

Review

Funding activities by the German Federal Environmental Foundation (Deutsche Bundesstiftung Umwelt) in the field of sustainable chemistry

Maximilian Hempel

German Federal Environmental Foundation (Deutsche Bundesstiftung Umwelt), An der Bornau 2, 49090, Osnabrück, Germany, e-mail: m.hempel@dbu.de

Abstract

Sustainable chemistry is the attempt to design chemical products and processes that reduce or eliminate the use and generation of hazardous substances, minimize waste and energy consumption, favor renewable resources and integrate aspects of recycling. Since 2004, the Deutsche Bundesstiftung Umwelt (German Federal Environmental Foundation) supported more than 70 projects in the area of “Sustainable Chemistry”. In this paper, current results and approaches of these sponsored projects are reviewed, e.g., substitution of fossil raw materials and biorefinery concepts, catalytic processes, novel solvent systems including solvent-free synthesis. The application of microstructured equipment and chemical reaction under novel process regimes, i.e., so-called “novel process windows” are summarized, and a brief look at concepts for inherently safe chemicals, new business models, e.g., chemical leasing and early stage eco-balancing, is given. It is revealed that industrial sustainable chemistry is no longer an emerging trend but already a reality.

Keywords: chemical leasing; chemical processing; novel process windows; solvent-free; sustainable chemistry.

1. Introduction

In 2004, the Deutsche Bundesstiftung Umwelt (DBU, German Federal Environmental Foundation) decided to institute the area of “sustainable chemistry” as a core funding issue. Since then, the DBU has supported more than 70 projects in this area. In this paper, current results and approaches of these sponsored projects are reviewed. The projects are divided into the following areas: (i) substitution of fossil raw materials and biorefinery concepts, (ii) catalytic processes, (iii) novel solvent systems including solvent-free synthesis, (iv) the application of microstructured equipment and chemical reaction under novel process regimes, (v) concepts for inherently safe chemicals, and (vi) new business models, e.g., chemical leasing. Most of the project results were acquired in cooperation between research institutes and small- and medium-sized enterprises.

2. Deutsche Bundesstiftung Umwelt (DBU)

The DBU is among the world’s largest environmental foundations; since its founding, nearly 7900 projects have received financial backing totaling approximately €1.4 billion, with a particular focus on small- and medium-sized enterprises.

A model for the promoting activities of the DBU is sustainable development. At the United Nations conference in Rio, 179 countries committed themselves to this model by signing an agenda for the 21st century. The concept of sustainable development requires sustained strategies, e.g.,

- by minimizing consumption rates of transitory resources through an improvement of efficiency, substitution of fossil resources by renewables and by recycling (life cycle assessment),
- by preventing the consumption rates of renewable materials and energies from exceeding their given rate of reproduction, and
- by preventing emissions from exceeding the capacity for regeneration and absorption by environmental media and human beings.

The DBU has two ways of support: with personal subsidies (postgraduate scholarships) and with subsidies for companies and institutes (cooperation projects).

2.1. Postgraduate – scholarships at German Universities

The DBU has established a German scholarship program to promote promising young scientists in the field of environmental protection, providing per annum approximately 60 scholarships for postgraduates from different disciplines – mostly in chemistry, biology and engineering.

2.2. Cooperation projects between institutions and small- and medium-sized enterprises

The DBU particularly encourages cooperation projects between small- and medium-sized enterprises and research institutions. The three main criteria for obtaining a subsidy are:

- Innovation: the project has to promise an advance compared with the state-of-the-art of current research and technology.
- Exemplary and model character: the innovation should be of interest to a broad segment of the players (e.g., a complete

industrial sector). It should also be possible to implement the innovation under commercial conditions within a brief time scale.

- Environmental benefit: the innovation should lead to new, supplementary measures for the protection of the environment.

The actual fields of promotion are laid down in the guidelines with nine special topics. One field of support is sustainable chemistry – procedures and products, aiming at promoting projects with the following objectives:

- Development of chemical techniques to convert renewable and waste materials to new products and materials.
- Development of new applications of micro- and nano-techniques (e.g., micro-reaction for synthesis).
- Development of optimized chemical processes, e.g., with new catalysts or separation techniques.

Projects in this field usually include a life cycle assessment or an eco-efficiency analysis. The decision to fund largely depends on the concrete contribution of the project towards environmental benefit. Within these projects, the DBU supports additional measures for dissemination and consolidation project results. Since 2004, the DBU has supported more than 70 projects in the area of “sustainable chemistry”.

3. Sustainable chemistry

The concept of sustainability is an integral part of public debate. The term is derived from the historical forestry in Germany in the 18th century, where sustainable forestry was the answer to the plight of forest destruction [1]. In 1987, the United Nations published the Brundtland Report, which included the following definition: “Sustainable development is development that meets the needs of the present without compromising the ability of future generations to meet their own needs” [2]. Here, the term is currently undergoing a variety of use and interpretation. In essence, sustainable development aims to meet human needs while preserving the environment so that these needs can be met not only in the present but also for generations to come [3]. To transform this general approach into practice for the chemical sector, several attempts have been made. There are different approaches and initiatives for sustainable chemistry, e.g., the 12 principles of green chemistry, developed by Anastas and Warner [4], the Suschem – Initiative of the European Chemical Industry and Chemical Societies [5–11]. Today, most of the international chemical companies have set environmental (and social) targets showing a high affinity for sustainable chemistry.

3.1. Substitution of fossil raw materials and biorefinery concepts

Approximately 90% of global oil production is not used as material resource, but for energy demands. In 2008, approximately 2.7 million tons of renewable raw materials are used in the German chemical industry, i.e., ~13% of organic raw

materials are renewable resources. Another 0.9 million tons of renewables are used in the German paper industry [12]. The main renewable resources are oil and fat, starch, cellulose and sugar. In 2004, the global demand for oils and fats was approximately 131 million tons, and in 2006 it was 265 million tons. Approximately 80% is used for food, 11% for chemical industry, and 9% for biofuels. It is obvious that the chemical industry will predominantly utilize non-food biomass, such as wood, waste material/byproducts of agro and wood industry as well as food and feed industry. The economical and environmentally friendly use of “non-food” biomass, however, requires advances in logistics, in the pretreatment and processing of raw materials and new approaches in chemical and biotechnological processing and conversion.

There are two basic options for the use of “non-food” biomass. On the one hand, component separation and subsequent biochemical and chemical processing of individual components, and on the other hand, thermo-chemical degradation (e.g., by gasification or pyrolysis), followed by classical refining, known from petro-based processing. In both cases, processing is of outstanding importance.

Developing biorefineries represents a key issue to the way forward to a more sustainable chemical industry and includes the integrated production of animal feed and chemicals, materials and biofuels (Figure 1). The availability, quality and price of biomass will play a decisive role for the industrial use of raw materials in biorefineries. The three main biorefinery concepts, using lignocellulosic biomass as feedstock, are: (i) lignocellulosic biorefinery, (ii) green biorefinery, and (iii) synthesis gas biorefinery.

One of the major challenges for the refining of lignocellulosic material and in particular for a profitable conversion to chemicals is the development of chemical catalysts and the search for suitable enzymes, yeasts or microorganisms. Further on, for all biomass conversion routes it is essential to develop effluent-free manufacturing.

Today, typical products in the chemical sector are paper, washing and cleaning agents, coatings, paints and polymers, as well as lubricants. Challenges arise by reason of limited land resources and the competitive production of agro products for energy (bio-fuel), chemicals (renewable resources) and food. In addition, some bio-based products, e.g., biodiesel show in their life cycle assessment a bad efficiency in terms of land and water use and greenhouse gases. Thus, there is a further focus on residues, e.g., from food production, integrated biorefinery systems and the cascaded use of renewable resources in material and energy flows. Chemists know how to synthesize fuels and chemicals from low molecular fossil resources. Renewable resources usually contain a number of high molecular hydrocarbons. Thus, it is a challenge for the chemist to develop methods to downgrade the hydrocarbons without losing the synthesis performance of nature. A very rough way is thermal depolymerization or hydrothermal liquefaction, where superheated water is used to downgrade the lignocellulosic fraction. Another attempt to turn various biomass waste into coal is hydrothermal carbonization. The process mimics the natural process of coalification in a few hours [14–17].

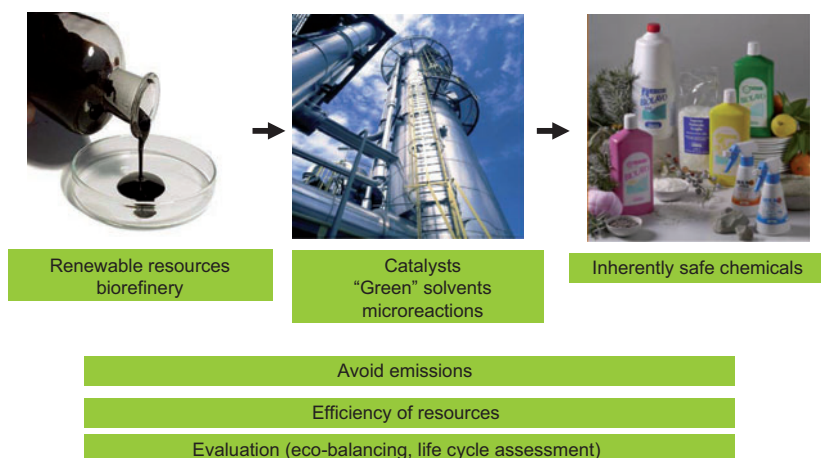


Figure 1 Pathway to a more sustainable chemical industry [13].

Use of the complex biobased molecules offers the chance to synthesize chemicals more efficiently than from fossil low molecular resources. In this context, future challenges for chemistry, biotechnology and process engineers are: (i) to develop methods to transfer lignocellulose material to bulk chemicals and bulk polymers, and (ii) to create biobased polymers with better performance and lower ecological impact.

3.2. Catalysts

In this field, catalytic processes are a key issue. Catalysis is a central technology in chemical industry: approximately 80% of current chemical processes make use of it. The main application fields are environmental protection, chemical and oil processing refineries and polymers. Catalysts are of outstanding importance for sustainability and profitability of chemical production processes: they enhance the selectivity of a synthesis and thus reduce waste, emissions and byproducts. Lower activation energy of a reaction scales down energy consumption. Identifying and developing selective working catalysts for relevant processes in chemical industry is a key issue for sustainable chemistry. The German Catalysis Society recently published a roadmap, summarizing seven main issues for a sustainable economic growth [18]:

1. Securing supplies of raw materials (crude oil, natural gas, coal, renewable materials and carbon dioxide usage).
2. Securing energy supply (fuels of the future, hydrogen production and storage, fuel cell technology, electro-catalysis).
3. Health and nutrition (active ingredients, human food products and animal feed, fertilizers).
4. Protection of the environment (catalytic water and waste water purification, treatment of industrial gases, waste gas treatment, catalytic combustion).
5. Material and energy efficient processes (production of monomers, tailor-made polymer and functional materials, conversion of synthesis gas to chemicals, fine chemicals).
6. New reactor concepts (multifunctional reactors and hybrid processes, microstructured reactors and new reaction media).

7. New analytical and preparation techniques (nano-structured catalysts, high-throughput experimentations, *in situ* methods, modeling).

From a sustainable chemistry point of view, special efforts are required in the areas of:

- Catalytic and chemical routes for the usage and separation of agriculture crops.
- Water and biocompatible catalysts and “mild” processes to transfer biomass with high efficiency, activity and yield to bulk chemicals.
- Converting renewable resources into chemicals, e.g., de-functionalization of cellulose, hemicellulose and chitin.
- New catalytic routes with a high eco-efficiency for bulk chemicals (e.g., propylene oxide, polyurethane) as well as pharmaceuticals and fine chemicals.

3.3. Solvents

Most of the chemical reactions are performed in the presence of a solvent. The world market for solvents is up to 17.9 million of tons per year, only in Europe are 4.5 million of tons merchandised.

Steinbach et al. [19] recently evaluated more than 400 processes in the chemical sector, mostly located in Germany (Table 1). The numbers correspond with earlier publications from Sheldon [20], who introduced an E-factor, which is the amount of waste produced in the process, defined as everything but the desired product. Whereas oil refining has an E-factor of <0.1 kg waste/kg product, fine chemicals finish up with an E-factor of 5 to >50 and pharmaceuticals with 25 to >100. Sheldon [21, 22] summarizes not only water and solvents but also other waste streams and byproducts, which partly explains the differences.

Organic solvents could have an impact on environment and health, as they often have an eco-toxicological potential. Chlorinated organic solvents show persistent and bio-accumulative characteristics. In addition, volatile organic compounds play a vital role in the formation of ozone. Novel

Table 1 Consumption of solvents and consumption of water in different branches in kg per ton product [19].

(kg/t product)	Consumption of solvents	Consumption of water
Pharmaceuticals	3200	5400
Dyes	700	71,200
Pesticides	250	6400
Fine chemicals	100	1500
Bulk chemicals	0	1900

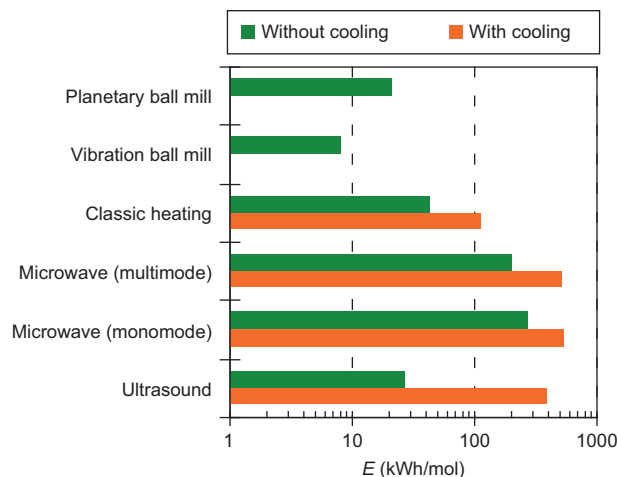
solvent systems as less harmful solvents, supercritical fluids, ionic liquids, solvent-less and -free processes may help to achieve environmental benefit.

Supercritical fluids (sc), as scCO_2 or scH_2O , are non-toxic, non-flammable and show interesting solvent agents, comparable to organic solvents. In addition, CO_2 is widely available and cheap. A disadvantage is the need of high-pressure and consequently costly pressure-resistant equipment and pressure work. Attempts of a CO_2 -recycling within a pressurized mode may overcome this drawback.

Industrial applications are known in the field of decaffeination, (chemical) cleaning of textiles, extraction of vitamins, sterilization of medical products and others.

Ionic liquids (ILs) are liquid compounds, containing essentially anions and cations. The attraction of the young substance group is partly due to its great variety of combinable anions and cations. ILs are electrically conductive and have extremely low vapor pressure. Currently, they are very common and used fairly diversely in industry. One of the first industrial applications of ILs was the BASIL process, using a 1-alkylimidazole to scavenge the acid from an existing process. ILs can dissolve cellulose and improve cellulosic fiber production and help to functionalize it. Other applications are known for gas transport. Some gases may be stored and transported in ILs, relying on their solubility in ILs. Other gases are insoluble in ILs, e.g., hydrogen, a property used for transportation of hydrogen, with an IL body replacing the piston. ILs also serve as homogeneous catalysts in reactive distillation, and as scavengers for separating sulfur from petroleum and natural gas. Owing to their electrical conductivity, ILs also suit the electroplating industry. Aluminum- and iron-electroplating has been technologically advanced. In the future, ILs may substitute perfluorinated compounds in this industry. Despite their non-volatility they are not necessarily environmentally benign. Actual results show that many of them are persistent and quite a few also are bio-accumulative and/or toxic [23].

With regard to solvent-free synthesis, the attempt to perform a chemical reaction without a solvent may be called a radical innovation. The Stolle and Ondruschka group [24–26] developed an elegant way for a solvent-free dehydrogenation synthesis in a planetary ball mill through direct oxidation of anilines to the corresponding azo and azoxy homocoupling products (Figure 2). This novel synthesis route without solvents shows some impressive advantages: it has a high energy efficiency, the product is not contaminated

**Figure 2** Energy efficiency for synthesis of azo dye from aniline with different methods of energy entry [24].

by a solvent, the risk of accident is reduced and upscaling is possible. Nevertheless, prior to a transfer to an industrial process, several objectives have to be pursued, e.g., the harmonization of reaction variables, a credible temperature and pressure control, and finally a continuous chemical process design [5, 6, 8].

3.4. Microstructured equipment

A higher level of automatization, the use of energy control systems and computer-aided simulation tools are frequently used items to optimize chemical processes. By changing the process from batch to continuous mode, the efficiency and safety of the process could be enhanced. Micro- and milli-reactions and fluidic systems can help to control exothermal reactions and enlarge the space time yield and energy efficiency. A Dutch government program has investigated the potential for process intensification in the Dutch chemical industry, leading to a “roadmap for process intensification” [27].

The application of micro-fluidic components and micro-reactors can offer advantages [28, 29]. One characteristic feature of micro-reactors is their significantly higher specific surface as compared with conventional reactors. Thus, heat transfer coefficients are very high and allow an extremely efficient heat exchange [30]. Microstructured reactors are hence especially suited for highly exothermic reactions conducted in a practically isothermic manner. A high surface-to-volume ratio also leads to higher yield and improved selectivity during the formulation of a chemical compound. Microstructured equipment can be used in a wide range of reactions, e.g., liquid, gas, gas-liquid, solid or even multiple phase reactions [30]. For production, a micro-reactor could be numbered up by simple replication, as demonstrated by Kralisch and Kreisel earlier [31]. Life cycle assessments show that the use of micro-reactors can involve a reduction of waste, emissions, needed feedstock and an increase of safety, product quality and yield.

3.5. Novel process windows

Actual progress in the field of micro-reaction and catalysis enables chemical reaction under novel process regimes. In these scenarios, temperature and concentration levels are feasible, which may lead to new and sustainable chemical processes. Pressure-resistant micro-reactors are essential to perform and control these new continuous chemical processes.

Micro- and milli-process technology enables new operations in process engineering. The main issues are minor volumes in the reactors, speeding up the heat transport and permitting operations under extreme conditions [32, 33]. Despite these advantages, experiments in the past also revealed reactions which were not suitable for micro- and milli-process technology as they were too slow or because plugging occurred. By contrast, a temperature rise can speed up the reaction in manifold ways and new solvent concepts could change the solubility dramatically. Sometimes, intermediates are instable and have to be separated very quickly. In addition, sometimes temperatures above the boiling point are required. Novel process windows is of benefit for these new technologies, and thereby far from former laboratory experience (Figure 3).

In 2007, the DBU initiated the program “novel process windows”. The aim of this program was to realize new continuously working chemical processes, which contribute to a more sustainable chemical industry and strengthen its innovation capacity in Germany. The projects are geared to develop chemical processes with high energy efficiency, minimized emissions, minimized waste output, inherent safe products, and enhanced space time yield.

The support program with its projects focuses on:

- high-temperature/high-pressure amination of hydrocarbons in continuous processes,
- high-temperature/high-pressure synthesis of chitosan from chitin in continuous processes,
- process intensification of the Kolbe-Schmitt synthesis by using novel process windows,
- using novel process windows for the solvent-less production of ink and highlighter,
- direct synthesis of hydrogen peroxide from hydrogen and oxygen in a membrane micro-reactor,
- anionic polymerization of styrene to polymeric organic semi-conductors in a micro-reactor,
- novel process windows for a safe synthesis of perester from butyl-peroxypivalate, and
- thermal catalytic defunctionalization of carbon hydrates.

The cluster “novel process windows” includes eight projects, started in 2007; most of the projects ended in 2011 or will end up soon. It covers project costs of €4.5 million with a subsidy



Figure 3 In 2007, the DBU initiated the program “novel process windows”.

of €2.2 million. Results of these projects are highlighted in several publications [34–40].

4. Inherently safe chemicals and the sustainable use of chemicals

The sustainable use of chemicals tends to provide socially necessary products while optimizing resource and energy efficiency, minimizing substance losses and controlling or better avoiding emissions and exposures. With regard to an evaluation, if a chemical is sustainable, the following criteria could help at first [41]:

- mentioned in a list of “problematic substances”,
- dangerous physicochemical properties,
- human toxicity,
- problematic properties related to the environment,
- mobility,
- origin of raw materials,
- emission of greenhouse gases, and
- resource consumption.

Higher security in the chemical industry is achievable by less dangerous chemicals, e.g., inherent safe chemicals. Inherent properties of a chemical are immanent physicochemical properties of the substance. Inherent safe chemicals only require the basic proper handling of chemicals. For example, from a viewpoint of occupational safety only those substances are inherently safe, which are classified as not hazardous. From an environmental point of view, inherently safe chemicals will not cause any short- or long-term problems after released into the environment. The dominating criteria for inherently safe chemicals are persistence, bioaccumulation and toxicity.

5. New business models: contracting and chemical leasing

Future scenarios predict that a great part of our economic growth will be based on services. Currently, the chemical industry implements service-orientated business models offering ecologic and economic benefits. Several models are conceivable, e.g., contracting and chemical leasing. Chemical leasing is a business model with a chemical producer supplying a product for a specific surface, e.g., coating or dyeing, with the ownership remaining with the chemical producer (Figure 4). This model shifts the focus from increasing sales volume towards a value added approach. The producer mainly sells the functions performed by the chemical and is paid for the functional units [43].

Some applications in the field of cleaning in the metal industry, in powder coating and catalyst use demonstrate the reduction of raw material charge and waste. Actually, ongoing projects, funded by the DBU, are verifying the potential of economic and ecologic savings by chemical leasing in the wood industry and printing industry. First results indicate that in the wood and wood processing industry existing standards

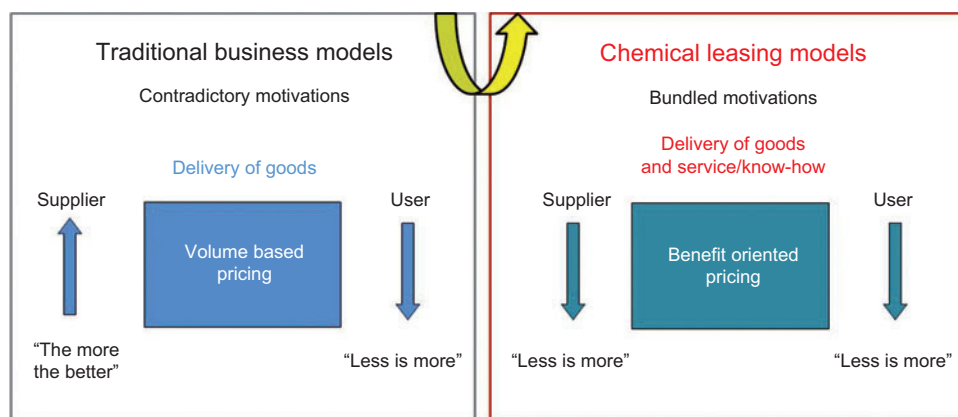


Figure 4 Concept of chemical leasing [42].

hinder the implementation of chemical leasing concepts (DBU Report 28210).

6. Early stage eco-balancing

How to design chemicals as environmentally benign as possible? The early stage of a development yields the largest ecological and sustainable optimization potential. If eco-toxicological aspects are considered during the development of a new chemical synthesis and processes, then chemicals with inherent safety and eco-efficient processes may result [44].

Nevertheless, there is a lack of a fast and easy-to-use software support for chemical and process engineers. The aim of an ongoing project is to overcome this deficiency and help estimate the ecological effects and costs in early stages of synthesis planning or development processing. IFU GmbH is developing a fast and easy-to-use software support for chemical and process engineers in cooperation with the University of Emden and ETH Zurich. With this software, the expected environmental impacts and costs are quantified in the early stages of planning the synthesis, process development and within existing processes. This helps to identify optima and environmentally friendly and cost-effective process routes (DBU report 25070).

Preiss et al. [45] and Cho et al. [46] suggest a general rule to predict the physicochemical property of new chemicals by using a structure-response relationship. The authors choose ILs to verify their hypothesis. The results are fairly promising. With the developed equation it is possible to predict the critical micelle concentration and other properties with good agreement with experimental results.

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Maximilian Hempel is Head of the Department of Environmental Chemistry for the German Federal Environmental Foundation (DBU). His main fields of interest are green and sustainable chemistry, environmental technology, ecotoxicology, green pharmacy, innovation management.