Diamond and Related Materials 11 (2002) 237–241

DIAMOND AND RELATED MATERIALS

www.elsevier.com/locate/diamond

Fabrication of diamond flow controller micronozzles

S. Silva[a], M.C. Salvadori[a,*], K. Kawakita[b], M.T. Pereira[b], W. Rossi[c], M. Cattani[a]

[a]*Institute of Physics, University of São Paulo, C.P.66318, CEP: 05315-970, São Paulo, SP, Brazil*

[b]*Institute for Technological Research (IPT), Cidade Universitária, São Paulo, SP, Brazil*

[c]*Laser Center and Applications, Institute of Energetic and Nuclear Research, Cidade Universitária, São Paulo, SP, Brazil*

Received 8 June 2001; received in revised form 24 August 2001; accepted 11 November 2001

Abstract

In this work we show how to manufacture diamond sonic micronozzles. The advantage in using diamond to fabricate the micronozzles, compared with other materials, is that it is the hardest material known, has low thermal expansion, thermal conductivity is very high and it is chemically inert. In our process, tungsten wires, used as molds, have been fabricated by using an electrochemical etching process. These molds have been coated with diamond films by Chemical Vapor Deposition. By a wet etching, the tungsten mold was removed, obtaining the micronozzle. Flow measurements have shown that the diamond micronozzles work as passive flow controllers and flowmeters under choking conditions. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Diamond; Micronozzle; Sonic flow; Flow controller

1. Introduction

The flow metrology was an active research area during the second part of the 20th century. This area promoted a great knowledge to the operation principles of many fluid flowmeters. Micromachining techniques were used to fabricate flowmeters to measure small flow rates, to be used in several industrial and scientific activities [1].

The small restriction elements, such as critical orifice nozzles, can control the flow rates, allowing their use as critical passive flow controllers and flowmeters [2]. The nozzles have the advantage that there are no moving parts, avoiding wear effects and thermal perturbation. In this work we show how to manufacture diamond micronozzles. Flow measurements have shown that they work as passive flow controllers and flowmeters under choking conditions. The strategical choice of the material used to fabricate the nozzles is an important feature to achieve interesting properties. Diamond is an excellent material for manufacturing nozzles due to its hardness (1.0×10^6 kg/cm^2), low thermal expansion (1.1×10^{-6} K^{-1}), high thermal conductivity (20 W/cm K) and chemical inertness [3]. The hardness is responsible for mechanical stability, and low thermal expansion guarantees the stability of the opening size and throat diameter for a large temperature range. The high thermal conductivity allows a precise temperature control and the chemical inertness permits the device use in corrosive environments.

The flow rate, in micronozzles and microtubes, can be controlled and measured under choking conditions [2] that occur when the gas flow velocity in the throat nozzle is equal to the speed of sound. In these conditions, the flow rate can be controlled and measured, determining the inlet pressure and temperature in despite of the outlet pressure and temperature.

2. Experimental procedures

A method for producing diamond structures by the replication of molds has been used before by others [4–6]. More specifically the production of diamond tubes using tungsten wires as a mold had been described by May et al. [4]. The production of diamond microtubes, used as a flow controller and flowmeter, is described by Silva et al. [7]. In this work we used a method somewhat

* Corresponding author. Tel.: +55-11-3818-6857; fax: +55-11-3818-6749.

E-mail address: mcsalvadori@if.usp.br (M.C. Salvadori).

0925-9635/02/$ - see front matter

PII: S0925-9635(01)00693-8

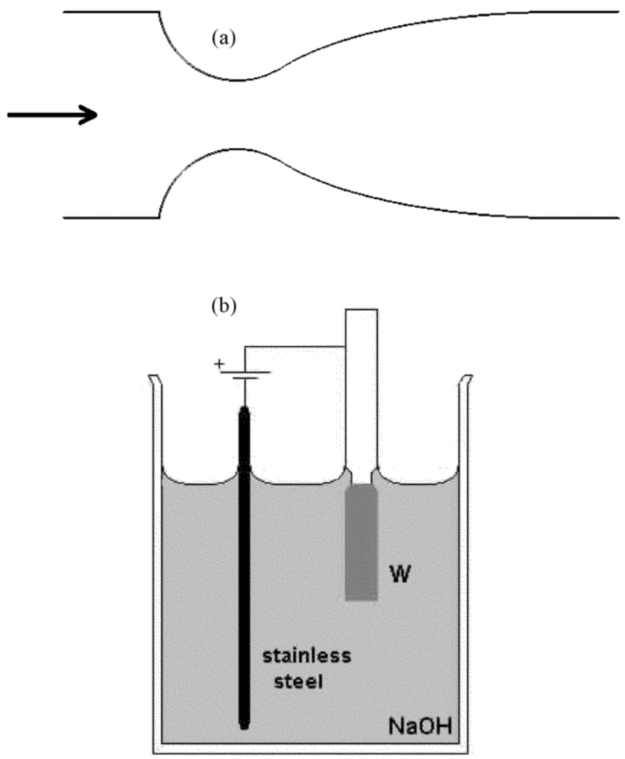

Fig. 1. (a) Typical nozzle convergent–divergent shape. (b) Scheme of the electrolytic cell for tungsten etching.

similar to those mentioned above, but now aiming the production of diamond micronozzle.

The molds utilized in this fabrication technique consist of tungsten wires with initial diameter of approximately 500 μm. The idea is to etch the wires, obtaining for them a convergent–divergent geometry. In Fig. 1a, a typical nozzle shape obtained in this paper is shown schematically.

Our electrochemical etching process follows those commonly adopted to make sharp tips for Scanning Tunneling Microscopy (STM) [8–11]. A scheme of the etching apparatus can be seen in Fig. 1b, where a NaOH solution was used. In the electrolytic cell, the tungsten wire was the anode and a stainless steel rod was the cathode. The stainless steel rod had a 5-mm diameter and 75-mm length. The distance between cathode and anode was approximately 30 mm. The volume of the NaOH solution was 400 ml, the applied potential was 10 V, the immersed wire length was 20 mm and the etching time ranging from 5 to 20 min.

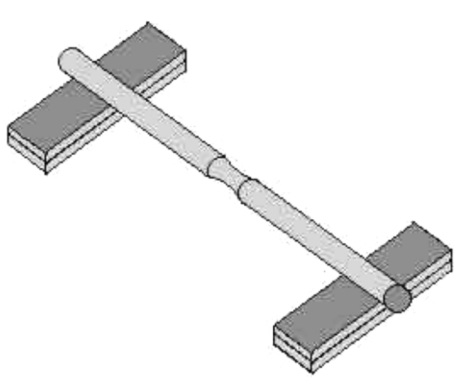

Fig. 2. 20-mm length tungsten wire, supported by two silicon pieces over the sample holder of the CVD system.

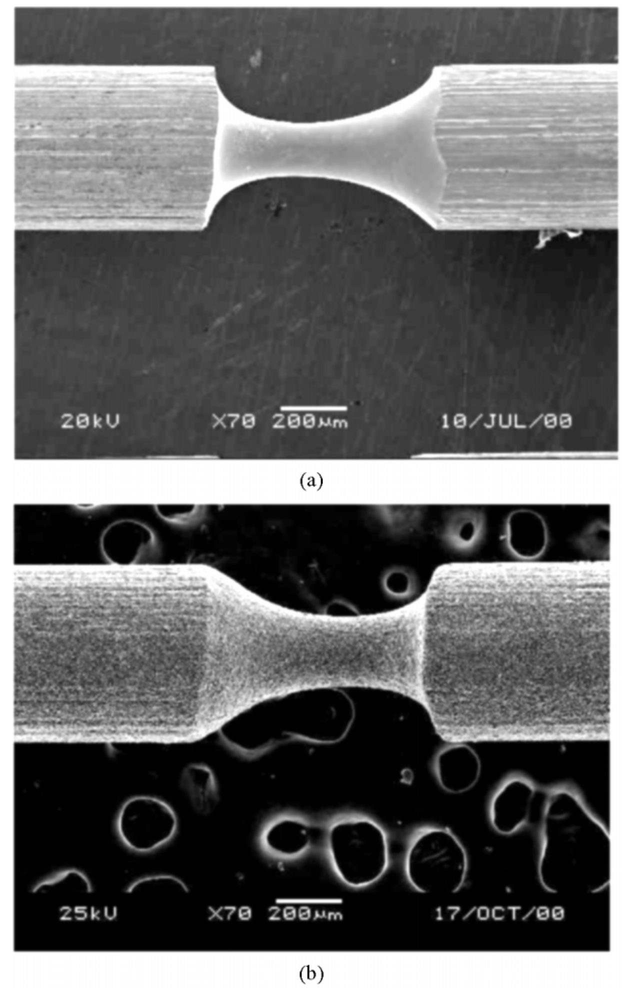

Fig. 3. (a) Convergent–divergent nozzle shape around the tungsten wire with 217-μm throat diameter. (b) Micrograph of a nozzle shape tungsten mold coated with continuous diamond film.

In order to avoid a uniform corrosion of the immersed wire, part of it was masked by painting with an etch resistant material (acetate). In this way, only a small area of the wire was exposed to the etching solution. This small area of the wire in contact with the solution was etched, producing the nozzle shape. The etching time was controlled in order to obtain the desired throat dimension. In this way we achieve the convergent–divergent shape for the tungsten wire molds.

In the next step, the surface of the nozzle-shaped wire was treated to improve the diamond nucleation. In order to do this, it was immersed in an ultrasonic bath

Table 1
Dimensions and correlated parameters of the samples

Samples	Length (L) (μm)	Initial diameter (D).(μm)	Throat diameter (d_t) (μm)	Ratio (D/d_t)	Area ratio ($A/A_t = D^2/d_t^2$)	Ratio (L/d_t)
Tube	5000	500	–	–	–	–
Nozzle 1	5000	500	480	1.0	1.1	10.4
Nozzle 2	5000	500	293	1.7	2.9	17.1
Nozzle 3	5000	500	217	2.3	5.3	23.0

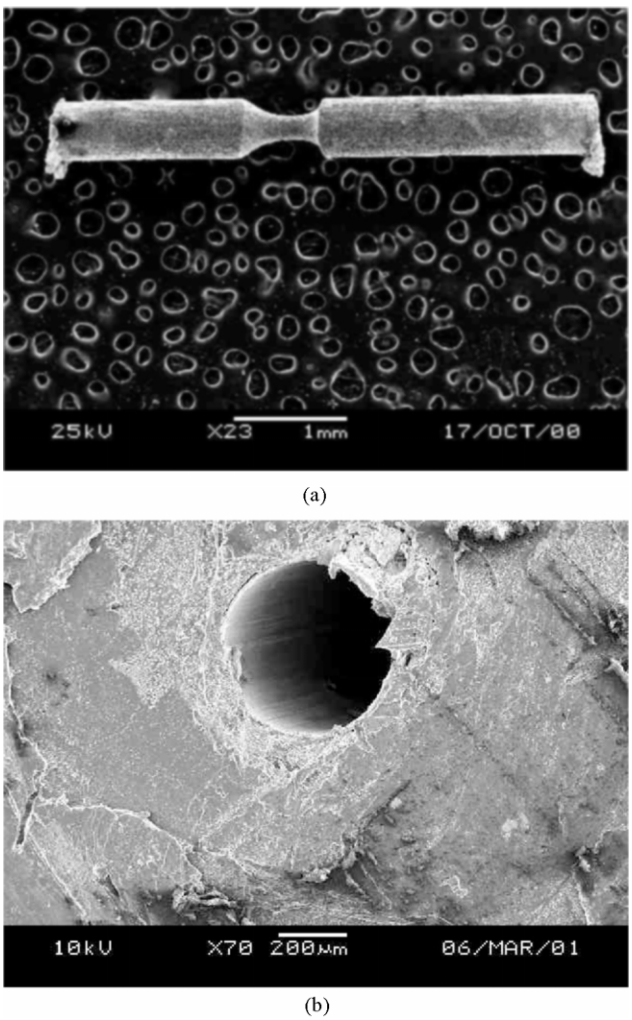

Fig. 4. (a) Tungsten mold with the diamond coating already cut with laser. (b) The inlet of the diamond micronozzles involved in beeswax.

Table 2
Critical properties of the four samples

Samples	Critical pressure ratio (P^*_{out}/P_{in})	Critical flow rate (cm^3/min)
Tube	0.5	1813
Nozzle 1	0.6	1557
Nozzle 2	0.6	627
Nozzle 3	0.6	319

containing water, alcohol and diamond powder (grain size of 1 μm), for 5 min. Then it was dried in air, in vertical position and a brush was used to homogenize the diamond powder distribution on the wire surface.

After that, the diamond deposition was performed, using a plasma assisted Chemical Vapor Deposition (CVD) system [12]. We coated 20-mm lengths of tungsten wires with polycrystalline diamond film, supporting the wire with two silicon pieces, according to Fig. 2, over the sample holder of the CVD system. Note that, in this way, the tungsten wire is about 0.8 mm from the holder. The diamond growth parameters were 300 sccm hydrogen flow rate, 3 sccm methane flow rate, 90 torr chamber pressure, 850 °C substrate temperature, 850 W microwave power and 24 h growth time.

The tungsten wires coated with polycrystalline diamond film were cut using a neodymium laser. The wires were cut to 5 mm in length, with the nozzle shape approximately in the center. The cutting parameters were 0.6 J energy pulse, 0.4 ms timing length, 119 Hz repetition rate, 300 mm/min cutting velocity, 150 μm beam diameter and O_2 was used as working gas.

Each wire piece, coated with diamond, was involved in beeswax with the ends of the tungsten wire exposed to be etched. The tungsten was etched using a mixture of hydrofluoric acid and nitric acid, in a volume ratio of 1:1, at room temperature, in 24 h. In this way, we got diamond micronozzles, involved in beeswax, that work as a matrix support.

We have prepared four samples in this paper: three diamond micronozzles, with different throat diameters and one diamond microtube. The diamond microtube fabrication was performed with similar techniques used for the micronozzles [4,7]. The dimensions and correlated parameters of the samples are summarized in Table 1.

The samples characterization as flow controllers has been performed using air in two different experimental settings. In the first one (low-pressure condition), we had atmospheric pressure at the inlet and at the outlet pressure ranged from 10 to 100 kPa. In the second case (high-pressure condition), we had atmospheric pressure at the outlet and at the inlet the pressure ranged from 100 to 800 kPa. We have measured the air pressure and temperature in the inlet and only the pressure in the

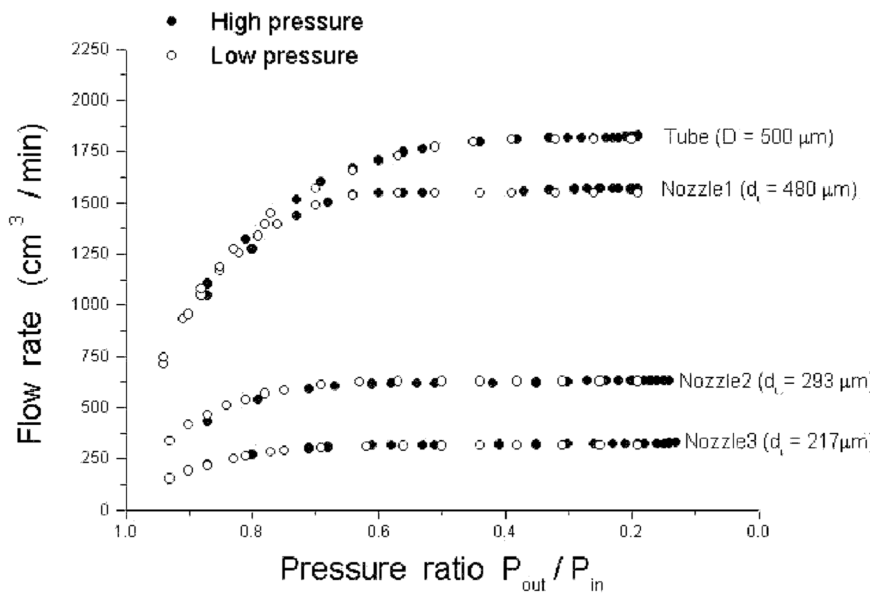

Fig. 5. Curves of the volumetric flow rate as a function of pressure ratio for the microtube and the nozzles.

outlet. The samples were tested under choking conditions and the data were described in curves of volumetric flow rate (cm^3/min) as a function of the pressure ratio P_{out}/P_{in} , where P_{in} is the inlet pressure and P_{out} is the outlet pressure. Note that the volumetric flow rate is the gas volume, at the inlet pressure (P_{in}), per minute (it is not sccm).

3. Results and discussion

3.1. Nozzles fabrication

The nozzle shape was successfully obtained, submitting the tungsten wires to an electrochemical etching process. In Fig. 3a typical convergent–divergent nozzle shape is shown. In this figure, it is possible to see grooves on the tungsten wire surface, due to its fabrication method. However, from Fig. 3a again, we verify that the nozzle surface region is smoother, due to the etching process. The tungsten surface roughness, measured by atomic force microscopy was found to be 270 nm in the grooved region.

Fig. 3b shows a micrograph of a tungsten wire mold coated with continuous diamond film. The polycrystalline film presents faceted diamond crystals with grain sizes from 5 to 10 μm. It is possible to observe that the diamond film is continuous, covering completely the tungsten mold surface.

In Fig. 4a diamond coated tungsten mold is shown, already cut with laser. In this figure, it is possible to observe melted tungsten in the cut regions.

Then, the tungsten mold was involved in beeswax and it was etched, using a mixture of hydrofluoric acid and nitric acid. In Fig. 4b the inlet of the obtained diamond micronozzles is presented. The diamond walls of the micronozzles do not have a uniform thickness [7], having approximately 25 μm in the thinner region and approximately 45 μm in the thicker region. In spite of this, it is important to note that, the inner cylindrical convergent–divergent symmetry of the nozzle is maintained, according to Fig. 1a.

3.2. Flow characterization

The nozzles and the microtube have been manufactured in order to work as passive flow controllers and flowmeters in choking conditions. The characterization of these devices has been performed in two different cases: low pressure and high pressure, as described in the experimental procedures.

In Fig. 5, for the microtube and the nozzles, we present the curves of the volumetric flow rate (cm^3/min) as a function of the pressure ratio P_{out}/P_{in}. As clearly seen, the choking conditions are obeyed in all cases, that is, for the four devices, in low and high pressures.

The critical pressure ratio P^*_{out}/P_{in} is defined as the starting point of the critical volumetric flow rate plateau. The critical pressure ratio P^*_{out}/P_{in} was approximately 0.5 in the diamond microtube and approximately 0.6 in the diamond micronozzles. Note that the critical flow rate decreases with nozzle throat diameter reduction. The critical properties of the four samples are summarized in Table 2. Based on several measurements performed for each device, we observed that the critical pressure ratio P^*_{out}/P_{in} is independent of the inlet and outlet pressure values. The flow rate, in choking conditions, can be controlled and measured when the critical pressure ratio P^*_{out}/P_{in} is reached. This implies that the nozzles, under choking conditions, can be used as critical passive flow controllers and flowmeters [13–15]. The mass flow rate (dm/dt) can be obtained by the critical volumetric flow rate plateau of Fig. 5 and measuring the inlet pressure and temperature [2,13].

Taking into account the devices dimensions, air temperatures (around 300 K) and pressures in our experimental conditions, we can verify, using the well-known fluid mechanics approach [14,15], that the gas flow is described by a 'flow with friction', where slip-flow boundary effects are negligible [16].

4. Conclusions

In this work we have shown how to fabricate diamond sonic micronozzles utilizing tungsten wires as molds. The advantage in fabricating diamond micronozzles, compared with other materials, is that: (i) it is the hardest material known, providing high mechanical stability; (ii) its low thermal expansion guarantees maintainance of the throat size for a large temperature range; (iii) its high thermal conductivity allows a precise temperature control; and (iv) the chemical inertness permits the device to be used in corrosive environments. The convergent–divergent nozzle shape was obtained by an electrochemical etching process. A good axial symmetry has been got for the nozzles shapes. A faithful diamond replica of the tungsten molds has been obtained. The experimental flow measurements have shown that the choking conditions were reached for our devices. In this way, they can be used as passive sonic flow controllers and flowmeters.

Acknowledgements

We are grateful to the 'Laboratório de Microscopia Eletrônica do LNLS' (LME-LNLS) for microscopy tasks and to the FAPESP and CNPq for financial support.

References

[1] C.-M. Ho, Y.-C. Tai, Micro-Electro-Mechanical-Systems (MEMS) and fluid flows, Ann. Rev. Fluid Mech. 30 (1998) 579–612.

[2] Arnberg B.T, Review of critical flowmeters for gas flow measurements. Transactions of the ASME (1962) 447–459.

[3] M. Seal, The current status of CVD diamond applications and prospects for the future. Applications of Diamond Films and Related Materials Third International Conference, Gaithersburg, USA, 1995, p. 3–10.

[4] P.W. May, C.A. Rego, R.M. Thomas, M.N.R. Ashfold, K.N. Rosser, CVD diamond wires and tubes, Diamond Rel. Mater. 3 (1994) 810–813.

[5] V.P. Mammana, M.C. Salvadori, I.G. Brown, Study of porosity in permeable diamond membranes, Thin Solid Films 308 (1997) 258–262.

[6] A.M. Pizzo Passaro, M.C. Salvadori, D.R. Martins, M. Cattani, Fidelity of diamond replica on silicon substrate, Thin Solid Films 377–378 (2000) 285–289.

[7] S. Silva, M.C. Salvadori, K. Kawakita, M.T. Pereira, R.M. Galvão, M. Cattani, Diamond flow controllers microtubes. Accepted for publication Journal of Micromechanics and Microengineering in March, 2001.

[8] A.J. Melmed, The art and science and other aspects of making sharp tips, J. Vac. Sci. Technol. B 9 (1991) 601–608.

[9] Y.-G. Kim, E.-H. Choi, S.-O. Kang, G. Cho, Computer-controlled fabrication of ultra-sharp tungsten tips, J. Vac. Sci. Technol. B 16 (1998) 2079–2081.

[10] J.P. Ibe, P.P. Bey Jr, S.L. Brandow, et al., On the electrochemical etching of tips for scanning tunneling microscopy, J. Vac. Sci. Technol. A 6 (1990) 3570–3575.

[11] H. Lemke, T. Göddenhenrich, H.P. Bochem, U. Hartmann, C. Heiden, Improved microtips for scanning probe microscopy, Rev. Sci. Instrum. 61 (1990) 2538–2541.

[12] M.C. Salvadori, V.P. Mammana, O.G. Martins, F.T. Degasperi, Plasma-assisted chemical vapor deposition in a tunable microwave cavity, Plasma Sources Sci. Technol. 4 (1995) 489–493.

[13] A.H. Shapiro, The Dynamics and Thermodynamics of Compressible Fluid Flow, The Ronald Press Company, New York, 1953.

[14] R.W. Fox, A.T. McDonald, Introduction to Fluid Mechanics, John Wiley & Sons, Inc, 1981.

[15] J.D. Anderson Jr, Modern Compressive Flow, McGraw-Hill Publishing Company, 1990.

[16] C.I. Weng, W.L. Li, C.C. Hwang, Gaseous flow in microtubes at arbitrary Knudsen numbers, Nanotechnology 10 (1999) 373–379.