

analytical chemistry

April 15, 2014 Volume 86 Number 8



**World-to-Digital-Microfluidic Interface
Enabling Extraction and Purification
of RNA from Human Whole Blood**



ACS Publications
MOST TRUSTED. MOST CITED. MOST READ.

www.acs.org

APRIL 15, 2014

VOLUME 86 ISSUE 8

ANCHAM 86(8) 3683–4066 (2014)

ISSN 0003-2700

Registered in the U.S. Patent and Trademark Office

© 2014 by the American Chemical Society

ON THE COVER: Illustration of a world-to-digital-microfluidics interface that integrates off-chip RNA extraction from 100 μL of human whole blood with on-chip RNA purification/concentration in 5–15 μL droplets. Image created by Mais Jebrail.

Letters to Analytical Chemistry

3683 

UV Photoelectron Spectroscopy at Near Ambient Pressures: Mapping Valence Band Electronic Structure Changes from Cu to CuO

Kanak Roy and Chinnakonda S. Gopinath*

[dx.doi.org/10.1021/ac4041026](https://doi.org/10.1021/ac4041026)

3688 

Click Chemistry Mediated Eu-Tagging: Activity-Based Specific Quantification and Simultaneous Activity Evaluation of CYP3A4 Using ^{153}Eu Species-Unspecific Isotope Dilution Inductively Coupled Plasma Mass Spectrometry

Yong Liang, Xiaowen Yan, Zhaoxin Li, Limin Yang, Bo Zhang, and Qiuquan Wang*

[dx.doi.org/10.1021/ac500123z](https://doi.org/10.1021/ac500123z)

3693 

Improved Separate Solution Method for Determination of Low Selectivity Coefficients

Vladimir V. Egorov,* Elena A. Zdrachek, and Valentine A. Nazarov

[dx.doi.org/10.1021/ac500439m](https://doi.org/10.1021/ac500439m)

3697

Cytosine DNA Methylation Is Found in *Drosophila melanogaster* but Absent in *Saccharomyces cerevisiae*, *Schizosaccharomyces pombe*, and Other Yeast Species

Floriana Capuano, Michael Mülleder, Robert Kok, Henk J Blom, and Markus Ralser*

[dx.doi.org/10.1021/ac500447w](https://doi.org/10.1021/ac500447w)

3703 

Bimodal Imprint Chips for Peptide Screening: Integration of High-Throughput Sequencing by MS and Affinity Analyses by Surface Plasmon Resonance Imaging

Weizhi Wang, Menglin Li, Zewen Wei, Zihua Wang, Xiangli Bu, Wenjia Lai, Shu Yang, He Gong, Hui Zheng, Yuqiao Wang, Ying Liu, Qin Li,* Qiaojun Fang,* and Zhiyuan Hu*

[dx.doi.org/10.1021/ac500465e](https://doi.org/10.1021/ac500465e)

3708 

Imaging Redox Activity at Bipolar Electrodes by Indirect Fluorescence Modulation

Laurent Bouffier,* Thomas Doneux, Bertrand Goudeau, and Alexander Kuhn

[dx.doi.org/10.1021/ac500623v](https://doi.org/10.1021/ac500623v)

Technical Notes

3712

[dx.doi.org/10.1021/ac404015j](https://doi.org/10.1021/ac404015j)

Magnetic Detection of Mercuric Ion Using Giant Magnetoresistance-Based Biosensing System

Wei Wang, Yi Wang, Liang Tu, Todd Klein, Yinglong Feng, Qin Li, and Jian-Ping Wang*

3717

[dx.doi.org/10.1021/ac5004129](https://doi.org/10.1021/ac5004129)

Tuning the Selectivity of Ionic Liquid Stationary Phases for Enhanced Separation of Nonpolar Analytes in Kerosene Using Multidimensional Gas Chromatography

Leandro W. Hantao, Ali Najafi, Cheng Zhang, Fabio Augusto, and Jared L. Anderson*

3722

[dx.doi.org/10.1021/ac500446z](https://doi.org/10.1021/ac500446z)

Analysis of Transmembrane Domains and Lipid Modified Peptides with Matrix-Assisted Laser Desorption Ionization-Time-of-Flight Mass Spectrometry

Mathias J. Gerl,* Timo Sachsenheimer, Michał Grzybek, Ünal Coskun, Felix T. Wieland, and Britta Brügger

Articles

3727

[dx.doi.org/10.1021/ac404251w](https://doi.org/10.1021/ac404251w)

Cavity-Enhanced Absorption Measurements Across Broad Absorbance and Reflectivity Ranges

Purnendu K. Dasgupta,* Ruchika P. Bhawal, Yin-Huan Li, and Akif Ibragimov

3735

[dx.doi.org/10.1021/ac403456t](https://doi.org/10.1021/ac403456t)

Fully Automated Circulating Tumor Cell Isolation Platform with Large-Volume Capacity Based on Lab-on-a-Disc

Jong-Myeon Park,* Minseok S. Kim,* Hui-Sung Moon, Chang Eun Yoo, Donghyun Park, Yeon Jeong Kim, Kyung-Yeon Han, June-Young Lee, Jin Ho Oh, Sun Soo Kim, Woong-Yang Park, Won-Yong Lee,* and Nam Huh

3743

[dx.doi.org/10.1021/ac404088c](https://doi.org/10.1021/ac404088c)

Automated Dispersive Liquid–Liquid Microextraction–Gas Chromatography–Mass Spectrometry

Liang Guo and Hian Kee Lee*

3750

[dx.doi.org/10.1021/ac4030472](https://doi.org/10.1021/ac4030472)

Reduction of the SIMS Matrix Effect Using the Storing Matter Technique: A Case Study on Ti in Different Matrices

B. Kasel and T. Wirtz*

3756

[dx.doi.org/10.1021/ac5007816](https://doi.org/10.1021/ac5007816)

Contrast-Enhanced Differential Mobility-Desorption Electrospray Ionization-Mass Spectrometry Imaging of Biological Tissues

Rachel V. Bennett, Chaminda M. Gamage, Asiri S. Galhena, and Facundo M. Fernández*

3764 [dx.doi.org/10.1021/ac500513t](https://doi.org/10.1021/ac500513t)**Raman Spectroscopy Enables Noninvasive Biochemical Characterization and Identification of the Stage of Healing of a Wound**

Rishabh Jain, Diego Calderon, Patricia R. Kierski, Michael J. Schurr, Charles J. Czuprynski, Christopher J. Murphy, Jonathan F. McAnulty,* and Nicholas L. Abbott*

3773 [dx.doi.org/10.1021/ac500793e](https://doi.org/10.1021/ac500793e)**Rapid Prototyping of Electrochromatography Chips for Improved Two-Photon Excited Fluorescence Detection**

Claudia Hackl, Reinhold Beyreiss, David Geissler, Stefan Jezierski, and Detlev Belder*

3780 [dx.doi.org/10.1021/ac4035746](https://doi.org/10.1021/ac4035746)**Determination of Cellulose Crystallinity by Terahertz-Time Domain Spectroscopy**

Francisco Senna Vieira* and Celio Pasquini

3787 [dx.doi.org/10.1021/ac403756u](https://doi.org/10.1021/ac403756u)**Chemical Method for Nitrogen Isotopic Analysis of Ammonium at Natural Abundance**

Dongwei Liu, Yunting Fang,* Ying Tu, and Yuepeng Pan

3793 [dx.doi.org/10.1021/ac500878x](https://doi.org/10.1021/ac500878x)**Ion Fusion of High-Resolution LC–MS-Based Metabolomics Data to Discover More Reliable Biomarkers**

Zhongda Zeng, Xinyu Liu, Weidong Dai, Peiyuan Yin, Lina Zhou, Qiang Huang, Xiaohui Lin, and Guowang Xu*

3801 [dx.doi.org/10.1021/ac500316x](https://doi.org/10.1021/ac500316x)**Analytical Differentiation of 1-Alkyl-3-acylindoles and 1-Acyl-3-alkylindoles: Isomeric Synthetic Cannabinoids**

Jack DeRuiter, Forrest T. Smith, Karim Abdel-Hay, and C. Randall Clark*

3809 [dx.doi.org/10.1021/ac500882e](https://doi.org/10.1021/ac500882e)**Single Cell Analysis with Probe ESI-Mass Spectrometry: Detection of Metabolites at Cellular and Subcellular Levels**

Xiaoyun Gong, Yaoyao Zhao, Shaoqing Cai, Shujie Fu, Chengdui Yang, Sichun Zhang, and Xinrong Zhang*

3817 [dx.doi.org/10.1021/ac4038625](https://doi.org/10.1021/ac4038625)**Hydrophilic Interaction Chromatography Using a Meter-Scale Monolithic Silica Capillary Column for Proteomics LC–MS**

Kanta Horie,* Takeo Kamakura, Tohru Ikegami, Masaki Wakabayashi, Takashi Kato, Nobuo Tanaka, and Yasushi Ishihama*

3825 [dx.doi.org/10.1021/ac4039064](https://doi.org/10.1021/ac4039064)**Gold Nanoparticles-Based Colorimetric Assay for Cathepsin B Activity and the Efficiency of Its Inhibitors**

Chan-Jin Kim, Dong-Ik Lee, Cheonghee Kim, Kangtaek Lee, Chang-Ha Lee, and Ik-Sung Ahn*

3834

[dx.doi.org/10.1021/ac403960g](https://doi.org/10.1021/ac403960g)

Signal-On Dual-Potential Electrochemiluminescence Based on Luminol–Gold Bifunctional Nanoparticles for Telomerase Detection

Huai-Rong Zhang, Mei-Sheng Wu, Jing-Juan Xu,* and Hong-Yuan Chen

3841

[dx.doi.org/10.1021/ac403971h](https://doi.org/10.1021/ac403971h)

Fully Integrated Lab-on-a-Disc for Nucleic Acid Analysis of Food-Borne Pathogens

Tae-Hyeong Kim, Juhee Park, Chi-Ju Kim, and Yoon-Kyoung Cho*

3849

[dx.doi.org/10.1021/ac403992r](https://doi.org/10.1021/ac403992r)

Real-Time Study of Interactions between Cytosine–Cytosine Pairs in DNA Oligonucleotides and Silver Ions Using Dual Polarization Interferometry

Yu Zheng, Cheng Yang, Fan Yang,* and Xiurong Yang*

3856

[dx.doi.org/10.1021/ac404085p](https://doi.org/10.1021/ac404085p)

World-to-Digital-Microfluidic Interface Enabling Extraction and Purification of RNA from Human Whole Blood

Mais J. Jebrail, Anupama Sinha, Samantha Vellucci, Ronald F. Renzi, Cesar Ambriz, Carmen Gondhalekar, Joseph S. Schoeniger, Kamlesh D. Patel, and Steven S. Branda*

3863

[dx.doi.org/10.1021/ac404051f](https://doi.org/10.1021/ac404051f)

Characterization of the Pharmaceutical Effect of Drugs on Atherosclerotic Lesions *In Vivo* Using Integrated Fluorescence Imaging and Raman Spectral Measurements

Yi-Cyun Yang, Wei-Tien Chang, Shao-Kang Huang, and Ian Liu*

3869

[dx.doi.org/10.1021/ac4040554](https://doi.org/10.1021/ac4040554)

Simple and Efficient Method to Purify DNA–Protein Conjugates and Its Sensing Applications

Zhaojuan Zhou, Yu Xiang, Aijun Tong,* and Yi Lu*

3876

[dx.doi.org/10.1021/ac4042312](https://doi.org/10.1021/ac4042312)

Quantitative Assessment of Protein Adsorption on Microparticles with Particle Mass Spectrometry

Caiqiao Xiong, Xiaoyu Zhou, Ning Zhang, Lingpeng Zhan, Suming Chen, Jiyun Wang, Wen-Ping Peng, Huan-Cheng Chang, and Zongxiu Nie*

3882

[dx.doi.org/10.1021/ac404101w](https://doi.org/10.1021/ac404101w)

Charge Transfer Kinetics from Surface Plasmon Resonance Voltammetry

Jin Lu and Jinghong Li*

3887

[dx.doi.org/10.1021/ac404175e](https://doi.org/10.1021/ac404175e)

Dynamic Vapor Generator That Simulates Transient Odor Emissions of Victims Entrapped in the Voids of Collapsed Buildings

M. Statheropoulos,* G. C. Pallis, K. Mikedj, S. Giannoukos, A. Agapiou, A. Pappa, A. Cole, W. Vautz, and C. L. Paul Thomas

3895

[dx.doi.org/10.1021/ac4042087](https://doi.org/10.1021/ac4042087)

Continuous and Simultaneous Electrochemical Measurements of Glucose, Lactate, and Ascorbate in Rat Brain Following Brain Ischemia

Yuqing Lin, Ping Yu, Jie Hao, Yuxiang Wang, Takeo Ohsaka, and Lanqun Mao*

3902

[dx.doi.org/10.1021/ac500640u](https://doi.org/10.1021/ac500640u)

Globally Visualizing the Microtubule-Dependent Transport Behaviors of Influenza Virus in Live Cells

Shu-Lin Liu, Li-Juan Zhang, Zhi-Gang Wang, Zhi-Ling Zhang, Qiu-Mei Wu, En-Ze Sun, Yun-Bo Shi, and Dai-Wen Pang*

3909

[dx.doi.org/10.1021/ac404232h](https://doi.org/10.1021/ac404232h)

Vertically Aligned Carbon Nanotube-Sheathed Carbon Fibers as Pristine Microelectrodes for Selective Monitoring of Ascorbate in Vivo

Ling Xiang, Ping Yu, Jie Hao, Meining Zhang,* Lin Zhu, Liming Dai,* and Lanqun Mao*

3915

[dx.doi.org/10.1021/ac404242q](https://doi.org/10.1021/ac404242q)

Monitoring of Selected Skin-Borne Volatile Markers of Entrapped Humans by Selective Reagent Ionization Time of Flight Mass Spectrometry in NO⁺ Mode

Pawel Mochalski,* Karl Unterkofler, Hartmann Hinterhuber, and Anton Amann*

3924

[dx.doi.org/10.1021/ac5000587](https://doi.org/10.1021/ac5000587)

A Highly Selective and Instantaneous Nanoprobe for Detection and Imaging of Ascorbic Acid in Living Cells and in Vivo

Na Li, Yanhua Li, Yaoyao Han, Wei Pan, Tingting Zhang, and Bo Tang*

3931

[dx.doi.org/10.1021/ac5000812](https://doi.org/10.1021/ac5000812)

Determination of Halide Impurities in Ionic Liquids by Total Reflection X-ray Fluorescence Spectrometry

Tom Vander Hoogerstraete, Steven Jamar, Sil Wellens, and Koen Binnemans*

3939

[dx.doi.org/10.1021/ac5004823](https://doi.org/10.1021/ac5004823)

Toward Understanding of Transfer Mechanism between Electrochemiluminescent Dyes and Luminescent Quantum Dots

Tao Hu, Xuefeng Liu, Shaoqin Liu,* Zhenlong Wang,* and Zhiyong Tang

3947

[dx.doi.org/10.1021/ac500148a](https://doi.org/10.1021/ac500148a)

Automatic Registration of Mass Spectrometry Imaging Data Sets to the Allen Brain Atlas

Walid M. Abdelmoula, Ricardo J. Carreira, Reinald Shytii, Benjamin Balluff, René J. M. van Zeijl, Else A. Tolner, Boudewijn F. P. Lelieveldt, Arn M. J. M. van den Maagdenberg, Liam A. McDonnell,* and Jouke Dijkstra

3955

[dx.doi.org/10.1021/ac5002355](https://doi.org/10.1021/ac5002355)

Fabrication of Gold Nanoparticle-Embedded Metal–Organic Framework for Highly Sensitive Surface-Enhanced Raman Scattering Detection

Yuling Hu,* Jia Liao, Dongmei Wang, and Gongke Li*

3964 S

[dx.doi.org/10.1021/ac500188j](https://doi.org/10.1021/ac500188j)

2D NMR Barcoding and Differential Analysis of Complex Mixtures for Chemical Identification: The *Actaea* Triterpenes

Feng Qiu, James B. McAlpine, David C. Lankin, Ian Burton, Tobias Karakach, Shao-Nong Chen, and Guido F. Pauli*

3973 S

[dx.doi.org/10.1021/ac5004344](https://doi.org/10.1021/ac5004344)

Enhancing Gas Chromatography—Time of Flight Mass Spectrometry Data Analysis Using Two-Dimensional Mass Channel Cluster Plots

Brian D. Fitz, Brooke C. Reaser, David K. Pinkerton, Jamin C. Hoggard, Kristen J. Skogerboe, and Robert E. Synovec*

3980

[dx.doi.org/10.1021/ac5003189](https://doi.org/10.1021/ac5003189)

Single-Step Total Fractionation of Single-Wall Carbon Nanotubes by Countercurrent Chromatography

Min Zhang, Constantine Y Khrapin, Jeffrey A. Fagan, Peter McPhie, Yoichiro Ito,* and Ming Zheng*

3985 S

[dx.doi.org/10.1021/ac500405x](https://doi.org/10.1021/ac500405x)

Ion Mobility Derived Collision Cross Sections to Support Metabolomics Applications

Giuseppe Paglia,* Jonathan P. Williams, Lochana Menikarachchi, J. Will Thompson, Richard Tyldesley-Worster, Skarphéðinn Halldórsson, Ottar Rolfsson, Arthur Moseley, David Grant, James Langridge, Bernhard O. Palsson, and Giuseppe Astarita*

3994

[dx.doi.org/10.1021/ac500353y](https://doi.org/10.1021/ac500353y)

A Fiber-Optic Sensor for Accurately Monitoring Biofilm Growth in a Hydrogen Production Photobioreactor

Nianbing Zhong, Qiang Liao,* Xun Zhu, and Rong Chen

4002

[dx.doi.org/10.1021/ac500415a](https://doi.org/10.1021/ac500415a)

Analysis of Krypton-85 and Krypton-81 in a Few Liters of Air

Le-Yi Tu, Guo-Min Yang, Cun-Feng Cheng, Gu-Liang Liu, Xiang-Yang Zhang, and Shui-Ming Hu*

4008 S

[dx.doi.org/10.1021/ac500426b](https://doi.org/10.1021/ac500426b)

Exonuclease III-Aided Autocatalytic DNA Biosensing Platform for Immobilization-Free and Ultrasensitive Electrochemical Detection of Nucleic Acid and Protein

Shufeng Liu, Ying Lin, Li Wang, Tao Liu, Chuanbin Cheng, Wenji Wei, and Bo Tang*

4016 S

[dx.doi.org/10.1021/ac500435d](https://doi.org/10.1021/ac500435d)

Densified Electrochemical Sensors Based on Local Redox Cycling between Vertically Separated Electrodes in Substrate Generation/Chip Collection and Extended Feedback Modes

Kosuke Ino,* Yusuke Kanno, Taku Nishijo, Hirokazu Komaki, Yuta Yamada, Shinya Yoshida, Yasufumi Takahashi, Hitoshi Shiku, and Tomokazu Matsue*

4024 S

[dx.doi.org/10.1021/ac500527q](https://doi.org/10.1021/ac500527q)

Development of Solvent-Free Ambient Mass Spectrometry for Green Chemistry Applications

Pengyuan Liu, Amanda Forni, and Hao Chen*

4033

[dx.doi.org/10.1021/ac500530n](https://doi.org/10.1021/ac500530n)

Optimization and Simulation of Tandem Column Supercritical Fluid Chromatography Separations Using Column Back Pressure as a Unique Parameter

Chunlei Wang,* Adrienne A. Tymiak, and Yingru Zhang

4041

[dx.doi.org/10.1021/ac500567g](https://doi.org/10.1021/ac500567g)

Polyion Selective Polymeric Membrane-Based Pulsed Electrode as a Detector in Flow-Injection Analysis

Andrea K. Bell-Vlasov, Joanna Zajda, Ayman Eldourghamy, Elzbieta Malinowska, and Mark E. Meyerhoff*

4047



[dx.doi.org/10.1021/ac500627r](https://doi.org/10.1021/ac500627r)

Size-Dependent Programming of the Dynamic Range of Graphene Oxide–DNA Interaction-Based Ion Sensors

Huan Zhang, Sisi Jia, Min Lv, Jiye Shi, Xiaolei Zuo, Shao Su, Lianhui Wang, Wei Huang, Chunhai Fan, and Qing Huang*

4052



[dx.doi.org/10.1021/ac500657k](https://doi.org/10.1021/ac500657k)

Label-Free Direct Visual Analysis of Hydrolytic Enzyme Activity Using Aqueous Two-Phase System Droplet Phase Transitions

David Lai, John P. Frampton, Michael Tsuei, Albert Kao, and Shuichi Takayama*

4058



[dx.doi.org/10.1021/ac5007766](https://doi.org/10.1021/ac5007766)

Parallel Affinity-Based Isolation of Leukocyte Subsets Using Microfluidics: Application for Stroke Diagnosis

Swathi R. Pullagurla, Małgorzata A. Witek, Joshua M. Jackson, Maria A. M. Lindell, Mateusz L. Hupert, Irina V. Nesterova, Alison E. Baird, and Steven A. Soper*