Chapter 4
Green and Sustainable Chemistry

Klaus Kümmerer and James Clark

Abstract The products of the chemical and pharmaceutical industries are indispensable for our high standard of living and health. Estimations say that about 100,000 chemicals are available on the market, mostly used in combination with other chemicals. Consumers may be unaware that the products of chemical industries provide the functionality they expect when buying or using a certain product. Often the contribution of chemistry is not clear to the consumer, as chemicals are used to improve or enable certain production processes, to improve the efficacy or the lifetime of a product or to generate a specific colour or taste (e.g. food additives, preservatives). In other words, the benefit of modern chemistry and pharmacy cannot be overestimated.

Contrary to current perception, which is dominated by the legacies of the past, chemistry can and will contribute in many ways to sustainability through its products and processes. However, it is important that chemistry itself becomes more sustainable. Sustainable chemistry encompasses green chemistry but is much more than that. An overview of green and sustainable chemistry and its important achievements are presented, and some possible future contributions are outlined.

Keywords Chemistry • Sustainability • Design • Resource • End of life • Biorefinery

1 Introduction

The products of the chemical and pharmaceutical industries are an indispensable basis of our high standard of living and health. Estimations say that about 100,000 chemicals are available on the market, most of them used in combination with other chemicals and often constituting complex products. Sometimes consumers are not even aware that the products of chemical industries provide the functionality they

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expect when they buy or use a certain product, the contribution of chemistry often going unnoticed as chemicals are used to improve or enable certain production processes, to improve the efficiency or the lifetime of a product and to generate a specific colour or taste (e.g. food additives, preservatives). In other words, the benefit of modern chemistry and pharmacy can hardly be overestimated. In many areas, chemistry and pharmacy make up the backbone for sustainable development. This includes among others a pivotal role in the so called megatrends:

– Natural resources and environment
– Demographics- Globalization
– Technology and Innovation
– Consumption patterns

Chemistry is fundamental for challenges related to these megatrends such as alternative feedstock, environmental technology, nutrition and health, clean air and water, intelligent and efficient materials, renewable energy to mention just a few. Thereby chemistry can contribute much to sustainability. However, at the same time chemistry itself has to become sustainable.

According to the OECD (2008), the value of chemical production will be roughly $4000 billion (US) in 2015 and rise to $5500 billion by 2030. Most of this increase is expected for non-OECD countries. However, there are also challenges and a backside to the coin. Population growth and climate change will place great pressure on resources in the future. Increasing income and health will result in an increase in products and wastes.

Nowadays, most western countries have measures for proper and effective treatment and the prevention of emissions into air, water and soil stemming directly from production and manufacturing in place (Kümmerer 2010a, 2011; Schwarzenbach et al. 2006). That is often not the case in less developed countries where the products used in developed countries are synthesised and manufactured (Larsson et al. 2007). Interestingly, the introduction of chemicals into the environment is often unavoidably connected to the proper use of certain products of the chemical and pharmaceutical industries, such as pharmaceuticals, disinfectants, contrast media, laundry detergents, surfactants, anticorrosives used in dishwashers, personal care products and pesticides, to name just a few. It has been learned in recent years that even if advanced effluent treatment were to be applied, a significant portion of these chemicals would still remain in the wastewater. Incomplete removal of the chemicals leads to introduction into the aquatic cycle, where they can undergo further distribution and transformation (Fig. 4.1). Follow-up problems of such an end-of-the-pipe measure are increased energy demand and formation of unwanted reaction products that can even be more toxic and persistent in the environment than the parent compounds. Additionally, such advanced technologies often cannot be applied in developing countries.

Other chemicals, such as flame retardants or textile chemicals, are washed out during laundering, and still others, again stemming from, e.g., furniture, carpets, computers and other items, enter the indoor air and the environment because of their volatility. In the air, chemicals may be distributed globally if their lifetime is higher than approximately 10 days.

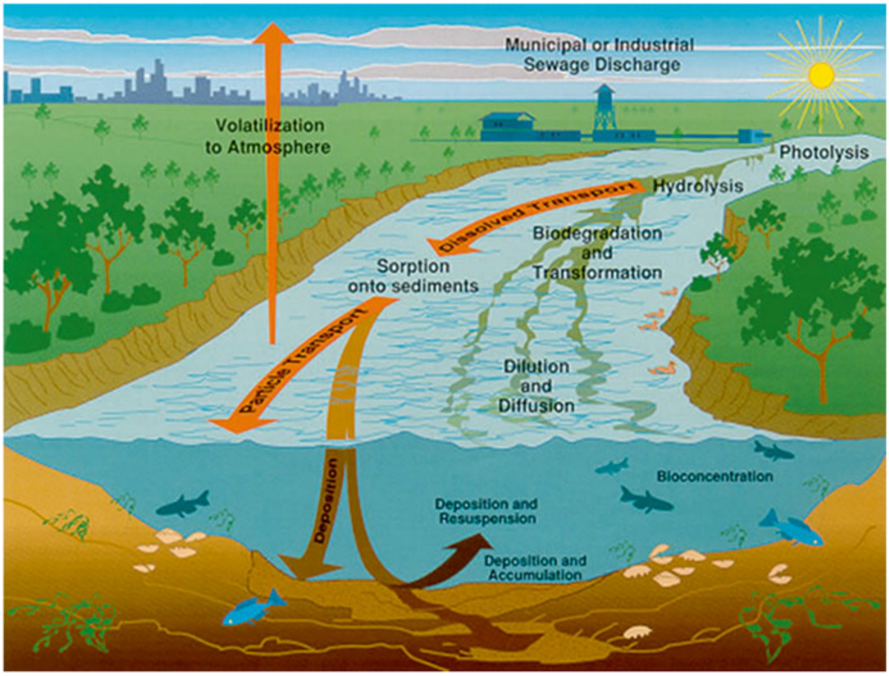


Fig. 4.1 Fate of pollutants in the aquatic environment (Source: U.S. Geological Survey, http://toxics.usgs.gov/regional/emc/transport_fate.html)

In some instances, it is not just the individual molecules but also the products themselves that pose a risk to the environment. An example is the pollution of the sea by plastics stemming from packaging such as bottles or bags, as well as from other plastic products such as rope. They are present as tiny particles (Andrady 2011) that adsorb other toxic chemicals and can cause the death of animals after ingestion by mechanically injuring them as well as by poisoning them through release of the formerly adsorbed chemicals. This pollution has economic consequences too.

2 Green and Sustainable Chemistry

Both "green" and "sustainable" chemistry embrace the full life cycle of chemicals and not just one stage of that cycle:

- Raw materials
- Synthesis
- Production
- Use
- Fate after use ("end of life")

Sustainable chemistry includes economical, social and other aspects related to manufacturing and application of chemicals and products. It aims not only at green synthesis or manufacturing of chemical products but also includes the contribution of such products to sustainability itself. In the Rio Declaration within Agenda 21 adopted in Rio de Janeiro in 1992, it was stated that it is important for research to intensify for the development of safe substitutes for chemicals with long life cycles (Agenda 21, # 19.21). Principles that address a more integrative view were subsequently established in the European Union in 1996 by a council directive (EC 1996). In general, use of the best available techniques, efficient energy use and prevention of accidents and limitations of their consequences were addressed. In Annex IV of the directive, specific measures were specified:

1. The use of low-waste technology;
2. The use of less hazardous substances;
3. The furthering of recovery and recycling of substances generated and used in the process, and of waste, where appropriate;
4. Comparable processes, facilities or methods of operation which have been tried with success on an industrial scale.
5. Technological advances and changes in scientific knowledge and understanding.
6. The nature, effects and volume of the emissions concerned.
7. The commissioning dates for new or existing installations.
8. The length of time needed to introduce the best available technique.
9. The consumption and nature of raw materials (including water) used in the process and their energy efficiency.
10. The need to prevent or reduce to a minimum the overall impact of the emissions on the environment and the risks to it.
11. The need to prevent accidents and to minimize the consequences for the environment.

An amendment came into force in 2010 as 2010/75/EU (ABl. EG L 334, p. 17–119).

Anastas and Warner (1998) published some similar simple rules of thumb addressing more or less the same points. These rules (later called the "12 principles") had their roots in the United States' Pollution Prevention Act of 1990 (http://www2.epa.gov/green-chemistry/basics-green-chemistry#definition):

1. Prevent waste: Design chemical syntheses to prevent waste. Leave no waste to treat or clean up.
2. Maximize atom economy: Design syntheses so that the final product contains the maximum proportion of the starting materials. Waste few or no atoms.
3. Design less hazardous chemical syntheses: Design syntheses to use and generate substances with little or no toxicity to either humans or the environment.
4. Design safer chemicals and products: Design chemical products that are fully effective yet have little or no toxicity.
5. Use safer solvents and reaction conditions: Avoid using solvents, separation agents, or other auxiliary chemicals. If you must use these chemicals, use safer ones.

6. Increase energy efficiency: Run chemical reactions at room temperature and pressure whenever possible.
7. Use renewable feedstocks: Use starting materials (also known as feedstocks) that are renewable rather than depletable. The source of renewable feedstocks is often agricultural products or the wastes of other processes; the source of depletable feedstocks is often fossil fuels (petroleum, natural gas, or coal) or mining operations.
8. Avoid chemical derivatives: Avoid using blocking or protecting groups or any temporary modifications if possible. Derivatives use additional reagents and generate waste.
9. Use catalysts, not stoichiometric reagents: Minimize waste by using catalytic reactions. Catalysts are effective in small amounts and can carry out a single reaction many times. They are preferable to stoichiometric reagents, which are used in excess and carry out a reaction only once.
10. Design chemicals and products to degrade after use: Design chemical products to break down to innocuous substances after use so that they do not accumulate in the environment.
11. Analyze in real time to prevent pollution: Include in-process, real-time monitoring and control during syntheses to minimize or eliminate the formation of by-products.
12. Minimize the potential for accidents: Design chemicals and their physical forms (solid, liquid, or gas) to minimize the potential for chemical accidents, including explosions, fires, and releases into the environment.

At the Johannesburg World Summit in 2002, as part of the millennium goals set up, it was agreed upon to substitute dangerous compounds, to increase resource efficiency and to cooperate for the development of a better management of chemicals globally, including education and training. This resulted in the establishment of a Strategic Approach to International Chemicals Management (SAICM; http://www.saicm.org).

There are estimates that green chemicals will save industry $65.5 billion by 2020 (http://www.navigantresearch.com/newsroom/green-chemicals-will-save-industry--65-5-billion-by-2020). However, it was not clearly defined in this context what “green chemicals” would exactly mean – the ones that fulfil one or a few of the above rules of thumb or the ones that fulfil all of them.

In general, only rarely are aspects that go beyond the chemicals themselves and their technical issues addressed by green chemistry, whereas sustainable chemistry generally includes all aspects of a product related to sustainability, e.g. social and economic aspects related to the use of resources, the shareholders, the stakeholders and the consumers (Fig. 4.2).

Integrating the principles of green and sustainable chemistry into synthesis of chemicals as well as the manufacturing of new materiala and complex porducts requires the chemist doing his work to think in an open-minded interdisciplinary manner and to take into consideration the world outside the laboratory from the very

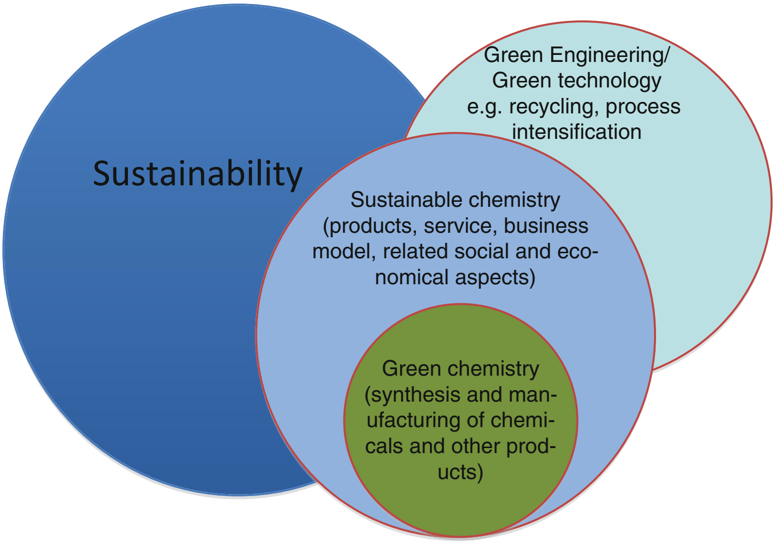


Fig. 4.2 The relationship of sustainability, sustainable chemistry green engineering, green technology, and green chemistry

beginning. This includes accounting for not only the functionalities of a molecule that are necessary for its application but also their impact and significance at the different stages of its life cycle.

3 Green Chemistry Metrics

It is important to be able to quantify the change when changes are made to chemical processes (Constable et al. 2007, Lapkin and Constable 2008). This enables us to quantify the benefit from the new technology introduced (if there are benefits). This can aid in in-house communication (to demonstrate the value to the workforce) as well as external communication. For yield improvements and selectivity increases, simple percentages are suitable, but this simplistic approach may not always be appropriate. For example, if a toxic reagent is replaced by a less toxic one, the benefit may not be captured by conventional methods of measuring reaction efficiency. Equally, these do not capture the mass efficiency of the process – a high-yielding process may consume large amounts of auxiliaries such as solvents and reagents, as well as those used in product separation and purification. Ideally, we also need to find a way to include energy and water, both of which have been commonly used in a rather cavalier way but which are now subject to considerable interest that can vary depending on the location of the manufacturing site.

Numerous metrics have been formulated over time and their suitability discussed at great length. The problem observed is that the more accurate and universally applicable the metric devised, the more complex and unemployable it becomes. A good metric must be clearly defined, simple, measurable, and objective rather than subjective and must ultimately drive the desired behaviour. Some of the most popular metrics are:

- E factor (which effectively measures the amount of product compared to the amount of waste – the larger the E factor, the less product-specific the process; the fine and pharmaceutical manufacturing sectors tend to have the highest E factors)
- Effective mass yield (the percentage of the mass of the desired product relative to the mass of all non-benign materials used in its synthesis – this includes an attempt to recognise that "not all chemicals are equal" – important and very real, but very difficult to quantify)
- Atom efficiency/economy (measures the efficiency in terms of all the atoms involved and is measured as the molecular weight of the desired product divided by the molecular weight of all of the reagents; this is especially valuable in the design "paper chemistry" stage, when low atom efficiency reactions can be easily spotted and discarded)
- Reaction mass efficiency (essentially the inverse of the E factor)

Of course, the ultimate metric is life cycle assessment (LCA), but this is a demanding exercise that requires a lot of input data, making it inappropriate for most decisions made in a process environment. However, some companies do include LCA impacts such as greenhouse gas production in their in-house assessment, for example, to rank solvents in terms of their greenness. It's also essential that we adopt a "life cycle thinking" approach to decision making so that we don't make matters worse when greening one stage in a manufacturing process without appreciating the effects of that change on the full process, including further up and down the supply. An integrated zero waste biorefinery that sequentially exploits an extraction, followed by biochemical and thermal processing, with internal recycling of energy and waste gases, is viewed as a model system. Extraction of secondary metabolites prior to their destruction in subsequent processes can significantly increase the overall financial returns.

4 Natural Resources and Chemistry

4.1 The Fossil Age

The resources of chemistry are inorganic materials, such as metals and minerals, that are gained by mining. Mining is often connected to severe environmental pollution and also has a social impact. For organic chemistry, oil is still by far the most important resource. It is also used as a resource for energy in chemical industry. This resource is limited, as are gas and coal. It is a hot bed of discussion nowadays

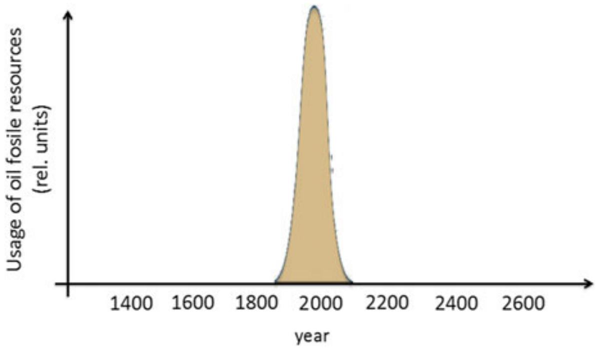


Fig. 4.3 The fossil age

as to how long these fossil resources will be available (Aleklett et al. 2012). However, even in the best case, it will be a matter of only decades or a century at the most (Fig. 4.3).

4.2 The Biorefinery and Its Potential for Replacing the Petrochemical Industry

The twentieth century saw a boom in the chemical industry with the emergence of an organic chemical manufacturing industry based on a cheap carbon feedstock, oil. This revolutionised the main energy source away from bio-resources, thereby creating the basis of the petroleum refinery we know today. It also helped create the chemical industry that has dominated the world for over 50 years.

Environmental and political concerns over the impact of continued fossil fuel use, their depletion and security of supply, combined with a growing population, have created a need for renewable sources of carbon. Over the last two decades, there has been a global policy shift back towards the use of biomass as a local, renewable and low-carbon feedstock. The "biorefinery" concept is a key tool in utilising biomass in a clean, efficient and holistic manner, whilst maximising value and minimising impact. However, the use of biomass as a source of energy, chemicals and materials is not new and has been taking place for millennia. The biorefinery concept is analogous to today's petroleum refineries. Biorefineries are ideally integrated facilities for conversion of biomass into multiple value-added products, including energy, chemicals and materials (Figs. 4.4 and 4.5). It is important that biorefineries utilise a range of low-value, locally sourced feedstocks, which don't compete with the food sector, including low-value plants such as trees, grasses and heathers, energy crop and food crop by-products (wheat straw), marine resource wastes, seaweeds and food wastes.

The main transformations available to the biorefinery can be classified as extraction, biochemical and thermochemical processes. The application of green chemical technologies (including supercritical fluid extraction, microwave processing, bio-

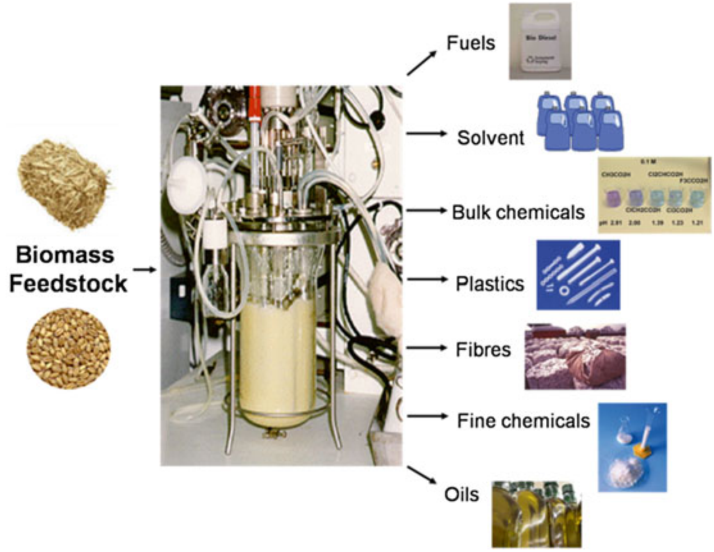


Fig. 4.4 Products from biomass

conversion, catalytic and clean synthesis methods) are all utilised with the aim of developing new, genuinely sustainable, low environmental impact routes to important chemical products, materials and bioenergy. These methodologies are usually studied independently of one another; however, the integration and blending of technologies and feedstocks is a way to increase the diversity of products and the socio-economic and environmental benefits of the biorefinery.

4.3 Valorisation of Waste

Modern society is based on a linear model of extraction of resources (oil, minerals, etc.), processing (e.g. chemical manufacturing, then formulation), use and disposal. This can only be sustainable if the disposal returns the resource to us in a useful form and on a reasonable timescale, typically measured within a human lifetime (<100 years). But with most types of resource, we have not been doing this. With metals, for example, we have been dispersing original virgin ores in the form of waste into landfills where recovery is difficult (Graedel 2011; Dodson et al. 2012). With organic materials, the situation is somewhat more complex – some are recycled by nature through biodegradation but many are not (e.g. non-biodegradable plastics). Our efforts at recycling are woefully inadequate (e.g. we recycle only about 1 % of the 260 million MT of plastics produced each year). Also, by stepping outside of the natural quick cycles of fast rotation bio-resources (plants, trees, etc.) and using a large proportion of bio-resources with very long cycles measured in millions of years (oil, coal, etc.), we have created an unsustainable economic model.

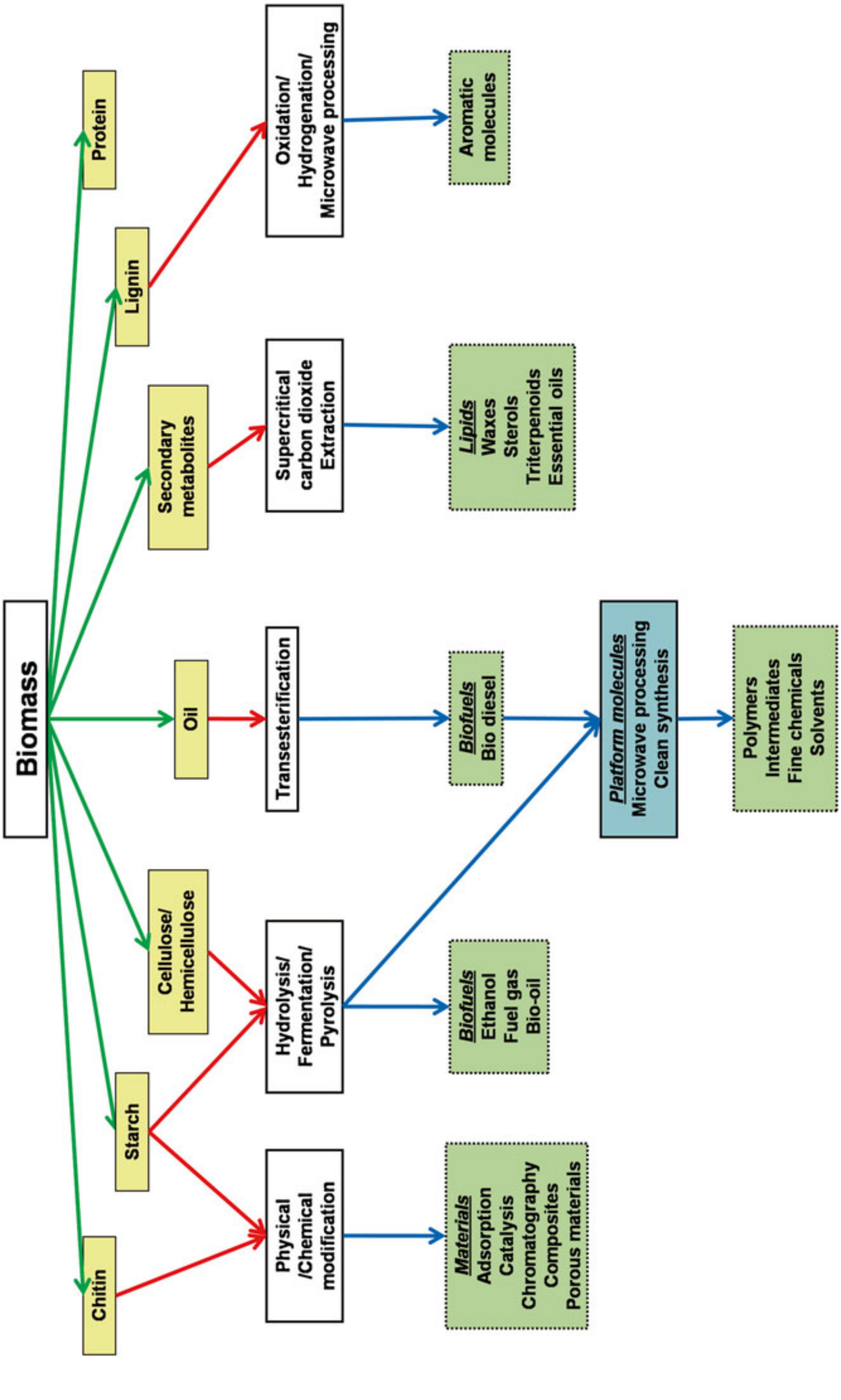


Fig. 4.5 Biomasses, chemical processes and products

We need to change our consumption pattern through adoption of a circular economy (Ellen MacArthur Foundation 2014) whereby we only consume resources in a way that can return them in a useful form on a sensible timescale. This is being "resource intelligent"! With a circular economy in place, our rate of consumption will then be limited by our efficiency – the closer we get to 100 % efficiency, the more we can enjoy the benefits of our planet!

5 Synthesis and Manufacturing

5.1 Solvent Selection

A significant number of organic solvents are strictly regulated and their use restricted (e.g. hexane, dichloromethane). This is set to increase, with new regulatory constraints being developed (REACH, VOC directives). It is likely that under REACH and other chemical-related legislation, many of the commonly used solvents in chemical and pharmaceutical manufacturing, as well as in other sectors, will be subject to authorisation or restrictions in use. As we seek alternatives, we should also be aware of incentives to encourage the use of bio-based chemicals, such as the EU prioritising some groups of chemicals, including solvents, for example, in the production of new standards.

In order to justify a replacement solvent, reaction efficiency must not be compromised simply to reduce the burden on the environment. Inferior reaction performance is not appealing on the grounds of increased waste and energy consumption, not to mention any economic implications. The search for greener solvent replacements can be systemised, thereby also providing additional justification for any proposed substitution.

Pharmaceutical manufacturing is one very important area in which new solvent restrictions could have dramatic effects. Typically, for the production of one kilogram of final API (active pharmaceutical ingredient), about 25–100 kg of waste are produced, meaning that 96–99 % of the overall process mass is discarded and requires, therefore, appropriate disposal. According to a report published by GlaxoSmithKline Pharmaceuticals in 2007, solvent usage accounts for 85–90 % of the total input material of a process. Besides, these figures do not include water usage, which is extensive in workup steps and as a reaction medium (Constable et al. 2007). The large volume of solvents used in drug manufacture, and the nature thereof, has become a matter of major concern in recent years. There is now a major search for greener solvents for use in industries like pharmaceuticals (Kerton 2013). This includes new bio-based solvents such as limonene (used in cleaning, but also a possible reaction solvent), cymene, cyrene (dihydrolevoglucosenone) and organic carbonates, as well as nonconventional solvents like water and liquid or supercritical carbon dioxide. There was a lot of research into ionic liquids as non-volatile, powerful solvents, but these have proven to be severely limited by factors including cost, some evidence for toxicity and difficulty in purification.

An example of the development of a new route of synthesis with fewer steps is given in Figs. 4.6 and 4.7.

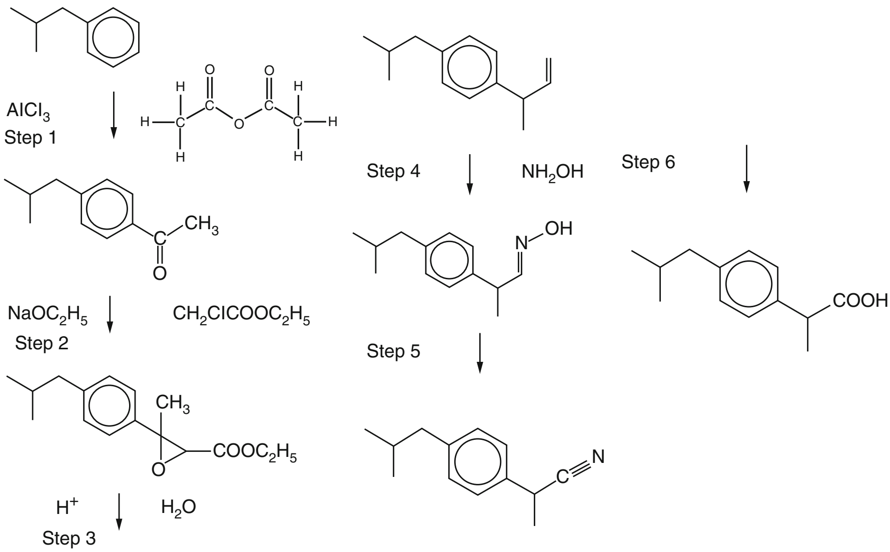


Fig. 4.6 Old route of synthesis of the pain killer ibuprofen (Source: http://www.rsc.org/learn-chemistry/resources/chemistry-in-your-cupboard/nurofen/5)

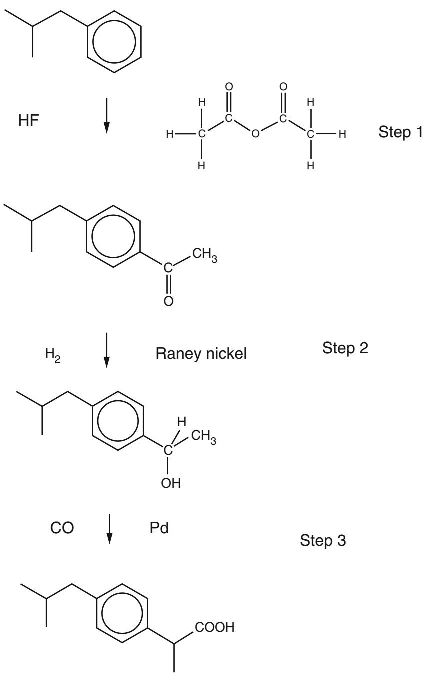


Fig. 4.7 New route of synthesis of the pain killer ibuprofen (Source: http://www.rsc.org/learn-chemistry/resources/chemistry-in-your-cupboard/nurofen/6)

6 Products and End of Life

6.1 Benign by Design

Many chemical products end up in the environment not because of improper use but because of proper use (see above). One of the biggest challenges nowadays related to chemicals is persistence in the environment (see, e.g. http://chm.pops.int/default.aspx). The correlation of structure and composition of a chemical, encoded in a formula, and its properties is at the core of chemistry and chemical language. A change in the structure of a chemical will result in different properties. The consideration of the functionalities of a molecule and the properties that are correlated with them and their significance and impact along their entire life cycle brings into the foreground the targeted design of new chemicals at the very earliest stage of their conceptualisation (Fig. 4.8) including end of life. This approach is called "benign by design" (Kümmerer 2007).

Substructures and functional groups are already known that may improve degradability by chemical processes such as hydrolysis, photolysis and biodegradation under environmental conditions (see Table 4.1). With the help of computer-based models such as (quantitative) structure activity relationships ((Q)SARs), a more systematic assessment can be done, e.g. of biodegradability (Rücker and Kümmerer 2012) or toxicity and physical chemical properties of molecules (Cronin and Madden 2010; Ekins 2007; Boethling and Mackay 2000). A big advantage then is that molecules can be assessed even before synthesis. This not only saves money and time, it also gives guidance as to which molecules may possess the desired low toxicity and fast and complete mineralization when they are introduced into the environment at the end of their life. The first steps on the road to greener pharmaceuticals are already done (Rastogi et al. 2014).

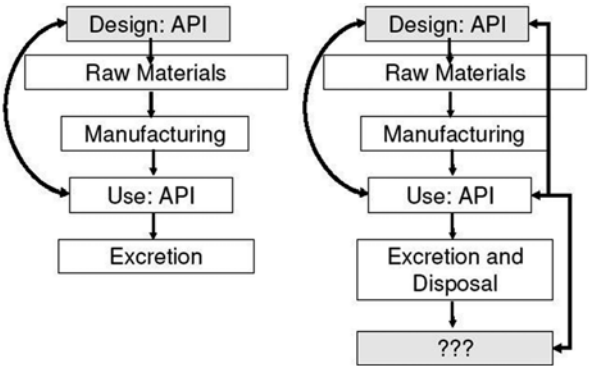
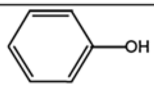
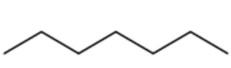
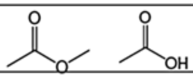
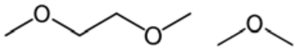
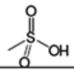
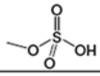


Fig. 4.8 Conventional (*left*) and sustainable approaches (*right*) for the design of new chemicals and active pharmaceutical ingredients (APIs): the end of life of the molecule is already taken into account at the very beginning (Source: Kümmerer 2010a)

Table 4.1 Examples of chemical functionalities and their impact on biodegradability in the environment

favorable	less favorable
	
	
	
	
	
—H	—F —Cl

(Source: Kümmerer 2010b)

6.2 Limits of Recycling and Material Flows

Inorganic molecules, metals and complex products are different from the above-described "small" organic molecules. Chemical products such as plastics often contain a mix of molecules, such as the polymer itself, and other molecules for the modification of their properties, such as softness or resistance against light. For them, as well as for inorganic materials and products, recycling is an option for recollecting the constituents. However, if the products are not designed for recycling, it may be difficult or even impossible to recycle the products themselves or extract components for further use. Furthermore, recycling needs additional energy and good logistics. Most often, the so-called recycling, in fact, is down cycling, that is, the quality of the regained products/and or constituents is lower than the original one. The laws of thermodynamics tell us that there is always a loss of material and quality through recycling.

In general, independent of recycling, the more complex the materials themselves, the higher diversity of products and chemicals that constitute a given material's flow, and the bigger those flows are, the bigger the loss. Even if the synthesis of a chemical or the manufacturing of products can be called green or sustainable, in the end, non-sustainability may result if the related material flows are too diverse and too big.

6.3 New Business Models

Thinking in terms of functionality or offering a service can avoid some of these pitfalls. For example, if the functionality “wood preservation” is needed, a wood preservative can deliver it. However, a wise construction may avoid giving water the access that makes the preservative necessary.

Another example would be a company that does not buy solvents, but instead leases them and returns them to the deliverer after usage. This has the advantage for the deliverer that, in taking back the solvent, they can use all their solvent-related knowledge and experience to make them most effective (e.g. solvent selection). Now, they have a specific interest in having the most efficient use of solvents. The leasing company has the same interest. This is a win-win situation – just selling a solvent is a win-lose situation: the provider wants to sell as much solvent as possible, the customer to buy as little as possible.

Another example is the use of disinfectants (Schülke 2011): A provider wants to sell as much disinfectant as possible. However, the goal behind the application of disinfectants is to safeguard a proper standard of hygiene. If the provider of the disinfectant is responsible for providing the necessary standard of hygiene, they will aim to use as little disinfectant as possible. As the manufacturer of a disinfectant has lots of knowledge about disinfectants and regulation on hygiene, they can provide training and education on the right use of disinfectants and application of other measures to maintain the necessary hygienic standards. In fact, they can save money by spending less money for the raw materials and synthesis/manufacturing of the disinfectant and earn money by selling a service – the maintenance of the appropriate standard of hygiene. Furthermore, fewer physical resources were needed; less energy for synthesis and delivery, less packaging material and less introduction of chemicals into the aquatic environment will result.

7 Conclusions

Chemistry has been – in modern words – a success story starting with the advent of chemical science working in close interaction with the beginning of chemical industries. Chemistry has been, is and will be a fundamental part of the modern way of life, contributing, for instance, to health benefits. But along the way, the other side of the coin has become apparent related to the intoxication of humans and pollution of the environment. Green and sustainable chemistry takes this into account through life cycle thinking and measures to prevent such negative impact from the very beginning and through all stages. It provides various goods and services that are needed for a successful transition to sustainable societies. Both the chemistry of the past and the present and even more so that of the future, that is, sustainable chemistry, are examples for successful interaction between disciplines (interdisciplinarity), basic science and everyday life (transdisciplinarity). There will be no future without chemistry. As to the challenges we face nowadays, however, we assert that chemistry needs to significantly change its ways in order to be part of a sustainable future.

Questions

1. What are the typical stages of life of a chemical product and what connection does each one have with sustainability?
2. What is a biorefinery and what would be its role in the future?
3. Where and how does chemistry contribute to sustainability?
4. Is there a difference between sustainable chemical products (molecules and materials) and sustainable chemistry?
5. What are typical environmental problems related to chemistry? What are solutions to these provided by sustainable chemistry?
6. What is peak oil and what is its significance for chemistry?

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