

Short thesis for the degree of doctor of philosophy (PhD)

**Investigation of physical and chemical effects of ion
irradiation on different materials and their possible
application**

*(Functionalization of microfluidic devices by microstructures
created with proton beam lithography)*

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1. Introduction

Microfluidic devices have become important part of technology nowadays in numerous fields of application, such as chemical reactions, biosensors, synthesis of nanomaterials, diagnostic device for detection of biological samples, etc.

In our work, we investigated the difficulty of the laminar flow mixing phenomena with fabricated microfluidic devices, functionalized them as passive micromixers. Our aim was to find structures, designs or any way to induce perturbation in the laminar flow, to make the mixing more efficient. This work deals with the computer simulation, the design and the creation of various microfluidic devices with and without integrated microstructures. As a new fabrication approach, PDMS microstructures were integrated into PDMS microdevices by combining proton beam lithography (direct writing technique utilizing focused, few MeV proton beams, to create micro- and nanoscale structures) and conventional UV lithography fabrication techniques. This way, the

microstructures and the micro-devices can be made from the same material, which is not only useful, but in case of certain applications (e.g. biomedical), it is essential.

2. Backgrounds and goals

It is well known, that the mixing of two fluid flow at microscale, even if they are the same material (e.g. water and a diluted aqueous solution), is still a great challenge for researchers because of the behavior of the laminar flow. Because of that, working with microfluidic devices, at certain applications, is still difficult, for example in chemical synthesis at microscale, analysis of chemical content of biological or biochemical applications, which gives importance to this area of research.

Aims during my doctoral work can be summerized as following:

- Literature overviewing, about simulation of microfluidic devices with different layouts, obstacles and designs to investigate the mixing properties of them using two different concentration liquids, as inlets.
- Finding good structures layout or design candidates for mixing, by using finite element

method (FEM) simulations, then fabricating them as real chips.

- Learn the steps of the fabrication of the microfluidic devices, first applying only macro lithography using UV lithography.
- Learn the microfabrication method using the focused proton beam microlithography with PDMS negative resist.
- Combining the two methods by integrating the microstructures, created with proton microlithography, to microfluidic devices and finalizing them with macro elements created with UV lithography.
- Finally, to investigate the mixing performance of the fabricated mixer devices and to compare their mixing efficiency values, then draw conclusions.

1. Thesis points

- I. **[P1]** According to my literature knowledge, mixing of fluids in microfluidic devices is essential in many chemical and biological applications, there is a difficulty of fluids mixing in microscale and there is low mixing by molecular diffusion depending on the geometry of the micromixer. Therefore, the mixing efficiencies of different passive micromixers (channel, circle, circle-with-pillars and circle-with-walls) were investigated using the simulation of different microstructures by Comsol software.

I proved, that the mixing efficiencies of passive micromixers can be improved by integrated proton beam written microstructures ensuring increased fluidic and decreased diffusion lengths in the same area / size microfluidic channels. Therefore, the mixing of fluids can be improved by decreasing the diffusion path of molecules or increasing the fluids contact area.

Publications related to this point are **[A1]** and **[C1]**.

II. [P2] The simulated microstructure were fabricated to be real microfluidic chips, and their mixing performance were evaluated. The real chips were created using lithography techniques (proton beam lithography, UV lithography).

Using FEM simulation technique and optical characterisation methods I proved, that the poor mixing efficiency of simple microfluidic channels can be improved by applying PB written microstructures, as additional walls to reach the acceptable (*min. 80%*) limit.

It was found that the mixing efficiency of the fabricated chips, channel, circle and circle-with-pillars, is low and cannot achieve an acceptable value (*80-100 %*). On the other hand, acceptable value (*82 %*) of mixing can be achieved using wall array.

Publications related to this point are [A1] and [C1].

III. [P3] Enhancements the mixing efficiency of the wall array micromixer can be achieved by changing the geometry of the walls (length and number). Comsol simulations were carried out for

different walls microstructures (simple circle [SC], circle with low number and same level walls [CLSW], circle with high number and same level walls [CHSW], circle with low number and tall (length) walls [CLTW], circle with high number and tall (length) walls [CHTW]).

It was found that enhancement the mixing efficiency of the proposed passive micromixers depends on the length and number of walls. So the mixing efficiency was enhanced to an excellent values (99.45 %) for CHSW micromixer and (99.99 %) for CHTW micromixer.

IV. [P4]The fabrication of the real micromixers with different walls-array were created using lithography techniques (PBL, UV).

It was found that the mixing efficiency was enhanced to excellent values, due to the used different geometry (length and density). The mixing efficiency value reach to values as high as 95.86 % for CHSW and 97.08 % for CHTW.

Publications related to this point are [A2] and [C2].

V. [P5] Comparison of the mixing efficiency results, simulation and experimental, was studied for the different walls microstructures.

It was found that there is a matching to some extent between the simulated and experimented mixing efficiency as summarized in the following table and figure:

Micromixer type	SC	CLSW	CHSW	CLTW	CHTW
Experimental mixing efficiency	29.11 %	63.67 %	95.86 %	81.72 %	97.08 %
Simulated mixing efficiency	38.17 %	73.18 %	99.45 %	91.36 %	99.99 %

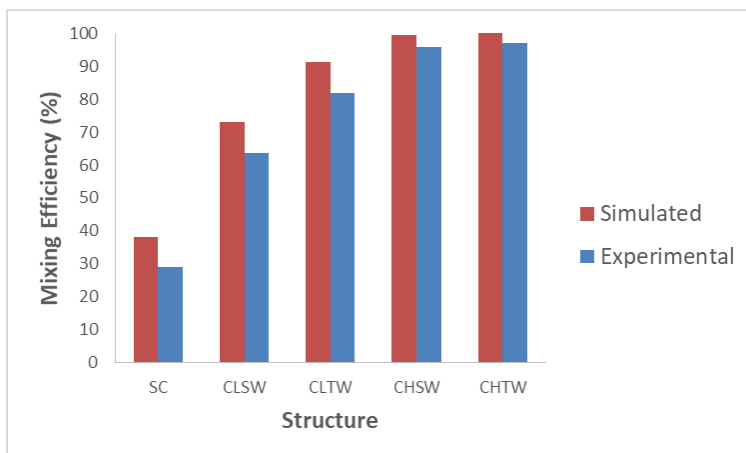


Figure 1. Comparison of simulated and experimental mixing efficiency for different microstructures micromixers.

Publications related to this point are [A2] and [C2].

3. Methods

The difficulty of the problem of fluid mixing in microscale through our microstructures of the designed passive micromixers is the backbone of our study and our main research challenge.

The applied methods can be summarized as following:

- The COMSOL simulations of designed micromixers were performed using the Microfluidics Modules (creeping flow and transport of diluted species), which were applied to perform the mixing simulations for two-dimensional designs like straight channel, simple circle, and circle with micropillars and with different walls.
- Mask-less lithography technique with a focused proton beam (PBL: Proton Beam Lithography) was used to fabricate microstructures on PDMS resist material.
- Masked UV-photolithography technique was used to make patterns on SU-8 photoresist materials, those

patterns were used to create the PDMS caps of our micromixers.

- Air plasma treatment was used to bond together the fabricated PDMS cap and PDMS microstructure to finalize the build-up of our microfluidic chips for passive micromixing test.
- The mixing efficiency values of our different passive micromixers were investigated numerically using COMSOL simulations and were compared for different designs microstructures. Also the values of mixing efficiency were calculated experimentally for the same simulated microstructures, using UV/vis spectrophotometry and mixing test system, at different flow rates. The simulated and experimental results of mixing efficiency have been compared to measure the extent of their compatibility.

4. Results

Various microfluidic devices with and without integrated microstructures were designed, optimized and created for micro-mixing purposes. As a new fabrication approach, poly (dimethyl-siloxane) (PDMS) microstructures were integrated into PDMS microdevices by the combination of conventional UV lithography and proton beam lithography fabrication method. This way, the microstructures and the micro-devices can be made from the same material, which is important in case of certain applications.

To compare the mixing capabilities of the proposed microstructure designs, real mixing tests were carried out. And the results can be categorized to two parts *A* and *B*, compared as following:

- A) The results of different microstructures like (channel, circle, circle-with-pillars and circle-with-walls micromixers), can be summerized as following:
- The simulation results show that the mixing efficiency of the channel and the circle

micromixers was low. However, the mixing efficiency of circle with pillars was improved a little bit, and the best mixing result was obtained using circle-with-walls structure.

- By experimental tests for the real chips (channel, circle, circle-with-pillars and circle-with-walls micromixers), the mixing efficiencies were calculated and found to be 58 %, 29 %, 72 % and 82 % respectively, at flow rate $1 \mu\text{l}/\text{min}$.
- Low pressure drop is very important for industrial production because it decreases energy requirements. The pressure drop for our simulated structures was calculated through the inlets and outlets for all the mentioned structures and its range was $(0.139 - 1.74 \text{ kPa})$.
- For the real fabricated structures, the drop pressure was calculated at flow rate range $(1-6) \mu\text{l}/\text{min}$ for the different micromixers using the pressure drop equation, and the results are listed in the following table.

Micromixer type	Flow rate (μl/min)	Pressure drop ΔP (kPa)	Micromixer type	Flow rate (μl/min)	Pressure drop ΔP (kPa)
Channel	1	0.05	Circle	1	0.01
	2	0.09		2	0.01
	3	0.14		3	0.02
	4	0.18		4	0.02
	5	0.23		5	0.03
	6	0.27		6	0.03
Pillars	1	0.03	Walls	1	1.19
	2	0.06		2	2.38
	3	0.1		3	3.56
	4	0.13		4	4.75
	5	0.16		5	5.94
	6	0.19		6	7.13

B) The results of different micro wall arrays (simple circle [SC], circle with low number and same level walls [CLSW], circle with high number and same level walls [CHSW], circle with low number and tall (length) walls [CLTW], circle with high number and tall (length) walls [CHTW]) can be summarized as following.

- For simulation results, the efficiency of mixing was poor and its value was 38.17% for simple circle [SC], while it improves for higher values

73.18 % for circle with low number and same level walls [CLSW] but still below the accepted range of mixing efficiency. The accepted value 91.36 % of mixing efficiency can be obtained by low number and tall (length) walls [CLTW], while excellent values of mixing efficiency 99.45 % and 99.99 % were achieved using micromixer circle with high number and same level walls [CHSW] and circle with high number and tall (length) walls [CHTW] respectively.

- For the real chips tests, the values of mixing efficiency at flow rate $1 \mu\text{l}/\text{min}$ were 29.11 %, 63.67 %, 95.86 %, 81.72 % and 97.08 % for the micromixers SC, CLSW, CHSW, CLTW and CHTW respectively. The highest mixing efficiency can be achieved using CLTW and CHTW micromixers due to the fact of its geometry, which increases the surface area between the different fluids and decreases the diffusion path.
- The Reynolds number were calculated for all design and geometries as a function of the flow

rates ($1-6 \mu\text{l}/\text{min}$). The calculations showed that Re ranges ($0.19-1.15$), ($0.43-2.61$), ($0.49-2.96$), ($0.51-3.06$) and ($0.49-2.96$) for SC, CLSW, CHSW, CLTW and CHTW, respectively. This low value of Re indicates that in our work the flow is laminar. The drop pressure was calculated at flow rate range ($1-6 \mu\text{l}/\text{min}$) for the different walls micromixers using the pressure drop equation, and the results are listed in the following table.

Micromixer type	Flow rate ($\mu\text{l}/\text{min}$)	Pressure drop ΔP (kPa)	Micromixer type	Flow rate ($\mu\text{l}/\text{min}$)	Pressure drop ΔP (kPa)
CLSW	1	0.58	CHSW	1	1.26
	2	1.16		2	2.51
	3	1.74		3	3.77
	4	2.33		4	5.03
	5	2.91		5	6.29
	6	3.49		6	7.54
CLTW	1	1.19	CHTW	1	1.87
	2	2.38		2	3.74
	3	3.56		3	5.61
	4	4.75		4	7.48
	5	5.94		5	9.34
	6	7.13		6	11.21

As a conclusion of the different microstructures, we successfully integrated PDMS microstructures into PDMS microfluidic devices. With a proper design of the microstructures, the mixing efficiencies could be significantly improved, and reached as high as 82%, keeping the size of the chip in the order of a few millimetres. The presented method opens the opportunity to further improve mixing efficiencies and to further decrease chip dimensions, by the development of new designs and by fabricating smaller and finer structures, for example, by nano-lithography (nano sized proton beam lithography).

As a conclusion of the different micro wall arrays, the length and the areal density of these walls were varied in order to find a better mixing performance. The results showed that, generally, the more densely distributed and the longer walls are more efficient and their combination (long walls, densely distributed) showed the highest performance.

The devices were then created with ion microlithography and UV lithography techniques. Mixing test were

performed from which the mixing efficiencies were determined.

It was found, that the mixing efficiency of the fabricated passive micromixers, the densely distributed short walls and the more densely distributed long walls, improves to an excellent value of 95.86 % and 97.08 %, 1 μ l/min flow rate. From these results, we can conclude that the distribution of the walls has the highest effect on the mixing.

5. Scientific Publications

Publications on the results of the present thesis

- **[A1]: Emad Nady**, Gyula Nagy, Róbert Huszánk, (2021). Functionalization of microfluidic devices by microstructures created with proton beam lithography. *Vacuum*, 190, 110295. <https://doi.org/10.1016/j.vacuum.2021.110295>
IF²⁰²¹=3.627
- **[A2]: Emad Nady**, Gyula Nagy, Róbert Huszánk, (2021). Improvement in mixing efficiency of microfluidic passive mixers functionalized by microstructures created with proton beam lithography, *Chemical Engineering Science Journal*.
<https://doi.org/10.1016/j.ces.2021.117006>
IF²⁰²¹=4.311

Publications not related to the dissertation

- **Emad N. Saad**, M. F. Eissa, Emad A. Badawi, M. A. K. El-Fayoumi (2014); Investigation of the physical properties of polymeric materials induced by alpha radiation Int. J. of Adv. Res. 2 (Dec). (ISSN 2320-5407).
- **Emad.N. Saad**, M. F. Eissa, Emad A. Badawi, M. A. K. El-Fayoumi. Study the effect of irradiation angle on the microstructure of CR-39 polymer, Applied Radiation and Isotopes Journal, submitted.

Conferences

- **[C1]: Emad Nady**, Róbert Huszánk, Gyula Nagy: A Novel Passive Micromixer with Microstructure by Proton Beam Lithography for High Mixing Efficiency at Low Reynold Number, 17th International Conference on Nuclear Microprobe Technology and Applications (ICNMTA

2020),14-15 September 2020, Beld, Slovenia, on-line, poster.

- **[C2]: Emad Nady**, Gyula Nagy, Róbert Huszánk: Functionalization of microfluidic devices by microstructures created by proton beam lithography, 18th International Conference on Thin Films and 18th Joint Vacuum Conference (ICTF-JVC 2020), 22-26th November 2020, Budapest, Hungary, on-line, poster.



Registry number: DEENK/87/2022.PL
Subject: PhD Publication List

Candidate: Ahmed Emad Nady Saad
Doctoral School: Doctoral School of Physics
MTMT ID: 10068199

List of publications related to the dissertation

Foreign language scientific articles in international journals (2)

1. **Nady, E.**, Nagy, G., Huszánk, R.: Improvement in mixing efficiency of microfluidic passive mixers functionalized by microstructures created with proton beam lithography.
Chem. Eng. Sci. 247, 1-8, 2022. ISSN: 0009-2509.
DOI: <http://dx.doi.org/10.1016/j.ces.2021.117006>
IF: 4.311 (2020)
2. **Nady, E.**, Nagy, G., Huszánk, R.: Functionalization of microfluidic devices by microstructures created with proton beam lithography.
Vacuum. 190, 1-9, 2021. ISSN: 0042-207X.
DOI: <http://dx.doi.org/10.1016/j.vacuum.2021.110295>
IF: 3.627 (2020)

Total IF of journals (all publications): 7,938

Total IF of journals (publications related to the dissertation): 7,938

The Candidate's publication data submitted to the iDEa Tudóstér have been validated by DEENK on the basis of the Journal Citation Report (Impact Factor) database.

17 February, 2022





Nyilvántartási szám: DEENK/87/2022.PL
Tárgy: PhD Publikációs Lista

Jelölt: Ahmed Emad Nady Saad
Doktori Iskola: Fizikai Tudományok Doktori Iskola
MTMT azonosító: 10068199

A PhD értekezés alapjául szolgáló közlemények

Idegen nyelvű tudományos közlemények külföldi folyóiratban (2)

1. **Nady, E.**, Nagy, G., Huszánk, R.: Improvement in mixing efficiency of microfluidic passive mixers functionalized by microstructures created with proton beam lithography.
Chem. Eng. Sci. 247, 1-8, 2022. ISSN: 0009-2509.
DOI: <http://dx.doi.org/10.1016/j.ces.2021.117006>
IF: 4.311 (2020)
2. **Nady, E.**, Nagy, G., Huszánk, R.: Functionalization of microfluidic devices by microstructures created with proton beam lithography.
Vacuum. 190, 1-9, 2021. ISSN: 0042-207X.
DOI: <http://dx.doi.org/10.1016/j.vacuum.2021.110295>
IF: 3.627 (2020)

A közlő folyóiratok összesített impakt faktora: 7,938

**A közlő folyóiratok összesített impakt faktora (az értekezés alapjául szolgáló közleményekre):
7,938**

A DEENK a Jelölt által az iDEa Tudóstérbe feltöltött adatok bibliográfiai és tudományometriai ellenőrzését a tudományos adatbázisok és a Journal Citation Reports Impact Factor lista alapján elvégezte.

Debrecen, 2022.02.17.

