Development of microfluidic nozzles with asymmetric tips for sample delivery

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Abstract

The study focused on enhancing the performance of gas dynamic virual nozzles. A new technique of reproducibly fabricating the inner capillary with a FIB was developed. This also allows for new designs and the production of capillaries with an asymmetric tip. It was found that an asymmetric tip allowed for the reduction of the liquid flow rate compared to a flat tip, or mechanically ground tip. Furthermore, jets of water solutions containing up to 15% polyethylene glycol (PEG) were achieved with this design.

Contents

1 Introduction

The delivery of biological samples for x-ray free-electron laser diffraction experiments has to satisfy a lot of conditions. Protein crystals have to be in a fully solvated state during the delivery into the x-ray beam. Each pulse has to hit different crystals due to the high radiation damage. In addition, consumption of the sample should be low, and the "hit rate" high, for efficient experiments. Gas dynamic virtual nozzles seem to be the best candidate for this kind of experiment. Sample is delivered directly from solution into the experimental chamber via a microscopic liquid jet. The goal of this work is to develop new designs of GDVNs in order to reduce jet diameter and liquid flow rate.

2 Gas dynamic virtual nozzle

2.1 Design

The basic design is shown in Figure 1. It consists of two main parts. The inner glass capillary supplies the liquid and the outer glass capillary focuses the gas. A co-flowing gas is used to reduce the diameter of liquid jet rather than a solid nozzle wall. Moreover, jet diameter can be adjusted by changing the applied gas pressure and liquid pressure [1].

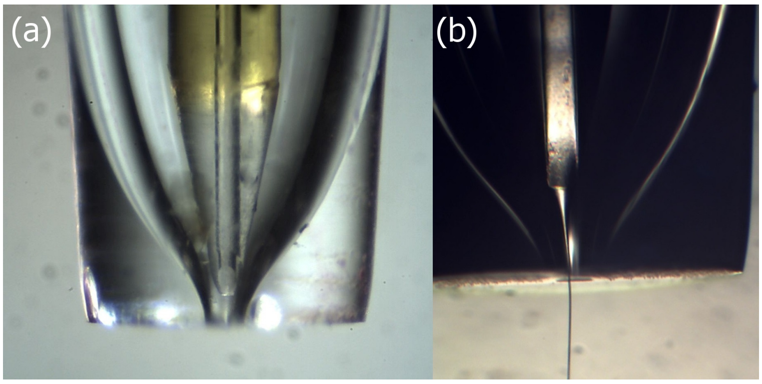

Figure 1: Typical design of GDVN

The liquid is accelerated by a co-flowing gas, forming a cone at the end of the inner capillary. A liquid jet is ejected out of the aperture, and in a short distance, it breaks up into droplets due to the Rayleigh instability.

2.2 Fabrication

The materials used for the fabrication of nozzles are shown in Table 1. Liquid and gas lines were supplied by MicroQuartz, and the outer glass capillaries were supplied by Science Products.

Table 1: The materials used for fabrication of nozzles

Capillary	Inner diameter	Outer diameter	Material
Liquid line	50 μm	350 μm	fused silica
Gas line	100 μm	350 μm	fused silica
Outer capillary	0.78 mm	1.0 mm	borosilicate glass

The typical fabrication procedure consists of three basic steps:

- melting the outer capillary
- grinding the inner capillary
- assembling the nozzle

To form the outer glass capillary to the shape shown in Figure 1, it is necessary to hold it horizontally and rotate around its axis while the end of the capillary is heated by a propane torch. The shape of the exit channel is dependent on the speed of melting. The inner capillary (liquid line) is sharpened at a grinding machine, usually at an angle of 15°. The last step is to assemble everything together. A complete nozzle is shown in Figure 2 (a). We found out that it is necessary to use spacers for our new designs of inner capillaries in order to align them axially. Spacers are shown in Figure 2 (b). In the next part we will discuss the use of a focused ion beam system for inner capillary modification.

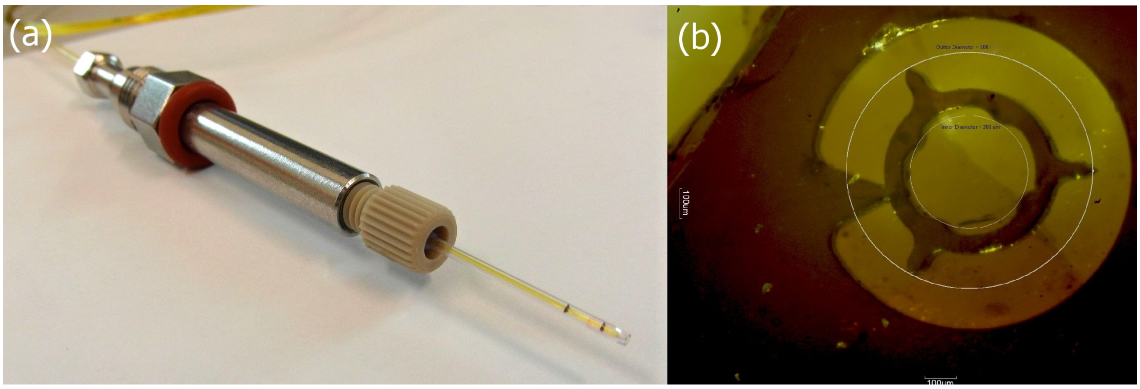

Figure 2: (a) Assembled GDVN. (b) Spacer with inner diameter 350 μm and outer diameter 500 μm

2.3 Focused Ion Beam modification

In the last chapter we explained common steps in GDVN fabrication. Here we will introduce a new approach in fabrication of new GDVN designs based on the use of focused ion beam system (FIB). The FIB system has four basic functions:

- milling
- deposition
- implantation
- imagining

Milling is a process that allows digging into the sample surface as a result of the use of relatively heavy ions in the beam [2]. This function can be used for modification of tips of inner capillaries which were previously ground into conical shapes. It allows us to fabricate various designs which could improve crucial properties of nozzles. For this purpose we used a Dual beam system from FEI company shown in Figure 3.

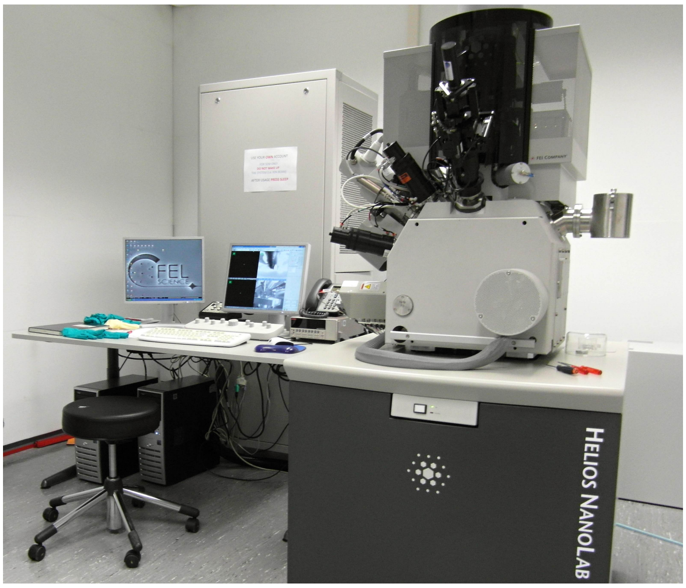

Figure 3: The dual beam system combines functions of scanning electron microscope and focused ion beam

The motivation for the usage of the FIB is also to make nozzles reproducible. This is the main problem with ground inner capillaries — each one is different. Moreover ground tips have irregular shapes, which can be easily eliminated with the FIB. The typical shape of a ground tip is shown in Figure 4 (a). Figure 4 (b) shows the results of a flat cut made with the FIB to clean the edge of the tip. The whole process took around 20 minutes. The ion current was set to its max value of 21 nA, and the accelerating voltage was 30 kV. These milling conditions were kept for every FIB modification done in this study.

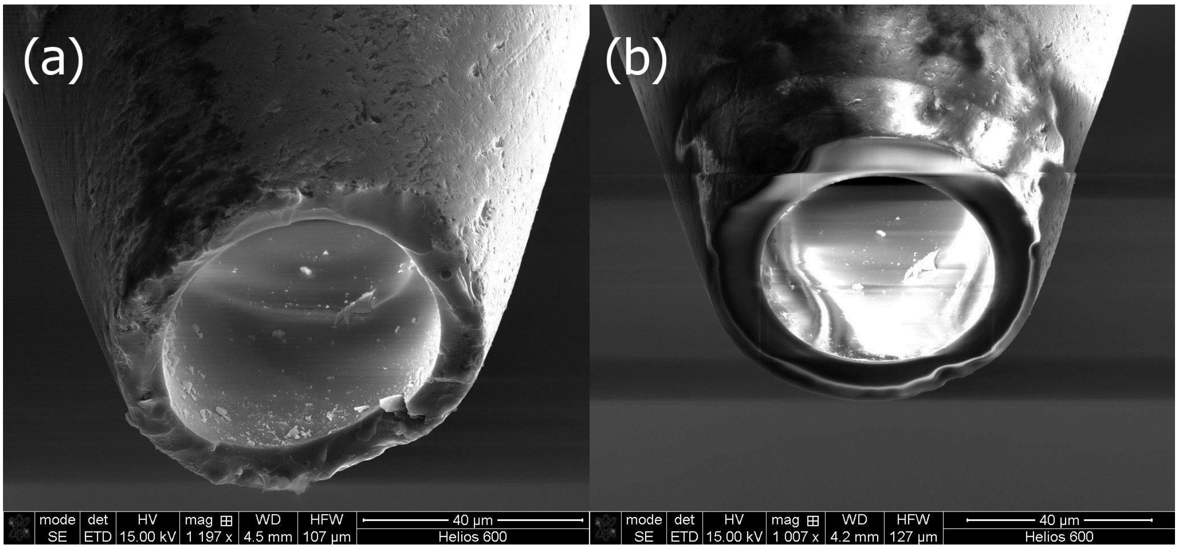

Figure 4: (a) An SEM image of a ground inner capillary. (b) The same capillary after FIB modification: 90° flat cut

This shape is more stable and reproducible, however, properties are similar to ground capillaries. Based on the paper by Acero *et al.* [3], asymmetric designs are very promising. A set of inner capillaries with asymmetric cuts of varying angles is shown in Figure 5. Their properties are discussed in next chapter. With a FIB it is possible to make more exotic designs like those in Figure 6. The performance of these designs need further characterization.

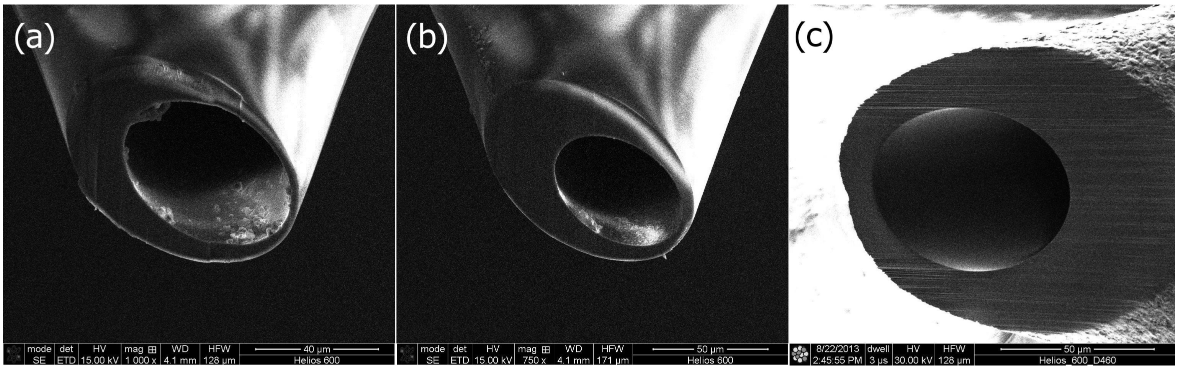

Figure 5: Images of capillaries with asymmetric cuts: (a) 65°, (b) 55°, (c) 38°

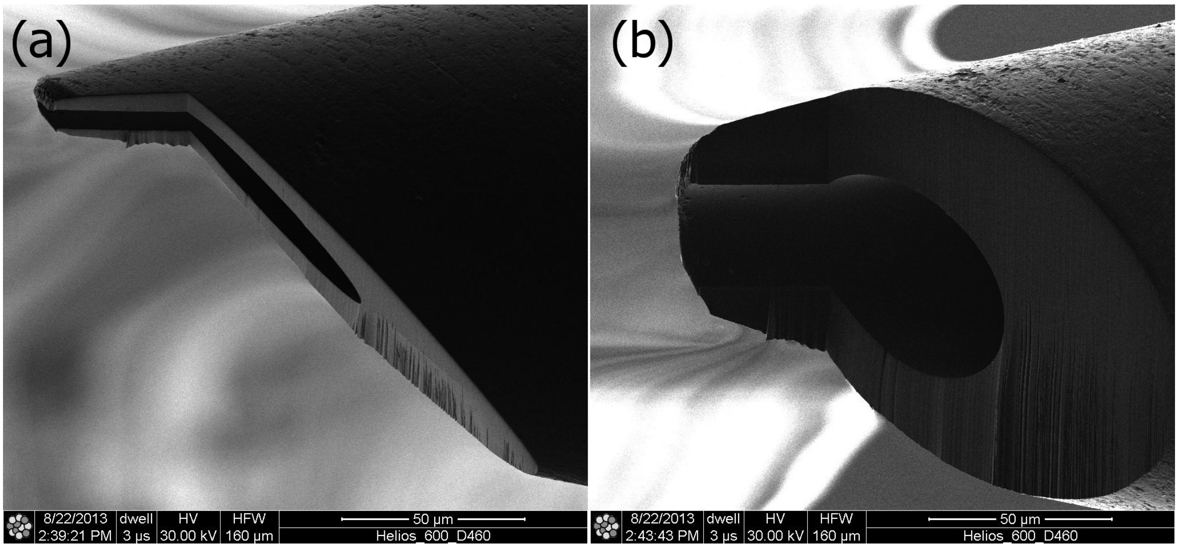

Figure 6: FIB images of a capillary from different angles

With increasing angle of the asymmetric cut, significantly more time is needed for the procedure. It took 3 hours to cut trough a capillary at the angle of 38°. Fortunately, it is possible to make asymmetric cuts on grinder, which can reduce time needed in the FIB. We successfully ground cuts even at angle of 20°.
Charging of the sample is a problem since the capillaries are made out of glass. Nevertheless, a gold coating isn't necessary for FIBing. Moreover, it is vital to ensure mechanical stability of the sample inside the FIB. We found out that the best way how to do this is to use carbon stickers to support the weight of capillaries, and to use conductive carbon cement at the same time to fix fine mechanical movement. In addition, taping capillaries together with ordinary tape is a good way to distribute tension equally among them.

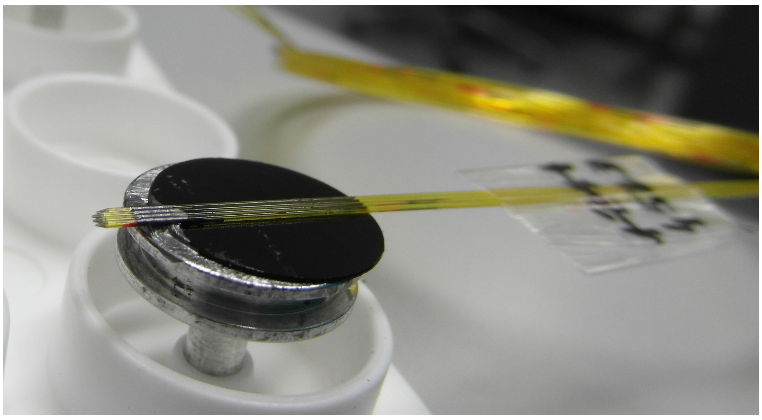

Figure 7: Fixation of the sample on the stage

3 Results

The characterization of FIB modified nozzles was done with deionized water in the testing chamber shown in Figure 8. The maximum applied gas and liquid pressures are both 850 psi. This is achieved by the use of Proportion Air GP1 valves and a common gas cylinder. Helium is used as the accelerating gas. We found that the performance of the nozzle depends highly on the position of the inner capillary. For this purpose, we installed a stage which allowed us to finely adjust the placement of the liquid line. An experimentally important parameter is the minimum liquid flow rate. We examined the stability of the jets under optical microscope at various conditions. The best parameters of the nozzles are shown in Table 2.

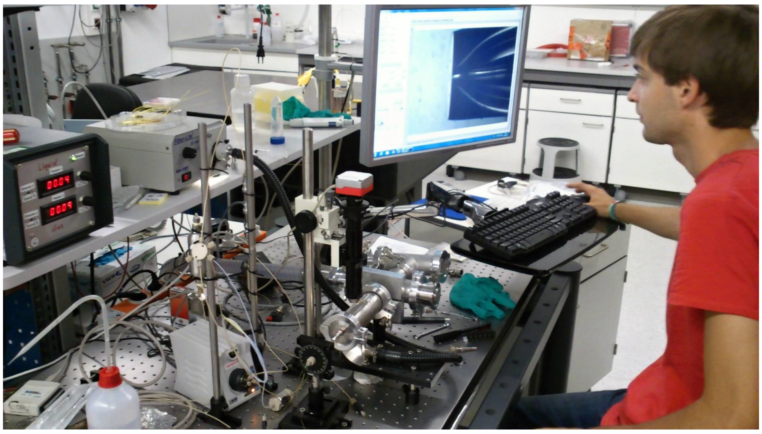

Figure 8: Testing chamber

Table 2: Minimum flow rates of different GDVN designs

Angle of cut	Minimum liquid flow rate
90° (flat cut)	12 μl/min
65°	10 μl/min
55°	7.5 μl/min
38°	4 μl/min

Dependence of the minimum liquid flow rate on the shape of tip is clearly visible. It is possible to achieve liquid flow rates under 10 μl/min with asymmetric tips which is not common in this gemoetry with mechanically ground inner capillaries of 50 μm inner diameter.

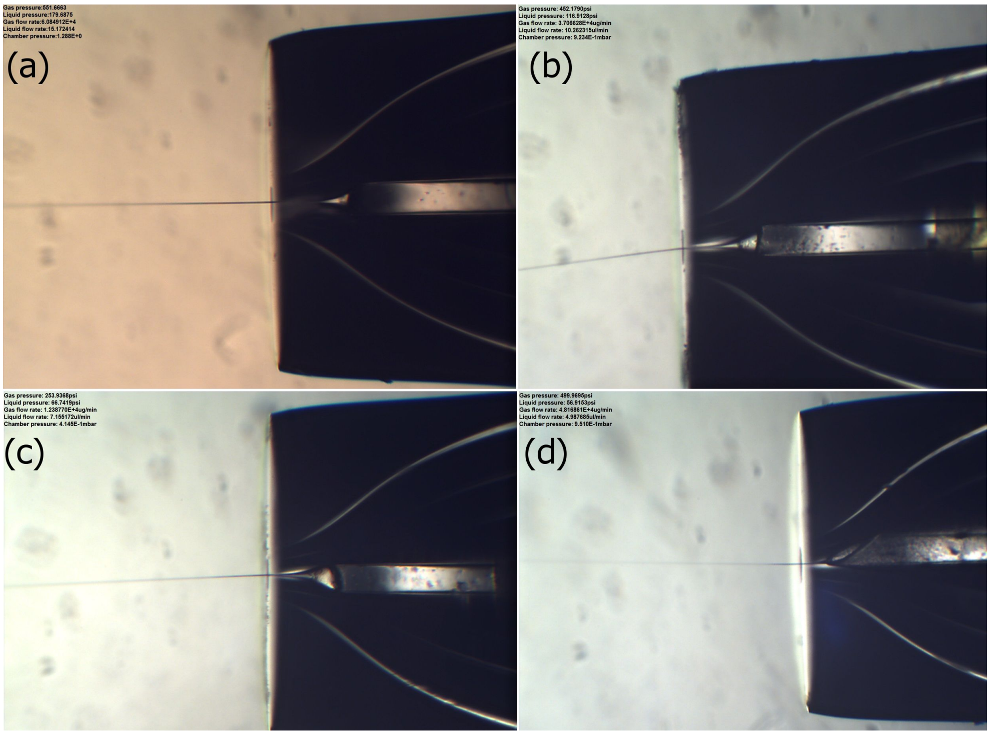

Figure 9: Images of jets from nozzles with different tips: (a) 90°, (b) 65°, (c) 55° and (d) 38°

Since the nozzle with 38 degree tip has best results we decided to characterize it more deeply. In Figure 10 is shown phase diagram. This nozzle has very broad stable region compared to common GDVN. Also there is an interesting region with gas pressure around 250 psi where the jet is stable at very low liquid pressure. There are three regimes of jet behavior:

- stable - continuous jet emitted from nozzle
- dripping - large single drops are emitted at low gas and liquid pressures
- flickering - instability at high gas pressures

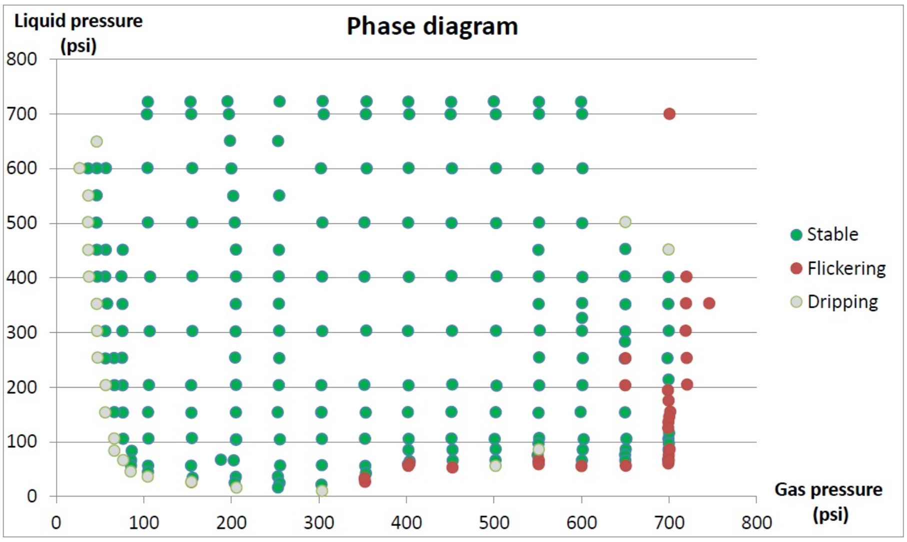

Figure 10: Phase diagram of 38° nozzle

These tests were done using deionized water. However GDVN have to run with real samples at the experiments. Samples normally consist of solutions with different viscosities usually higher than viscosity of water. Figure 11 shows the behavior of the 38° nozzle with different liquids.

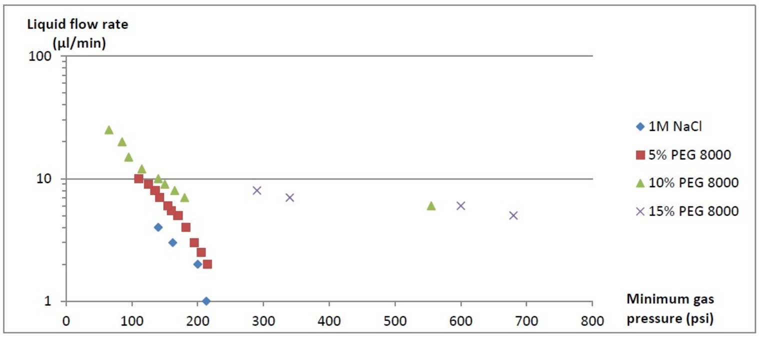

Figure 11: Dependence of liquid flow rate on minimum gas pressure for different solutions

4 Conclusions

Asymmetric designs of the inner capillary tips made by FIB technique significantly improved the performance of GDVN, especially the minimum liquid flow rate. There is a clear dependence of the minimum liquid flow rate on the angle of the asymmetric cut, as shown in Table 2. This technique allows for the fabracation of highly reproducible designs, which are also able to jet viscous solutions. Further investigation of more complicated designs is needed. Such as, a sharp tip on the end of an asymmetric cut may improve the nozzle performance. Furthermore, the deposition of platinum by the FIB offers new possibilities for tip designs.

References

[1] Injector for scattering measurements on fully solvated biospecies *U. Weierstall et al.*

[2] Introduction to the focused ion beam system *N. Yao et al.*

[3] A new flow focusing technique to produce very thin jets *A. J. Acero et al.*