THERMO-FLUID-DYNAMIC EVALUATION OF A MICROSYSTEM TO ANALYSE RADIOACTIVE SOLUTIONS

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ABSTRACT

It has become common place to use micro-electromechanical system (MEMS) to evaluate the chemical properties of solutions. However, such microchips have not yet been applied to analyse radioactive solutions, for the purpose of nuclear safeguards, in the nuclear reprocessing industry. Implementing MEMS in this area results in a reduced volume of the sample to be analysed. This brings about a reduction in radiation and many advantages over conventional methods. The radioactive solution is sucked into through a microchannel (300 x 300 µm) in this chip; the solution will release heat due to radioactive decay. The flow and heat characteristics of microchannels have been observed to deviate from conventional and well established theory. This work initially evaluates these differences with an in-depth literature study examining the reasons for these differences. The microscopic effect of the electrical double layer (EDL) is focused on. These investigations on the validity of the traditional macroscopic models allowed application of classical theories within a well defined validity range and the adaptation of these theories to suit microscopic models. It was concluded that the EDL was the most influential on the flow. FEMLAB 3 (COMSOL AB 2004) was then utilised to model the microchip for both fluid dynamics and thermodynamics. Subsequent thermal stresses were also investigated. From this study conditions on the released heat were derived that guarantee no deformation of the chip and no temperature shift for the absorptiometry measurements.

INTRODUCTION

Significant progress in microfabrication techniques enables the manufacturing of today's microsystems, with a large and steadily growing application potential in the area of microelectronics, analytical chemistry, pharmaceutics, and biochemistry. These "systems on a microchip" are realised by integrating on a substrate mechanical actuators, sensors and (opto-)electronics, all devices with the size of 1 to 500 μm.

The starting key element is the Micro-Electro-Mechanical System (MEMS) as actuator, which is a sophisticated functional, fluid manipulating device on a small silicon chip, produced at low costs. A cheap mass production of miniaturised single-use analysis microchips is possible by for instance the hot embossing technique.

Since microsystems proved to be very efficient, economic and performant to analyse chemical solutions in microvials, a research project at IPSC also introduced these microsystems in the Safeguards area. So far microchips have not been applied to analyse radioactive solutions in the nuclear reprocessing industry. For each nuclear reprocessing facility bookkeeping of the inventory nuclear material is obliged and carried out by accurately measuring the total content U and Pu in the input and output accountancy tanks. Therefore samples of several milliliters of the solution are taken, to determine the density, chemical composition and concentration in U and Pu. It is proposed to replace a milliliter sample with a microliter sample, which is then analysed by spectrophotometry and densitometry. Obviously the relevant volume reduction with a factor 6321 by replacing the 7 millilitre size with 0.62 microlitre size, implies an almost proportional dose reduction with a factor 6000 at 1m distance, as proven by Buyst et al. (2005). The reduced radiation has several advantages. A direct analysis on the real solution can take place without the need for dilution: This fastens and simplifies the analytical analysis procedure; and the direct results allow an interpretation with a smaller uncertainty range. The analysis can take place in a glove box, no hot cell is needed any longer. The reduction in shielding allows to make the analytical device transportable. The radioactive waste is also reduced, not only because of the volume of the microchip itself but mainly because of the reduction or even omission of the sample preparation.

THE MEMS DESIGN

The design of the microchip results as a compromise between the requirements for manufacturing and those for the special treatment of the radioactive solutions. The MEMS of the microchip has to allow reproducible spectrophotometric and densitometric measurements along the microchannel, in which an electrophoretic flow of the radioactive solution is established. The spectrophotometric aspects have been

investigated by Nucifora (2004)[8], the densitometric aspects by Macerata (2004)[7].

As shown in Fig.1 the MEMS base consists of three microchannels, all with 300μm x 300μm cross section. The first channel contains the reference solution, the middle one is the optical positioning channel and the third represents the chemical analysis channel, through which the subject radioactive solution is pumped based on the principle of electrophoresis. In a cover inlets and outlets are foreseen and between the two layers a series of electrodes are inserted to create an electric field along the microchannel length axis. The complete MEMS is embedded in a positioning frame and kept at constant temperature by means of a cooling Peltier element.

As substrate material a High Density Polyethylene (HDPE) has been selected because this polymer is (i) resistant to acidity and irradiation during the measurement time of maximum 1 day, (ii) transparent for the photospectrometric analysing technique, and (iii) compatible with the material requirements for hot embossing. A first prototype and the mould will be made of Si-compounds.

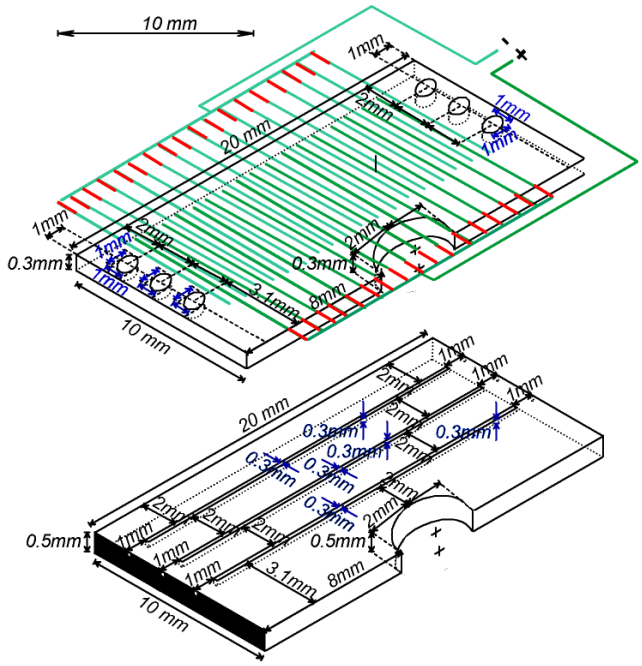


Fig.1: MEMS design with 3 microchannels for densitometric and spectrophotometric analysis of radioactive solutions. The green grid represents the pairs of electrodes (composed of one left and the closest subsequent electrode right), through which a voltage over each pair is imposed in alternating mode by subsequent activation of one left and one right red connection.

THE FLUID FLOW OF THE RADIOACTIVE SOLUTION

Fluid properties

To evaluate the fluid flow it is needed to know the density and viscosity of the radioactive solution in function of the lanthanide concentration. The radioactive solution is prepared with 3 molar nitric acid and the dissolved spent fuel, which contains typically 2 up to 250 grams lanthanide nitrate. Its properties in function of the lanthanide concentration and at varying temperatures and acidities of the solution are determined experimentally.

The density varies linearly with the concentration as given by the relations of Tanaka & Hosoma (2003)[9]. Typical input solutions have a reference density of 1.4g/cm³ with 150g/l U in the 3 molar nitric acid and at 25°C and a reference viscosity of 1.9 mPas with 150g/l U in the 3 molar nitric acid at 25°C.

Capillary flow

The flow in microchannels differs strongly from the classical one in macrochannels, as described by M. Gad-el-Hak (1999)[3] amongst others. Based on the electrical double layer (EDL) theory, presented a.o. by Israelachvili (1998)[4], we assume that the boundary layer of the liquid is composed of a nanolayer and a microlayer, as shown in Fig. 3. The nanolayer includes all molecular effects and is for simplicity modelled by an infinitely thin layer with a surface energy density γ_0 and with a surface charge density q. This surface charge density causes a long-range Coulomb effect as described in the EDL theory with a concentration of ions in the microlayer, and thus with an electric energy in it.

The surface energy of the microlayer becomes significantly larger than the one of the nanolayer in the case of a liquid interface in contact with a solid wall. Under the assumption that the wall is completely inert, a wall surface charge q_W induces an ion concentration in the microlayer with an additional electric energy.

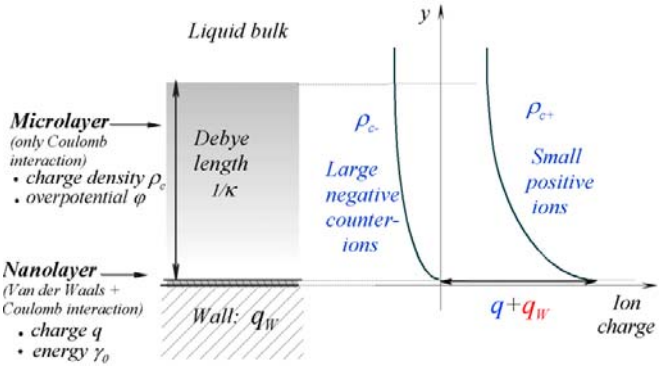


Fig. 3: Microscopic model of liquid/solid interface

To estimate the thickness of the microlayer, the Debye length has been calculated in function of the salt concentration with the formula:

$$\kappa^{-1} = \left(\frac{\sum z_i^2 e^2 n_{i\infty}}{\varepsilon \varepsilon_0 kT} \right)^{-1/2} \qquad (1)$$

Thereby the dielectric constant has been derived with the relationships of Wang & Anderko (2001)[10]. The Debye length varies linearly from 0.9nm for a 3M niric acid with 2 g/l lanthanide to 0.07nm for a nitric acid with 200g/l lanthanide, which is in agreement with Atkin's data [2].

The EDL-layer yields a self-induced EDL-potential φ, which fulfils the Poisson-Boltzmann equation and a microlayer at the wall boundary with a charge density gradient, as described in detail by Janssens-Maenhout & Schulenberg (2003)[5]. The calculated Debye length in nm range allows to model the EDL with vanishing thickness but with a given surface charge density, that differs from the bulk charge density.

This means that the microchannel flow can be calculated as a macroscopic laminar flow but with a slip condition at the

boundary, simulating the EDL. The slip velocity is calculated with the following characteristic number (dimensionless)

$$N_E = \frac{8zen_\infty l}{(\kappa a)^2}\frac{E_0}{\Delta p} \qquad (2)$$

that expresses the ratio of electric forces to hydraulic forces in the microlayer.

FEMLAB SIMULATION OF THE FLOW

The laminar flow in the microchannel has been simulated with a 3D hybrid mesh of which cross section is given in Fig.4 and with a Δp as driving force.

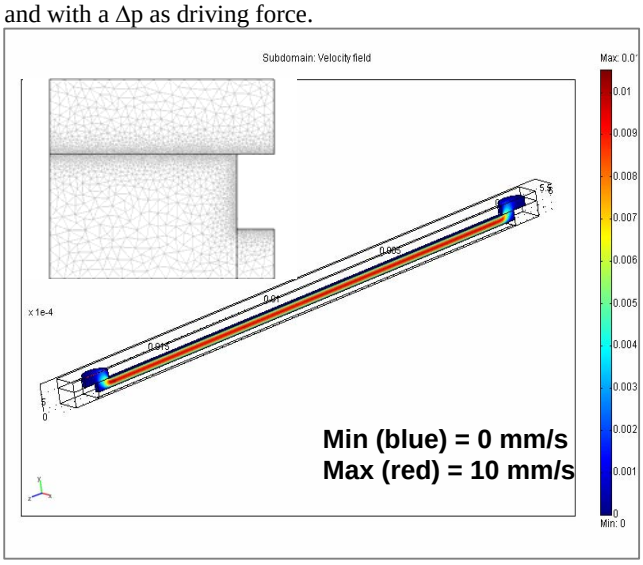
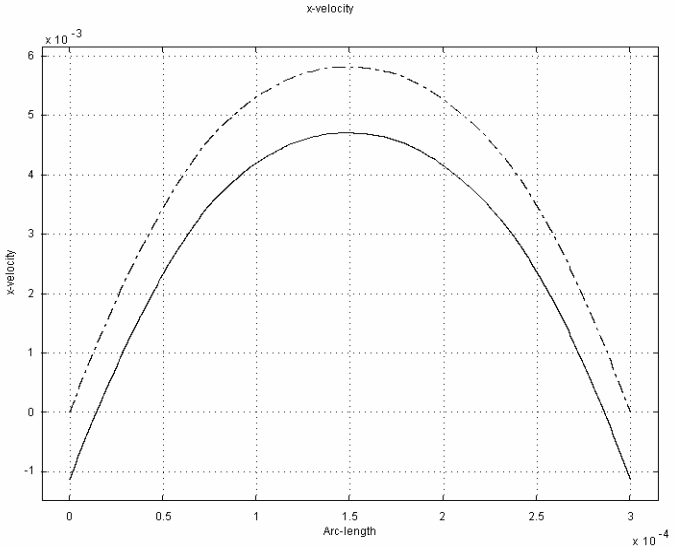


Fig. 4: Setup for microchannel flow simulation

The boundary conditions are:

- slip condition at the wall of -1.1mm/s
- symmetry at the central vertical plane of the horizontal microchannel
- inlet/outlet boundary specified with Δp = 50 Pa

The results, as shown in Fig. 5, show a Poiseuille profile of the laminar flow shifted down with 20% corresponding to the slip condition. The flow was fully developed at 150 μm from the inlet. No death zones have been indicated by the simulation. Therefore it can be assumed that concentration profiles averaged over the cross section are representative.



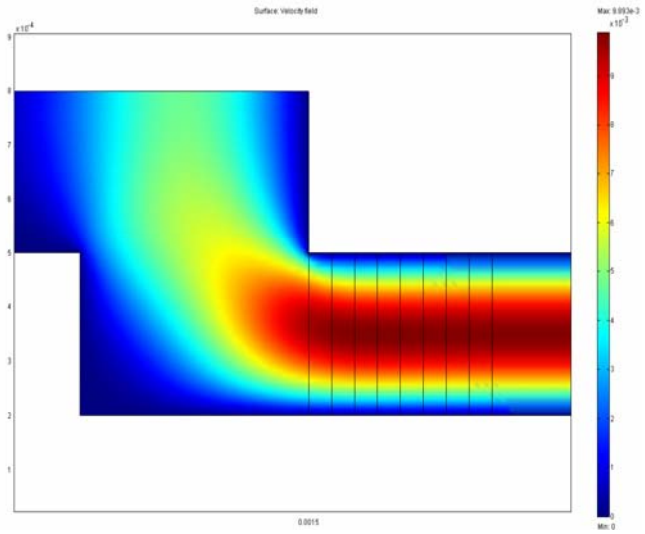


Fig. 5: Results of the simulated microchannel flow:
a) Poiseuille profile over the flow cross section;
b) Development of the flow in flow direction from inlet.

FILLING OF THE MICROCHANNEL

Taking advantage of the capillary effects of the microchannel flow, the technique of filling by means of electrophoresis has been selected because it was the most suitable method to move the radioactive liquid.

Other possibilities, by means of micro-pumps or by the surface acoustic wave principle have been evaluated as not appropriate. Movement with mechanical parts is always avoided in radioactive environments. Moreover, the liquid from the input accountancy tank may contain micro-size particles. As the liquid can be considered as a homogeneous emulsion, the properties, density, viscosity and surface tension remain valid. However due to the presence of particles the sound acoustic wave principle could not be applied for moving the liquid while determining the density.

The fluid will not be moved into the microchannel with a smooth constant flow but an electrophoretic peristaltic flow will be induced by a series of electrodes with constant potential difference and alternatively becoming active. With all the liquid properties of the radioactive solution, the pressure drop was calculated by superposition of the static and the dynamic pressure drop. The capillary action, resulting from the adhesion and surface tension leads to a dragging of the liquid into the microchannel with a concave meniscus. With the wetting behaviour of the liquid and its meniscus this upward force compensates a part of the static and dynamic pressure force.

To fill the microchannel by electrophoresis, an external axial electric field has to be applied such that the electric forces counteract the hydraulic forces. These electric forces increase with increasing salt concentration because of the growing hydrostatic and dynamic pressure drop for higher density liquids and because of the diminishing capillary action. The liquid movement into the channel can be expressed in relation to the required electric field by means of the electro-osmotic mobility

$$\mu_{eo}\big|_{150g/l\ Nd} = \frac{u}{E} = \frac{1\,mm/s}{19.8\,V/mm} = 0.05\,mm^2/Vs \qquad (3)$$

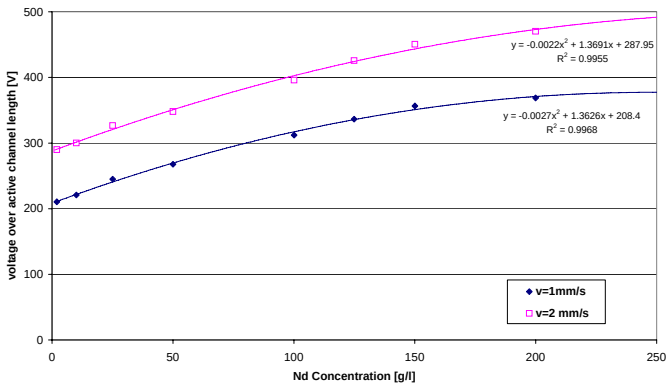


Fig. 6: Externally applied electric field forces the liquid into the microchannel.

This means that to fill the microchannel with a reference solution of 150 g/l Nd an electric potential of 356.52V is to be applied over the total active microchannel length of 18 mm in order to have a mean velocity of 1 mm/s. For 2 mm/s an electric potential of 450.66V is needed. Fig. 6 represents the total electric potential, necessary to move the liquid in the microchannel with mean velocities of 1mm/s and 2mm/s. Practically the electric potential will be established by inserting 10 micro-electrodes with a potential difference of 50V. This 50V potential difference is applied between succeeding electrodes in an alternating way from the first two till the 9th and 10th. The alternation of voltage between succeeding electrode pairs will cause a suction of the liquid in the microchannel in a peristaltic way.

With FEMLAB the electrophoretic flow has been simulated for the case of an external potential of 356.52V over the 18mm flow length. The Poiseuille profile, now shifted up towards values between 1.434mm/s and 1.438mm/s has been obtained, as shown in Fig. 7.

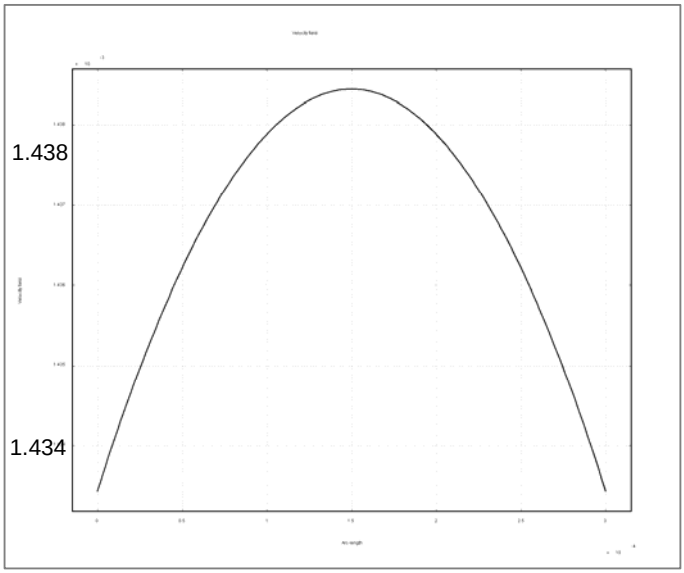


Fig. 7: Simulated cross section profile of the electrophoretic flow: the typical Poiseuille profile for the microchannel flow is again observed, shifted towards higher velocities because of the driving electrical field.

THERMO-DYNAMIC BEHAVIOUR OF THE FILLED MICROCHANNEL

FEMLAB simulations

It was needed to analyse as well the thermo-dynamic effects of the microchannel, because it will be filled with a radioactive and therefore hot solution. For these analyses the radioactive fluid can be considered as a thin heat heat source consist in the specific heat of the hot solution with the decay heat. Only a very small heat source (in the order of mWatt) needs to be taken into account because of the microvolume that contains this specific heat.

The heat transfer from the hot microchannel fluid towards the ambient has been simulated with FEMLAB by modelling

- conduction over the microchip material
- and free convection at the outer chip wall.

The temperature evolution over the surface of the microchip has been calculated with FEMLAB utilising the following boundary conditions:

- over the microchip: $q = \frac{T_1 - T_2}{L} = \lambda \frac{\Delta T}{L}$ (4)
- at the symmetry axis: insulation: $\mathrm{n} \cdot (\lambda \nabla T) = 0$ (5)
- at the outer wall: $q = h(T_s - T_\infty)$ (6a)

  with $\frac{Ts - T}{Ts - T_\infty} = 0.99$ and $h = \lambda / \delta$ (6b)

For a heat source of 50mW a maximum temperature difference of 5˚C with a the evolution of the temperature above and below the microchip as given in Fig. 8

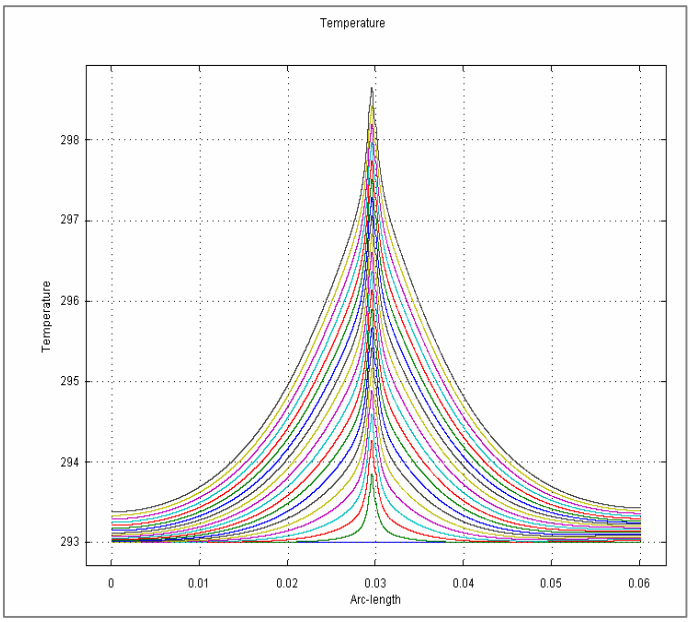


Fig. 8: Evolution of the heat transfer below the microchip (left) and above (right) the microchip with a heat source of 50 mW and an ambient temperature of 20˚C towards a thermodynamic equilibrium.

For a realistic case of 1.57mW heat source the temperature distribution after thermodynamic equilibrium is reached has been simulated and the different heat transfer coefficients calculated. The temperature distribution over the microchip is shown in Fig. 9 and the results for the heat transfer coefficients are given in Table 1.

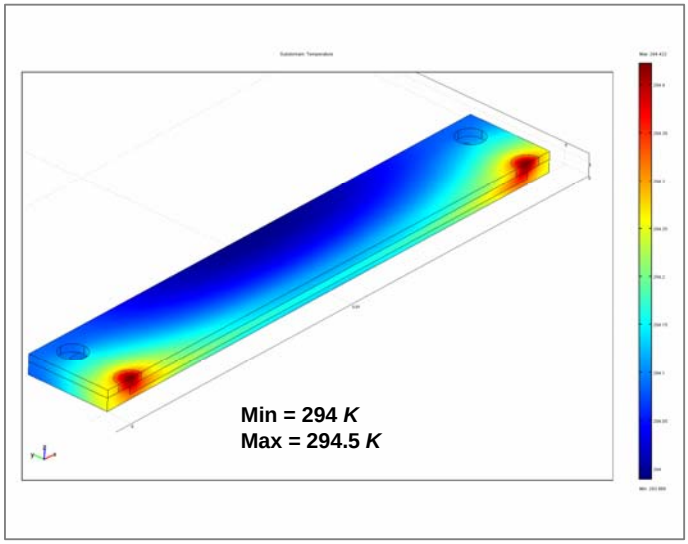


Fig. 9: Temperature distribution for a heat source in the microchannel of 1.57 mW. The temperature shows a maximum difference of 0.5˚C and the inlet and outlet channel which represent relative large volumes can be considered as the hot spots.

Heat transfer coefficient *h*	W/m/K
At the upper microchip cover	12.3
At the substrate bottom	10.9
At the side of the glass cover	8.9
At the side of the HDPE substrate	6

.Table 1: heat transfer coefficients for the different outer walls of the microchip

Comparison with FLUENT simulations

In order to verify the simulated temperature distribution the results for the temperature have been compared with the results of a FLUENT simulation by Uyttenhove (2005)[13] for a heat source of 1 mW, shown in Fig. 10. A similar behaviour has been observed with hot spots at the inlet and outlet and a maximum temperature distribution of 0.2 ˚C is of the same order as the 0.5 ˚C with FEMLAB for a 50% larger heat source. It can be concluded that the temperature distribution is qualitatively acceptable.

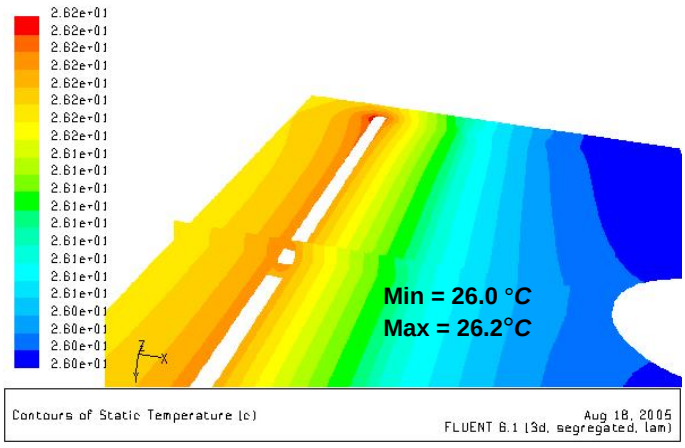


Fig.10: FLUENT simulation of the temperature distribution in the microchip for a heat source of 1 mW.

Comparison with experiments

In order to validation of the thermodynamic simulation model of the microchip it is needed to compare with real experiments. A thermographical experiment has been carried out by Uyttenhove (2005)[13] in which the heat source of the radioactive solution has been simulated with a copper wire in the microchannel. For increasing power, the surface of the upper cover and substrate bottom has been registered with a thermographical camera and the results of Fig. 11 have been found.

The experimental results indicate that below 1 mW no temperature difference over the chip larger than 0.5˚ can be observed. Both the FEMLAB as well as the FLUENT calculations are in agreement with this observation. A somewhat larger calculated temperature difference than measured can be accepted because the measured temperature distribution might be perturbed by a probable air layer between the copper wire and the microchannel.

It could definitely be concluded that the thermodynamic simulations are modelling the real microchip within acceptable variation range. Moreover it could be concluded for the microchip that a Peltier element for cooling the microchip is not needed, given the very low heat source.

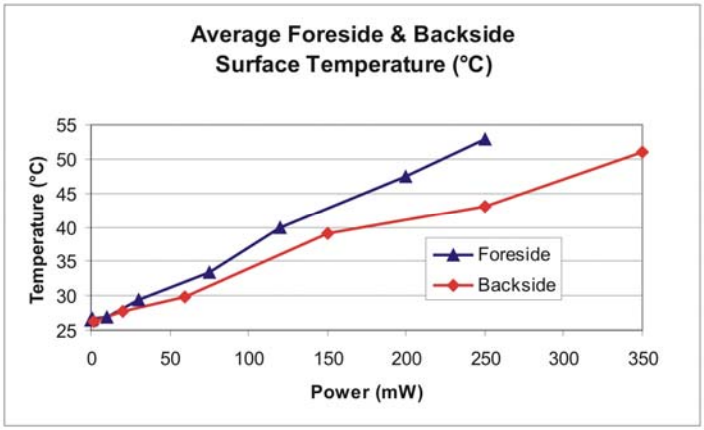


Fig. 11: temperature measured at the surface of the microchip (in which a copper wire is inserted to simulate the radioactive solution heat source) by a thermographical camera for increasing power.

Thermal stresses

Finally it is important to analyse of the microchip undergoes deformation due to temperature changes in the microchannel. A deformation would perturb the absorptiometry substantially because the light needs to transverse the complete channel length. A deformation of the channel itself or in relation to the positioning channel could make the absoprtiometry results invalid. Moreover the dependence of the absorption peaks on the wavelength is sensitive to the temperature. A change in temperature could induce a shift of the absorption peaks towards other wavelengths.

With the FEMLAB temperature distribution for a heat flux of 60 mW/cm^2 (corresponding to the case of 1.57 mW heat source) the distribution of the stresses has been calculated. The resulting distribution of stresses are represented in Fig. 12 and can be summarised with a maximum tensile stress of 48.3MPa and a maximum compressive stress of 109.82MPa.

The von Mises stresses of 29 MPa on the glass cover and 20MPa on the HDPE substrate needed to be compared with the maximum yield stresses for the glass (pyrex), i.e. 3500

MPa respectively for the HDPE (plastic), 28 MPa. It could be concluded that the stresses are acceptable for the materials, even without water bath or cooling element (Peltier). However it is recommended to utilise materials with similar expansion coefficients. Large stresses are observed between the pyrex cover and HDPE substrate because of strongly different volumetric expansion.

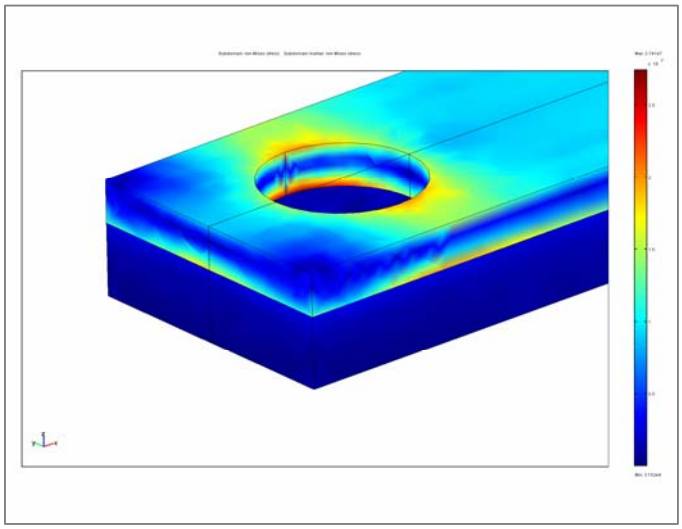


Fig. 12: Distribution of the thermal stresses in the microchip of HDPE and with glass cover.

CONCLUSIONS

This research work contributes to the feasibility study of the innovative application of a microchip to analyse a radioactive solution for nuclear safeguards purposes. This paper focuses on the micro-fluiddynamic and thermo-dynamic aspects of the microchip by means of FEMLAB simulations.

The micro-fluiddynamic simulations demonstrated that:

- the radioactive solution can be sucked into the microchannel by means of electrophoresis with an external field of 356V/18mm.
- the microchannel flow is laminar and shows a typical Poiseuille profile shifted down because of the slip boundary condition that is needed to model the Electrical Double Layer at the surface wall of the microchannel.
- No death zones could be indicated. This allows to assume that the average over the cross-section is representative for the entire solution
- The flow is fully developed after 150mm from the inlet.Therefore the flow can be assumed to be homogenous of the complete flow path along the axis of the microchannel

The thermo-dynamic simulations and the comparison with experiments and FLUENT simulations demonstrated:

- a quantitative good agreement with the experimental results and a qualitative good agreement with the FLUENT simulations. Therefore the thermo-dynamic model can be accepted as valid to predict the temperature distribution in similar microchip designs
- The microchannel with radioactive hot solution can be considered as almost isothermal because of the very low specific heat with decay heat of the radioactive solution.
- The thermal stresses observed in the microchip are acceptable in comparison to the yield stresses of the material. No special cooling with water bath or Peltier cooling element is necessary. The deformation will not perturb significantly the absorptiometry results.
- It is recommended to select for the substrate and the cover materials with similar expansion coefficients, in order to reduce the stresses between substrate and cover.

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