US006831274B2

(12) **United States Patent**
Smith et al.

(10) **Patent No.: US 6,831,274 B2**
(45) **Date of Patent: Dec. 14, 2004**

(54) **METHOD AND APPARATUS FOR MULTISPRAY EMITTER FOR MASS SPECTROMETRY**

(75) Inventors: **Richard D. Smith**, Richland, WA (US); **Keqi Tang**, Richland, WA (US); **Yuehe Lin**, Richland, WA (US)

(73) Assignee: **Battelle Memorial Institute**, Richland, WA (US)

(*) Notice: Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 0 days.

(21) Appl. No.: **10/091,833**

(22) Filed: **Mar. 5, 2002**

(65) **Prior Publication Data**

US 2003/0168591 A1 Sep. 11, 2003

(51) **Int. Cl.**[7] **B01D 59/44**; H01J 49/00
(52) **U.S. Cl.** **250/288**; 250/281; 250/282
(58) **Field of Search** ..... 250/281–300, 250/423 R, 424, 423 F

(56) **References Cited**

U.S. PATENT DOCUMENTS

4,524,121	A	*	6/1985	Gleim et al.	430/176
4,898,775	A	*	2/1990	Oka et al.	428/336
6,107,628	A	*	8/2000	Smith et al.	250/292
2001/0032929	A1	*	10/2001	Fuhrer et al.	250/281
2002/0000517	A1	*	1/2002	Corso et al.	250/288
2002/0185595	A1	*	12/2002	Smith et al.	250/288
2003/0111599	A1	*	6/2003	Staats	250/288

OTHER PUBLICATIONS

Tang, et al. "Generation of Multiple Electrosprays Using Microfabricated Emitter Arrays for Improved Mass Spectrometric Sensitivity", Analytical Chemistry, Apr. 15, 2000, pp 1658–1663, vol. 73, No. 8, American Chemical Society.*

Nicholas H. Bings, et al., "Microfluidic Devices Connected to Fused–Silica Capillaries with Minimal Dead Volume," Analytical Chemistry, Aug. 1, 1999, pp. 3292–3296, vol. 71, No. 15., American Chemical Society.

J. Fernandez De La Mora, et al., "The Current Emitted by Highly Conducting Taylor Cones," J. Fluid Mechanics, 1994, pp. 155–184, vol. 260. Cambridge University Press.

Frantisek Foret, et al., "Subatmospheric Electrospray Interface for Coupling of Microcolumn Separations with Mass Spectrometry," Electroopphoresis 2000, pp. 1363–1371, vol. 21, Wiley–VCH Verlag GmbH.

Larry Licklider, et al., "A Micromachined Chip–Based Electrospray Source for Mass Spectrometry," Analytical Chemistry, Jan. 15, 2000, pp. 367–375, vol. 72, No. 2, American Chemical Society.

R.S. Ramsey, et al., "Generating Electrospray from Microchip Devices Using Electroosmotic Pumping," Analytical Chemistry, Marcy 15, 1997, pp. 1174–1178, vol. 69, No. 6, American Chemical Society.

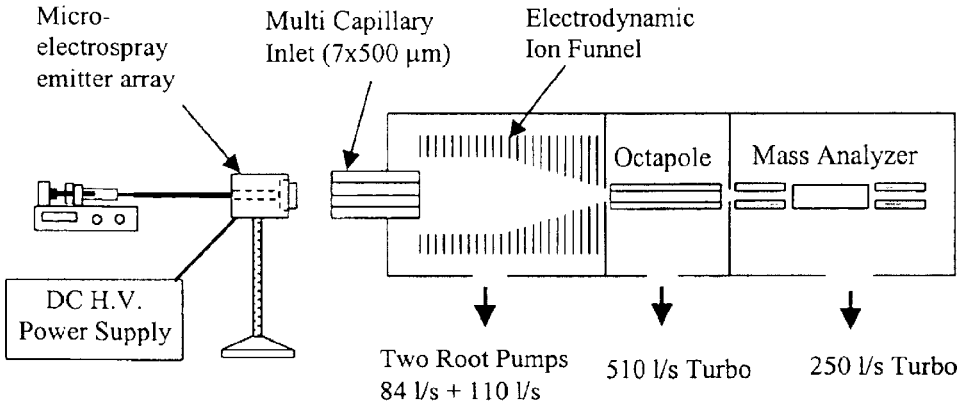
Gary A. Schultz, et al., "A Fully Integrated Monolithic Microchip Electrospary Device for Mass Spectrometry," Analytical Chemistry, Sep. 1, 2000, pp. 4058–4063, vol. 72, No. 17, American Chemical Society.

"Improved Ionization Source Utilizing a Multi–Capillary Inlet and Method of Making," Patent Application Ser. No., filed, Group art unit.

(List continued on next page.)

Primary Examiner—Nikita Wells
Assistant Examiner—Christopher M. Kalivoda
(74) *Attorney, Agent, or Firm*—Douglas E. McKinley, Jr.

(57) **ABSTRACT**

A method and apparatus that utilizes two or more emitters simultaneously to form an electrospray of a sample that is then directed into a mass spectrometer, thereby increasing the total ion current introduced into an electrospray ionization mass spectrometer, given a liquid flow rate of a sample. The method and apparatus are most conveniently constructed as an array of spray emitters fabricated on a single chip, however, the present invention encompasses any apparatus wherein two or more emitters are simultaneously utilized to form an electrospray of a sample that is then directed into a mass spectrometer.

16 Claims, 7 Drawing Sheets