}	$\frac{\sum_{i=1}^{nInd} m_{4,Ind,i}}{m_P}$	quota of mass indirectly used (e.g., for energy supply)

fication section. This condition is not always fulfilled (e.g., if acids or bases are used during extraction processes). Corresponding modifications are then required. If there are no reactions, for each individual component of the system the mass flow in equals the mass flow out. We define

$$m_{P, Loss} = m_{WT,P} + m_{PT,P} + m_{ED,P} \quad (6)$$

which gives

$$m_P = m_{PR,P} - m_{P, Loss} \quad (7)$$

Assuming no recycling of product, coupled products, impurities, and byproducts and after substitution of all

corresponding expressions into eq 4, the modified expression is

$$m_{3,out} = m_{PR,P} + m_{PR,CP} + m_{PR,BP} + m_{PR,S} - m_{RR,S} + m_{PR,Imp} + m_{PR,Cat} - m_{RR,Cat} + m_{PR,Solv} - m_{RR,Solv} + m_{PR,Aux} + m_{APR,Aux} \quad (8)$$

The indice 3 indicates the reaction and product recovery and purification part of the system (Figure 1). Division by $m_{PR,P}$ and grouping of components give modified mass-loss indices as given in Table 2. $MI_{3,noLoss}$ is the mass index for region 3 not considering any loss of product during the product recovery and purification:

$$MI_{3,\text{noLoss}} = \frac{m_{3,\text{in}}}{m_{\text{PR,P}}} = \frac{m_{3,\text{out}}}{m_{\text{PR,P}}} = 1 + MLI_{3,\text{CP}} + MLI_{3,\text{BP}} + MLI_{3,\text{S}} + MLI_{3,\text{Imp}} + MLI_{3,\text{Cat}} + MLI_{3,\text{Solv}} + MLI_{3,\text{Aux}} \quad (9)$$

The next step is to include product loss during recovery and purification. The indices calculated according to Tables 1 and 2 are not changed. A new mass index, MI_3 , is introduced. Equation 9 is first multiplied with $m_{\text{PR,P}}$ and then divided by m_{P} , the net production rate of the product. The new mass index is therefore

$$MI_3 = MI_{3,\text{noLoss}} \frac{m_{\text{PR,P}}}{m_{\text{PR,P}} - m_{\text{P,Loss}}} \quad (10)$$

The difference between MI_3 and $MI_{3,\text{noLoss}}$ accounts for all mass losses caused by an imperfect product recovery and purification and is called $MLI_{3,\text{Loss}}$. Alternatively, MI_3 can be calculated directly by dividing mass flows to the waste treatment and directly to the environment by the amount of product actually recovered, m_{P} .

For the inclusion of the waste pretreatment section (balance region 4) MI_5 is calculated for region 5 as follows:

$$MI_5 = MI_3 + MLI_{4,\text{Aux}} \quad (11)$$

$MLI_{4,\text{Aux}}$ as well as corresponding indices for equipment aliquot and mass needed for energy supply are given in Table 2. If necessary, the latter indices are added to eq 11. For multiple reactions or synthesis steps an analogous procedure leads to stepwise and overall indices.

Definition of Ecological Indices

Mass indices give only a first rough measure of the ecological impact of a reaction system. It is desirable to weigh all streams entering as well as those leaving the system according to their environmental relevance. Environmental problems are associated with emissions and with supply of raw materials and utilities as shown in Figure 1. In actual production the streams crossing the boundary of region 7 are relevant, but these are hardly known in early development phases. Therefore, the effluents of balance regions 1, 3, and 5 of Figure 1 are used for the assessment.

We assign each compound used as well as each compound released to the downstream processes or directly to the environment an environmental weighing factor, EF_i , the estimation of which will be described later. This weighing factor is taking into account potential effects in any of the environmental compartments. Then we combine these with mass balances and relate the calculated environmental effects to the unit amount of product formed corresponding to a modification of mass indices given in Tables 1 and 2. For example, the ecological index for byproduct formation considering only region 1 of Figure 1, $EI_{1,\text{BP}}$, which originates from product losses caused by nonideal selectivity is

$$EI_{1,\text{BP}} = \frac{EF_{\text{BP}} m_{\text{PR,BP}}}{m_{\text{PR,P}}} \quad (12)$$

The major problems of such a straightforward approach are the determination of the environmental factors and the allocation of compounds to waste streams. Especially in the early design phases this is characterized by a high degree of uncertainty.

ABC Analysis for the Definition of Environmental Factors. Because of the inherently high degree of uncertainty in environmental assessment, a classification method with three classes is used (ABC analysis). This method is particularly suited to identify significant problems. A similar methodology was used by Schaltegger and Sturm (1994). First, relevant categories of problems are identified to characterize possible impacts on human health and on the environment. These categories are defined in Tables 3 and 4. Second, for each category, three classes, A, B, and C, are defined. Class A characterizes serious problems which may later on stop process development or may create large costs. Class C is noncritical and class B is somewhere in between. All classification is based on best-available present knowledge and may therefore change in the future.

All streams entering and leaving are assessed using this method. The assessment can be purely qualitative with classification of all streams into the defined classes. First, the number of class A and class B memberships of processes can be compared, similar to the method of Mak et al. (1997). Any class A membership should immediately be analyzed carefully whether it can be converted to a less critical class during further process development. If a compound is reclassified as acceptable for process development continuation, the reasons should be specified clearly (e.g., the technical problem solution expected should be defined).

Knowing the limitations for quantification and knowing that there is a high and unknown degree of uncertainty, a weighing factor, EF , was assigned to each compound based on the ABC classification. This weighing factor is obtained by multiplying the weighing factors of each category. Class C membership was assigned a value of 1. Three class C memberships still give a value of 1. For class A the category weighing factor was arbitrarily given a value of 4, amplifying strongly multiple class A memberships. The value assigned to class B membership was chosen from the difference between the case of one class A and two class C memberships compared to the case of three class B memberships. It was decided to take a ratio of a factor 2 which resulted in a value of 1.3 for a class B membership in each category. Taking a higher value between 1.3 and 1.5 reduces this ratio and therefore reduces the importance of a class A membership. Values above 1.5 make class A membership no longer dominate class B memberships. A smaller value between 1 and 1.3 makes the influences of class B memberships disappear. The resulting factor EF has 10 unequally spaced levels from 1 to 64. Examples for compounds used in the case studies of part 2 of this paper are given in Table 5.

Valuation of Feed Streams. The supply of feed streams requires exploration of raw materials from the earth, uses energy, and produces emissions during production and transport. In these supply chains environmental problems may be hidden and these are sometimes called gray inputs. An ecological assessment of input streams is useful to early discover and identify future problems (e.g., transfer of a problem to another

Table 3. Tentative ABC Environmental Classification of Input Streams

	class			refs
	A	B	C	
complexity of synthesis of raw materials used	>10 stages	3 < stages < 10	<3 stages	Weissermehl and Arpe, 1994; Keim, 1986; Witcoff and Reuben, 1996; Ullmann, 1985; Kirk and Othmer, 1991; McKetta, 1994
critical materials used for production of raw materials	critical materials like AOX, heavy metals used in stoichiometric amounts	critical materials involved in substoichiometric amounts; stoichiometric amounts of undesirable material	no critical compounds involved	Ullmann, 1985; Kirk and Othmer, 1991; McKetta, 1994
availability of raw material resources	only fossil with predicted exhaustion within 30 years	partly renewable, or with estimated deposits lasting for 30–100 years	exclusively renewable, or guaranteed long term supply (>100 years)	World Resources Institute, 1986ff; U.S. Department of the Interior, Bureau of Mines, 1970ff)

Table 4. ABC Characterization of Effluent Streams^a

	class		
	A	B	C
air pollution	organic compounds LRV I inorganic compounds LRV I and II	LRV II LRV III	LRV III LRV IV
water pollution	WGK 2,3	WGK 1	WGK 0
special problems (e.g., local issues, complexity of downstream processing); special landfill problem	materials produced unacceptable at specific location, downstream processing very complex and with unknown reliability. Not permitted for landfill.	materials produced possibly critical at specific location suitable for residual material landfill because of limited leaching potential.	no special problems foreseen no landfill required or inert material (only silicates and carbonates).

^a LRV, Swiss clean air act: (Luftreinhalte-Verordnung, LRV, 1996). Organic compounds: class I—toxic, persistent, accumulating, intensive odor; class II and III—less critical. Inorganic gases: classes I–IV. WGK: water hazard class (e.g., Hommel, 1987)—2,3 is toxic, persistent, accumulating; 1, 0 is less critical. Landfill classification from Swiss environmental act (Technische Verordnung Abfälle, TVA, 1996).

Table 5. ABC Characterization of Selected Compounds (Used in Part 2 of This Paper) Following Guidelines Given in Tables 3 and 4

substance	input				output			
	complexity of synthesis of raw materials	critical materials used for production	availability of raw material resources	EF	air pollution	water pollution	special problems	EF
acetic acid	1.00	1.00	1.30	1.30	1.30	1.30	1.00	1.69
ammonia	1.00	1.00	1.30	1.30	1.30	4.00	1.00	5.20
methylacrylate	1.30	1.30	1.30	2.20	4.00	4.00	1.00	16.00
dimethylsulfate	1.30	4.00	1.30	6.76	1.30	4.00	1.00	5.20
DMSO	1.30	1.30	1.30	2.20	1.00	1.30	1.00	1.30
2,6-di- <i>tert</i> -butylphenol	1.30	1.00	1.30	1.69	1.30	4.00	1.00	5.20
methyl-3-(4-hydroxy-3,5-di- <i>tert</i> -butylphenyl)propanoate	4.00	1.30	1.30	6.76	1.30	4.00	1.00	5.20
stearyl-3-(4-hydroxy-3,5-di- <i>tert</i> -butylphenyl)propanoate	4.00	1.30	1.30	6.76	1.30	4.00	1.00	5.20
ethanol	1.30	1.00	1.30	1.69	1.00	1.00	1.00	1.00
formaldehyde	1.00	1.00	1.30	1.30	1.30	1.30	1.00	1.69
HCl	1.00	1.30	1.30	1.69	4.00	1.30	1.00	5.20
HJ	1.00	1.00	1.00	1.00	1.30	1.30	1.00	1.69
2-propanol	1.30	1.00	1.30	1.69	1.00	1.30	1.00	1.30
lithiumamide	1.00	1.00	1.30	1.30	1.30 ^a	4.00 ^a	1.00	5.20
nitrotoluene	1.30	1.30	1.30	2.20	4.00	4.00	1.00	16.0
<i>o</i> -chlorophenol	1.30	1.00	1.30	1.69	1.30	4.00	1.00	5.20
phosphoric acid	1.00	1.00	1.30	1.30	1.00	1.00	1.00	1.00
sodium hydroxide	1.00	1.00	1.00	1.00	1.00	1.30	1.00	1.30
sodium nitrite	1.30	1.00	1.30	1.69	1.00	4.00	1.00	4.00
styrene	1.30	1.00	1.30	1.69	1.30	1.30	1.00	1.69
sulfuric acid	1.00	1.00	1.30	1.30	1.00	1.30	1.00	1.30

^a Assumes instantaneous reaction of lithiumamide with water creating LiOH and NH₃.

sphere). The quick method for the assessment of these effects proposed here uses three categories of effects as given in Table 3. It is assumed that the complexity of a synthesis is a significant indication of the degree of pollution associated with it. The number of synthesis

steps can be easily identified from textbooks or from handbooks on technical chemistry (e.g., Weissermehl and Arpe, 1994; Keim et al., 1986; Witcoff and Reuben, 1996; Ullmann, 1985; Kirk and Othmer, 1991; McKetta and Cunningham, 1976). In the second group the

occurrence of particular environmentally critical materials in significant amounts causes class A membership. Large amounts of undesirable materials or subcritical amounts of otherwise critical compounds cause class B membership. These critical compounds include AOX, dioxins, heavy metals, and carcinogens. The identification of these problems is more difficult and requires some experience and background knowledge. The information was collected from sources cited above. In the absence of information, class C membership was assumed, based on the assumption that significant problems in manufacturing were already discovered and described. In the third group the type of raw material source is characterized. The exclusive use of fossil materials with only small known deposits is considered to be particularly critical, whereas the use of renewable materials is considered to be most acceptable. Renewable materials are those harvested from the biosphere in a sustainable way. Here, materials with known deposits lasting for more than 100 years based on present forecasts are also considered renewable. Materials whose supply is guaranteed for more than 30 but less than 100 years, based on present forecasts, belong to class B (World Resources Institute, 1986ff; U.S. Department of the Interior, Bureau of Mines, 1970ff). All materials whose supply is not guaranteed for more than 30 years are considered very critical and are therefore members of class A. Examples are as follows: Class A, Cd, Hg, and Zn; class B, natural gas and crude oil.

Valuation of Output Streams. As outlined above we use output streams leaving balance regions 1, 3, or 5 of Figure 1 for effluent assessment and not the actual emissions. We decided to use the same classification methods as used for actual emissions. This can, of course, only be used within a company for internal purposes of comparison of process alternatives and for the identification of significant potential problems. The assessment can be done in three steps corresponding to balance regions 1, 3, and 5.

Three categories were defined, air pollution, water pollution, and other significant, special problems. Generally the characterization method tries to use well-accessible data already prepared for direct use. For air pollution the classification from Swiss legislation (LRV, 1996) was used. All critical components are classified into three groups. The assessment is quick and easy and is based on broad expert knowledge and social acceptance. This is also true for water pollution (Hommel, 1987). Often special problems not considered in the two previous categories have to be considered. These may be otherwise noncritical salt solutions or special problems concerning landfill (TVA, 1996). These problems are all context specific (e.g., factory location, complex mixtures which cannot be incinerated). Compounds not given in these lists are classified by comparison with similar compounds. The Swiss legislation background can easily be replaced by that of other countries.

In many cases, the assessment of regions 1 and 3 in such a way is not very meaningful because of unrealistically high penalties for compounds reliably removed by processes of regions 4 and 6 in Figure 1. Then preliminary information about the fate of these compounds in these regions may give a more realistic picture of the process. In these regions, critical compounds are

converted to less critical ones. In region 4, specific pretreatment steps make stream compositions suitable for standard treatment techniques available or even for direct release to the environment. Such pretreatment requires additional materials, characterized by m_{APT} , as well as energy. Examples of pretreatment steps are neutralization, hydrolyzation, precipitation with filtration or sedimentation, and absorption of gases. The best solution would always be to give complete mass balances of these balance regions and use total input and output streams for the assessment. Since this is usually not possible in these early phases, a more qualitative approach is suggested. If the fate of critical compounds in regions 4 and 6 can be reasonably well-predicted and if the treatment leads to significantly less harmful compounds, these predictions are used to characterize balance regions 4 and 6. This should, however, be done with great care and the assumptions for doing it should be explicitly specified and documented. In this procedure it is essential to allocate compounds to streams and streams to particular waste pretreatment techniques (e.g., gas absorption, biological wastewater treatment, landfill, incineration (Figure 2)). An example is the absorption of released SO_2 in an alkali absorber yielding SO_4^{2-} after oxidation with air. Such an absorber would be part of balance region 4, containing special waste stream treating devices. Similarly, the occurrence of a toxic but biodegradable phenolic compound could be moderately critical provided it will be submitted to a wastewater treatment plant (balance region 6) below toxic levels. A critical point in the special problems category is the landfill assessment. Organic materials are generally considered as critical. Most of the organic waste does, however, never end up in a landfill but is incinerated. Class A is therefore only used, if the organic materials are mixed with salts or metals to be deposited.

The environmental characterization using the ABC method, as used here, is not a quantitative method, although it provides numbers. The calculated relative values may be used to identify major problems. Process discrimination on this basis seems only possible, if the differences are very significant. Expert knowledge is essential to make this method reliable.

Definition of Economic Indices

A third series of indices describes some essential economic facts. Economic or cost indices (CI) are calculated first only on the basis of input streams (substrates and auxiliary materials) and later including output streams (e.g., waste treatment costs as well as equipment costs).

Raw Material Cost Indices. It is not easy to find a well-defined cost reference point which should represent the absolute minimum costs required. Here, a fictive minimum product price is defined as

$$P_{\text{P,min}} = \frac{-\sum_{i=1}^{nS} v_{\text{S},i} \text{MM}_{\text{S},i} P_{\text{S},i}}{(1 + \text{MLI}_{\text{CP}}) v_{\text{P}} \text{MM}_{\text{P}}} = \frac{\sum_{i=1}^{nS} v_{\text{S},i} \text{MM}_{\text{S},i} P_{\text{S},i}}{\sum_{i=1}^{nS} v_{\text{S},i} \text{MM}_{\text{S},i}} \quad (13)$$

Table 6. Economic Indices, CI, Defined for the Reaction/Reactor System (Region 1 of Figure 1) without Consideration of Recycling

economic index	definition	description
$CI_{1,CP}$	$\frac{\sum_{i=1}^{nCP} \nu_{CP,i} MM_{CP,i}}{\nu_p MM_p} = \frac{\sum_{i=1}^{nCP} m_{PR,CP,i}}{m_{PR,P}}$	formation of nCP coupled products
$CI_{1,S}$	$\frac{\sum_{i=1}^{nS} m_{PR,S,i} P_{S,i}}{m_{PR,P} P_{P,min}}$	loss of nonreacted reactants S_i caused by incomplete conversion
$CI_{1,BP}$	$\frac{\sum_{i=1}^{nBP} m_{PR,BP,i} P_{P,min}}{m_{PR,P} P_{P,min}}$	formation of byproducts BP_i by other reactions
	$\frac{\sum_{i=1}^{nS} \left(m_{RM,S,i} - m_{PR,S,i} - \frac{\nu_S MM_{S,i} m_{PR,P}}{\nu_{p,i} MM_p} \right) P_{S,i}}{m_{PR,P} P_{P,min}}$	calculated from substrate loss in reaction/reactor
	$\frac{\sum_{j=1}^{nBP} \sum_{i=1}^{nS} \lambda_{SR,j} \nu_{Sj,i} MM_{Sj,i} P_{Sj,i}}{M_{PR,P} P_{P,min}}$	calculation from substrates actually consumed for byproduct formation
$CI_{1,Imp}$	$\frac{\sum_{i=1}^{nImp} m_{PR,Imp,i} P_{S,i}}{m_{PR,P} P_{P,min}}$	impurities, Imp_i , contained in the substrates
$CI_{1,Solv}$	$\frac{\sum_{i=1}^{nSolv} m_{PR,Solv,i} P_{Solv,i}}{m_{PR,P} P_{P,min}}$	solvent consumption
$CI_{1,Cat}$	$\frac{\sum_{i=1}^{nCat} m_{PR,Cat,i} P_{Cat,i}}{m_{PR,P} P_{P,min}}$	catalyst consumption
$CI_{1,Aux}$	$\frac{\sum_{i=1}^{nAux} m_{PR,Aux,i} P_{Aux,i}}{m_{PR,P} P_{P,min}}$	consumption of other auxiliary materials (e.g., neutralization agents)
$CI_{1,Equip}$	$\frac{\sum_{i=1}^{nEquip} C_{1,Equip,i} \frac{t_{1,i}}{t_{Life,i}}}{M_{PR,P} P_{P,min}}$	equipment cost quota for reactor, balance region 1. $C_{1,Equip,i}$: cost of equipment i . $M_{PR,P}$: mass of product produced in the reactor in a considered period. $t_{1,i}$: time of utilization of equipment i in this period. $t_{Life,i}$: total lifetime of equipment i .
$CI_{1,Ind}$	$\frac{\sum_{i=1}^{nInd} C_{1,Ind,i}}{M_{PR,P} P_{P,min}}$	indirect costs for unit i not considered elsewhere. $C_{1,Ind,i}$ (e.g., for energy supply).

where P is the price of a raw material per unit mass. In this fictive product price only costs for the substrates used for the synthesis are concerned and 100% yield is assumed. Equation 13 actually assigns both, the product and the coupled products, the same value. This is

obviously not correct since coupled products are usually waste, even creating additional costs. It gives, however, some idea about the potential savings when finding another reaction which avoids the formation of coupled products. The corresponding index, CI, for the forma-

Table 7. Modified Economic Indices Including Product Recovery and Purification Section as Well as Recycling of Substrate, Solvents, and Catalysts, CI_3 (Balance Region 3 of Figure 1); Modified Economic Indices Also Including Waste Pretreatment (Balance Region 4), CI_4 ; Other Indices Are Unchanged from Table 6

economic index	definition	description
$CI_{3,S}$	$\frac{\sum_{i=1}^{nS} (m_{PR,S,i} - m_{RR,S,i}) P_{S,i}}{m_{PR,P} P_{P,min}}$	loss of nonreacted reactants S_i caused by incomplete conversion
$CI_{3,Solv}$	$\frac{\sum_{i=1}^{nSolv} (m_{PR,Solv,i} - m_{RR,Solv,i}) P_{Solv,i}}{m_{PR,P} P_{P,min}}$	solvent consumption
$CI_{3,Cat}$	$\frac{\sum_{i=1}^{nCat} (m_{PR,Cat,i} - m_{RR,Cat,i}) P_{Cat,i}}{m_{PR,P} P_{P,min}}$	catalyst consumption
$CI_{3,Aux}$	$\frac{\sum_{i=1}^{nAux} (m_{PR,Aux,i} + m_{APR,Aux,i}) P_{Aux,i}}{m_{PR,P} P_{P,min}}$	other auxiliary materials (e.g., neutralization agents)
$CI_{3,Equip}$	$\frac{\sum_{i=1}^{nEquip} C_{3,Equip,i} \frac{t_{3,i}}{t_{Life,i}}}{M_{PR,P} P_{P,min}}$	equipment quota for reactor and product recovery and purification, region 3
$CI_{3,Ind}$	$\frac{\sum_{i=1}^{nInd} C_{3,Ind,i}}{M_{PR,P} P_{P,min}}$	indirect costs for unit i in balance region 3 not considered elsewhere, $C_{3,Ind,i}$ (e.g., for energy supply)
$CI_{4,Aux}$	$\frac{\sum_{i=1}^{nAux} m_{APT,Aux,i} P_{Aux,i}}{m_{PR,P} P_{P,min}}$	other auxiliary materials (e.g., neutralization agents) balance region 4.
$CI_{4,Equip}$	$\frac{\sum_{i=1}^{nEquip} C_{4,Equip,i} \frac{t_{4,i}}{t_{Life,i}}}{M_{PR,P} P_{P,min}}$	equipment quota for balance region 4. These costs may also include maintenance costs as well as cost for down time. $t_{4,i}$: time of utilization of equipment i in this period.
$CI_{4,Ind}$	$\frac{\sum_{i=1}^{nInd} C_{4,Ind,i}}{M_{PR,P} P_{P,min}}$	indirect costs for unit i in balance region 4 not considered elsewhere, $C_{4,Ind,i}$ (e.g., for energy supply).

tion of coupled products is

$$CI_{1,CP} = \frac{\sum_{i=1}^{nCP} \nu_{CP,i} MM_{CP,i} P_{P,min}}{\nu_P MM_P P_{P,min}} = \frac{\sum_{i=1}^{nCP} \nu_{CP,i} MM_{CP,i}}{\nu_P MM_P} = \frac{\sum_{i=1}^{nCP} m_{PR,CP,i}}{m_{PR,P}} = MLI_{1,CP} \quad (14)$$

From this equation we can again see that the price of coupled products is assumed identical to the hypotheti-

cal minimum price of the product. One could say that this definition punishes the formation of coupled products. This is justified since the formation of coupled products is usually one of the major origins of downstream product recovery and purification costs. Coupled products are inherently bound to a particular synthesis and cannot be reduced by process optimization but only by changing the chemical synthesis.

In early phases of process development, side reactions are usually not known. A first estimation of a cost index for byproduct formation is therefore made without knowing the actual reaction stoichiometries of byproduct formation. Raw material requirements are assumed to be identical with those for the product formation. The

index for the losses due to the formation of byproducts is then

$$CI_{1,BP} = \frac{\sum_{i=1}^{nBP} m_{PR,BP,i} P_{P,min}}{m_{PR,P} P_{P,min}} = \frac{\sum_{i=1}^{nBP} m_{PR,BP,i}}{m_{PR,P}} = MLI_{1,BP} \quad (15)$$

If the total amount of byproducts formed is not known, which is the typical case, the raw material costs for byproduct formation can be calculated from the difference between total consumed substrates and substrates used for the actual production formation. Detailed definitions of cost indices are given in Table 6.

$$CI_{1,BP} = \frac{\sum_{i=1}^{nS} \left(m_{RM,S,i} - m_{PR,S,i} - \frac{\nu_{S,i} MM_{S,i} m_{PR,P}}{\nu_P MM_P} \right) P_{S,i}}{m_{PR,P} P_{P,min}} \quad (16)$$

Other economic indices are given in Tables 6 and 7. The cost of the equipment use as given in these tables is based on average costs of equipment per unit time of occupation. These costs will usually include also maintenance, labor, analytics, and equipment utilities. Other utility costs (e.g., electric and thermal energy and cooling water) are summarized in the indirect costs index, $CI_{1,Ind}$.

The total input-oriented cost index based on the reactor balance only (region 1), $CI_{1,in}$, is then

$$CI_{1,in} = 1 + CI_{1,CP} + CI_{1,BP} + CI_{1,S} + CI_{1,Imp} + CI_{1,Cat} + CI_{1,Solv} + CI_{1,Aux} = 1 + \sum_{i=1}^{nI} CI_{1,i} \quad (17)$$

The definition of the cost indices is modified when moving to balance regions 3 or 5 analogously to the mass indices and is given in Table 7. Multiple reactions are treated analogously.

Costs of Waste Treatment. Additional costs are created by the waste streams created. They usually need treatment before release to the environment. The first task is the allocation of waste materials in different waste streams (Figure 2), a task also necessary for the final ABC assessment of waste streams. Knowing volatility and solubility of system components, they can be allocated to distinct waste streams, either gases, organic liquids, water, or solids. Each waste stream created has to be characterized in terms of necessary treatment technology and corresponding costs. This is discussed in more detail in part 2 of this paper which is also presenting some case studies.

Conclusions

Summarizing, we present a general framework for assessing ecological and economic impacts in early development phases of fine chemical processes (Figure 3). The whole procedure starts with the definition of development goals. Then, a first preliminary version of the process has to be defined, which could only consist of the desired chemical reaction. Later on, side reactions and reaction auxiliaries will be included. The core of any successful assessment are clearly specified mass balances. Starting with the exclusive knowledge of the

stoichiometry of this reaction, more and more detailed mass indices (MLIs) are calculated which provide condensed information about process mass flows. In the case of severe problems the design has to be changed again. In a next step, economic and ecological indices are calculated. Identified ecological and economic problems have to be addressed by improving design. If everything looks satisfactory, design has to be refined until arriving at the final process version. This requires redesign or extension of balance regions (i.e., inclusion of product recovery and purification processes, waste pretreatment steps, and eventually waste treatment processes).

With this method, in any step of process development an assessment is possible and new available information is easily incorporated into the existing set of data as well as into data representation. The indices can be presented in graphical form which helps in reviewing process characteristics and also aids in communication with management and legal authorities. Examples of the application of the methodology are presented in part 2 of this paper together with two industrial case studies. The definition of environmental factors, EF, which characterize the ecological significance of both process input and output is a first attempt to weigh process streams according to their ecological significance. Further work is necessary to validate the definitions made here, especially about ecological assessment. Eventually, a databank of most important compounds can be created. It will be necessary that each company using such an approach extends the list with its own specific compounds and considers its own specific production environment. The attempt to allocate the source of problems from the very beginning should help to quickly focus development work to major points, allowing the most significant savings of materials, energy, and wastes. Such a screening methodology is very suitable to help designing and developing clean and economic processes without losing time and without significant additional efforts.

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Nomenclature

CI = cost index (economic index)
E = environmental factor as defined by Sheldon (1994)
 EF = environmental factor
 EI = environmental index
 EQ = environmental quotient
M = mass
m = mass flow rate
 MI = mass index
 MLI = mass loss index
 MM = molar mass
 P = product
P = price
 S = substrate
t = time
 λ = extent of reaction
 ν = stoichiometric factor

Indices

1, 2, 3, 4, 5, 6, 7 = balance regions 1–7 in Figure 1
 A = total auxiliary material input

APR = utilities for product recovery and purification processes
 APT = auxiliary material input to waste pretreatment
 AR = auxiliary supply to the reactor
 Aux = auxiliary materials
 AWT = auxiliary material input to waste treatment
 BP = byproduct
 Cat = catalyst
 CP = coupled product
 ED = direct emission to the environment
 EG = emission into gas sphere
 EPT = emissions from waste pretreatment
 Equip = equipment
 ES = emission into soil sphere
 EW = emission into water sphere
 EWT = emissions from waste treatment
 Imp = impurities contained in the substrates
 in = input
 Ind = indirect (e.g., for energy supply)
 Life = lifetime of equipment
 Loss = loss
 noLoss = loss of product during product recovery and purification not considered
 out = output
 P = product
 PR = reactor outlet
 PT = dedicated waste pretreatment process
 PTWT = stream from waste pretreatment to waste treatment
 RM = raw material
 RR = recycle to the reactor
 S = substrate
 Solv = solvent
 UCBP = utilizable coupled products and byproducts
 WT = general waste treatment process

Literature Cited

- Bretz, R.; Frankhauser, P. Screening LCA for large numbers of products. Estimation tools to fill data gaps. *Int. J. LCA* **1996**, *1*, 139–146.
- ESU (Gruppe Energie–Stoffe–Umwelt (ESU)). *Oekoinventare von Energiesystemen. Eidgenoessische Technische Hochschule, Zürich, Sektion Ganzheitliche Systemanalysen*, 3rd ed.; Bundesamt für Energiewirtschaft: Bern, 1996.
- Hommel, G. *Handbuch der gefährlichen Güter*; Springer-Verlag: Berlin, 1987.
- Keim, W.; Behr, A.; Schmitt, G. *Grundlagen der industriellen Chemie: technische Produkte und Prozesse*; Salle/Sauerländer: Frankfurt am Main, 1986.
- Keller, A.; Stark, D.; Fierz, H.; Heinzle, E.; Hungerbühler, K. Estimation of the time to maximum rate using dynamic DSC experiments. *J. Loss Prev. Process Ind.* **1997**, *10*, 31–41.
- Kirk, R. E.; Othmer, D. E. *Encyclopedia of Chemical Technology*; John Wiley & Sons: New York, 1991.
- Linninger, A. A.; Stephanopoulos, E.; Ali, S. A.; Han, C.; Stephanopoulos, G. Generation and assessment of batch processes with ecological consideration. *Comput. Chem. Eng.* **1995**, *19*, S7–S13.
- Linninger, A. A.; Stephanopoulos, E.; Ali, S. A.; Han, C.; Stephanopoulos, G. Knowledge-based validation and waste management of batch pharmaceutical process designs. *Comput. Chem. Eng.* **1996**, *20*, S1431–S1436.
- Luftreinhalte-Verordnung (LRV). *Die eidgenoessische Luftreinhalteverordnung (Dec. 16, 1985)*; SVG-Verlag: Zürich, 1986.
- Mak, C.-P.; Muehle, H.; Achini, R. Integrated solutions to environmental protection in process R&D. *Chimia* **1997**, 184–188.
- Mallik, S. K.; Cabezas, H.; Bare, J. C.; Sikdar, S. K. A pollution reduction methodology for chemical process simulators. *Ind. Eng. Chem. Res.* **1996**, *35*, 4128–4238.
- McKetta, J. J.; Cunningham, W. A. *Encyclopedia of chemical processing*; Marcel Dekker: New York, 1976.
- Peters, M. S.; Timmerhaus, K. D. *Plant design and economics for chemical engineers*; McGraw-Hill: New York, 1991.
- Phoenix. *ABB Lieferantentagung bei Phoenix Contact AG in Tagelswangen*; Press release; H. Christeler + Partner: Uster, Switzerland, 1997 (Jan 7).
- Schaltegger, S.; Sturm, A. *Oekologieorientierte Entscheidungen in Unternehmen: oekologisches Rechnungswesen statt Oekobilanzierung: Notwendigkeit, Kriterien, Konzepte*. ISBN 3-258-05039-2, Haupt, Bern, 1994.
- Schmidt, A.; Christiansen, K.; Pommer, K. *Livcyclusmodel til vurdering an nye materialer. Metoder, vurderingsgrundlag og fremgangsmåde*; (Life cycle model for the assessment of new materials. Methods, basis and procedure of assessment.) dk-Teknik: Søborg, Denmark, 1993.
- Schmidt-Bleek, F. MIPS. A universal ecological measure? *Fresenius Environ. Bull.* **1993**, *2*, 306–311.
- Sheldon, R. A. Consider the environmental quotient. *Chemtech* **1994**, Mar, 38–47.
- Technische Verordnung für Abfälle (TVA). *Systematische Sammlung des Bundesrechts SR814015*, Bern, 1996.
- Ullmann, F. *Ullmann's Encyclopedia of Industrial Chemistry*; VCH: Weinheim, 1985.
- U.S. Department of the Interior, Bureau of Mines. *Minerals Yearbook*; U.S. Government Printing Office: Washington, DC, 1970ff.
- Weidenhaupt, A.; Hungerbühler, K. Integrated product design in chemical industry. A plea for adequate life-cycle screening indicators. *Chimia* **1997**, *51*, 217–221.
- Weissermel, K.; Arpe, H.-J. *Industrielle organische Chemie: bedeutende Vor- und Zwischenprodukte*; VCH: Weinheim, 1994.
- Wittcoff, H. A.; Reuben, B. G. *Industrial organic chemicals*; John Wiley & Sons: New York, 1996.
- World Resources Institute. *World resources*; Oxford University Press: New York, 1986–1997.
- Züst, R. *Sustainable products and processes. ECO-Performance '96*; Züst, R., Caduff, G., Frei, M., Eds.; Verlag Industrielle Organisation: Zürich, 1996; pp 5–10.

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