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Miniaturized spray injection systems

Sistemas de injeção por nebulizador miniaturizado

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Abstract

Spray production and its miniaturization are common need and goal in Chemical Engineering and Chemical fields. This work aims the production of miniaturized low cost spray injection systems using planar and three-dimensional microchannels, both 73 cm long and 100 µm diameter, and acrylic as substrate. Tests and simulation used fluid flow rates up to 10 standard mL/min, for gaseous samples, and up to 1 mL/min to liquid ones; tracers and filming were used to understanding fluid behavior. Simulations and experimental results showed good agreement: planar channels seem to be useful for mixing two liquids whereas three-dimensional channels produce small droplets useful for sample pretreatment in miniaturized impactors.

Keywords: Miniaturized structures; Spray; Impactors; Sample pretreatment.

Resumo

A produção e miniaturização de nebulizadores é uma necessidade além de um objetivo na área de Engenharia Química e Química. Este trabalho objetiva a produção de sistema de injeção por nebulizador miniaturizado usando, para tanto, microcanais planares e tridimensionais. Tais canais têm 73 cm de comprimento, 100 µm de diâmetro e foram produzidos usando acrílico como substrato. Simulação e testes usaram velocidade de fluido de até 10 ml/min, para amostras gasosas, e 1 ml/min, para amostras líquidas, além de traçadores e filmagem para entender o comportamento do fluido. As simulações e medidas experimentais apresentaram boa concordância: parece ser mais adequado canais planares para misturas de dois líquidos enquanto canais tridimensionais produzem gotas pequenas, úteis para proceder a pré-tratamento de amostras em impactadores miniaturizados.

Palavras-chave: *Estruturas miniaturizadas; Spray; Impactadores; Pré-tratamento de amostras.*

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Introduction

Spray production is a very common need on Chemical Engineering and Chemical fields. On Chemical Engineering, several different unit operations will rely on spray formation; therefore, dryers or cooling towers and extraction or leaching equipment are quite dependent on spray formation[1]. Spray formation and manipulation is part of aerosol science and also fundamental to irrigation processes[2]. Spray aerosol generators are applied to the formation of thin films and fine powders[3] and even on domestic environments scrubbers, sprinklers and atomizers are common day-to-day tools. Thus, Liu considered that atomization of liquids is a technology used in almost all industrial operations, in applications such as evaporative cooling, combustion, gasification, fire suppression, agriculture and spray drying[4].

Many times, chemical analysis also requires spray formation, such as atomizers in flame atomic absorption spectrometry, which Wu quoted as a traditional analytical technique for trace element determination but with poor sensitivity due to nebulization efficiency, that could be improved by thermospray[5]. Krucker stated that hyphenation, such as HPLC-MS (high-performance liquid chromatography to mass spectrometry), requires as interface an efficient spray formation; furthermore, liquid chromatography also uses electrospray ionization (ESI) for soft ionization of the analytes[6], which makes electrospray and thermospray important tools also on mass spectrometry hyphenation[7]. On these electrosprays, droplets occur in a wide size range, which means bigger droplets might be analyzed by optical imaging[8,9] but not the smaller ones[10]. Thus, as remarked by Arruda, since nebulization and atomization efficiency are directly correlated with the sensitivity of analytical methods, nebulizer and atomizer systems are the key tools on chemical analysis[11].

Furthermore, miniaturization has been a driving force for new developments and some examples can be collected on micro electrospray manufacturing aiming the use on mass spectrometry equipment[12-15] or capillary electrophoresis and mass spectrometry hyphenated methods[16]. However, even with the huge development on MEMS (Microelectromechanical Systems) field, many nebulizers and electrospray devices are based on capillary tubes and flow interactions and just a few present dimensions on microdevice range[17,18].

This possible gap among the nano-, micro- and macro-technologies on the chemical engineering field is a main concern for some authors[19,20] since miniaturization is an important driving force on development of green chemical process and a meaningful step for the development of a sustainable world. Thus, in a final analysis, there is a gap on devices with meso scale dimensions, i.e., manufactured in an intermediary size range between macro and microdevices. Nonetheless, devices with meso structures present several advantages because they can be manufactured using conventional tools; therefore, this dimension range was addressed in our former works by the manufacturing and testing of microreactors[21,22].

The production of liquid sprays, according to Pougatch[23], can be successfully done by two main methods: jets or fluid interactions. First method requires that the liquid be pressured against an orifice whereas in the second one gas and liquid interact and pass through a constriction. The design of such constriction and the location of gas insertion define, among other features, droplet size, which is usually small on such devices. Thus, liquid sprays can easily be miniaturized by the production of microchannels. Moreover, on MEMS devices, microchannels are widely spread on transportation and mixing of fluids and can also perform tasks as microreactors, from relatively simple devices to completely automatic equipment[24]. Due to the use of microelectronics technology for production of MEMS devices, microchannels are normally obtained in a planar shape. An important use for microchannels that normally relies on a planar structure is the manufacturing of chromatographic columns for gas or liquid analysis, most of them built on a silicon wafer[25-28]. In our former works, not only planar but also three-dimensional microchannels were machined and sealed using conventional tools, i.e. mechanical lathe/milling cutter and adhesives[29,30]. Whereas planar microchannels may present retention of organic compounds due to secondary phenomena, such as capillary effects[31], three-dimensional microchannels may act as a chromatographic column[30]. Since planar microchannels can present advection zones, this might also produces spray tiny particles, whereas simple changes in design of three-dimensional microchannels allow multiple interactions in the fluid[21]. However, to the best of our knowledge, no attempts in producing miniaturized spray using such approach are available. Therefore, the aim of this work was the production of miniaturized low cost spray injection systems based in the interaction between gas and liquid flows in planar (spiral) and three-dimensional (helical) microchannels.

Experimental

The microstructures that form the spray systems are composed of two parts: microchannels and external packing. These structures were produced to allow gas and liquid injection, i.e., fluid interactions liquid sprays according to Pougatch[23]. Two distinct microchannels, both 73 cm long and 100 µm diameter, and similar to previously evaluated ones[30,31], were modified to form the sprays. Figure 1 shows schematics of spray microstructures and microchannels on the details. In order to insert gas on these structures, secondary channels or orifices were machined. The planar channel received an orifice in the center and, just for comparison with three-dimensional channel, was also divided in two parts. The three-dimensional channel, which corresponds to a microthread machined on one cylinder, was scratched out perpendicularly. Thus, the general idea is the simultaneous admission of gas and liquid on a single long channel but with multiple interactions among fluids. The external sealing packing does not present channels; therefore, the internal volume and area is 1.1×10^{-3} cm^3 and $1.4 cm^2$, respectively, with area/volume ratio of 1300. Sealing was performed using a 5µm thick tape (3M, YR-9767) on the spiral planar shape microchannel and silicone glue on three-dimensional microchannel. The structures were manufactured in poly(methyl methacrylate) – PMMA 5 mm thickness bars (Plastotal Ltda., Brazil), with milling cutter and mechanical lathe, respectively. PMMA was used due to the optical transparency that allows photographic tests. The integrity of both structures was evaluated and correspond to an overpressure up to 3 atm, either in liquid or gaseous flow.

Tests and simulation used fluid flow rates up to 10 standard mL/min for gaseous samples an up to 1 mL/min to liquid ones. The fluids were nitrogen or water used on gas or liquid phase, respectively. Tracers and filming (Sony, DSCW7 – P72, USA) were used for evaluation of fluid behavior and/or spray formation. Tracers are composed by aqueous solutions and particles suspensions. Solutions are 10% wt. aniline, with several colors. Dispersions used 13µm particles (Carborundum Abrasivos Ltda, Brazil) suspended in water and, if necessary for enhancement of the visualization process, the suspension was also dyed with aniline. The reactants are all P.A (Casa Americana SA, Brazil) and only distilled water was used on flow tests and solutions preparation. Optical microscopy used conventional equipment (Baush&Lomb, model 311871). In order to understand the flow behavior inside the structures, incompressible Navier-Stokes 2D or 3D simulations were performed using FEMLAB® 3.2b in a Pentium IV platform (2.4 GHz, 2 GB of RAM).

Tests of spray formation were carried out in a setup shown in Fig. 2. The setup comprises a small centrifugal pump with two simultaneous outlets and a capillary that works as recipient; therefore the maximum amount of liquid turned into spray is controlled by its volume. A paper sheet is maintained 10 cm apart from the spray outlet to evaluate droplet formation and dimension. On such condition only the biggest droplets will be determined (formed by Rayleigh mechanism)[32].

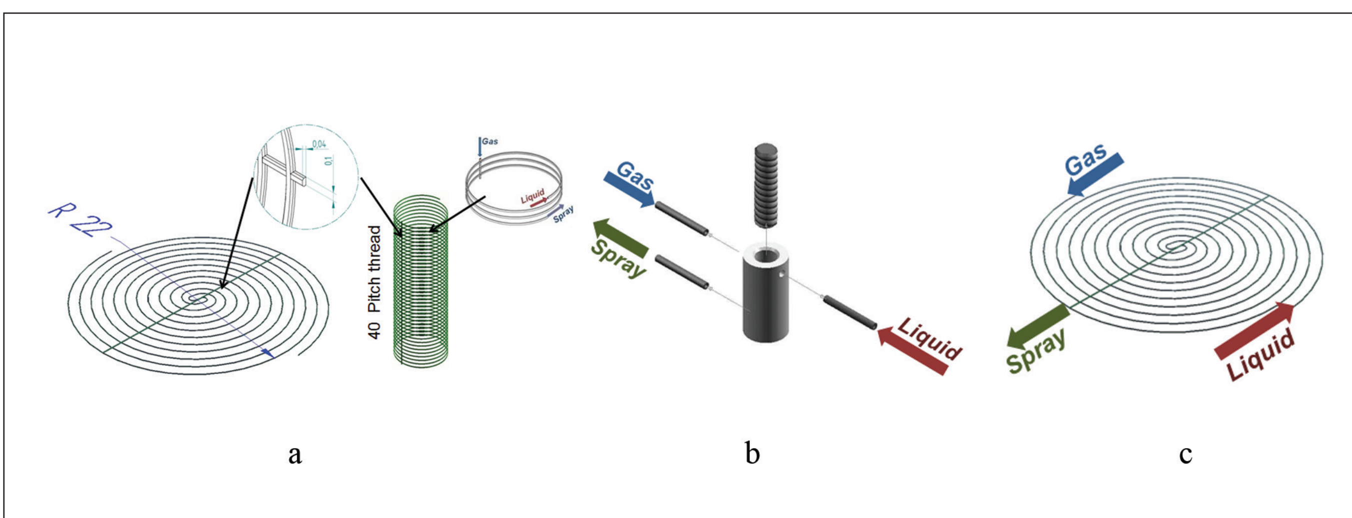

Figure 1. Spray manufacturing (a). Details (mm) for three-dimensional microchannels (b) and planar (c) setups.

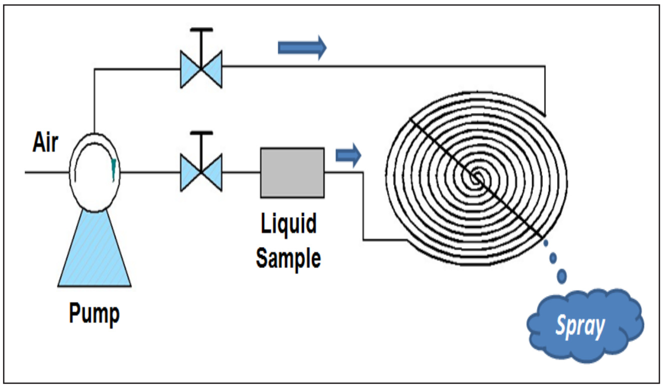

Figure 2. Setup for spray formation and a planar structure

Results and Discussion

This section comprises the spray behavior, simulation and experimental, on planar and three-dimensional channels.

Planar microchannels

On planar microchannels, as can be seen in Fig. 3, there are periodic variations on velocity, vorticity and Reynolds numbers that might help on spray formation. Thus, due to the high flow dispersion and main variations of velocity and Reynolds numbers that occur on the center of the structure (see details in Fig. 3), an orifice on this area probably favors spray formation and three different possibilities were simulated, as can be seen in Fig. 4. If sample is removed on such region advections zones can be produced (1st option). On the other hand, due to differences on length, each turn on the spiral presents a slight different velocity profile, thus, addition of liquid sample on the center can disturb the flow significantly (2nd option). Addition of gas on the center might pressurize the whole structure, which is also useful for spray production (3rd option). Nonetheless, all possibilities presented the same behavior, with extremely high velocity profile and vorticity, which is characteristic on spray systems.

On the other hand, multiple gas/liquid interactions are difficult to implement. If a long channel is scratched perpendicularly to the inlet, as can be seen in Fig. 5, there will be low interaction between the two phases. Nonetheless, several orifices (one on each intersection between structure channel and perpendicular channel) show a better interaction among the fluids in the perpendicular channel, with a higher vorticity if compared with only one orifice; therefore, this design might be useful in miniaturized mixers.

Experimental results show good agreement with simulations. All situations described in Fig 4 produced spray droplets; nonetheless, the smaller drops are obtained if dro-

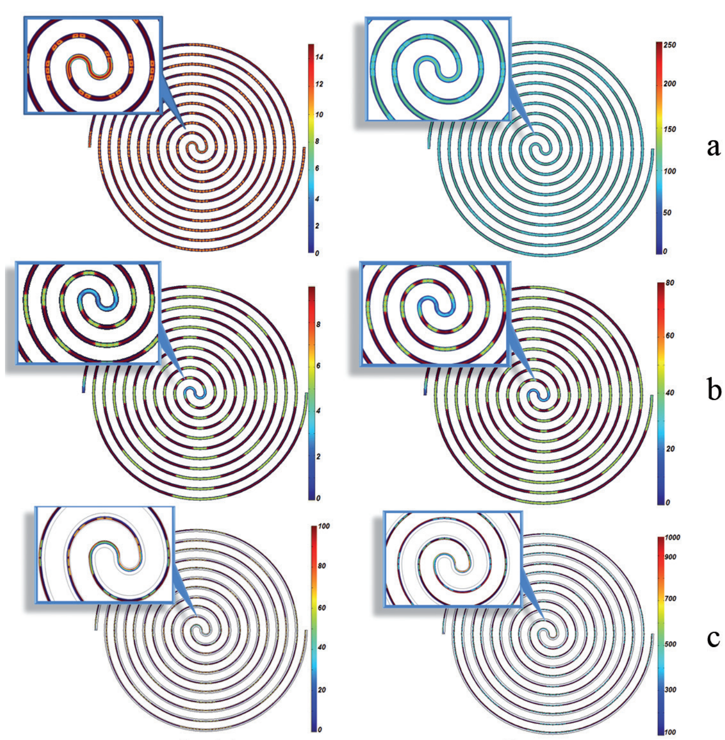

Figure 3. Velocity profile (cm.s^{-1}) (a), Reynolds number (b) and vorticity (c) to gaseous or liquid flow inserted in a planar channel.

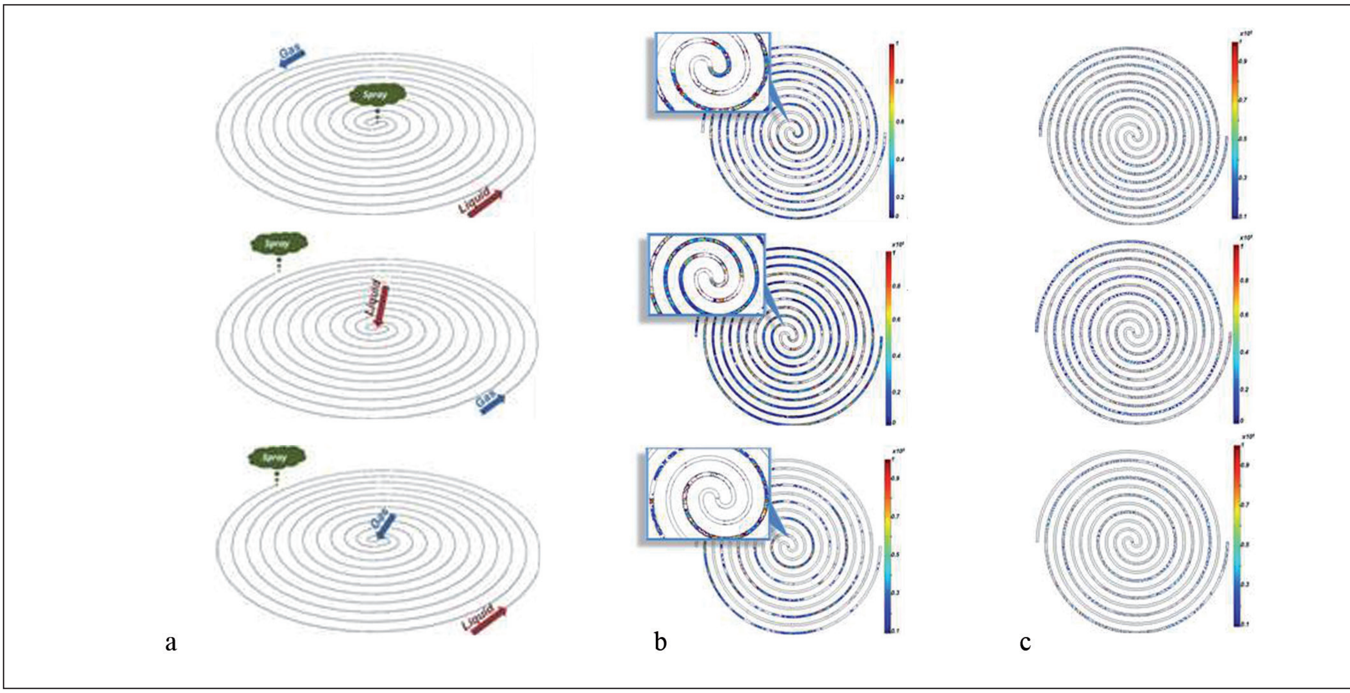

Figure 4. Spray production using planar spiral channels and an orifice in the center of the structure: (a) schematics, (b) velocity profile ($cm.s^{-1}$) and (c) vorticity.

plets' collecting is in the center of the structure. On such condition, it is not observed droplets, i.e., the biggest diameter drop is less than 20 µm. However, on the other two conditions, at the beginning and ending of liquid sample, big droplets, up to 0.1mm diameter, can be formed and Fig. 6 shows typical results.

Tests were carried out on the best configuration (spray formed in the center of the structure) and Fig. 7 shows typical results for mixing and spray formation using suspensions. Mixture occurs if two different liquid samples are simultaneously injected in the structure and the detail in Fig. 7 shows the aspect of an adsorbent paper placed 10 cm far from the outlet and using a sample that completely fulfill the channel, i.e., minimum volume of 1 mL. The obtained droplets are at least 1 mm diameter and were formed for several small drops reaching the same place. Although some color differences can be observed, the main trend is the formation of dark spots (mixture of red and blue dyes). Particle suspensions show an increase in droplets diameter and the maximum dimension observed seems to be dependent on the particle diameter, with mixtures presenting the biggest droplets. A possible mechanism that explains this behavior is particle coalescence, probably during droplet formation inside the channel and due to pressure variations, which indicates this spray configuration to suspensions with less than 10% wt particles; furthermore, it is worthing to note that coalescence probably implies in clogging inside the channel and must be avoided.

Three-dimensional microchannels

Three-dimensional channels do not favor vorticity and/or velocity variation, as can be observed in Fig. 8. On the other hand, multiple gas/liquid interactions can be easily implemented. Since the increase on step screw raises the drift on velocity[21], two different situations were simulated: interaction between two channels with different step screws and a scratch perpendicular to the screw axis of symmetry. Figure 9 shows the interaction on the first lap of two channels; if one of them has a step screw four times bigger than the other (Fig. 9a), although gaseous flow presents a high velocity variation, the velocity on the liquid flow is only slight perturbed, which indicates spray will hardly be produced on such circumstances. However, a higher interaction can be perceived with the two jets interacting in 90º angle, the velocity profile changes although the maximum velocity remains the same and, probably as a consequence of these interactions, vorticity and Reynolds number also increase Fig. 9b). Simulation of interaction in three screw laps with 90º angle is shown in Fig. 9c, where is possible to observe a high variation on velocity profile and vorticity.

Experimental measurements show good agreement with simulation results, as can be seen in Fig. 10 that depicted spray jet and droplets obtained 10 cm apart from the jet nozzle, in the detail. The biggest droplets do not exceed

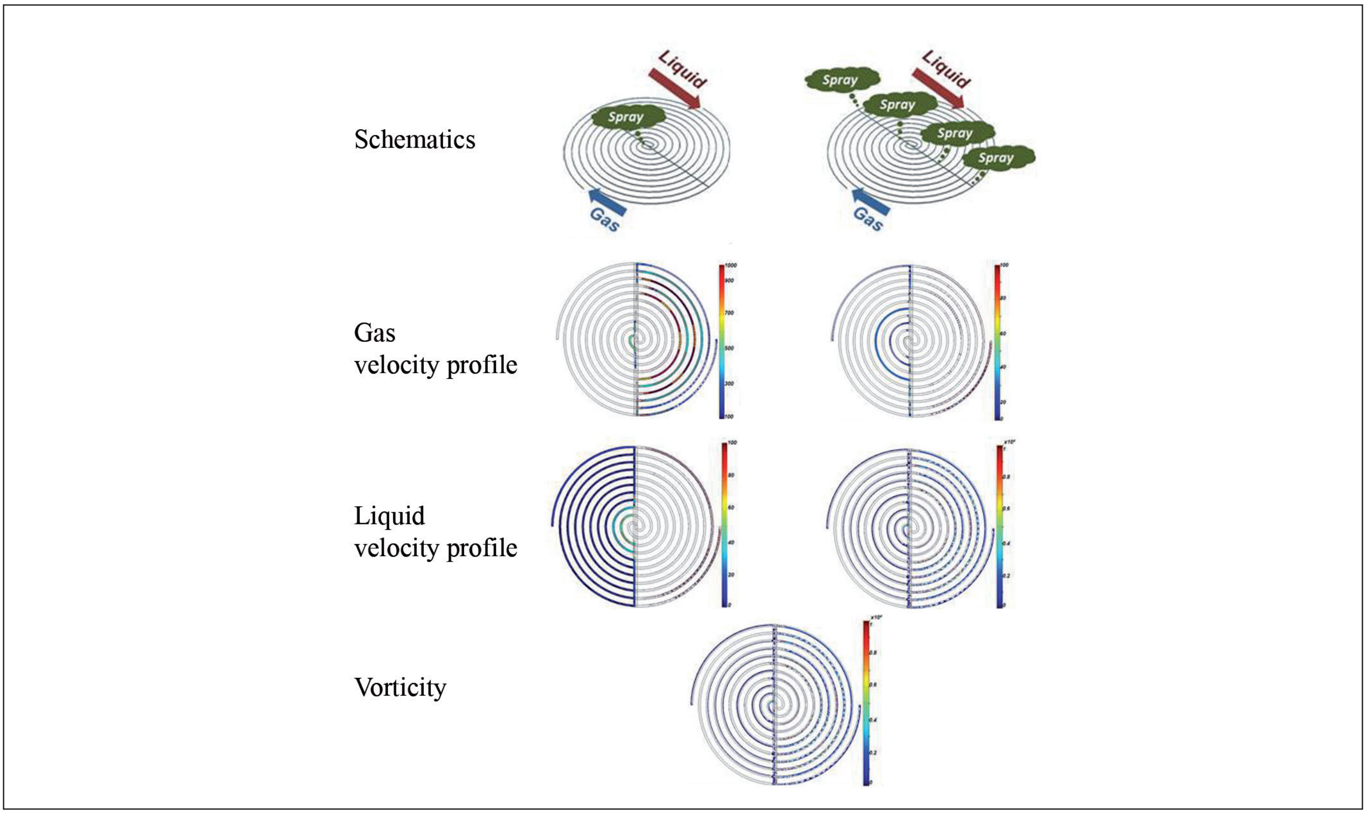

Figure 5. Multiple gas/liquid interactions. Schematics; velocity field (cm.s^{-1}) for gaseous and liquid flow and vorticity on planar structures.

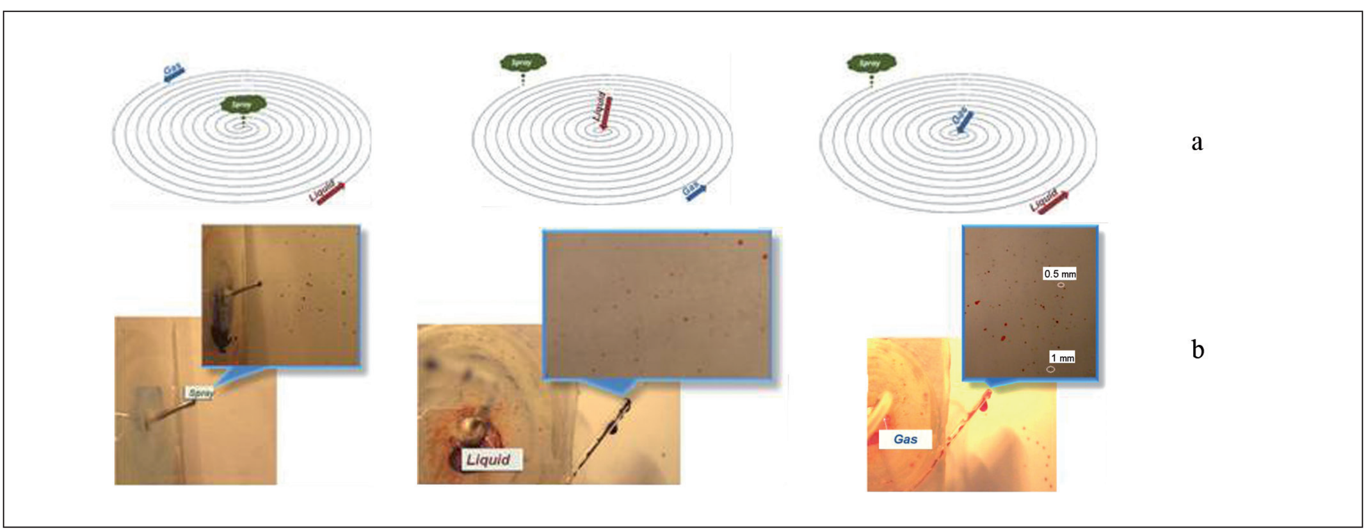

Figure 6. Spray schematics (a) and the corresponding droplet formation using aniline aqueous solution (b).

10 µm and it is visualized only if sample with 10 µm particles is used on the spray system, i.e., the droplet size is determine probably mainly by the particle diameter. Furthermore, the spray jet cannot be distinguished in the photograph, which indicates formation of tiny particles. Thus, a simple mechanism that can explain such results is the multiple interactions of gas/liquid inside the channel. Due to these sequential interactions coalescence is hindered and even a heavily contaminated dispersion (10% wt.) can produce small droplets.

An important characteristic on this three-dimensional channel is the small pressure decay, i.e., no difference on pressure was found during tests and simulations indicate a pressure decay of 1.1 atm in the whole structure. Thus, considering that sprays are useful on several unit operations and suspensions are a main issue in sample pretreatment,

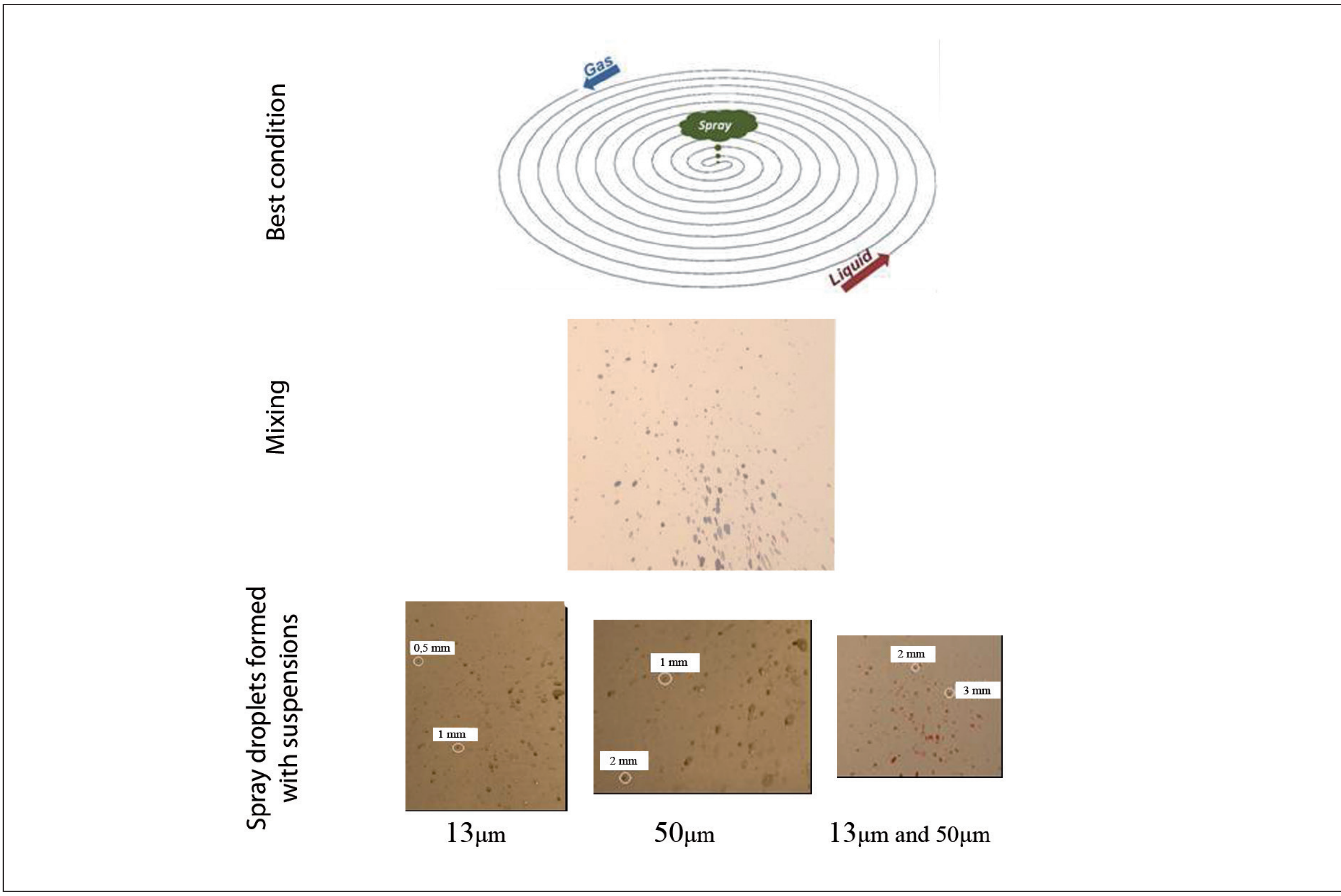

Figure 7. The best configuration for spray formation; mixing and droplets obtained with suspensions of 13μm and/or 50μm.

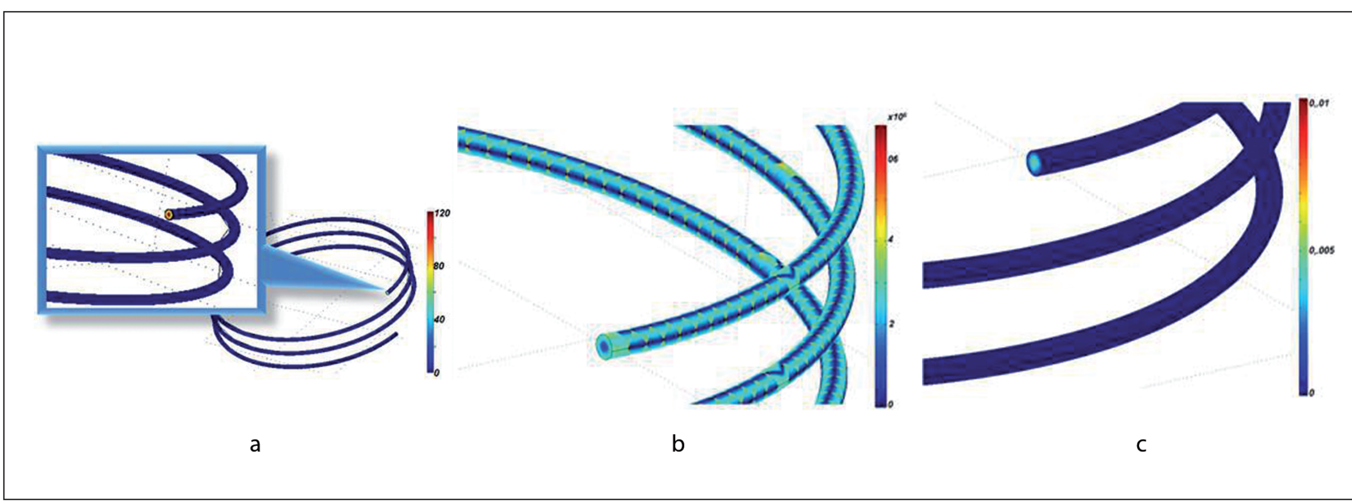

Figure 8. Velocity profile ($cm.s^{-1}$) (a), vorticty (b) Reynolds number (c) and to liquid flow inserted in a three-dimensional channel.

tests were carried out using a miniaturized impactor. Impactors are commonly used to collect particles and/or simultaneously separate them according with their dimensions. One impactor was miniaturized and used for collecting particles from air[33] showing high capacity; however, it was not useful on liquid sample. Therefore, the design was modified and tested to remove particles from liquid but show low capacity[34]. The main issue on particle removal on such cases is the strong interaction between the liquid and the collecting wall, which can be diminished by the use of a spray. Therefore, the miniaturized impactor was coupled to the spray system and Fig. 10d shows photos, in a frame sequence, if the sample is heavily contaminated with particles (10%wt.). The clear liquid obtained in the final frames of the sequence indicates that it is possible to remove such particles using these two structures coupled.

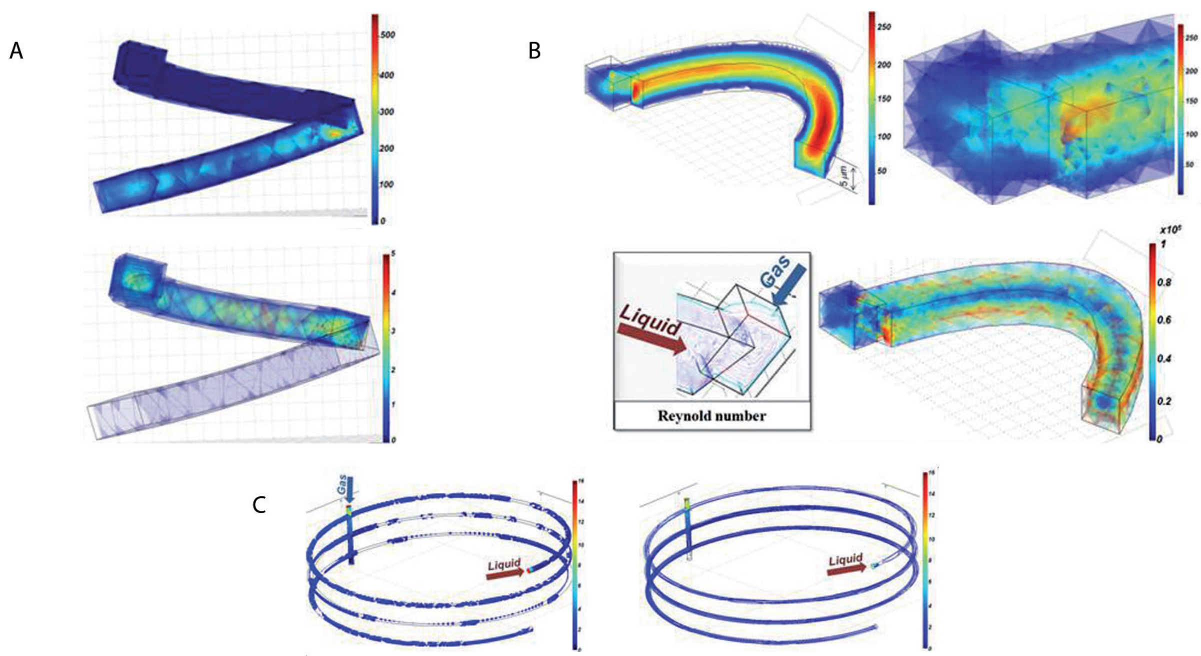

Figure 9. Interaction on the first lap of two channels: velocity profile (cm.s^{-1}) - liquid and gas - if one of them has a step screw 4 times bigger than the other (a) or in 90º angle (b), with respective vorticity and Reynold number. Interaction in three laps and 90º angle (c).

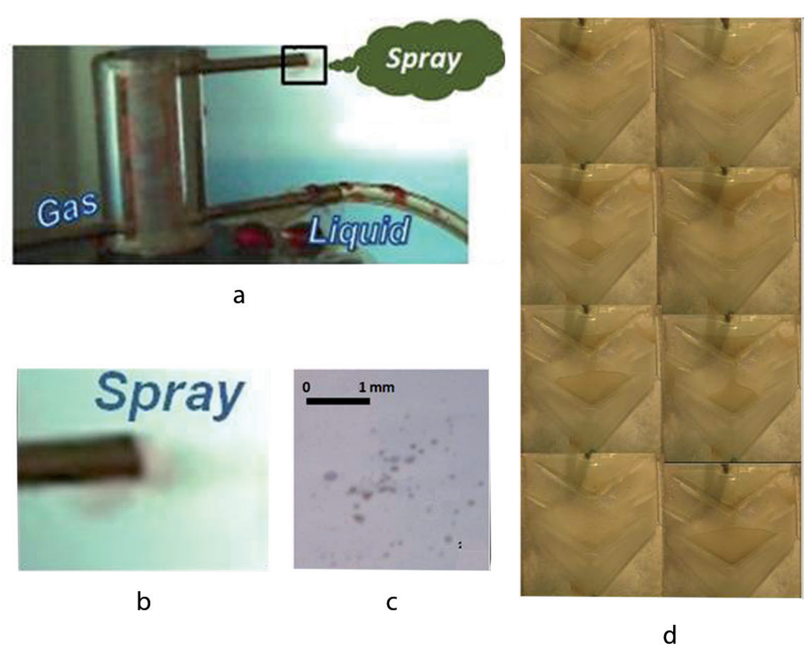

Figure 10. Spray (a), jet spray (b), droplets obtained 10 cm apart from the jet nozzle (c) and several frames showing particle removal in a miniaturized impactor (d).

Conclusions

This work evaluated qualitatively the spray formation using planar and three-dimensional microchannels in order to propose low cost mesostructures. The simulated and tested structures are simple to manufacturing and can present several uses not only in spray production but also in mixing two liquids. Therefore, several miniaturized equipment and unit operations, such as microreactors, could use these small devices. Thus, whereas especially due to the small dimension of the whole structure, three-dimensional channels are a good choice for spray formation, planar channels are more adequate to mixture small amount of liquids due to the high velocity variations.

Although the evaluation was only qualitative, it is worthing to note that three-dimensional channels used as spray and coupled to miniaturized impactors show good results in particle removal.

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