Theory and Tools of Physical Separation/Recycling

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Materials for recycling may consist of end-of-life (EOL) product streams, byproducts and waste streams from original equipment manufacturing and the production of components, and finally also rejects, byproducts and waste streams from raw-material producers. A common feature is that all consist of compounds. The elements of the compounds can be recycled only by chemical or metallurgical means.

Recycled products can be characterized by the properties they have as a function of size. Properties can be physical or chemical in nature. They arise from the mass/area distribution of the compounds of a stream. These compounds can either be dominant in the product or form varying parts of them. As an example, a freight railroad car consists mostly of different carbon steels made by alloying three or four elements, but a mobile phone consists of a multitude of compounds made out of approximately 60 elements.

The chemical complexity of a compound as well as a metal alloy is not a function of particle size, and thus cannot be reduced by physical means. In practice, mixtures of very fine matter in a continuous matrix, such as pigments or flame retardants in plastic, also show a similar behavior, in that the complexity is not a function of particle size. Some of the most miniaturized electronic devices also belong to this group, because the required particle size is too small for practical purposes.

Most of the products manufactured are characterized by a variable scale showing a degree of particle size dependency. Some of them are complex and often contain components that fall into the first category in which complexity cannot be reduced. Good examples of this are EOL electronics or cars. At the other end of the spectrum are simple products that consist of few materials, and in which the joints between compounds (alloys) are easily breakable: for example, the freight railroad car.

Many waste streams, like metal production slag, have voluminous matrix components that cannot be reduced in complexity. These can be treated by the removal of the matrix into streams where the valuable components are concentrated.

Compounds in a recyclable stream are distributed and connected in different ways, affecting the size dependency. This is termed *liberation*. If the particle size is made finer, the liberation will change in a way that is typical to the recyclable material and method of particle reduction.

This characteristic is termed a *liberation curve*, named for the similar phenomenon in natural minerals. Particles in a recycled set will contain different mass fractions of different compounds. This is discussed later more in detail. These particles will also exhibit different physical and chemical properties that react to physical forces and the chemical environment in different ways. The chemical interactions may become complex and lead to unwanted reactive results, affecting recycling rates negatively.

This essential feature can be captured in a product-based approach to recycling, in contrast to a material-based approach. The former implicitly takes into account the whole cycle from the EOL phase to the production of renewed raw material. It also allows the interactions caused by the stream complexities to be modeling, to achieve improved recycling rates by technology and systems development. The latter is adequate for bookkeeping but does not address the effects of complexity. As an example, an error often made in material-based recycling discussions is to mix steel recycling with iron recycling. Steel forms a wide family of iron-based alloys. Many of the over 6000 alloys can be recycled together. However, for the production of recycled steel, there are limits to the content of several other metals included in the scrap, which either need to be diluted with primary material, separated away from the feed scrap stream or lost into production slag, fumes and dusts. Examples are copper and tin.

In addition to the complexity of a single product, practical recycling streams have another level of complexity arising from the collection phase where different products are collected into a combined stream. This may dilute the economic value of a stream substantially by changing the mass ratio of high-value compounds relative to low-value compounds. Because different products have different liberation curve characteristics, combining different kinds of streams will also increase the complexity of the liberation characteristics.

A complex set of properties makes recycling difficult. As an example, freight railroad car recycling does not pose difficulties, whereas mobile phone recycling does.

5.1 RECYCLING PROCESS

The collection of recyclable materials should be designed so that an unnecessary increase in stream complexity is avoided. Much can be done at the origin of the recycling process. In industry cuttings, turnings, rejects, etc., should be sorted carefully. The same applies to EOL goods from households, a much more difficult task. The optimal degree of presorting is dictated by the collection system costs and structures, location and process capabilities of treatment facilities, and economic incentives available for different actors.

After collection, the streams often tend to be too complex and unsuitable for final processing. Many valuable elements and compounds may be present at too low a value to merit the high cost of final processing. Thus, the normal step following collection is first to reduce the particle size to a more suitable finer size, and then to perform a mechanical separation step or several steps using the physical property differences between the particles in the feed stream. These streams are either further purified physically or sent for further processing by chemical or metallurgical means. Some material streams may already have reached a saleable commercial quality after physical treatment. Physical treatments range from manual sorting to sophisticated automated systems.

In all of these steps, the particle size, shape and density of individual pieces is important and affects the recycling process outcome.

5.2 PARTICLE SIZE

Any mass of material to be recycled consists of particles. The particles can be characterized by

their size. Usually the shape of particles differs from a sphere, which is the only geometric form that has a well-defined unique size, its diameter. All other geometric and irregular forms have different sizes, depending on the technique used for measurement. The most common method for scrap sizing is sieving. The particle size x_A is characteristic of an aperture through which the particle passes. The sieve surfaces are usually woven wire cloths with square apertures. In this case, the size is the side length of the square. The surface can also be made with a punched square or round hole. Another common method for measuring particle size is to measure its settling velocity in liquid or air. Then, the size is given as the size of a sphere with the same settling velocity as the particle. This is called Stokes diameter, x_s, for small particles settling at laminar velocities (Reynolds number < 0.2), or more generally, drag diameter, x_d. Particles can be illuminated by light and their projections are measured to obtain the projected area diameter, d_p. Other image-processing diameters are the Feret diameter, d_F, and the Martin diameter, d_M. They are respectively the distance between parallel tangents to the image and the cord length of the particle image measured in a defined direction. Particle volume and particle surface can also be measured to yield the volume diameter, d_v, and surface diameter, d_s, which are the diameters of a sphere with the same volume or area as the particle. For fine fractions, laser diffraction is currently the most common method. The size is the equivalent diameter of a sphere, with the same optical properties, that produces a diffraction pattern similar to the particle. It is, however, more complex than that, because the diffraction pattern measured is the total light energy falling on the sensor array and the particle size distribution is inversely computed from those data using known material optical models and diffraction theories. As the particles are suspended in a fluid and pass the measurement cell at some velocity, the measured light energy is an average over some time, and thus the obtained particle size is also a time average of the mass passed through the measurement cell.

Several average particle sizes can be used to estimate recycling system behavior. The most common is the average volume/surface diameter, the Sauter mean diameter (x_{32}). It is the diameter of a sphere that has the same volume-to-area ratio as all particles in the whole sample.

Particle sizes and their distributions are most often given in discrete classes for historical reasons, because sieving naturally creates classes. The class divisions are given usually in geometric series. The traditional series is the Tyler series, in which the subsequent size of an aperture between sieves has a ratio $\sqrt{2}$. The base size is a 74-μm sieve. This is often called the 200 mesh (200#) sieve, because there are 200 wires per inch in the woven sieve cloth. The international ISO-565/ISO 3310 standard is based on a base sieve of 1 mm and has a geometric ratio of R20/3, i.e., every third in a series of $\sqrt[20]{10}$ from 125 mm to 32 μm. Finer sizes follow R10 series ($\sqrt[10]{10}$) down to 20 μm.

Any set of particles (the sample) will have a particle size distribution. It can be expressed either as the frequency distribution $q(x)$ or as a cumulative distribution finer than size x, $Q(x)$.

$$Q_r(x) = \int_{x_{\min}}^{x_{\max}} q_r(x)dx \approx \sum_{i=1}^{n} q_{ir}(x). \qquad (5.1)$$

There are four types of particle size distributions distinguished by the subscript r in the equation above: number, length, area and volume. The most common are number and volume (mass) distribution. The convention is that the subscript r is given a value from 0 to 3 subsequently from number to volume distribution. So, $Q_3(x)$ means a cumulative volume distribution and $q_0(x)$ is a number frequency distribution.

Number distributions are important to planning sampling campaigns because statistical errors and analytical confidence limits depend on the number of particles sampled. Often,

reaction rates are related to the reactive surface and the mass balances of such reactions correlate with the mass distributions.

Real distributions can be estimated with simple mathematical distributions. All are suitable only for mono-modal distributions. The simplest representation is an exponential function called the Gates−Gaudin−Schuhmann (GGS) equation

$$Q_r(x) = \left(\frac{x}{x_0}\right)^\alpha, \qquad (5.2)$$

where x_0 is the size at which all particles are finer and α is the slope (width) of the distribution. A second widely used form of distribution is the Rosin−Rammler−Sperling−Bennett equation

$$Q_r(x) = 1 - \text{Exp} - \left\{ -\left(\frac{x}{x_n}\right)^m \right\}, \qquad (5.3)$$

which is capable of describing the ends of the distribution better than GGS. In the equation, x_n is the size at which 62.3% of particles are finer. Parameter m is the slope (width) of the distribution.

The third equation given here is the log-normal distribution

$$q_r(x) = \frac{1}{\sigma_{\ln}\sqrt{2\pi}} Exp\left\{ -\frac{1}{2}\left(\frac{\ln(x_r/x_{\mu,r})}{\ln s_g}\right)^2 \right\}, \qquad (5.4)$$

where $x_{\mu,r}$ is the median and $\ln(s_g)$ is $\ln\left(\frac{x_{84,r}}{x_{50,r}}\right)$ (x_{50} = size at which 50% is finer).

For multi-modal particle distributions, one has to combine two or more distributions. Multi-modality is often observed in recycled feeds because the material properties (for example, brittleness) in a product can vary a lot.

5.2.1 Translational Velocity of Particles

A single particle moves in a fluid medium (liquid or gas) obeying classical mechanics. Two dimensionless numbers are useful for evaluating the behavior of particles settling a fluid.

Reynolds number Re is the ratio of inertial forces to viscous forces. For particles, we can write it as

$$\text{Re} = \frac{\rho v d}{\mu}, \qquad (5.5)$$

where d is the characteristic length, i.e., particle diameter and μ the dynamic viscosity (Pa s). At low Reynolds numbers, the viscous forces dominate and the flow around the particle is smooth. A limit is considered typically to be $\text{Re} \leq 0.2$. When the Reynolds number exceeds $\text{Re} \geq 1000$, the inertial forces dominate and the fluid forms a distinct turbulent wake at the aft of the particle.

The other important dimensionless variable is the drag coefficient C_d. When a particle moves through a fluid, it must displace fluid elements from its path. This consumes energy, which can be understood as a force F_d slowing particle velocity. This drag force has two components that are important to velocities used in recycling. The first; skin friction, is caused by the fluid viscosity; and the second, form drag, is caused by the pressure difference between the fore and aft of the particle

$$C_d = F_d \frac{2}{\rho_f v^2 A}. \qquad (5.6)$$

Drag coefficient varies as a function of velocity, particle size and shape, fluid density and viscosity. Drag coefficient is a function of the Reynolds number Re.

The drag force, buoyancy and gravitational force are the main forces controlling the settling of a particle in a quiescent fluid. The acceleration of a particle is

$$m\frac{dv}{dt} = mg + mg\frac{\rho_f}{\rho_s} + \rho_f v^2 C_d A/2, \qquad (5.7)$$

where m is the mass; v, the velocity of the particles; ρ density (subscript f for fluid and s for solids); C_d, drag coefficient; and A, the area of

the particle perpendicular to the direction of movement. As the velocity increases, the drag force will also increase until the acceleration comes to zero and the particle has obtained terminal velocity.

For fine particles (typically below 60 μm for solid particles), the drag coefficient can be estimated to be $C_d = 24/Re$ (for spherical particles). For terminal velocity, this leads to the well-known Stokes equation

$$v_{St} = \frac{d^2 g(\rho_s - \rho_f)}{18\mu}. \tag{5.8}$$

For very high Reynolds numbers, the drag coefficient is essentially a constant $C_d \approx 0.44$. This leads to a terminal settling velocity equation for large particles (also known as Newton's equation)

$$v_N^2 \approx \frac{3 d_p (\rho_s - \rho_f)}{\rho_f}. \tag{5.9}$$

For intermediate sizes, no closed solutions exist. Turton and Clark (Turton and Clark, 1987) presented a useful approximation using dimensionless numbers. For dimensionless velocity v^*, they give as a function of dimensionless size d^*

$$v^* = \left[\left(\frac{18}{d^{*2}} \right)^{0.824} + \left(\frac{0.321}{d^*} \right)^{0.412} \right]^{-1.214}. \tag{5.10}$$

The dimensionless size d^* can be obtained from

$$d^* = \left(\frac{3}{4} C_d Re^2 \right)^{\left(\frac{1}{3} \right)} = d_p \left(\frac{g(\rho_s - \rho_f)\rho_f}{\eta^2} \right)^{\left(\frac{1}{3} \right)} \tag{5.11}$$

and dimensionless velocity v^* from

$$v^* = \left(\frac{4 Re}{3 C_d} \right)^{\left(\frac{1}{3} \right)} = v_s \left(\frac{\rho_f^2}{\eta g(\rho_s - \rho_f)} \right)^{\left(\frac{1}{3} \right)}. \tag{5.12}$$
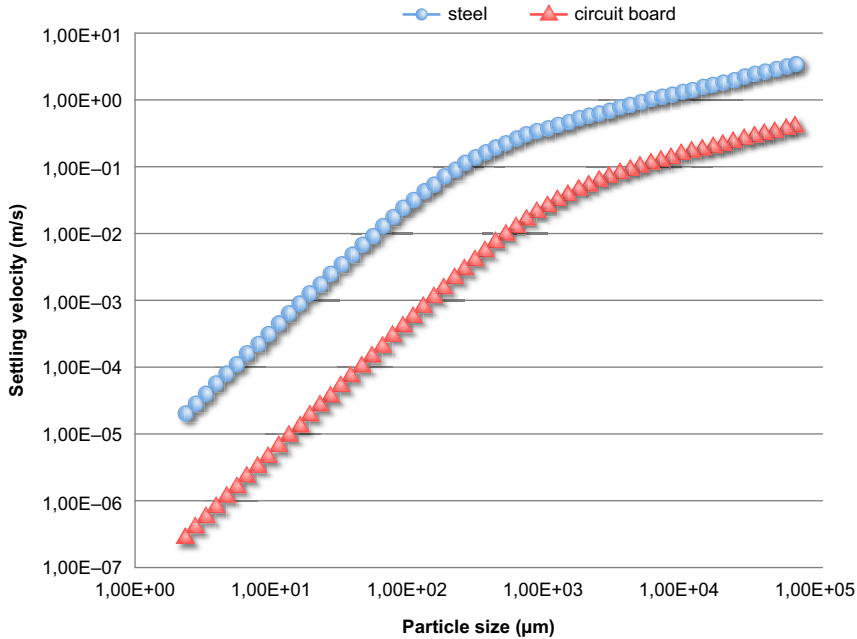

FIGURE 5.1 Particle settling.

For a known size, first calculate the dimensionless size (Eqn (5.11)) and use it to estimate dimensionless velocity (Eqn (5.10)). Then, solve the real velocity from Eqn (5.12).

The differences in settling velocity are large when comparing a piece of steel or a piece of circuit board resin, as can be seen in Figure 5.1. A resin piece with a diameter of 4.3 mm obtains a terminal velocity of 0.1 m/s. A steel particle is only 0.23 mm in diameter.

Non-spherical particles often behave in an erratic way, depending on their Reynolds number and the shape itself. In laminar conditions, the particles tend to become oriented so that the total drag force is minimized. Platy particles tend to wobble and flow in an erratic way. The drag coefficient tends to be a decade higher than for spheres of the same density and mass. Needle-shaped particles translate in a laminar flow with the longest dimension aligned with the flow.

In turbulent flow conditions, the particles tend to become oriented so that skin friction is minimized. Platy particles are moving wobbling in a position where the highest surface area is perpendicular to the flow. This causes the drag coefficient to be up to 2 decades higher than for a respective sphere. Needle-shaped particles wobble in a turbulent flow.

As a general rule of thumb, platy particles may be an order or even two orders of magnitude larger (largest dimension) to obtain the same terminal velocity.

5.3 PULP RHEOLOGY

Pulp rheology substantially affects the flow behavior of a separator. There, the most important variable is the volume concentration of solids ϕ in the suspension given by

$$\phi = \frac{\theta_p - 1}{\theta_s - 1}, \quad (5.13)$$

where specific weight $\theta_s = \rho_s / \rho_{water}$.

5.3.1 Apparent Density

In separators, where solid particles are dispersed in a fluid, the apparent specific density of the dispersion increases as

$$\theta_p = \frac{100}{\left(\frac{P}{\theta_s}\right) + 100 - P}, \quad (5.14)$$

where P is the suspension solid content percentage by mass.

5.3.2 Apparent Viscosity

Most pure fluids are Newtonian in their behavior. Any small stress will cause a shear and the fluid moves. The ratio is called viscosity. When the solids content increases in a fluid, the behavior of the fluid resembles increasing viscosity effects. Thomas (Thomas, 1965) proposed the following equation for the viscosity effects of suspended solids

$$\frac{\mu_a}{\mu_0} = 1 + 2,5\phi + A\phi^2 + B \exp(C\phi), \quad (5.15)$$

where ϕ is the solids volume concentration. Heiskanen and Laapas (Heiskanen and Laapas, 1979) proposed slightly different parameters to the equation. (Table 5.1).

The difference between the predictions is typically below 4% when ϕ is below 30% but then increases quickly, because the Thomas equation predicts substantially higher apparent viscosities at higher volume concentrations.

5.3.3 Hindered Settling

When the solids content of a fluid, notably water, increases, the translational velocity of

TABLE 5.1   Parameters for the Apparent Viscosity Equation

	A	B	C
Thomas (Thomas, 1965)	10.05	0.00273	16.6
Heiskanen and Laapas (Heiskanen and Laapas, 1979)	14.1	0.0274	−16.6

TABLE 5.2 Parameters for the Richardson and Zaki Equation

Re < 0.2	$b = 4.65$
0.2 < Re < 1.0	$b = 4.36/Re^{-0.03}$
1.0 < Re < 500	$b = 4.4/Re^{0.1}$
Re > 500	$b = 2.39$

the particles will decrease. For spherical particles, we get as an experimental equation for the ratio for hindered settling (Richardson and Zaki (Richardson and Zaki, 1954))

$$\frac{v_h}{v_{St}} = (1 - \phi)^b, \quad (5.16)$$

where b is a function of Re. (Table 5.2).

A small particle with a density of 3000 kg/m^3 in a 30% by weight (12.5% by volume) slurry has only about 55% of the free settling velocity. Increasing the solids fraction by weight to 50% reduces the settling velocity further to about 28%. For very large particles, the ratios are 72 and 50%, respectively.

To evaluate the effect of particle density in a solid suspension, the following approximate equation can be used for small particles

$$\frac{x_1}{x_2} \approx \sqrt{\frac{\theta_2 - \theta_p}{\theta_1 - \theta_p}} \quad (5.17)$$

and

$$\frac{x_1}{x_2} \approx \frac{\theta_2 - \theta_p}{\theta_1 - \theta_p} \quad (5.18)$$

for large particles.

The size ratio between a steel particle and an aluminum one increases by 12% for fine particles and 24% for coarse particles, when the specific gravity increases from 1.0 to 1.4.

5.4 PROPERTIES AND PROPERTY SPACES

Any particle in a size class can be characterized by its properties averaged over its volume or surface. Of course, this property can be simply its chemical composition, but the composition of compounds and the set of consequent physical properties are more useful. Any set of particles will have a distribution of property values (Figure 5.2). This distribution can be treated as any statistical distribution with a mean and a variance. Let us denote the particle size as **i** and the properties as **j,k**,.... The size classes can be single ISO-565 fractions or any combination deemed applicable, or any other sizes. For example, it can be too fine for processing at −4 mm, optimal for processing at 4−64 mm, and too coarse and unliberated at +64 mm.

Properties must be considered by their utility. Only those properties that are important to the separation stage at hand and the requirements of further processing need to be considered. It is advisable that there be as few property classes as possible.

For example, if low purity steel scrap is to be treated, the simplest technology is to perform magnetic separation. Because most carbon steels exhibit ferromagnetism, magnetic susceptibility can consist of two classes: with or without this property. Because steel scrap also contains elements detrimental to the steel-making process, for high-quality steels we need to add composition properties such as a fractional content of copper in, say, 10%-unit steps (in any single particle). However, this approach quickly leads to a high number of combinations of properties.

The property space shows a way to model recycling (Figure 5.3). Models that track the changes in numbers of particles between the different volumes of property space are called population models. The simplest space is a one-dimensional binomial space of similar-size white and red beads; a description of a mobile phone after complete shredding will need an N-dimensional space with a large number of classes for every property, which is impossible to model. However, these
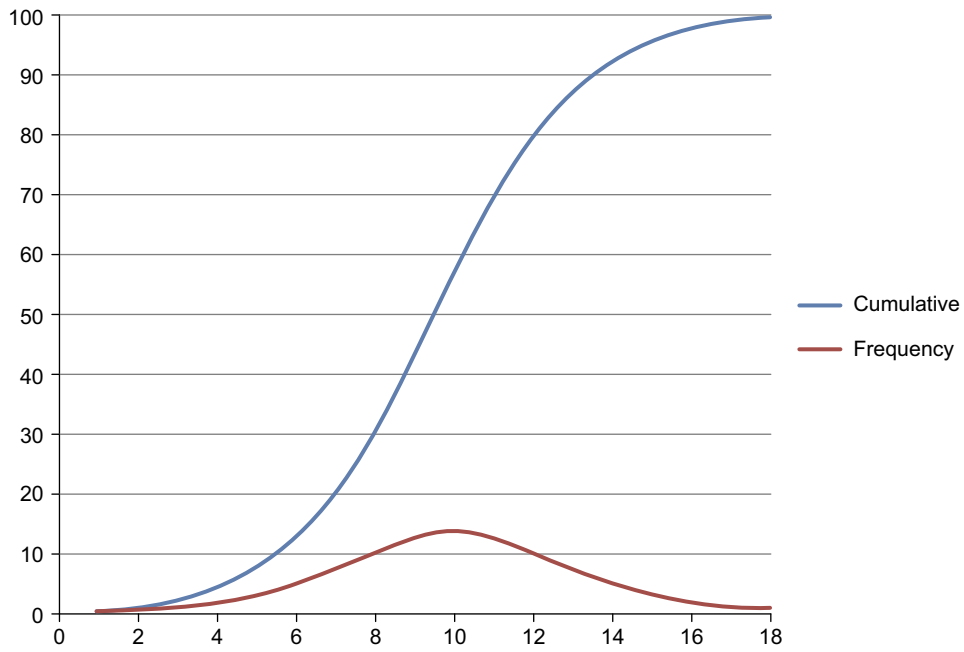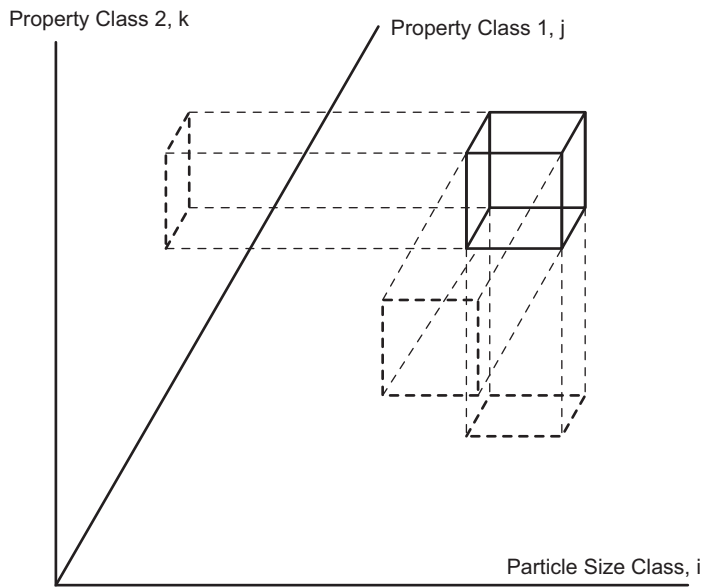
FIGURE 5.2 Property distribution.

FIGURE 5.3 A three-dimensional property space R with a volume of particles belonging a class $R_{i,j,k}$ (with a surface area of S_c).

population models can still be useful in modeling size reduction and separation in recycling.

Particles can move to and from the property class volume by several mechanisms. First, they can move through the boundary by a convective motion. This can be by mechanical abrasion, by chemical reactions starting to take place as the temperature or chemical environment changes, by melting, or by any such process. They can also move by finite steps as a result of breakage or agglomeration. They can be destroyed in one volume for the progeny particles to arrive into several others. There can also be physical additions and withdrawals of particles into and from a given property volume.

$$\frac{\partial}{\partial t} \int_{R_c} N\Phi(x)dx = -\int_{S_c} N\Phi\mathbf{u}\,\mathbf{n}\,d\theta - D \\ + B - Q + A, \tag{5.19}$$

where N is the number of particles, $\Phi(x)$ is the frequency size distribution of particles, $\mathbf{u}$ is a vector describing the rate of particles passing the boundary S_c, D is the disappearance and B is the birth of particles resulting from some physical or chemical action, and Q and A are the removal and addition, respectively, of the particles' byproduct and feed streams.

For many practical applications, the number distribution can be substituted with a mass distribution.

One way to use the property space for mass balancing and data reconciliation is to define for each stream f a flow rate Q_{fj} of a phase j (property class 1). Each phase consists of components k (property class 2). We can define $\mathbf{P_{kjf}}$ to be the fraction of component k in phase j in stream f. We can define X_{ijf} to be the fraction weight of particle size class 1 of phase j in stream f. As the last definition, we have T_{kijf} as the fraction of component k in particle size class i of phase j in stream f.

For many technical purposes, a one-dimensional cut from the mass-based property space, i.e., a frequency distribution or histogram of a single property mass, is interesting, as will be discussed next.

5.5 SAMPLING

Properties described earlier can be treated as property distributions with a mean and standard deviation. The variability is always a function of particle numbers sampled, not mass. There are properties that are integrative, such as the chemical composition. They are independent of particle size and can therefore be shredded and comminuted to finer sizes to increase the number of particles sampled. There are also properties that are size dependent, such as specific surface area of the material.

Even if the unknown real distribution is skewed, the sampled distribution tends to be closer to a normal distribution. It is usually assumed that the sampled distribution of the mean is normally distributed. Then, the limit clause

$$\mathrm{var}(\overline{x}) = \frac{\mathrm{var}(x)}{n} \tag{5.20}$$

stipulates that the variance of the mean is an nth fraction of the variance of a single measurement. It also says that doubling the sample size will reduce the variance to half. For a small number of samples (<30), the distribution follows Student t distribution, which is a wider distribution than the normal distribution because of the uncertainty in estimating the standard deviation.

Gy (Gy, 1979) developed a sampling theory that is in general use. The variance that is caused by the inhomogeneity of the material itself is called the fundamental variance. This error will remain even when the sampling is performed in an ideal way. The fundamental variance is related to the third power of the largest particles present in the sampled material

$$\text{var}(x_{\text{fundam}}) = \frac{Cx_{95}^3}{n\Delta m}, \qquad (5.21)$$

where x_{95} is the size at which 95% are finer, Δm is the increment size, n is the number of increments, and C is a constant depending on the property distribution, liberation, particle shape and width of the size distribution. As can be seen in Eqn (5.21), a reduction in size of the largest pieces reduces the fundamental variance rapidly.

There are several sources for error in performing the sampling. The total variance of sampling consists of the fundamental variance and variances of error taking place in assaying and sample selection owing to wrong delimitation of the sample (for example, loss of material from increment) and owing to integration errors caused by the discrete sampling of a continuous variability.

$$\text{var}(z) = \text{var}(\text{fundamental}) + \text{var}(\text{assaying})$$
$$+ \sum \text{var}(\text{sampleselection})$$
$$(5.22)$$

The dimension of sampling can be defined as the spatial directions of a sampler to obtain a representative sample. A one-dimensional example is a material stream falling from a transport belt by a sampler that traverses it. A three-dimensional example is a heap of material, in which sampling points need to be distributed in three dimensions, a practical impossibility.

Materials for recycling can be sampled by

- Random sampling
- Systematic sampling
- Stratified sampling.

These can be also performed as a two-stage process or as a sequential process. Random sampling is discouraged in most cases because true randomness is difficult to obtain. This is especially the case for three-dimensional examples. A random grab sample will have a high variance and will often be prone to errors caused

by nonideal sampling. The most accurate sampling method is systematic sampling from a one-dimensional case, i.e., an automatic sampler sampling a moving stream of material. This also applies to all secondary sampling before assaying.

If the material has a tendency to segregate, one can try stratified sampling, in which different strata of the material are sampled separately and the result is obtained by weighing the results by stratum masses.

For cheap materials, sampling can be performed using a two-stage process, in which the first sampling may be, for example, first selecting randomly the wagons (m) from a train of M wagons to be sampled in more detail

$$s_{\text{ts}}^2 = \left(\frac{M-m}{M-1}\right)\frac{\sigma_{\text{b}}^2}{m} + \frac{\sigma_{\text{w}}^2}{mn}, \qquad (5.23)$$

where σ_{b} is the standard deviation between wagons, and σ_{w}, within wagons.

A sequential process is often used for quality standards. If the first set is clearly within or outside defined limits, the sampling is discontinued and the lot is either accepted or rejected. If the result falls between the defined limits, sampling is continued with a second set.

For a variable (y) computed from a set of measured variables as $y = f(x_1, x_2, ..., x_n)$, the variance is computed as follows

$$\text{var}(y) = \left(\frac{\partial y}{\partial x_1}\right)^2 \text{var}(x_1) + \left(\frac{\partial y}{\partial x_2}\right)^2 \text{var}(x_2) + ...$$
$$+ \left(\frac{\partial y}{\partial x_n}\right)^2 \text{var}(x_n).$$
$$(5.24)$$

5.6 MASS BALANCES AND PROCESS DYNAMICS

Recycling can also be understood as a materials handling operation, where material is

transported, concentrated and purified during the treatment. The process always consists of the units handling the material with temporal holdups, units of moving the material, and storage units. They all are combined by an average flow of material.

5.6.1 Mass Balances

Mass balances can be written either over a single unit in the process or over larger parts of the process. In mechanical recycling, one often starts with steady-state mass balances, where the recycled mass and its constituents are assumed to be constant in any flow.

As will be discussed later, these equations never hold completely, but contain random sampling and assay errors, and therefore do not close completely and will require data reconciliation.

For a separation as in Figure 5.4, recovery of material with the property of interest can be calculated from the mass balance

$$R_a = \frac{Cc_{ac}}{Fc_{af}} = \frac{a_{af}(a_{ac} - c_{at})}{c_{ac}(c_{af} - c_{at})}.$$ (5.26)

For a two-product case (Figure 5.5), in which properties a and b are divided to both C and S product streams, the recoveries become

$$R_a = \frac{c_{ac}[(c_{af} - c_{as})(c_{bs} - c_{bt}) - (c_{bf} - c_{bs})(c_{as} - c_{at})]}{c_{af}[(c_{ac} - c_{as})(c_{bs} - c_{bt}) - (c_{ac} - c_{bs})(c_{as} - c_{at})]}$$

$$R_b = \frac{c_{bs}[(c_{ac} - c_{af})(c_{af} - c_{bt}) - (c_{bc} - c_{bf})(c_{as} - c_{at})]}{c_{bf}[(c_{ac} - c_{as})(c_{bs} - c_{bt}) - (c_{ac} - c_{bs})(c_{as} - c_{at})]}.$$ (5.27)

$$F = C + T$$
$$Fc_{af} = Cc_{ac} + Tc_{at}$$
$$Fb_{bf} = Cc_{bc} + Tc_{bt} \quad\quad (5.25)$$
$$\ldots$$
$$Fm_{mf} = Cc_{mc} + Tc_{mt},$$

where capital letters denote the total mass flow of feed (F), product (C), and tails (T), and c_{af}, c_{ac}, c_{at}, c_{bf}, c_{bc}, c_{bt},...,c_{mf}, c_{mc}, and c_{mt} are the concentrations of the property of interest ($a,b,...m$) in streams f, c, and t, respectively.
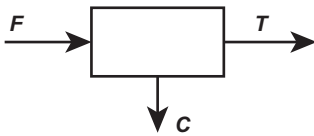
For dynamic situations, where a property is changing in time, we have to use dynamic mass balance equations. Examples are shredding, smelting, and leaching.

$$\frac{d}{dt}(Vc_a) = Fc_{af} - Pc_{ap} - Vr_a$$ (5.28)

$$\frac{d}{dt}(Vc_g) = Fc_{gf} - Pc_{gp} - Vr_g,$$

where V is the volume of reacting space, F is the feed, P is the product, and r is the reaction rate. These equations can be written in state-space
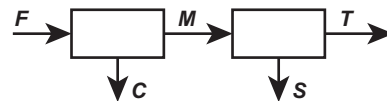

FIGURE 5.4 A simple separator.

FIGURE 5.5 A two-stream separation case.

notation. If the reaction between a and g is of first order $r = kc_a$, we get

$$\dot{X}(t) = \mathbf{A}(t)\mathbf{x}(t) + \mathbf{B}(t)\mathbf{u}(t)$$

$$\mathbf{x} = \begin{bmatrix} c_a \\ c_g \end{bmatrix}, \mathbf{A} = \begin{bmatrix} -\left(k + \dfrac{1}{\tau}\right) & 0 \\ k & -\dfrac{1}{\tau} \end{bmatrix},$$

$$\mathbf{u} = \frac{1}{\tau}\begin{bmatrix} c_{af} \\ c_{gf} \end{bmatrix}, \mathbf{B} = \mathbf{I}. \qquad (5.29)$$

Equation (5.28) can be used as a starting point for population balance modeling of recycling.

5.6.2 Process Dynamics

The holdup or storage variation (mass stored W) can be expressed as the difference between incoming and outgoing flows in the ideal case.

$$\frac{dW(t)}{dt} = Q_i(t) - Q_o(t). \qquad (5.30)$$

The change in buildup is mathematically an integrating process, but for our needs the important point is that it takes time to change the inventory.

If material is transported a given distance L at a velocity v, it will show a transportation lag $\tau = L/v$. For a property $p(t)$ entering the transport system at time $t = 0$, we can write

$$p_{out}(t) = p_{in}(t - \tau). \qquad (5.31)$$

In Laplace domain[1] the transfer function of this is

$$G(s) = e^{-\Delta ts}. \qquad (5.32)$$

This is a transfer function of a pure time delay.

In recycling, we can use the well-known limiting cases. In the first case, the reactor is not mixed (plug flow reactor) (Eqn (5.32)); in the second case, it is instantaneously fully

mixed (an ideal [fast] reactor). For the fully mixed case, we have

$$G(s) = \frac{1}{\tau s + 1}. \qquad (5.33)$$

If such a mixer is disturbed by a change in the feed composition, we can multiply the transfer function with the Laplace transform of the disturbance to obtain the response. By performing the inverse Laplace transform back to time space, we get the response in time. If the property entering the fully mixed reactor is a step change with a Laplace transform of $(1/s)$, we get for the response

$$C(s) = \frac{1}{s}G(s) = \frac{1}{s}\frac{1}{\tau s + 1} => C(t) = 1 - e^{-t/\tau}. \qquad (5.34)$$

For n reactors in series with a constant residence time in all of them

$$G(s) = \frac{1}{(\tau s + 1)^n}. \qquad (5.35)$$

By using these simple components, we can predict the dynamic response of a flowsheet (Figure 5.6) by first constructing a signal flow diagram (Figure 5.7).

By summing all of the transport lags and taking into consideration only the two largest time constants (the third has only a minor effect on the dynamic response), we get the following Figure 5.8. Using the combined notation of the figure, the variation in the product $p(s)$ related to the required quality set point $q(s)$ is

$$p(s) = \frac{G_X(s)G_Y(s)}{1 + G_X(s)G_Y(s)G_M(s)}q(s). \qquad (5.36)$$

In designing the control circuit in Figure 5.8, one has to ensure that the system is stable and that the control result is adequate. The overall stability of the control circuit can be answered by solving the function $1 + G_X(s)$ $G_Y(s)G_M(s) = 0$. The roots must be negative or have negative real parts. For a constant set point, the dynamic response of

---

[1] Laplace transform $F(s) = \displaystyle\int_0^\infty f(t)e^{-st}dt.$
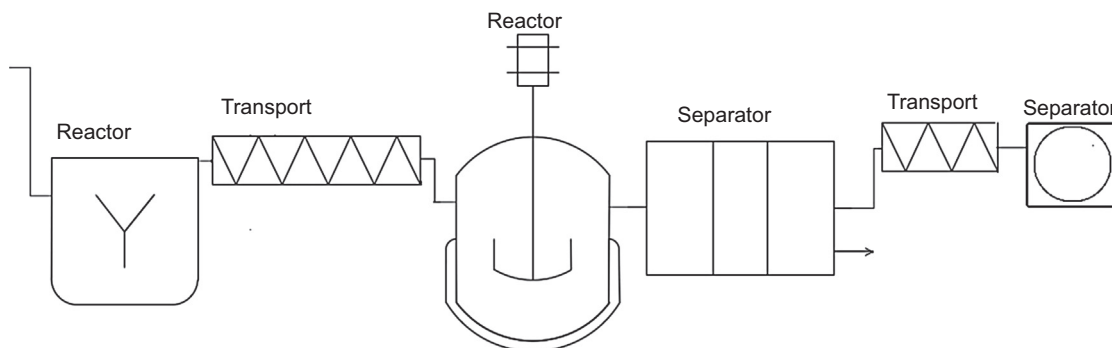
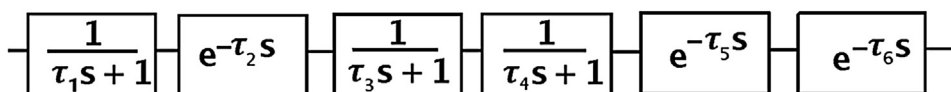FIGURE 5.6 A recycling flowsheet.
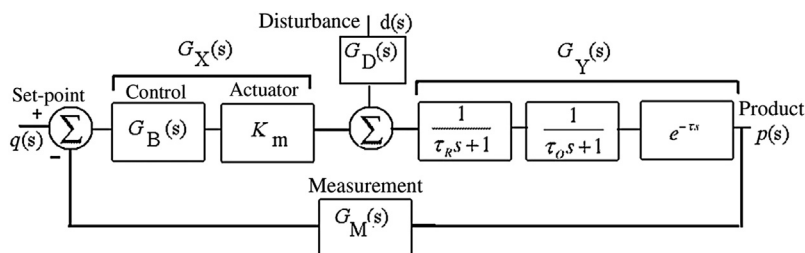

FIGURE 5.7 Flowsheet as a signal flow diagram.

FIGURE 5.8 Signal flow diagram for feedback control for product quality.

(product/disturbance) is defined by the open-loop transfer function $G_Y(s)G_X(s)G_M(s)$.

5.7 MATERIAL BALANCING

5.7.1 Linear Data Reconciliation

A prerequisite for meaningful recycling computations is to perform data reconciliation, which allows consistent and closed process balances to be obtained. This allows one to generate a good understanding of the operation and its trends for further process improvement. Closed balances are also needed for process accounting and performance estimates. The aim is obtain consistent estimates for recycling process variables subject to model constraints.

A recycling process can be thought as a network of nodes connected to perform the recycling task at hand.

The node behavior can be expressed by constraining state equations, which need to be fulfilled when performing data reconciliation. For the balance of the process, we can write in matrix form

$$\mathbf{Cx} = 0, \tag{5.37}$$

where $\mathbf{C}$ is the constraint equation matrix (with elements c) and $\mathbf{x}$ is the vector of flows connecting the units.

For this, we need a set of measurements of process variables such as particle size, material and chemical composition, and so forth, to write a constraining model for any single node. These are typically conservation equations. Nodes can consist of units combining or separating streams (physical separators), and reactors (chemical and metallurgical reactors and furnaces). The two first kinds are characterized by the conservation of all variables; the reactors always conserve the total mass and the masses of elements but may not conserve other variables.

We also need an estimate of the uncertainty of the measured variables. An assumption made in the reconciliation process and in formulating the previous equation is that the errors involved are not gross errors (bias) but are always randomly distributed.

It is typical that variables are measured only from some of the streams. Thus, we have measured and unmeasured variables. Some can also be calculated from information obtained from other streams. If a variable is not measured but can be calculated, it is called observable. Of course, if we have no way to obtain the value of a variable if it is unobservable.

called topological. It considers measurements from a stream. If the size distribution of all three streams of a size separator is measured, all measurements are redundant because we can compute the values of one stream from the two others and also have a direct measurement. If one of the measurements is not performed, the remaining stream variables become nonredundant. One of the streams is then nonmeasured but observable. If a second stream measurement is omitted, the two streams not measured become unobservable. A nonredundant variable becomes unobservable if its measurement fails. The second kind of redundancy arises from repeated measurements. This is important because it gives information about the standard deviation of the variable.

Estimability is a slightly broader definition than observability, which is reserved only for nonmeasured variables. A variable is estimable if it is measured or nonmeasured but observed.

A typical flowsheet in recycling is depicted in Figure 5.9.

For simple material flow constraints, we get for matrix **C**:

$$
C = \begin{array}{c}
\begin{array}{cccccccccccccc} S_1 & S_2 & S_3 & S_4 & S_5 & S_6 & S_7 & S_8 & S_9 & S_{10} & S_{11} & S_{12} & S_{13} & S_{14} \end{array} \\
\begin{bmatrix}
1 & -1 & -1 & & & & & & & & & & & \\
& 1 & & -1 & -1 & & & & & & & & & \\
& & 1 & & & -1 & -1 & & & & & & & \\
& & & 1 & & & & -1 & -1 & & & & & \\
& & & & 1 & & & & 1 & -1 & & & & \\
& & & & & 1 & & & & -1 & -1 & & & \\
& & & & & & & & & & 1 & -1 & -1 & \\
& & & & & & & & & & & 1 & -1 & -1
\end{bmatrix}
\end{array}.
\tag{5.38}
$$

If the variable is measured and can also be calculated, it is redundant; if it can be obtained only by the measurement itself, it is nonredundant. The first kind of redundancy can be

By first arranging the streams as unmeasured and measured and using the basic matrix operators, we can have the unmeasured part arranged into observable and unobservable parts
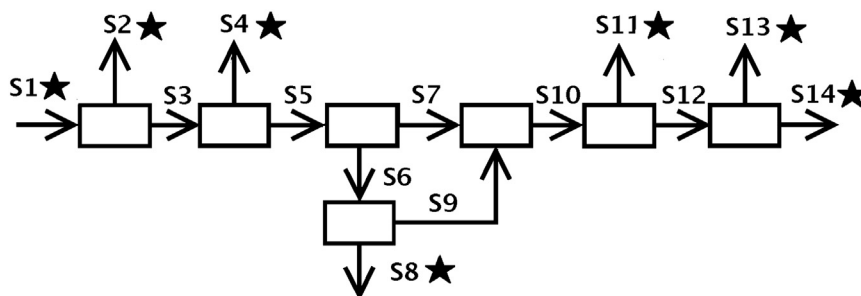
FIGURE 5.9 A typical flowsheet for recycling (*H* denotes sampled stream).

by developing the matrix canonical form. The canonical form is

$$
C = \begin{array}{c}
\begin{array}{ccccccccccccc}
S_3 & S_5 & S_{12} & S_{10} & S_6 & S_7 & S_9 & S_1 & S_2 & S_4 & S_8 & S_{11} & S_{13} & S_{14}
\end{array} \\
\left[\begin{array}{ccccccc|ccccccc}
1 & & & & & & & -1 & 1 & & & & & \\
& 1 & & & & & & -1 & 1 & 1 & & & & \\
& & 1 & & & & & & & & & 1 & & -1 \\
& & & 1 & & & & & & & & -1 & -1 & -1 \\
& & & & -1 & -1 & & -1 & 1 & 1 & & & & \\
& & & & 1 & & -1 & 1 & -1 & -1 & -1 & & & \\
& & & & & 1 & 1 & & & & & -1 & -1 & -1
\end{array}\right]
\end{array}
\tag{5.39}
$$

In the example, streams S_3, S_5, S_{12}, and S_{10} are observable because they only have one nonzero element in the column. Streams S_6, S_7, and S_9 are unobservable. All the measured streams are nonredundant. A material reconciliation cannot be performed for this sampling scheme.

Equation (5.37) can be written with the help of Eqn (5.39) to show the measured (M) and unmeasured (U) variables (Rao and Narasimhan (Rao and Narasimhan, 1996))

$$
\begin{bmatrix} \mathbf{C}_U & \mathbf{C}_M \end{bmatrix} \begin{bmatrix} \mathbf{x}_U \\ \mathbf{x}_M \end{bmatrix} = 0.
\tag{5.40}
$$

We get from the system[2]

$$
c_R x_R = 0.
\tag{5.41}
$$
$$
x_O = c_{RO} x_R + c_{NRO} x_{NR}
\tag{5.42}
$$
$$
c_{UO} x_{UO} = -c_{RUO} x_R - c_{NRUO} x_{NR}.
\tag{5.43}
$$

The first equation cannot be satisfied by measurements and forms the basis for data reconciliation. The second equation allows us to calculate the observable variables. The third equation cannot be solved.

The redundant variables will never satisfy Eqn (5.41). We have

$$
c_R x_R = r,
\tag{5.44}
$$

where **r** is a vector for residuals.

For linear systems (i.e., only simple mass flow conservation equations for nodes $x_i = Q_i$) reconciliation procedure is a minimization problem where the objective function is minimized subject to a set of constraints.

In matrix for the equation can be written as

$$
Min\left\{ \begin{bmatrix} \overline{Q}_R - Q_R \end{bmatrix}^T s_R^{-1} \begin{bmatrix} \overline{Q}_R - Q_R \end{bmatrix} \right\}
\tag{5.45}
$$

with the constraint $c_R \overline{Q}_r = 0$. This is a least-square minimization problem.

[2] R, redundant; NR, nonredundant; O, observable; UO, unobservable.

Standard Lagrange multipliers can solve this minimization.

$$L = \left[\overline{Q}_R - Q_R\right]^T s_R^{-1}\left[\overline{Q}_R - Q_R\right] - \lambda^T\left(c_R\overline{Q}_R\right). \tag{5.46}$$

The condition for optimality is that $\frac{\partial L}{\partial \overline{Q}_R} = 0$ and $\frac{\partial L}{\partial \lambda} = 0$.

The answer becomes

$$\overline{Q}_R = \left[I - s_R c_R^T\left(c_R s_R c_R^T\right)^{-1} c_R\right] Q_R. \tag{5.47}$$

This is the minimum level of data reconciliation to be performed.

5.7.2 Nonlinear Data Reconciliation

If the sampling has also given information about components, they can be included into data reconciliation. In addition to Eqn (5.37), we have

$$c_R M_j = 0, \tag{5.48}$$

where M is the component flow of P components $m_{jk} = Q_j c_{jk}$; $k = 1...p$. Components can be liberation classes, size classes, elements, etc.

The equation for data reconciliation now becomes (like Eqn (5.45))

$$\begin{aligned} Min\Big\{ &\left[\overline{Q}_R - Q_R\right]^T s_{QR}^{-1}\left[\overline{Q}_R - Q_R\right] \\ &+ \left[\overline{m}_R - m_R\right]^T s_{PR}^{-1}\left[\overline{m}_R - m_R\right]\Big\} \end{aligned} \tag{5.49}$$

subject to the constraints $c_R\overline{Q}_R = 0$; $c_R\overline{M}_j = 0$; $\sum_{j=1}^{K} \overline{y}_{R,j} = 0$. The procedure with Lagrange multipliers is analogous.

As it is written, Eqn (5.49) contains redundant flow rates and concentrations from the same redundant flows. However, concentrations can be measured without a subsequent flow rate measurement. These concentration measurements can sometimes be used to obtain flow rate values for flows that otherwise would be unobservable. This makes the computations complex, because the redundancy is affected. In programs designed for data reconciliation, this is often solved by taking into consideration all variables. The unmeasured variables are given a large variance. This approach can lead to large optimization systems and numerical problems prohibiting convergence.

For a single separator, where all streams are measured, we can either minimize the sum of squares of the closure residuals (to be shown here) or minimize the sum of squares of the component adjustments. For a steady state, we can start by dividing the balance equation by Q_f (Figure 5.4) to get

$$c_{fk} - C c_{ck} - (1 - C)c_{tk} = r_k. \tag{5.50}$$

The sum subject to minimization is now

$$S = \sum_{k=1}^{p} (r_2)^2. \tag{5.51}$$

We get as a result

$$\overline{C} = \frac{\sum_{k=1}^{p}(c_{fk} - c_{tk})(c_{ck} - c_{tk})}{\sum_{k=1}^{p}(c_{ck} - c_{tk})^2}. \tag{5.52}$$

To distribute the closure errors, we can write

$$\Delta_{fk} - \overline{C}\Delta_{ck} - (1 - \overline{C})\Delta_{tk} = r_k. \tag{5.53}$$

This can again be solved by using Lagrangian multipliers. The Lagrangian becomes

$$\begin{aligned} L = &\sum_{k=1}^{p}\left(\Delta_{fk}^2 + \Delta_{ck}^2 + \Delta_{tk}^2\right) \\ &+ 2\sum_{k=1}^{p}\lambda_k\left(r_k - \Delta_{fk} + \overline{C}\Delta_{ck} + (1 - \overline{C})\Delta_{tk}\right). \end{aligned} \tag{5.54}$$

Finding by derivation the minima for the variables (including λ_k) we get, after manipulation,

$$\Delta_{fk} = \frac{r_k}{1 + \overline{C}^2 + \left(1 - \overline{C}\right)^2}$$

$$\Delta_{ck} = \frac{-\overline{C}r_k}{1 + \overline{C}^2 + \left(1 - \overline{C}\right)^2} \tag{5.55}$$

$$\Delta_{tk} = \frac{-(1 - \overline{C})r_k}{1 + \overline{C}^2 + \left(1 - \overline{C}\right)^2}$$

5.8 LIBERATION

The mix of compounds in particles of various origins, from complete devices or parts of them, or any type of byproducts such as slag, may vary from a single compound to a mix of several compounds. A particle consisting of a single compound is called liberated. A mix of two compounds is called a binary, and with the same logic, ternary particles have three compounds.

For the optimal processing of compounds, the aim is to maximize the mass of liberated particles in a set of recycled material. As mentioned before, it poses an optimization problem, because breaking the material into too-fine particles causes the processes to operate under nonoptimal conditions.

All recycled materials have a specific way of breaking when exposed to an impacting, compressing, or shearing force large enough. By studying the progeny particles, one can form a

model of liberation breaking. This kernel function can be determined by either textural modeling or probabilistic methods (van Schaik et al. (van Schaik et al., 2004), Gay (Gay, 2004)).

As pointed out by Gay (Gay, 2004), the approach of direct liberation classes soon becomes numerically expensive for multiphase particles. It can be avoided by using parent particle to progeny particle relationships by using a liberation kernel function K.

$$g_j = \sum_i f_i K_{ij}, \tag{5.56}$$

where f_i and g_j are the composition frequencies of parent particle type i and progeny particle type j, respectively, and K_{ij} is the kernel function. When the kernel is known, it can be used to calculate the frequency distribution of type j progeny particles originating from type i parent particles
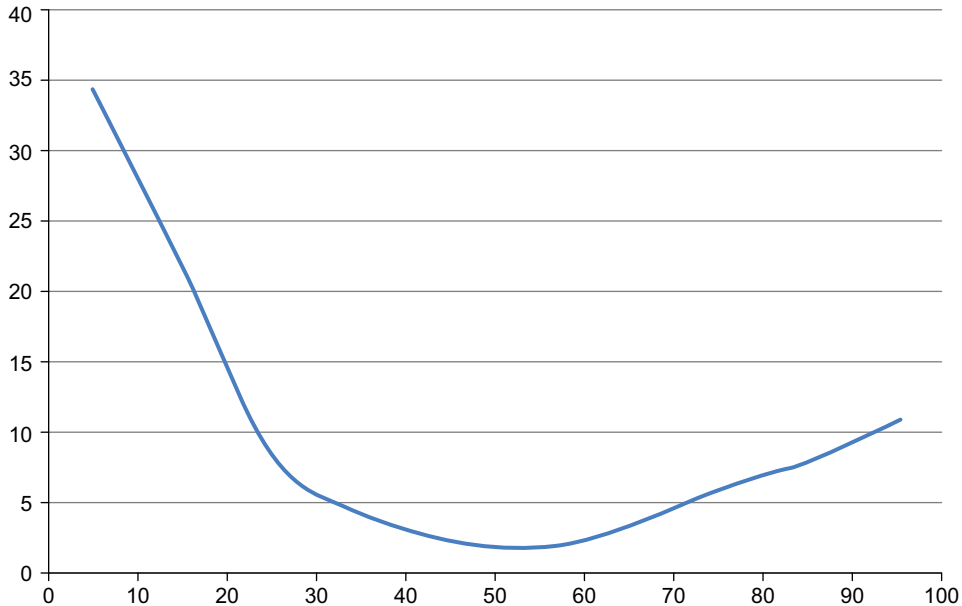
$$p_{ij} = f_i K_{ij}. \tag{5.57}$$

FIGURE 5.10 Frequency distribution of a single property.

Gay (Gay, 2004) proposed using the probability entropy method to solve Eqn (5.57) under the constraints that the original composition distribution of parent particles is satisfied, the original composition distribution of progeny particles is satisfied, and the average mineral composition of the resulting progeny particles is the same as the parent particles. The constraints can be formulated as follows

$$\sum_j p_{ij} = f_i (\text{for each } i)$$
$$\sum_i p_{ij} = g_j (\text{for each } j) \qquad (5.58)$$
$$\sum_j p_{ij}(c_{jm} - C_{im}) (\text{for each } i \text{ and } m)$$

where c_{jm} is the composition of the mth mineral for the jth progeny particle and C_{im} is the composition of the mth mineral for the ith parent particle.

He obtains as a solution

$$\ln(p_{ij}) - 1 = \lambda_{1i} + \lambda_{2j} + \lambda_{3im}(c_{jm} - C_{im}), \quad (5.59)$$

where the Lagrange multipliers λ_1, λ_2, and λ_3 are for the respective constraints.

After shredding, the frequency distribution of a property might look like the curve in Figure 5.10. One can see that there is a large mass of particles that does not possess the property, and a good mass of particles with a high degree of the defined property.

The distribution gives the possibility of separating the stream into a stream low in the property and a stream rich in the property. As an example, say, the property is aluminum (Al)
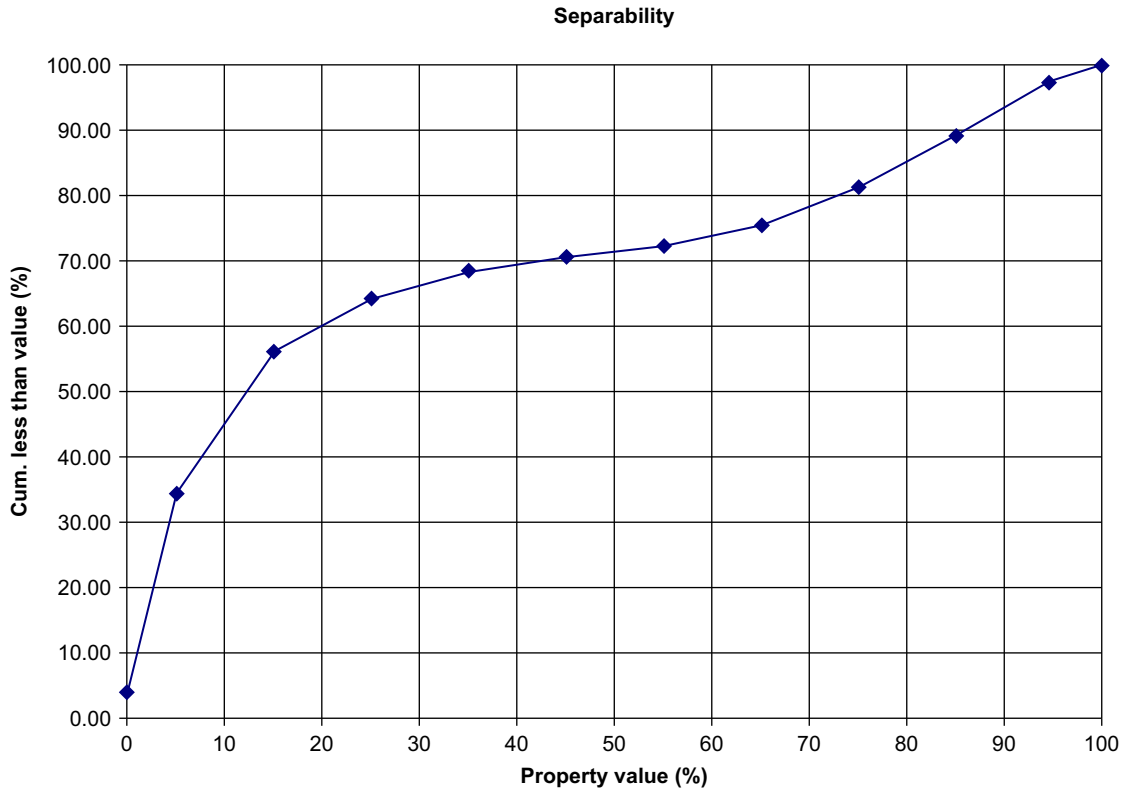

FIGURE 5.11   Separability curve.

mass content. We can separate an Al-rich and an Al-poor product from the feed.

By calculating the total mass content of, say, Al cumulatively, starting from the poorest fraction, we get a curve called the separability curve, which shows how much Al is present in the fraction below a property limit (Figure 5.11). The example reveals that 70% of Al values are particles, which carry more than 50% non-AL matter.

5.9 GRADE-RECOVERY CURVES

A relation always exists between the grade of a separated product and its recovery (recyclability). There are two basic reasons for that relationship. First, the liberation of particles subjected to separation is not complete. Second, the related response to a physical force or chemical potential gradient will cause different particles to react in different ways.

A liberation-based grade-recovery curve (Figure 5.12) can be constructed directly from the separability curve with easy computations. Taking the entire feed stream as a product, the recovery is 100% and the product grade is the same

as the feed: in our Al case, 33.7% Al. Leaving the fractions with less than 10% Al away reduces the mass almost to 50% according to Figure 5.10, but only loses 5% of the Al. It also increases the product quality to 49% Al. Continuing in this way yields a full grade-recovery curve owing to the lack of liberation. As can be seen, with this material a 90% Al purity product can only be obtained with a 55% recovery.

Breaking the material into smaller particles will improve the separability curve (Figure 5.13). This experimental value is from the same scrap sample but is substantially finer in particle size.

Compared with Figure 5.11, the central part of the curve is much flatter, indicating that no significant mass of middling particles exists. This leads to the following grade recovery curve (Figure 5.14).

The recovery for a 90% pure product has increased to 85%, a substantial improvement in the theoretically obtainable result.

The importance of the liberation-based grade-recovery curve depends on the property interactions and the nature of the following processes.
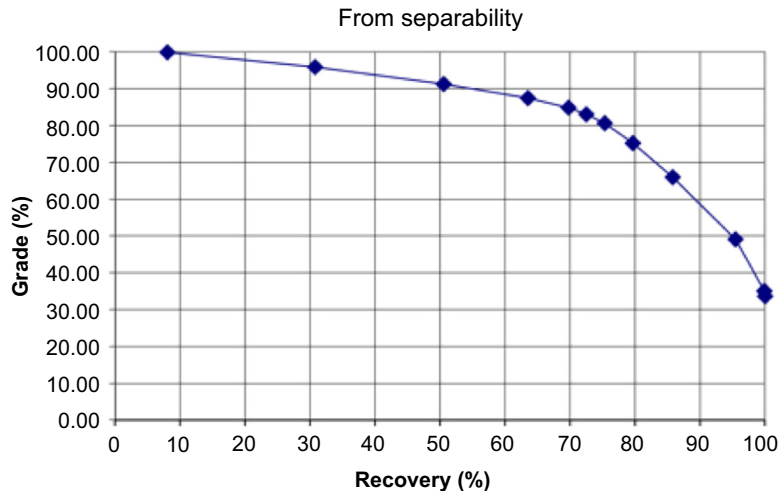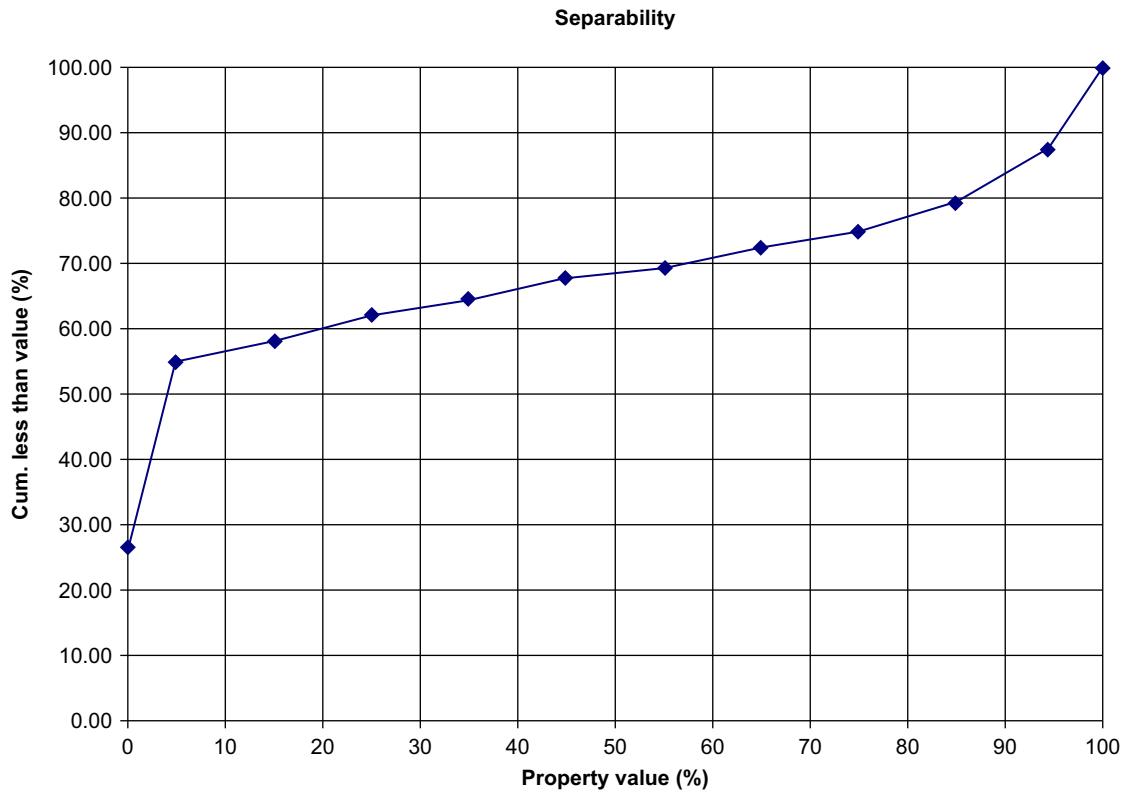

FIGURE 5.12 Grade-recovery curve.

Separability

FIGURE 5.13   Separability curve after further breakage.

5.9.1 Mechanical Separations

As will be discussed in more detail in the Appendix, mechanical separation is based on a balance of forces. The force for separation is chosen according to the properties of the particles to be separated. It can be a body force such as gravity, centripetal force or magnetic force, or a surface force induced by surface property modifications. This active separation force is directed by the separator design so that the trajectory of particles affected by the force becomes different from the trajectory of particles not affected by the force. Mass forces can be used to enhance the difference in particle trajectories.

Figure 5.15 depicts a separator with a flow from one feed point to two streams. All particles start from the same point, but the acting separation force (lower picture) takes the particles to the upper discharge, whereas a similar particle without the affecting force will report to the lower discharge.

As stated, separation is a particulate process in which particle size, shape, and density affect the outcome in addition to the active separation force. For large particles, the mass forces are the most important forces. As the mass decreases to the third power of diminishing particle size and area only to the second power, at some point the surface forces will become dominant. The
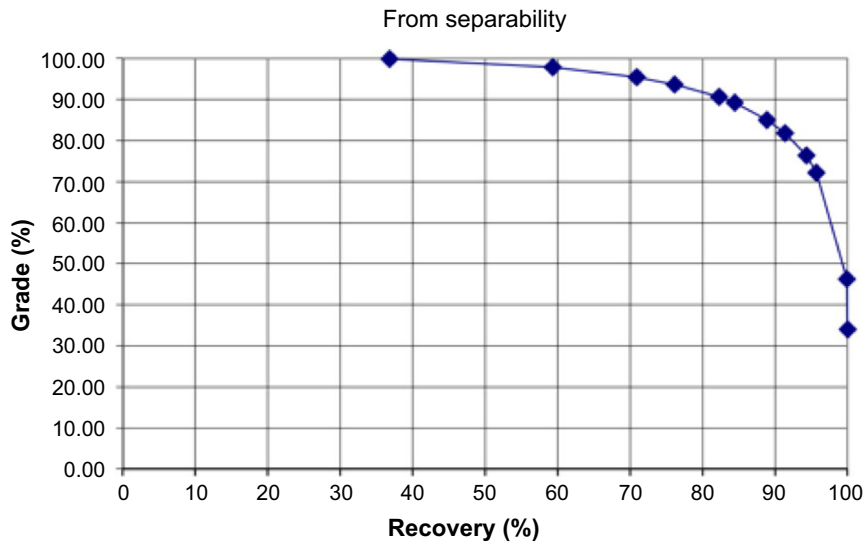
From separability

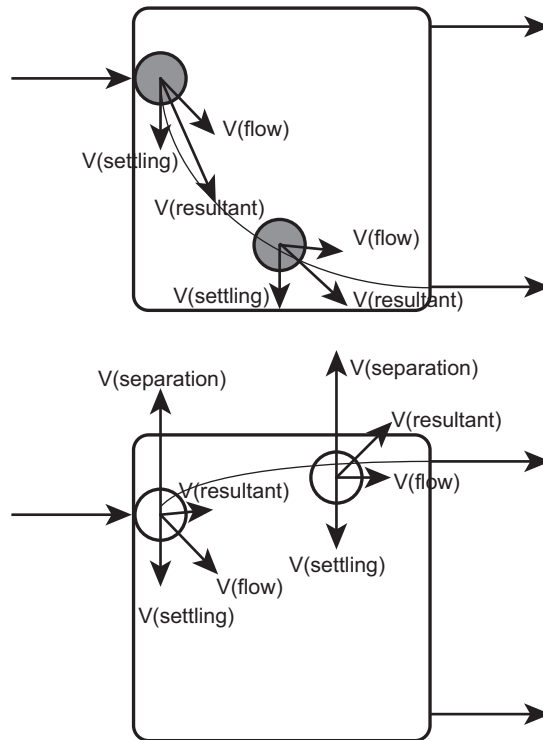**FIGURE 5.14** Grade-recovery curve after further breakage.

FIGURE 5.15 Particle trajectories in a separator, when a separating force acts at some particle classes.

smaller the particle, the more will surface forces such as drag and viscosity and even electrostatic and van der Waals forces affect the total force balance. The task is to optimize particle size for liberation and for efficient separation.

For mechanical separations, some operational deficiencies always exist owing to property and size distributions, apparent viscosity effects, turbulence, and boundary flows.

Any particle entering a separator will have a probability of entering one of the product streams. The Separation cut point is the value of the property in which particles have an equal probability of entering either of the two product streams. It is often denoted as ψ_{50}. In Figure 5.16, the cut point density is 2705 kg/m (Heiskanen and Laapas, 1979).

The separation efficiency curve can be calculated from reconciled data as the percentage of the mass in each property class to report to the chosen product. The shaded areas are misplaced particles. Vertical shading represents light particles reporting in heavies or sinks, and the horizontal shading is heavy particles found in the light fraction. The steepness of the curve can be used as a measure of quality, imperfection.

$$I = \frac{\psi_{75} - \psi_{25}}{2\psi_{50}}. \qquad (5.60)$$

A separation efficiency curve can also be computed for secondary effects. In Figure 5.16, the substance F probability curve has a similar slope with an offset. This indicates a slight concentration to the heavy fraction, but homogeneously.
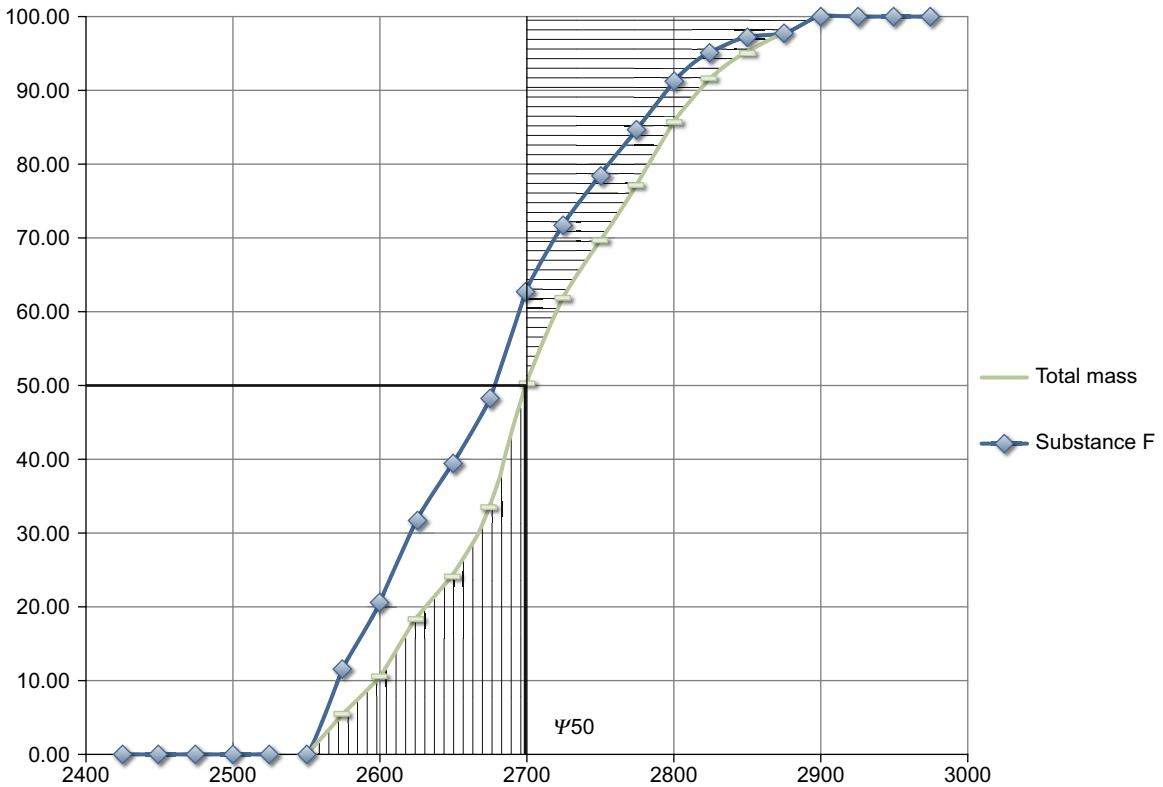

FIGURE 5.16 Separation efficiency curve for density (x-axis) versus probability of sinking (Tromp curve) ($I = 0.023$).

Chemical methods for recycling i.e. hydro- and pyrometallurgical recycling technology, theory and systems are discussed in various chapters of Part II.

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