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Economically and environmentally sustainable closed-loop chemical recycling of polyurethanes has been a goal of the polyurethanes industry for many years.

This paper shows how the ultimate recovery and recycling have been integrated into the design of a novel polyurethane cushioning material which still meets all the performance requirements of today. Recycling was one aspect of the product design process which considered also the environmental impact of the whole life-cycle, from raw material manufacture, product fabrication, use and recycling to ultimate disposal.

The practical implementation of closed-loop recycling involves more than chemistry. In this case, it requires commitment from furniture designers and manufacturers. Above all, it requires new processes for collection, dismantling and separation which become broader social responsibilities.

1. Introduction

Nearly four years ago, governments worldwide endorsed the concept of 'sustainable development' at the UN Conference on Environment and Development in Rio de Janeiro. The conference, which came to be known as the Earth Summit, brought to public attention the fact that enormous growth in world population together with rising affluence is taking place on a planet that is finite, both in size and in availability of resources (Sitarz 1993).

For example, since that conference took place the world population has grown by over 350 million people, equivalent to a new unified Germany in each subsequent year. To satisfy the world's growing timber demand, 23 million hectares have been logged, an area the size of the UK, and the global average temperature has risen by approximately 0.3 °C, although it must be stressed that year-to-year changes are erratic and difficult to interpret. Nevertheless, the atmospheric concentration of carbon dioxide, which will largely determine the scale of the greenhouse effect, has risen from 352 parts per million to 359, an increase of half a percent per year (Brown *et al.* 1995).

On the other hand, and despite the fact that oil production has also been rising in the last four years to keep up with a growing world economy, global emissions of nitrogen have been stabilized while those of sulphur have even fallen slightly. The most important reasons for the slow down in global nitrogen and sulphur pollution are improvements in industrial technologies and the international treaties and domestic legislation that encourage manufacturers to use these better, cleaner technologies.

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2. Environmental criteria for product development

The power of sustainable development as a framework within which the world can address its environmental and economic problems, and a driver of cleaner technologies, has grown strongly. The concept of sustainability reconciles three fundamental global issues: environmental protection, economic growth and social well being. From a business point of view, it means that the companies which will survive are those which fulfil the needs of consumers without compromising their environment and quality of life (UNWCED 1987).

In order to help companies achieve this, the World Business Council for Sustainable Development (WBCSD) has introduced the concept of 'eco-efficiency' as a means for companies to continuously add value while constantly reducing energy and material use, pollution and waste. Following the WBCSD terminology, there are seven core elements of eco-efficiency which need to be addressed over the entire life-cycle of goods and services. Expressed simply they are: (1) reduce material intensity; (2) reduce energy intensity; (3) enhance material recyclability; (4) maximize sustainable use of renewable resources; (5) extend product durability; (6) increase service intensity; and (7) reduce toxic dispersion (WBCSD and UNEP 1996).

The two central themes which emerge from the above and are now well accepted in environmental product development are dematerialization, using less material, and detoxification, using less harmful materials.

Moreover, it is now evident that product design which reduces the negative environmental impact of the product must encompass its whole life-cycle. Consequently, life-cycle design (LCD), design for environment (DFE) and related initiatives based on product life-cycle have emerged as systematic approaches for integrating environmental issues into product and process design (Keoleian & Menerey 1994).

Until recently, the life-cycle framework has been much more widely associated with environmental assessment than design. Life-cycle assessment (LCA), resource environmental profile analysis (REPA) and ecobalances are all methods which attempt to evaluate the life-cycle environmental consequences of a product from cradle to grave. Although LCA is considered to be an evaluation tool and not a design method, ICI believes that a simplified method for life-cycle assessments would be an invaluable aid to environmental product development and is actively involved in the development of an industry-standard methodology of this type.

A good example of this approach is provided by the recent development by ICI of a novel, recyclable polyurethane furniture cushioning material called 'Waterlily'.

First, however, it is worth briefly reviewing polyurethanes.

These versatile plastic materials are made from the reaction of two principal chemical components: an isocyanate and a polyol. Relatively few standard isocyanates and a wide range of polyols of different molecular weights and functionalities are used to produce the whole spectrum of polyurethane materials. These can vary in density from 6 to 1200 kg m^{-3} and in polymer stiffness from very flexible elastomers to rigid, hard plastics.

Over the last 40 years, polyurethane materials have been used in an ever increasing range of applications most of which are part of everyday life, for example, automotive steering wheels and dashboards, insulation for refrigerators, footwear soles, insulation for buildings and in furniture upholstery and mattresses. The furniture and mattress market constitutes around 50% of the world polyurethane consumption and is mainly based on an isocyanate called toluene di-isocyanate or TDI. Most of the other applications have been developed using another isocyanate of lower volatility called MDI or diphenyl methane di-isocyanate.

Table 1. *Life-cycle concept of flexible polyurethane foam slabstock*

	energy consumption	key environmental issues
raw material manufacture	−80%	climate change, acidification, water pollution and toxic chemicals
product fabrication	−5%	VOC, ozone depletion and waste
packaging and transport	−5%	climate change and acidification
use	0%	(consumer safety)
waste management	−10%	recyclability, volume and toxicity of waste

3. Redesigning mattresses

The initial objective in the 'Waterlily' project was to design a flexible foam which did not use any organic volatile solvents in production, and which replaced TDI by the less volatile MDI to improve workplace air quality. Flexible foams are materials of densities 10–80 kg m^{-3} composed of lightly cross-linked polyurethane with an open-cell structure. Essentially flexible and resilient padding materials, flexible foams are produced as continuous slabstock or as individually moulded cushions and pads.

However, it rapidly became clear that a more appropriate broader objective was to develop a product with reduced environmental impact during the whole life-cycle. The first step was therefore to undertake a broad-brush analysis of the main environmental impacts during the life-cycle of existing polyurethane slabstock (see table 1). The life-cycle of the product was divided into five main phases: raw material manufacture; product fabrication; packaging and transport; use; and, lastly, management of waste. An estimate of the energy consumption and the key environmental issues was made for each phase.

It was evident that nearly 80% of the total energy consumption for the product had already occurred by the first stage, namely from drilling the oil out of the ground up to the manufacture of the main raw materials. However, the analysis of the key environmental issues in the form of air and water pollution and solid waste did not point out clearly which phase of the life-cycle carried the greatest environmental burden. Instead, each phase had a different set of environmental problems associated with it and no universally accepted guidelines exist which placed these problems in terms of importance.

This is a difficult challenge for a typical research department of a manufacturing company trying to implement the principles of eco-efficiency or design for environment. Invariably, when dealing with one technical or environmental problem, a trade-off has to be made with another. Consequently, a set of tools or guidelines is needed to help focus the effort on the important issues.

In this project, the main benefit of such an exercise was to draw attention to the importance of the raw material manufacturing stage and hence focus effort on a design for recycling strategy.

Table 2. *Life-cycle design criteria for 'Waterlily' comfort cushioning*

raw material manufacture	less volatile raw materials simpler formulation
product fabrication	no isocyanate vapour no organic blowing agents no autocombustion
packaging and transport	returnable and recyclable minimum environmental cost of transportation
use	improved fire performance no halogens comfort performance
waste management	designed for chemical recycling suitable for physical recycling desirable feed for energy recovery

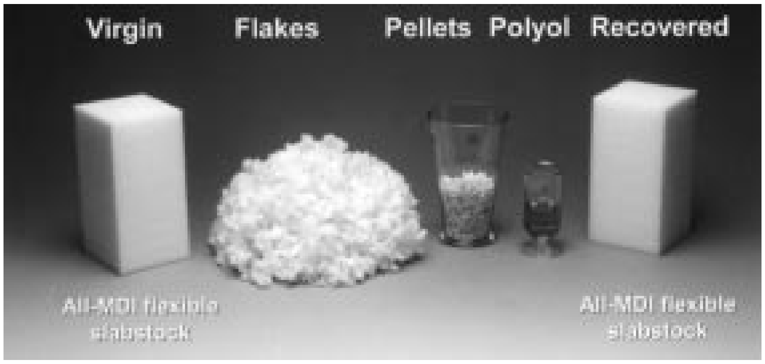

Figure 1. Recycling 'Waterlily' comfort cushioning.

4. Key safety, health and environmental requirements

As a second step in the project, various stakeholders were contacted along the product chain in order to verify the assumptions made and to define a set of requirements for each phase of the life-cycle.

Their main requirements are listed in table 2.

For the raw material manufacture stage, the main issues were the use of a less volatile isocyanate and the design of a simpler formulation.

At the product fabrication stage, the main requirements were no measurable isocyanate vapour in the workplace, no organic blowing agents and elimination of the possibility of autocombustion.

The packaging material should be returnable and recyclable and the environmental impact of transporting low density foam should be minimized.

The issues associated with the use of the product were primarily centred on improved fire performance without the use of halogenated fire retardants and comfort performance.

Once again it was interesting to note the importance of the management of the waste. Various stakeholders felt that the waste arising from both production and post consumer should be designed for chemical recycling, suitable for physical recycling or, as a last option, a desirable feed for energy recovery.

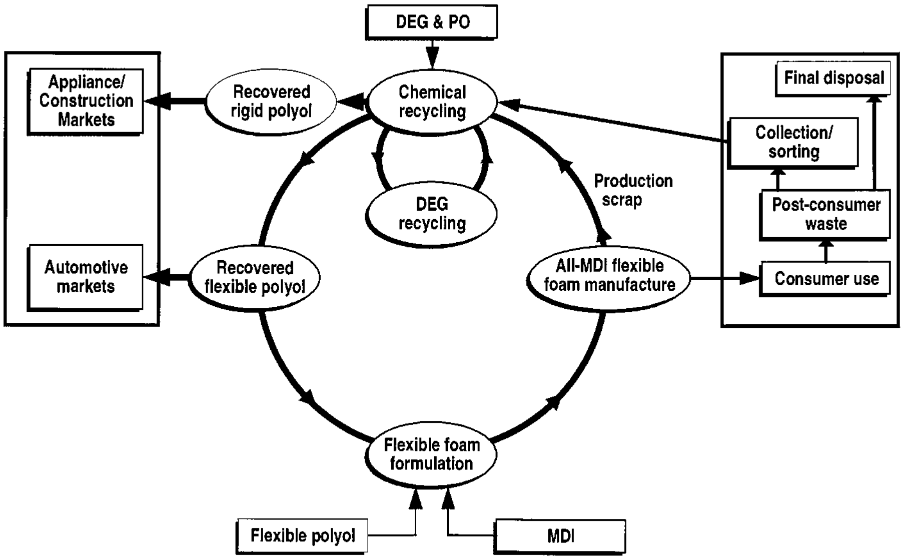

Figure 2. Schematic of closed-loop recycling of flexible polyurethane foam.

5. The importance of recycling

The management of the waste in the flexible foam industry has been a long standing issue primarily due to the large volume of production scrap which is generated when the slabstock is cut to shape for furniture upholstery and to a lesser extent mattresses (Hicks & Krommenhoek 1994). Historically, it has been exported from Europe to the United States of America, where a large market exists for rebonding scrap chips into carpet underlay.

There are also other methods for recycling flexible foam. Physical recycling consists of the pulverization of flexible foam to a fine powder which subsequently can be used as a filler. This filler is then added to the polyol component and used to produce flexible foam.

An alternative method, known as feedstock recycling, involves breaking down the plastic into basic chemicals, plastics monomers or hydrocarbon feedstocks. It is usually carried out on mixed plastic waste and the products are used as raw materials in the petrochemical industry.

Finally chemical recycling can be used to depolymerize polyurethanes and recover those materials which can be reused (figure 1). Several processes are applicable to polyurethanes, including hydrolysis, hydrogenation, aminolysis and glycolysis. These processes vary in terms of the requirements of quality of the polyurethane feed, the process complexity and the quality of final products generated.

However, there are several limitations associated with each of those recycling methods.

Firstly, there is concern that the supply of foam scrap into the United States may exceed the capacity of the rebond market to absorb it. Historically, the market for production scrap has been highly volatile and there is a belief that price levels of scrap will continuously decline.

Secondly, there are two problems associated with physical recycling: (1) the incorporation of filler in the formulation often affects the physical properties of the foam; and (2) the filler affects the viscosity of the formulation and hence only a limited amount, approximately 10–20%, can be added.

Thirdly, the presence of nitrogen in polyurethanes reduces their value in feedstock recycling which is ideally suited to polyolefins. Halogenated additives are perceived to create toxic species when the foam is incinerated to recover energy.

Considering the environmental burden associated with the raw material manufacturing stage and the limitations of the current methods of managing the waste, it is clear that the ideal solution would be closed-loop recycling of flexible polyurethane foam (figure 2). In other words, the challenge is to design a flexible foam which can be depolymerized into high value raw materials, at least one of which can be reused in the same application. This difficult challenge placed a new set of demands on ICI Polyurethanes and it decided to pursue a completely new chemical approach, in developing 'Waterlily' comfort cushioning.

6. Design for chemical recycling

The urethane group is produced by reacting an isocyanate group together with a polyol. If this reaction is carried out in the presence of water then the isocyanate reacts with the water to form carbon dioxide which acts as a blowing agent and expands the polymerizing material to form a foam. In addition, the reaction of water with the isocyanate produces heat and forms substituted polyureas. In order to produce an MDI-based flexible foam with acceptable properties, it is vitally important to control precisely the balance between the polyurethane and polyurea reactions.

In this project, this was achieved by reacting a special blend of MDI variants together with high reactivity polyols to form two novel intermediates or prepolymers. These, in turn, were reacted in the presence of water and a carefully selected catalyst to produce the foam.

Throughout the development of the 'Waterlily' formulation, the greatest technical challenge was to maintain the principles of eco-efficiency. For example, no additives were permitted which could in any way increase the environmental burden and complicate the recycling process. Similarly, no tin-based catalysts were used, nor were there any ozone-depleting blowing agents or halogenated fire retardants.

In addition, this novel flexible foam had to meet all the market requirements in terms of physical properties relating to comfort performance and durability.

In parallel with the research of the novel chemical formulation, the process technology for chemical recycling was being developed. The approach taken for recycling 'Waterlily' comfort cushioning was to aim for high quality, and therefore high value products. This meant controlling the composition of the feed as well as the process such that flexible polyol and isocyanate components could be separated and purified, and such that lower value mixtures of flexible polyols were avoided.

A process known as split-phase glycolysis was developed around the chemical formulation used for 'Waterlily' cushioning.

The feedstock for this chemical recycling process was therefore, restricted to only positively identified formulations of polyurethanes. The suitability for recycling of each new formulation of 'Waterlily' comfort cushioning by split-phase glycolysis was verified at laboratory scale.

In addition, recognizing the environmental and practical difficulties and costs of transporting low density foams, a compaction process was developed to convert 'Waterlily' comfort cushioning with densities as low as 30 $\mathrm{kg\ m^{-3}}$ into solid pellets with a bulk density of 500 $\mathrm{kg\ m^{-3}}$. This process was based on a standard design of a pellet mill.

The split-phase glycolysis process can be carried out in chemical plants consisting

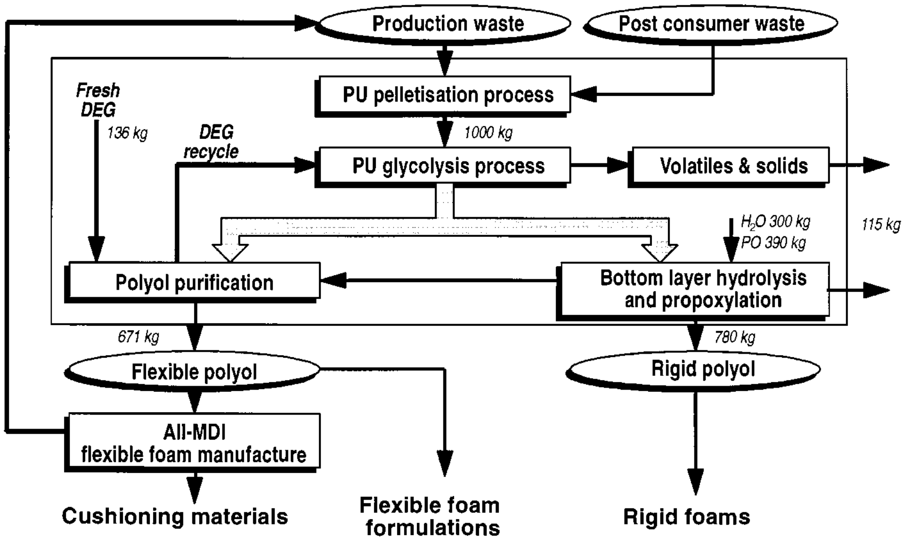

Figure 3. The split-phase glycolysis process.

of standard equipment, e.g. batch reactor and several storage tanks as shown in the process schematic in figure 3. The glycolysis reaction is carried out by charging compacted pellets made of 'Waterlily' comfort cushioning into a stirred batch reactor containing di ethylene glycol (DEG). A catalyst is added and the reactor is heated to 200 °C. Typically, the pellets dissolve within one hour and the reaction reaches equilibrium within two hours.

At this stage, the reactor contains a mixture of DEG, flexible polyol and reaction products from the isocyanate groups. The agitator on the reactor is stopped and the reaction mixture allowed to separate into two layers. The top layer consists of DEG and flexible polyol with a small quantity of impurities. One of the key requirements for designing 'Waterlily' comfort cushioning for recycling was the solubility of the flexible polyol in DEG which allows the separation of the polyol from other species in the foam. The bottom layer consists primarily of DEG and aromatic compounds derived from the isocyanate in the foam.

Since the goal is to produce recycled materials of equal quality to virgin materials, a further purification is carried out. The top layer is washed with more DEG, either in the batch reactor or in a liquid/liquid extraction column. After the final wash, DEG is removed by vacuum stripping to give pure flexible polyol.

Consequently, the flexible polyol obtained after split-phase glycolysis can be used to completely replace virgin polyol in 'Waterlily' comfort cushioning prepolymers. This results in a recycled content in the final foam of up to 70%. The recycled polyol has a lower level of unsaturation than virgin flexible polyol (see table 3). This low unsaturation is responsible for the improvement in humid and dry compression sets in foams made using only recycled polyol over foams made using virgin polyols. There is, however, a reduction in mean tear strength of foams made using recycled polyol (figure 4).

The mixture contained in the lower layer cannot be separated into the original isocyanates and therefore a different approach is followed. Steam is added to the bottom layer to form an aromatic amine in DEG which can be used as the precursor for a polyol used in rigid foam for insulation.

Split-phase glycolysis uses only small quantities of readily available chemicals.

Table 3. *Properties of flexible polyol produced in split-phase glycolysis process*

test	recycled polyol	virgin polyol
appearance	clear/straw coloured	clear/water white
OH value (mg KOH g^{-1})	27	28
viscosity (mPa s at 25 °C)	1355	1250
acidity (mg KOH g^{-1})	0.01	0.05
water (%)	0.02	0.05
unsaturation value (meq g^{-1})	0.013	0.080
DEG (%)	0.06	0.00

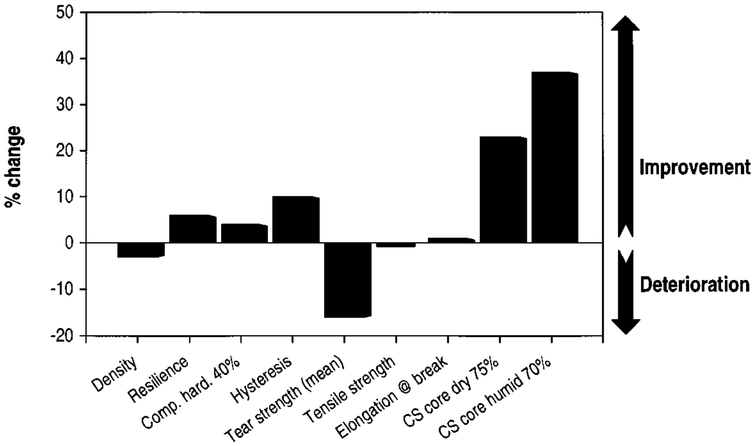

Figure 4. Energy requirements for recycling ‘Waterlily’ comfort cushioning.

The product to feed ratio was 1:1.5. The raw materials used per tonne of waste were 136 kg of DEG, 390 kg of PO, and 0.5 kg of catalyst. Any DEG which was stripped off during the process was suitable for reuse. Contaminated DEG from washing the flexible polyol could be used for the subsequent glycolysis batch. Each tonne of dry polyurethane yielded 671 kg of flexible polyol and 780 kg of rigid polyol. The yield of flexible polyol was around 90% of the theoretical value. The estimated energy consumption for the chemical recycling process is 5 MJ kg^{-1} polyurethane. This is roughly between 6 and 7% of the current estimates for the energy required to manufacture virgin polyol from oil.

7. The design check using LCA

In order to check that chemical recycling was indeed the optimum method of dealing with production and post-consumer waste, a full life-cycle inventory of the various recycling options was carried out for the case of a mattress containing ‘Waterlily’. The scope of the study included the manufacture of the slabstock, the production of the mattress, the collection of the used mattress in a segregated collection scheme and four different methods of waste management: chemical recycling; physical recycling; incineration with energy recovery; and rebonding of the foam. A brief summary of the results in terms of energy consumption is shown in figure 5.

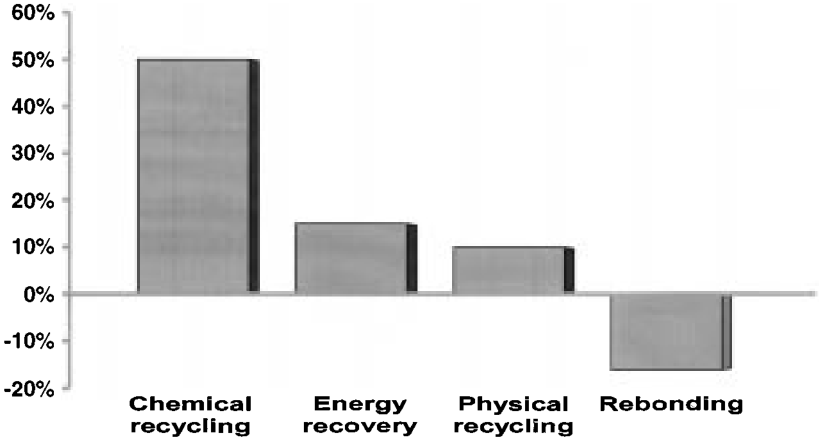

Figure 5. Analysis of base case for chemical recycling of 'Waterlily'comfort cushioning using split-phase glycolysis.

These results show that chemical recycling can save from 42 up to 50% of the original energy required to manufacture the mattress, depending upon the transport distances assumed. Although energy consumption due to transportation is clearly significant, it is more than offset by the energy reduction from the recovery of useful raw materials.

As a final option in the waste management hierarchy, a mattress containing 'Waterlily' comfort materials can be combusted safely in appropriately designed plant. The life-cycle inventory results show that this would result in a saving of up to 15% of the original energy used. Physically recycling 10% of waste foam into a new formulation can save nearly the same amount in energy terms.

On the other hand, rebonding small pieces of foam for carpet underlay consumes energy. This can be approximately 16% of the energy to manufacture the original mattress. It should be noted that it is difficult to compare this option which produces a new product, carpet underlay, to the three previous options in which virgin material is replaced by recycled material.

8. After product design: implementation

In order for a recycling loop to operate, a number of issues need to be addressed such as collection, sorting, cleaning, transportation, disassembly and finally feed preparation for recycling. For each of those processes there are technical problems to be resolved and often there is a high cost associated because of the small volumes involved.

These problems can only be resolved through the formation of new alliances in the product life-cycle chain, working together to create new business opportunities. Sustainable success will only come to those that find environmentally and economically viable solutions.

There is an additional, wider aspect to recycling in that it must be a requirement of society. It is important to realize that industry is part of society and can only succeed together with society. While it is the responsibility of industry to reduce the environmental impact of its economic activity, individuals, their political leaders and opinion formers have a crucial contribution to make through their choice of material standards and consumption.

From the beginning of the development of 'Waterlily', we have maintained a dialogue with decision-makers in the manufacturing chain, not only with our direct customers, the manufacturers of slabstock cushioning foam, but also the furniture manufacturers, retailers and consumers.

Particularly exciting was our collaboration with some of the most influential furniture designers in Europe, with whom we tested that our technological objectives, found echoes in their view of consumer needs. An exhibition of their furniture designs incorporating environmental thinking and inspired by 'Waterlily' was held to great acclaim in Milan in 1993.

9. Conclusion

The success of products such as 'Waterlily' comfort cushioning will only be sustainable if the public is encouraged to move beyond a general consciousness of environmental issues towards making informed and active choices about lifestyles and hence, the products it wishes to buy. It is now becoming clear that consumer attitudes are shaped not only by the communication but also by the credibility and the amount of public agreement on the product improvement in question. For that reason it is very important that some form of consensus on the goals of environmental product improvement is achieved. This would help the credibility of certain environmental claims as well as focus resource in research departments more effectively.

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