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# MAGNETICALLY ACTUATED POLYMER FLAP ARRAYS AS ARTIFICIAL CILIA

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## KEY WORDS

Cilia, hydrogel, photolithography, magnetic actuation, magnetic particles, photoreactive polymers

## ABSTRACT

*Handling of liquids in sub millimeter sized channels is essential for microfluidic systems. We developed microactuators inspired by small oscillating hairs called cilia found in nature. Our microactuators are based on polymers filled with magnetic nanoparticles and motion of the structures in liquids is achieved through exposure to varying magnetic fields. The overall cilia manufacturing process we developed is based on a simple and robust process for the generation of surface-attached polymer networks containing magnetic nanoparticles. A combination of two different variants of the photochemical process to generate such surface architectures has been used to manufacture surface-attached polymer micro-flaps that resemble a cilia covered surface. The photochemical process is based on the use of copolymers that contain a small amount of photoreactive groups. The polymers are deposited onto a surface and a subsequent illumination with UV light triggers a crosslinking reaction and at the same time also an attachment to the surface. Using polymers with different photoreactive groups that respond to UV light of different wavelengths one can use this general approach to generate arrays of small flaps on the surface. Each individual flap is only attached to the surface at one end. If the polymers are filled with several weight percent of magnetic nanoparticles the resulting flaps can be released from the surface and actuated by using magnetic fields. We discuss the overall fabrication process with an emphasis on the parameters that allow the generation of large area arrays as these are needed in a microfluidic device and on device integration.*

## 1. INTRODUCTION

Moving small amounts of liquids is often realized in nature by means of small hairs called cilia. One can exploit such principles by attaching ciliated microorganisms to surfaces to induce liquid flow (Bacterial Carpets) [1] On the experimental side, a number of such architectures have been realized and recently successful mixing was achieved using arrays of electrostatically actuated artificial cilia [2]. Flows of up to 600 µm/s were possible but the strong electric fields limited the use of such systems to non-aqueous solutions. Other systems are based on piezo actuators, liquid crystals and magnetically filled PDMS rubber [3-8].

For practical applications large arrays of cilia are needed to generate sufficient flow from magnetically actuated cilia systems. It was, hence, our goal to establish materials and processes that can be used together with standard microstructuring techniques to build such arrays and to also provide simple means to integrate these cilia into microfluidic devices.

## 2. RESULTS AND DISCUSSION

Biological cilia and many of its bio-mimetic counterparts are usually hairy structures that stand upright on a surface. Synthetically such a system is hard to realize and will often yield systems that ask for a tedious production process which is not compatible with existing technologies. Also the hairs will often be mechanically rather vulnerable which poses a special problem when it comes to storage or handling, i.e. during insertion into a microfluidic device. We were looking for a way to avoid such problems and decided to follow an approach in which the cilia structures are processed within the sample plane and then released from the surface to form flaps which bend up from the sample as shown in Figure 1.

The process is based on the use of photoreactive polymers [9-11] which can be processed into thin layers for example by spin-casting. Upon irradiation with UV light of an appropriate wavelength the polymers form crosslinks and are eventually attached to the surface provided that suitable groups are present there. These may be realized by using a silane carrying the same photoreactive group as the polymer or another polymer layer [12]. Surface attachment and crosslinking is then achieved simultaneously except in areas in which an underlying polymer is not crosslinked itself. In this case, the removal of the underlying polymer leads to the flap release. In summary, a two color process was used to generate cilia-like polymeric flaps.

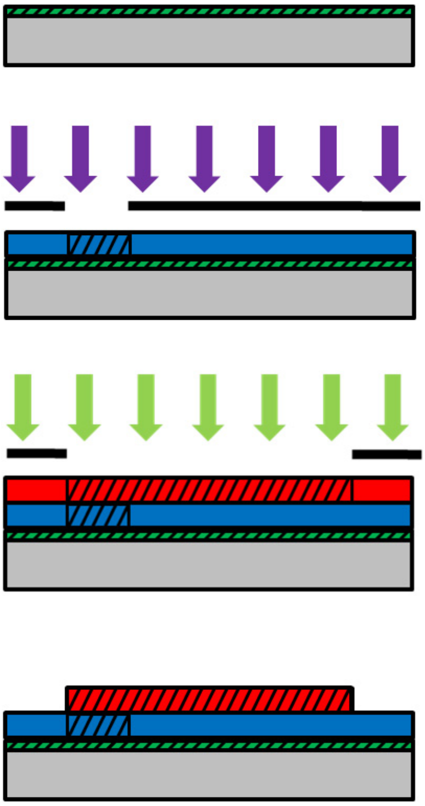
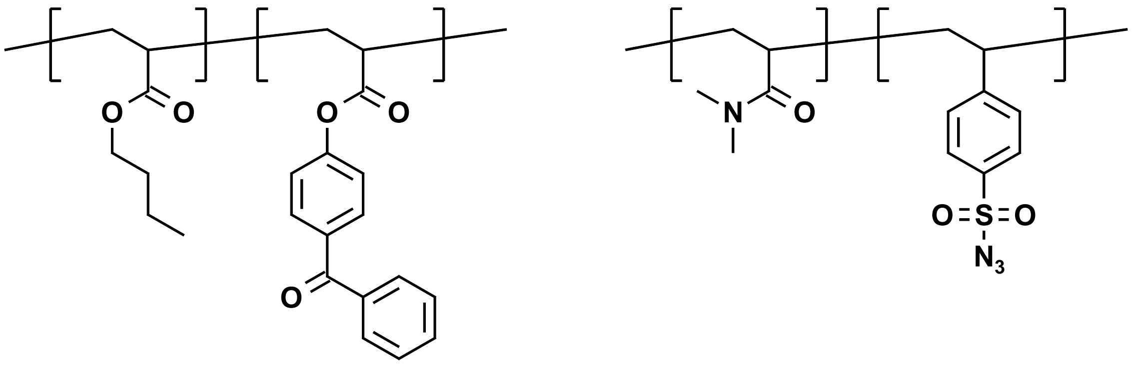


**Figure 1:** Scheme of the two-color lithographical process used to generate cilia arrays. A first layer (blue) generated at ~250 nm provides an anchoring patch for the subsequently deposited cilia material and a sacrificial layer which avoids the attachment of the cilia material (red). The cilia is generated at 365 nm. This wavelength does not trigger any photoreaction within the sacrificial layer.

The process is compatible with a variety of materials within some limits as will be discussed below.

**Flap material.** The most important requirement for the flap material is its elasticity. This may be achieved by two different means: Either the material is a rubber and elastic by its very nature or the material swells in the medium of interest to become elastic upon use. In the latter case, water-swellable materials (hydrogels) are the polymers of choice as microfluidic systems are usually most interesting for medical or biomedical purposes. We have investigated candidates for both systems and have chosen crosslinked poly-n-butylacrylate (PnBA) as the rubbery materials and poly-N,N-dimethylacrylamide (PDMAA) as the hydrogel material. In both cases the polymers carried small amounts (2 – 5 Mol-%) of a photoreactive comonomer to enable crosslinking during processing. Two typical variants are given in Figure 2.



**P(nBA-co-MABP)** **P(DMAA-co-SSAz)**

**Figure 2.** Polymers used in this study for the generation of cilia-like microflap arrays: P(nBA-co-MABP) is a copolymer of n-butylacrylate and the photocrosslinker methacryloyloxybenzophenone (MABP) and yields a rubbery material after photocrosslinking. P(DMAA-co-SSAz) is built from N,N-dimethylacrylamide and 4-styrenesulfonyl azide. It yields a hydrogel after crosslinking. The two crosslinkers are minor components (few Mol-%) within the polymers. Depending on the nature of the crosslinker, different wavelengths can be used to trigger the photoreaction.

All crosslinked materials yield highly flexible material with an G-modulus of approximately 20 kPa or less at 1 Hz and are therefore suitable for flap fabrication. Note that these values will increase when the material is filled with magnetic nanoparticles, however, the increase in stiffness caused by the inorganic component still leaves a sufficiently elastic flap.

Comparing these two general materials – rubber and hydrogel – it is obvious that the hydrogel flap will increase in volume during swelling. Typical degrees of swelling are around S = 2, i.e. a doubling of the volume of each individual flap takes place. This phenomenon can be accounted for by adjusting the mask design. However, the increase in volume renders the hydrogel flaps less stable and mechanical damage may occur more easily. On the contrary, the rubber flaps do not show any significant water up take. The hydrophobic nature of the material needs to be considered during use as a sticking of the flaps is at times encountered when two neighboring flaps touch each other or whenever one flap folds back onto itself. We have found that a hydrophilic coating deposited by a dry deposition process takes away this tackiness.

**Materials for anchor layer / sacrificial layer.** Depending on the choice as the material for the flaps, the polymer used as the anchoring layer and sacrificial layer needs to be insoluble in solvents used for flap polymer deposition and soluble in non-solvents for the flap polymer. We have used the same base polymers as shown in Figure 1 such that the water-soluable PDMAA-based system forms either the flap or the anchor / sacrificial layer with the PnBA-based material being used for the other layer, respectively. Water (or alcohols) can be used to release the flaps if the PDMAA-based polymers form the sacrificial layer. We expect that this choice is favorable for possible applications as any biomedical use would ask for such a simple way to release the cilia and also because it is not a simple task to build microfluidic gaskets that tolerate organic solvents. This limitation means in turn that most of the systems we investigated were based on the PnBA rubber flaps with PDMAA anchors even though the inverse configuration is in principle equally suitable to generate cilia arrays for magnetic actuation.

**Magnetic particles.** We have studied two general material systems for use in a magnetic actuation setup: One is ferromagnetic (cobalt ferrite) and shows a permanent magnetic behavior while the superparamagnetic magnetite particles are only magnetic in the presence of a respective field. Both systems were studied and standard synthetic procedures were used and adjusted to yield particles of a diameter between 10 and 20 nm. It was found that both classes can be coated with surfactants such that they would not agglomerate significantly during mixing and film generation with the polymeric materials mentioned above (Figure 3).

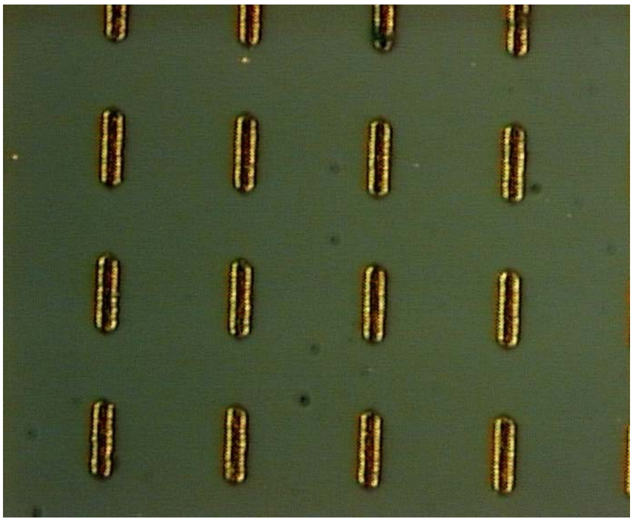


**Figure 3 .** Example of microstructures filled with magnetite particles. The length of the structures is 50 µm. Crosslinking was not significantly influenced by the presence of the particles and excessive agglomeration was not observed.

The filler content of the microstructures could be adjusted to values up to a magnetite content of 38% by weight. This value was detected using thermal gravimetric analysis and is based on the residual sample weight after combustion of all organic material including the surfactant coating. We found that these filler contents are high enough to enable magnetic actuation at moderate fields.

**Processes.** The processes (Figure 1) for flap generation are entirely based on standard photolithographic procedures. The substrates are at first modified with a silane bearing a photoreactive benzophenone unit. This substance is known to link to adjacent coatings when illuminated with UV light [12]. On top of this layer the material for the anchor strips and sacrificial layer is deposited using a spin coater. This layer is then illuminated with light of a wavelength of 254 nm through a mask to generate the anchor strips by crosslinking the polymers in the illuminated areas and by simultaneously attaching these strips to the surface through the photoreaction with the benzophenone silane monolayer. After illumination the anchor strips are not developed but the flap material is directly spun on top of it. This deposition is done using a toluene solution of the PnBa-based polymer because toluene is a non-solvent for the PDMAA-based layer. The crosslinking is again achieved using UV light and a photomask. Illumination is done at 360 nm which leads to a crosslinking of the flaps through the benzophenone moietes but leaves the less reactive SSAz groups within the sacrificial layer untouched. The presence of magnetic particles within the layer leads to slower crosslinking as compared to the pure material but the time necessary to achieve sufficient crosslinking remained acceptable. After the illumination step the cilia were developed using toluene again. This treatment left a sample on which the flaps are attached to the anchoring strips but still lay on the sacrificial layer. We found that this state is the preferred way to store the samples until further use.

**Microfluidic integration, release and actuation.** The samples were integrated into a microfluidic chamber (Figure 4a) using a vacuum channel and tested in terms of cilia release and magnetic interaction. Release was accomplished by simply pumping water through the channel. A high number of individual hairs detached from the surface but were kept in place by their anchor. Using an electromagnet we generated a magnetic field of 50 mT which was sufficient to bend the hairs completely upwards. Upon removal of the field the cilia returned to their horizontal position. Driving the field at frequencies up to 10 Hz we observed a full orthogonal displacement of the hairs with each cycle. Higher frequencies were possible but movement remained incomplete as the hairs were no longer able to follow the field oscillations quick enough. Long-term testing revealed that the cilia arrays are rather stable: No significant change was observed even after operating the cilia at 10 Hz for 4 hours which equals approximately 300.000 beats.

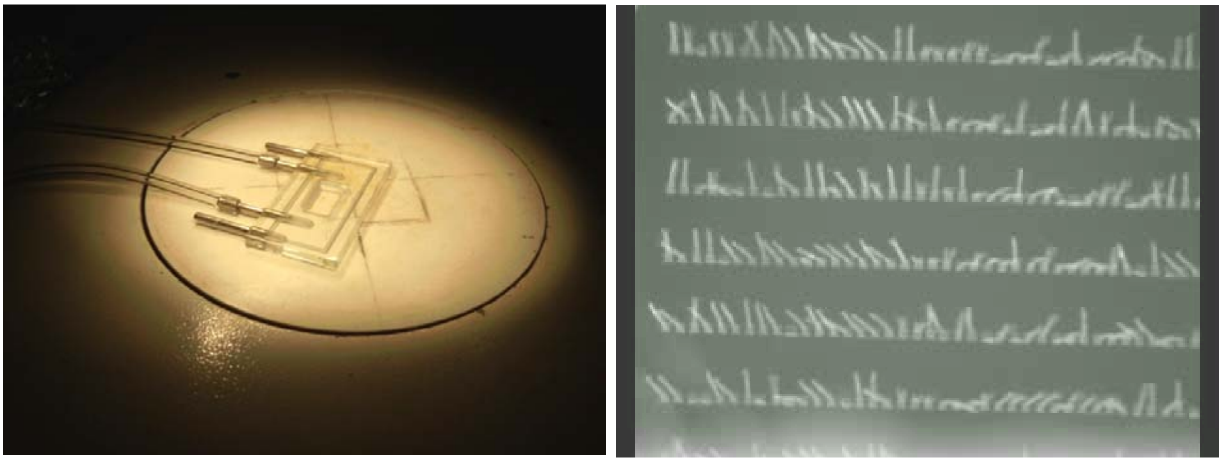


**Figure 4:** a) Microfluidic chamber used for cilia testing: The flap arrays are processed on a slide and the slide is integrated into a plastic gasket carrying all microfluidic inlets and outlets. b) Optical micrograph of a cilia array after release in the absence of a magnetic field.

## 3. CONCLUSIONS

Using standard photolithographic procedures to generate magnetically responsive mechanical microstructures is a very suitable approach for building artificial cilia structures. The most important features of the process described above is the use of two different polymers with two different photoreactive groups which can be processed to surface-attached polymer networks by using two different wavelengths in subsequent lithographic processing steps. This combination allows for the use of the first layer as the attachment strip and as a sacrificial layer onto which the flaps can be deposited. Two different polymeric systems were investigated as flap or cilia material: One is an elastic rubber the other one forms a hydrogel and gains its elasticity due to swelling in aqueous media. Both materials are highly flexible with the rubber being mechanically more stable. Key for the production of the cilia is the good compatibility between the polymers that form the matrix and the magnetic nanoparticles. For the latter both superparamagnetic as well as ferromagnetic materials are suitable. Typical sizes of the particles are between 10 and 20 nm in diameter and agglomeration is reduced by various surfactants. Filler contents of up to 38% magnetic particles (without the surfactant shell) can be realized. Such high contents are needed to allow for a sufficient magnetic actuation. All materials can be transferred into normal cleanroom facilities and the use of photolithographic standard techniques allows for the generation of large and uniform arrays of microstructures. The sizes of these structural features can be varied by using different mask layouts. One concern for using such architectures is that they might be damaged during handling, e.g. while they are built into a microfludic system. This problem is largely eliminated in our system as the cilia arrays are not developed after the second illumination step. This way the cilia structures rest on a sacrificial layer and are largely protected from damage during handling. The individual hairs are then released using an aqueous solution. Magnetic actuation using magnetic fields is possible and first results indicate that an asymmetric movement of the cilia can be realized. The cilia can be operated at rather high frequencies of several ten Hertz for hours without observing degradation of the array. Initial results indicate that a net flow can be realized using rotating fields but further research is needed to quantify the achievable flow rates.

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