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Fluidic bus system for chemical process engineering in the laboratory and for small-scale production

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Abstract

Within the framework of a BMBF-funded project, five research institutes are developing a standardized system for the combination of microstructured devices and laboratory equipment of various suppliers thus leading to the building of chemical plants. The concept is based upon the bus system and simultaneously handles a number of tasks such as mechanical stability, fluidic flow and signal transmission. A key feature of the backbone interface developed is its open architecture. It does not rely on standardized connections thus allowing the combination of devices from various suppliers. The interface shows robustness, withstands high pressures and temperatures while thermal cross-talk is minimized through the use of different materials. Its application in chemical synthesis has shown truly promising results.

Keywords: Small-scale production; Chemical microprocess engineering; Microstructured reactors; Microfluidic platform; Bus system; Microplant

1. Introduction

The use of microstructured components for process engineering has gained increasing importance in chemical, pharmaceutical and life science applications during the last years [1]. Small devices – reactors, heat exchangers, static mixers and other process components – can be fabricated in configurations measured in millimetres and embedded with micrometer-sized pores or channels. Due to large specific surface areas, devices with these small dimensions provide more efficient mass and heat transfer. This results in greater selectivity and higher yield for many chemical reactions. Microprocess engineering and the application of apparatus like microstructured reactors, heat exchangers and mixers developed into a self-dependent discipline. Especially in branches like the chemical industry, the automotive industry and the environmental technology an increasing number of chemical reactions and physical transformations in microstructures was executed under highly selective and inherently safe conditions.

Many users of microprocess technology complain about the costs for individually developed and manufactured devices. The non-existing compatibility of devices from different suppliers represents the main restraint especially because many suppliers offer only a limited number of devices and can thus not provide the complete range of unit operations.

In 2000, the industrial platform "modular microprocess engineering" was founded with up to now 45 research institutes, suppliers and industrial users. The aim of this platform was to create a concept for the establishment of an uniform standard and a modular approach to process technology which should avoid the above-mentioned disadvantages and offer a cost-efficient solution. A manufacturer spanning building-set which consists of compatible microprocess components had to be developed. For the transaction of the volu-

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minous development a BMBF-research project was launched in October 2001 [2,3] with the partners Dechema in Frankfurt/M., the Forschungszentrum Karlsruhe, the Fraunhofer Institut für Zuverlässigkeit und Mikrointegration (IZM) in Berlin, the Institut für Angewandte Chemie Berlin-Adlershof e.V. (ACA), the Institut für Mikrotechnik Mainz GmbH (IMM) and the Technische Universität Darmstadt (TUD). In the present paper, the concept of micromodular process engineering is introduced together with the backbone interface developed in order to realize this. The integration of sensors and an electronic bus system is also described while the physical characterisation of the backbone is discussed within the case study of enantioselective synthesis of organoboranes. Within the case study of the sulfonation of toluene with gaseous sulfur trioxide, the backbone system together with the microstructured devices used is finally assessed based on its application for chemical synthesis.

2. Conception of micromodular process engineering

2.1. Process design

Although a number of microstructured devices and process equipment like valves, pumps, etc., are currently on the market, only a limited number of applications have been reported so far which combine microstructured reactors from the same or even different suppliers. Aside from problems with capacity differences, one key limitation of these devices is that they use different kinds of interfaces which hamper their direct connection. Standardized interfaces would allow the execution of complex chemical processes. Through the collaboration of the above-mentioned five project partners a so-called *backbone interface* has been developed based on the bus concept where the flow is passed through a central spine (Fig. 1). Such a "microplant" is seen as an alternative or a supplement to the miniplant approach in plant engineering. Similar to this technology a closed loop set-up with a refeeding of side products is realized if unit operations for product separation can be integrated. At the present state of development this demands the integration of conventional miniplant glassware, for example, by the installation of a distillation column into the microplant set-up. Such a combination is feasible because the flow rates of a microplant are similar to typical flow rates of miniplant separation units.

Microplant components can also be looked at as a supplement to miniplant components if one has to deal with highly exothermic reactions or high-pressure environments. Due to their low internal volume, microstructured devices withstand large internal pressures. In such a way, a pressurized subregion of a plant can be kept at high pressure using microstructured devices (appropriate control equipment exists) whereas the rest of the plant is kept under low pressure using miniplant equipment. Examples [4,5] exist for very fast and/or highly exothermic reactions or reactions in the explosive regime (unthinkable with standard glassware).

In one of the herein presented processes – namely the organoborane synthesis – a combination of miniplant and microplant equipment is used as a production facility for fine chemicals. This approach differs from the usual application of a miniplant as a mediator between laboratory-scale and pilot plant-scale operation. In the second presented process a microplant for the sulfonation of toluene is presented and characterised.

The modular backbone introduced here allows both commercial and demonstration-type microstructured devices to be coupled in all three dimensions in a flexible and easy manner. Microstructured heat exchangers, reactors and mixers of different manufacturers are surface-mounted onto this back-

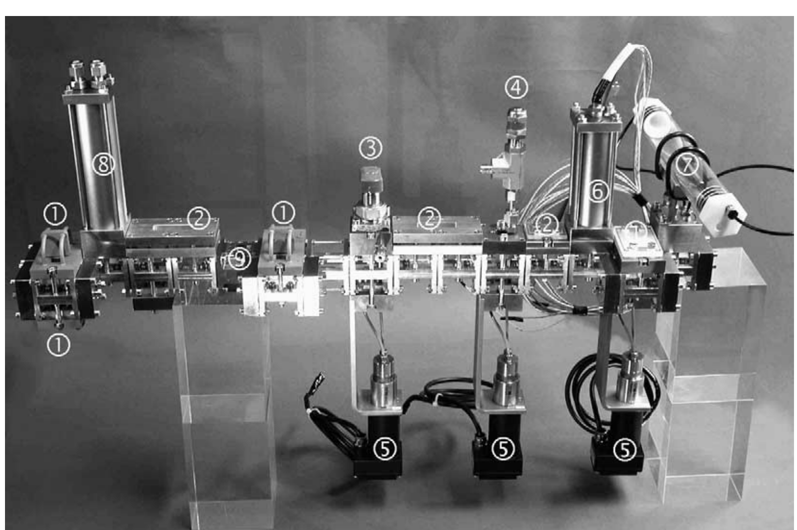

Fig. 1. Example of a microplant based upon the modular fluidic backbone ((1) heat exchanger, (2) mixer, (3) valve, (4) safety valve, (5) pump, (6) heated residence-time module, (7) mixer-settler extractor, (8) heated mixer-tube reactor, and (9) thermal decoupler).

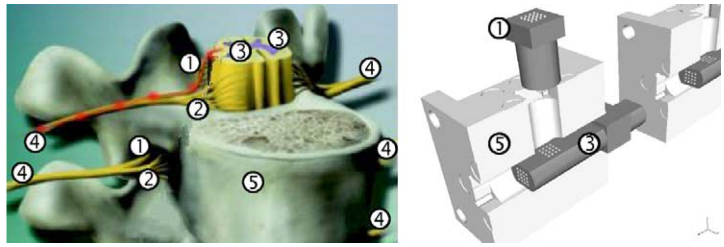

Fig. 2. Archetype human vertebra ((1) sensor nerves, (2) motoric nerves, (3) spinal cord, (4) spinal nerves, and (5) bone structure) and backbone element as electro-mechanical equivalent to a spinal cord ((1) sensor signals, (3) signal bus, and (5) mechanical support).

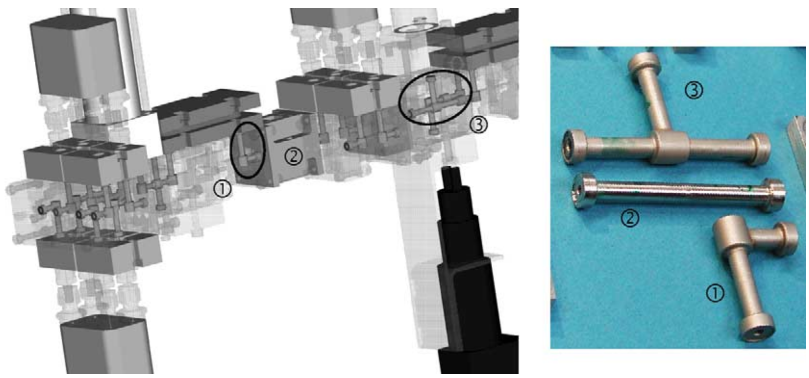

Fig. 3. Internal view of backbone structure ((1) elbow, (2) straight pipe or straight boring in the thermal separator (basic material PTFE), and (3) T-piece).

bone. Due to the standardized interfaces, devices can easily be exchanged, for example, to evaluate different types of mixers. The backbone itself consists of elements, which can be combined individually and flexibly in all directions, according to the demands of the plant to be built. The backbone provides the flow paths for fluids as well as electrical conduits for power supply and signal transmission of sensors and actuators.

Its construction leads to compact systems with low internal volume of 35.3–950.0 mm^3 (per backbone element) which can be operated up to 300 °C and 100 bar, being only limited by the physical properties of the gaskets used (e.g. steel, graphite, Viton®, Kalrez®). By exchanging tubes and borings of various diameters, a large range of volume flows is accessible. For liquid-phase processes, volume flows of up to 32 l/h at a pressure drop of 150 mbar (per spine unit, d_i = 1.5 mm) can be realised and extended to 130 l/h (70 mbar, d_i = 3.0 mm) by the exchange of the pipe inlets, thereby enabling pilot-plant operation.

The backbone interface has the additional advantage of being able to incorporate within its body sensors such as those developed within the *Match-X* consortium [6,7] and of performing as a platform for commercially available valves and mass flow controllers. Such a backbone consists of single standardised elements each incorporating a number of pipes and housing parts and will also be equipped with internal trace heating for the fluidic channels. Fig. 2 gives a comparison between a fluidic/mechanical backbone element and its archetype—a human vertebra.

Fig. 3 (left) shows the positioning within the backbone of typical tubing such as those shown in Fig. 3 (right) (a straight pipe, an elbow and a T-piece). The principle of both the fluidic

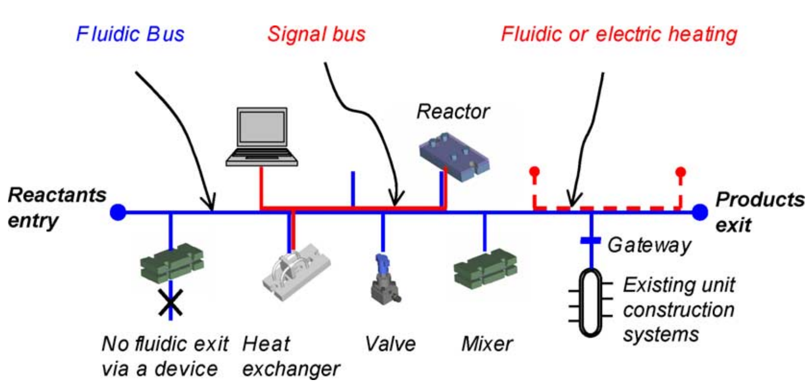

Fig. 4. Parallel bus concept.

Fig. 5. Laboratory-scale miniplant with microplant (within hatched line).

and the electric paths are based upon the bus concept as shown in Fig. 4.

During the experiments the microplant concept was also assessed by measuring properties such as seal reliability and thermal cross-talk between parallel fluidic channels. The performance of the microplant based on the backbone with respect to parameters such as product yield, selectivity and compactness, flexibility or robustness, shall be examined using exemplary chemical reactions. These are advanced multistep reactions in organo-metallic boron chemistry (l), fast and exothermic sulfonation of alkyl-substituted aromatics (g/l) and Darzens glycid-ester synthesis (l/l). Starting by connecting a few microstructured devices, entire set-ups shall be built based upon a 'plug and produce' plant-assembly systematic.

A microplant (in Fig. 5 integrated into a typical miniplant) will exhibit a larger complexity than the miniplant set-up but it nevertheless consumes a much smaller package volume of the plant set-up.

2.2. Integrated sensors and electronic bus system

A state-of-the-art plant concept must enable controlled process operations. This demands the integration of sensor and control equipment directly into the process. In a first attempt to obtain process parameters actually the values at the entry and the exits of devices like heat exchangers or reactors is sufficient information. The first step of sensor integration

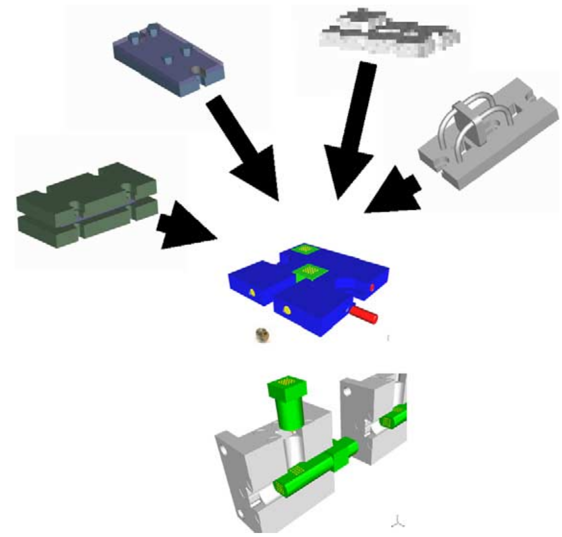

Fig. 6. Backbone elements, adapter plate incorporating sensors and standardized process technology elements.

is to position a so-called sensor-adapter plate between the backbone and the devices.

Fig. 6 shows such a combination of standardized elements together with an adapter plate and a backbone element. The adapter plate can incorporate, e.g. pressure and temperature sensors. A future extension will be the integration of chips for the convertion of analog sensor signals to digital signals directly neighbored to the sensor (Fig. 7). The electrical data and power bus is routed through the whole backbone system. By using the electrical bus, the control system communicates with the sensors.

The data transmission of the signals will be executed by an electronic bus consisting of standardized elements as shown in Fig. 8. The aperture in the middle of the plate is reserved for the vertical electrical connection. This connector system allows the horizontal connection from the beginning to the end of the complete backbone, whereas the vertical connection is either used for a branch connection

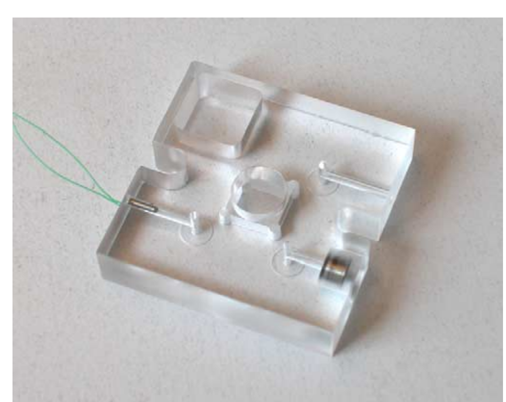

Fig. 7. Model of sensor adapter plate with temperature (left) and pressure (right) sensor (the cavity in the back of the plate is provided for the future integration of an A/D-converter chip).

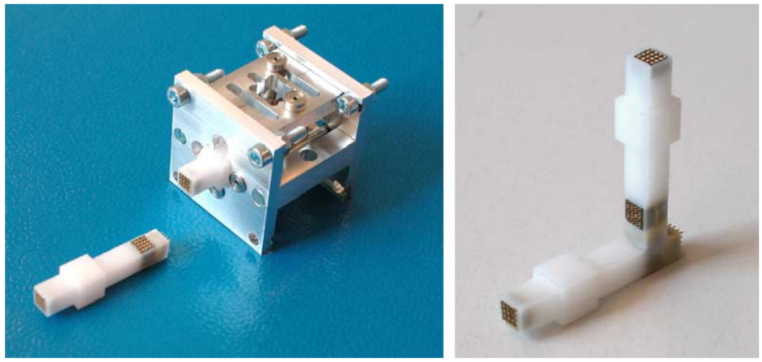

Fig. 8. Principle of backbone element with models of internal bus connectors (left) and the combination of the same bus connectors for an external bus set-up (right).

or for contacting the sensors and the miniaturized sensor electronics. These bus connectors can be positioned inside the mechanical backbone but also – for example in case of high-temperature applications, for example, heating inside the backbone – outside the backbone as a separate electronic bus. That means, there are no cables outside the chemical microplant, apart from high power cables, e.g. for electrical heaters.

3. Physical and chemical characterization

3.1. Physical characterization of the set-up for the enantioselective synthesis via organoborane

Driven by the rapidly growing demand for highly enantiomerically pure substances in the pharmaceutical industry in order to provide safer and cheaper drugs, the development of a process which can achieve enantioselectivities approaching 100% has become desirable and challenging.

One case study within the framework of this project is thus to test the concept of a microplant by applying the fast reaction of the enantioselective synthesis via organoboranes yielding chiral-substituted alcohols. This is typically a batch process carried out in the laboratory using conventional glassware and has been in the present case converted into a continuous process carried out by microstructured devices. This set-up has been used until now to characterise the physical properties of the backbone system.

As first test, a number of elements were assembled and the *spacing* of the housings measured. It turned out that the spacing is independent of the number of screwed elements. The small errors of the individual housing dimensions compensate each other. This is a prerequisite for the build-up of complex three-dimensional plants. The latter require a continuity of spacing also over long distances. The leak tightness was characterized using nitrogen (g) under a pressure of 76 bar. Three backbone elements were combined consisting of two straight pipes, two elbows and six PTFE seals ($d = 6$ mm, $t = 1$ mm). The pressure was recorded as a function of time (see Table 1).

The leak rate was calculated according to the German DIN standard. This leak rate is related to the lengths of all seals (90 mm) and the internal volume (837 mm^3). The specific leak rate obtained was 0.00076 mg/s m which corresponds to a leak rate class of $L_{0,001}$ comparable to commercial valves. For this first test the O-seals were manufactured manually by punching from a sheet of PTFE which often resulted in non-centric/non-circular seals. The sealing results are nevertheless satisfactory. It is to be expected that sealing performance will increase with future industrially manufactured seals. In a plant it is desired that the pressure drop along pipes and fittings is at least one order of magnitude smaller than the pressure drop of the devices. A typical microstructured device will possess a pressure drop of approximately 1 bar when operated with a volume flow (water assumed) of approximately 1 l/h. The calculated pressure drop in a straight pipe inlay under laminar conditions was calculated to be 1 mbar which is three orders of magnitude smaller than the typical pressure loss of a microstructured device. It is obvious that the pipes will not dominate the pressure loss of the plant as in fact their contribution to the pressure loss is nearly negligible.

Fig. 9 shows a single backbone element as the smallest unit of the backbone. The element is equipped with three different pipe inlays, a straight pipe, an elbow and a T-piece. All three inlays are inserted in the front plate in which case they also have a thermal contact via this plate. This could be a desired effect if, for example, trace heating is intended. However in most cases this is an unwanted situation.

In order to study the influence of thermal cross-talk, liquids with different temperatures were conveyed through the back-

Table 1
Pressure of three combined backbone elements recorded as a function of time

Time (min)	Pressure (bar)
0	75.9
10	75.9
24	75.8
58	75.8
81	75.8
1008	74.8

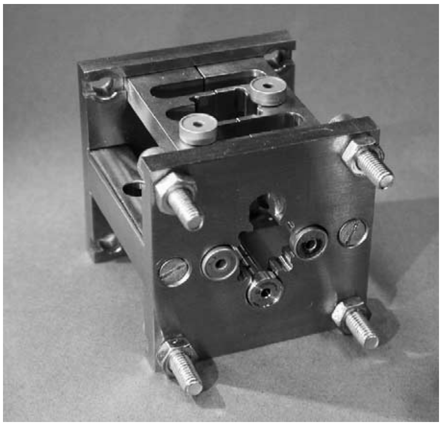

Fig. 9. Standardized single backbone element consisting of two face plates, two spacer bars and fork-type support plates equipped here with three pipe inlays (elbow, straight pipe and T-piece from left to right).

bone and their temperatures measured with thermocouples and with thermo graphic infrared measurements. To reach realistic measurements, elbows were equipped with thermocouples with direct contact to the fluids (see Fig. 10).

Fig. 10 shows the complete set-up used for the temperature tests. A realistic set-up was chosen by combining three heat exchangers, a glass mixer and a mixer-tube reactor with six backbone elements. Initially the microplant was at room temperature.

The test set-up chosen for the infrared measurements clearly shows regions with distinct temperature values. The heat exchangers 1 and 2 were supplied with the cooling agent *iso*-propanol cooled in a cryostat to −11 °C. The surface temperature as indicated in the thermo graphic image (Fig. 11) is much lower than the temperature of the neighboring backbone element, which is an indication of thermal decoupling between the backbone structure and the pipe inlays which deliver the cooling agent to the heat exchangers. Thermocouples inserted in the pipe inlays to the heat exchanger showed a temperature of −6 °C at the heat exchanger exit.

Nearly complete thermal decoupling is enabled by the use of a special backbone element used for insulation (see Fig. 10). This element is made of fibre-reinforced PTFE (Teflon®) which is chemically fairly inert and possesses an extremely small heat conductivity coefficient of 0.25 W/mK compared to the standard backbone materials steel (16 W/mK) and aluminium (204 W/mK). Such an insulation element enables the definition of distinct temperature regions. For example, in Fig. 11 a region on the left side (consisting of one element) with ambient temperature conditions is thermally separated from a region cooled down to −10 °C at the right side (consisting of four elements). As the temperatures – indicated in color code in the infrared image – are due to different surface emissivities only qualitative temperatures, these thermographic images were complemented by temperatures measured directly in the fluid with thermocouples inside the pipe inlays. All the devices exhibit individual temperatures different from the backbone temperature. This desired effect can as well be magnified by the usage of steel or PTFE as housing material for the backbone elements. In this contribution we used aluminium as housing material. The largest temperature difference exists between the mixer-tube reactor and the heat exchanger 3 with approximately 60 K. Remarkable is also that the glass mixer is well suited to insulate the fluid due to the low heat transfer coefficient of glass (see arrow in Fig. 7). This result was confirmed by measuring the temperature difference between product inlet and outlet of the glass mixer which amounted to only 3 K.

The insulation effect of the Teflon element is obvious if the temperature course is given as a function of the element

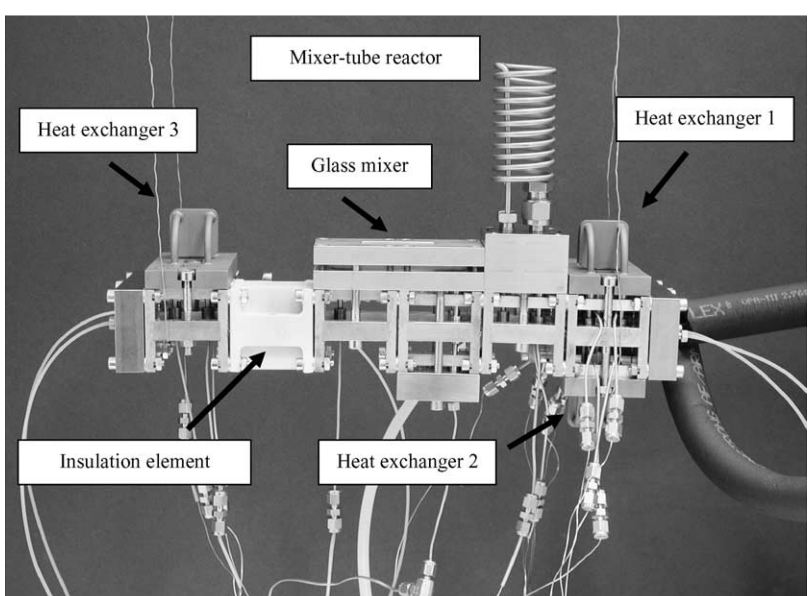

Fig. 10. Microplant set-up for studying cross-talk behaviour (the horizontal backbone consists here of six elements without branching).

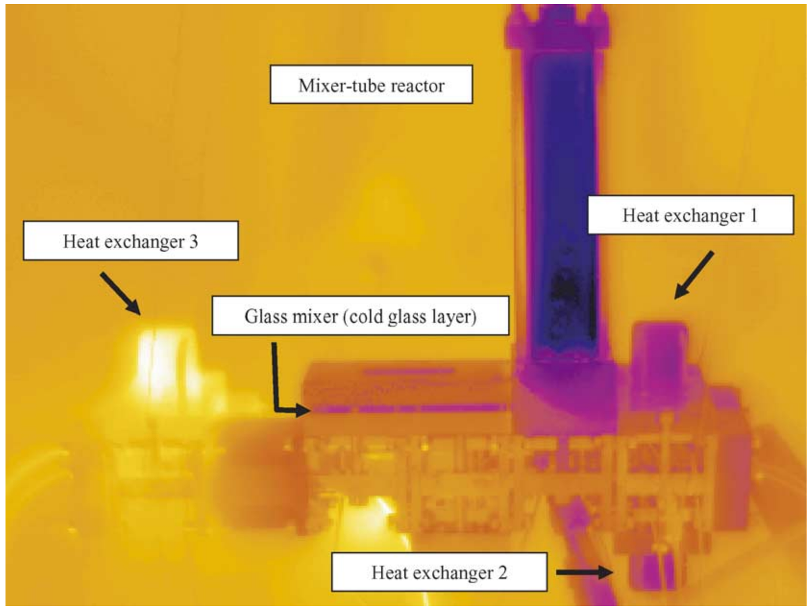

Fig. 11. Thermo graphic image of the set-up shown in Fig. 10 the temperatures range from approximately −10 °C (surface temperature of the perpendicular tube of the mixer-tube reactor) to 50 °C (surface temperature of heat exchanger 3).

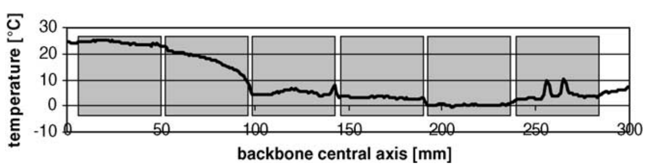

Fig. 12. Temperature course along the central axis of the backbone (the grey boxes indicate the position of the individual backbone elements, the peaks in the right box are due to cables lying in front of the element, temperatures are surface temperatures measured at the spacer bars with corrected material emissivities contrary to Fig. 11).

position (see Fig. 12). For this reason the surface temperature of the six horizontal spacer bars was recorded. The surface temperatures of the spacer bars depend on the emissivity coefficient of the materials and on the surface characteristics. To eliminate these effects the spacer bars were painted.

The temperatures reflect only the housing temperatures and not the fluid temperatures inside the pipes. It is nevertheless a strong hint that a thermal separation of different plant sections is possible. Here, the left heated section is separated from the colder right section, as the PTFE element (second

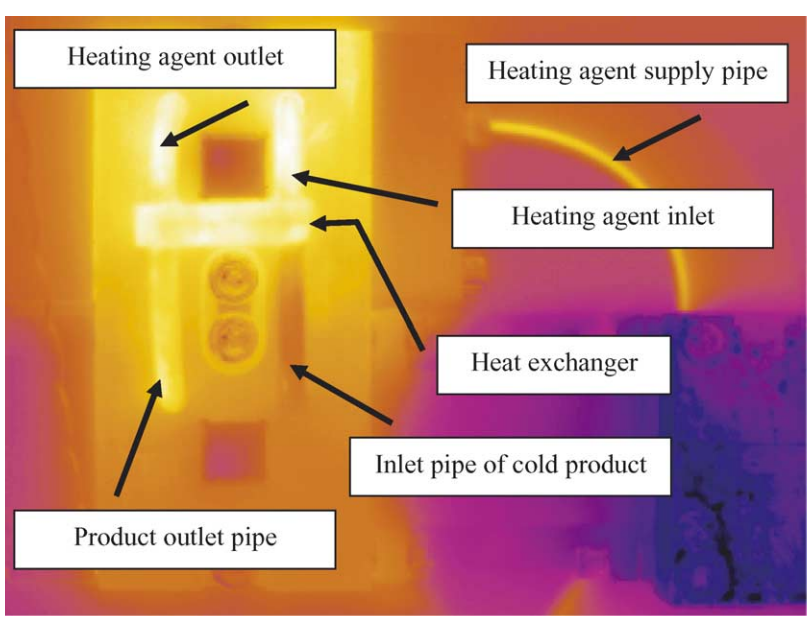

Fig. 13. Surface temperature variation at heat exchanger 3 (largest temperature range indicated in the thermo graphic image varies from approximately 10–50 °C).

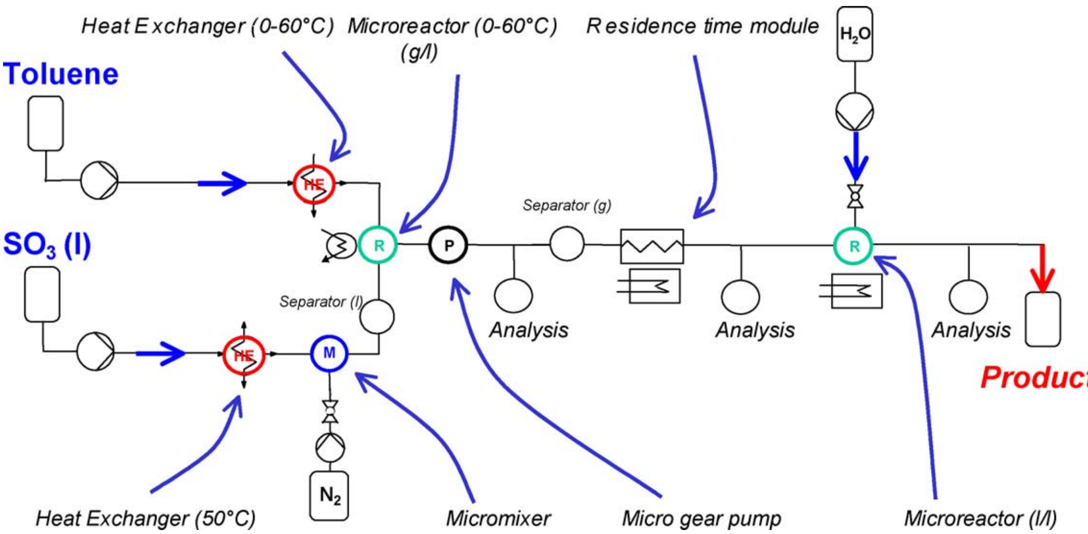

Fig. 14. Process flow sheet for sulfonation of toluene with gaseous SO_3.

element from the left side) enables the development of a sufficiently large temperature gradient of approximately 20 °C in this example. The temperature of the liquid (*iso*-propanol) led through the piping reflects a similar behaviour. Directly before it reaches the insulation element the temperature was 2.6 °C and after the element it amounted to 27.4 °C.

The heat exchanger in Fig. 13 was supplied with 1 l/h water as the heat transfer medium at an inlet temperature of 60 °C and *iso*-propanol as the product with a volume flow of 60 ml/h and an inlet temperature of 27.4 °C. In the heat exchanger it was heated up to 46.1 °C.

3.2. Chemical characterization of the backbone using the sulfonation of toluene with gaseous SO_3

The highly exothermic sulfonation of toluene with gaseous sulfur trioxide is one reaction which has been investigated in the microplant. Fig. 14 shows the process flow sheet of the microplant used at the ACA.

Toluene is heated up to 40 °C using a microstructured heat exchanger while at the same time liquid sulfur trioxide is heated up to 60 °C in order to evaporate it. Nitrogen is further added so as to dilute the system and the stream is then passed into a separator with the purpose of removing any traces of liquid. Thus, a gas stream is allowed to flow through to a microstructured reactor where it reacts with the liquid toluene. As shown in Fig. 15, reaction 1, sulfonic acid is produced here via the desired reaction step. At the same time, though, sulfone (reaction 2), a mixed anhydride and sulfonic acid anhydride are also formed by side reactions. Sulfone cannot be converted further but the mixed anhydride reacts in the residence-time module with toluene and forms the desired product, sulfonic acid, as shown in Fig. 15, reaction 3. To convert the sulfonic acid anhydride to sulfonic acid, a hydration step is required (Fig. 15, reaction 4). To achieve this, water is added to the reaction mixture after the residence-time module as shown in Fig. 14.

Fig. 16 gives an overview of the microplant used. Emerging from the front, is a typical microtooth gear pump used, while on the left-hand side of the photo the falling film microstructured reactor can be seen. This is used to carry out the gas/liquid reaction at the start of the process. Further along the backbone a large cylindrical residence-time module is visible in the background. The macro-scale residence-time

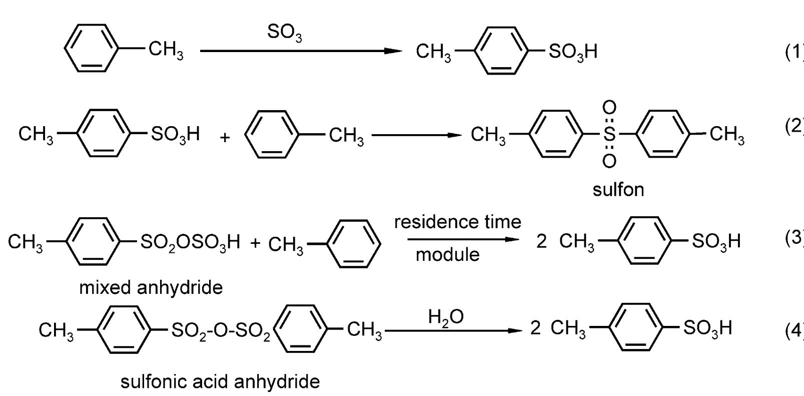

Fig. 15. Reaction paths during sulfonation of toluene with gaseous SO_3.

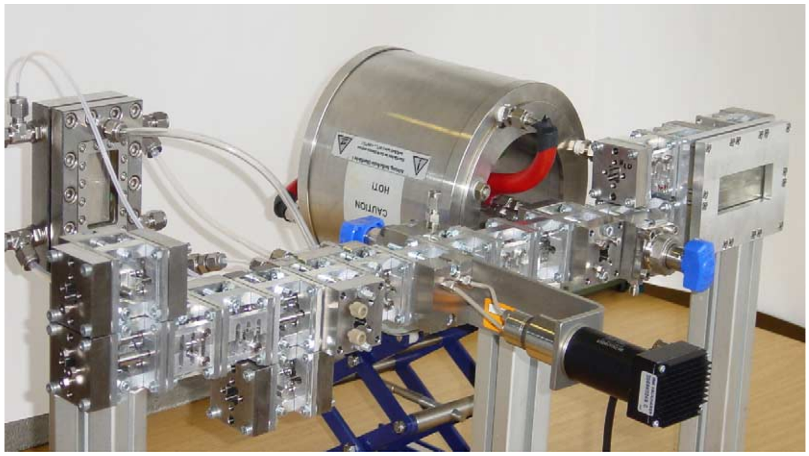

Fig. 16. Microplant used for the sulfonation of toluene.

module consists of a wound tube heated by a heat transfer agent. The final reactor seen at the end of the backbone on the right hand side of the picture is the reactor where the final hydration step takes place. The entire set-up has been tested for leak tightness up to 5 bar, the pressure limit for the glass reactors.

To-date, the reaction has been carried out up until the residence-time module. The final hydration step (Fig. 15, reaction 4) has not taken place. Even so, first results are very encouraging as shown in Fig. 17. In order to evaluate reaction conditions, the mole ratio of the two reactants, sulfur trioxide and toluene, was varied and the selectivity of the desired product (sulfonic acid) and of the by-products (sulfon and the anhydride mixture) was determined. Evidently, with increasing SO_3/toluene mole ratio, the selectivity of the undesired by-products decreases while the selectivity of sulfonic acid stays nearly constant. At a mole ratio of 13/100, the selectivity of sulfonic acid is approximately 80% while that of sulfone decreases to approximately 3% and that of the sulfonic acid anhydride to approximately 1.3%.

The isomer selectivity was also determined to be 8.1% for the *ortho*-sulfonic acid, 1.5% for the *meta*-sulfonic acid and 90.4% for the *para*-sulfonic acid. From literature, at a SO_3/toluene mole ratio of 13.4, the selectivity of the *ortho*-sulfonic acid was 17.6%, of the *meta*-sulfonic acid, 1.2% and that of the *para*-sulfonic acid was 81.2% [8]. Very recently also the last hydration step was executed successfully. These results together with the chemical characterization of the organoborane case study will be given in a second paper.

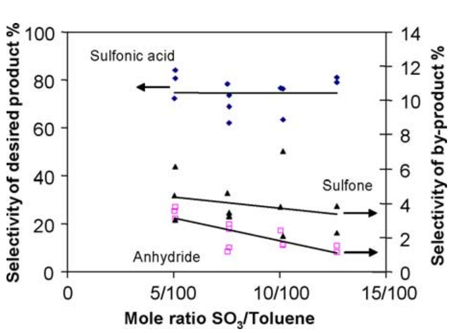

Fig. 17. Selectivity of products at various SO_3/toluene mole ratios.

4. Conclusions

First attempts in the development of a standardised modular system for microreaction technology have been illustrated. The concept is based upon the human spine which in a similar manner also incorporates the simultaneous handling of a number of very different tasks including mechanical stability and signal transmission. The system does not rely on standardised reactors but instead on standardised connections thus allowing the use of commercially available non-uniform reactors.

Leak tightness up to high pressures has been proved and temperatures are only constrained by the material of the gaskets. Thermal cross-talk is minimised by the use of Teflon backbone elements. First results obtained during the sulfonation of toluene with gaseous SO_3 show a high selectivity of the desired product. Further investigations are needed in order to achieve a complete picture of the advantages of microstructured reactors and the backbone system but first signs are certainly encouraging.

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