

PROSPECTIVES

Review and outlook: mechanical, thermodynamic, and kinetic continuum modeling of metallic materials at the grain scale

Martin Diehl, Max-Planck-Institut für Eisenforschung GmbH, Germany, and National Institute for Materials Science, Japan

Continuum modeling approaches are well established in materials science and engineering of metals. They enable the quantitative investigation of diverse questions related to the improved understanding of mechanics and microstructure evolution of various material classes. Applicable to time and length scales relevant in manufacturing and service, continuum modeling approaches are widely used to study engineering-related phenomena such as recrystallization, strain localization, fracture initiation, and phase transformations. However, focusing on individual physical aspects hampers the wider routine use of continuum modeling tools for many engineering applications. With the advent of multi-physics modeling tools developed with the help of and parametrized by (sub-)micrometer-scale simulations and experiments, a huge variety of applications such as hot rolling, bake-hardening, and case-hardening comes within reach for full-field integrated computational materials engineering. Moreover, the integration of experimentally characterized microstructures and the use of user friendly simulation and evaluation tools render powerful modeling approaches feasible for a broad materials science user community. The state of the art and future trends of mechanical, thermodynamic, and kinetic continuum modeling of metallic materials at the grain scale are outlined in this prospective article. DOI:10.1557/mrc.2017.98

Enhanced shape memory and superelasticity in small-volume ceramics: a perspective on the controlling factors

Xiaomei Zeng and **Zehui Du**, Nanyang Technological University, Singapore; **Christopher A. Schuh**, Massachusetts Institute of Technology, USA; and **Chee Lip Gan**, Nanyang Technological University, Singapore

Shape memory ceramics show potential for energy damping and actuation applications. In particular, small-scale structures of zirconia-based ceramics demonstrate significantly enhanced shape memory and superelastic properties compared with their bulk counterparts, mainly because an oligocrystalline or single-crystal microscale structure reduces mismatch stresses amongst grains. In this prospective article, we review recent experiments that explore the shape memory

properties of small-scale zirconia-based ceramics, including the effects of composition, sample and grain size, and cyclic loading. These factors are reviewed with an eye toward rendering shape memory ceramics more useful in future applications. DOI:10.1557/mrc.2017.99

High-frequency organic rectifiers through interface engineering

Chan-mo Kang, Electronics and Telecommunications Research Institute, South Korea; and **Hyeonwoo Shin** and **Changhee Lee**, Seoul National University, South Korea

The demand for high-frequency (HF) and low-cost rectifiers has encouraged many researchers to investigate organic rectifiers. Recently, organic rectifiers with enhanced intrinsic carrier mobility and charge injection efficiency have enabled operating frequencies to reach up to a gigahertz (GHz). The metal/organic and organic/organic interfaces have played a significant role in determining the electrical properties of the organic rectifiers. In this prospective article, we review the structure of organic rectifiers and present the current state-of-the-art to attain their HF performance. We discuss methods for improving their electrical properties using interface engineering and present future prospects for practical use of GHz-operable organic rectifiers. DOI:10.1557/mrc.2016.100

Magnesium–sulfur battery: its beginning and recent progress

Zhirong Zhao-Karger, Helmholtz Institute Ulm, Germany; and **Maximilian Fichtner**, Karlsruhe Institute of Technology, Germany

Rechargeable magnesium (Mg) battery has been considered as a promising candidate for future battery generations because of its potential high-energy density, its safety features and low cost. The challenges lying ahead for the realization of Mg battery in general are to develop proper electrolytes fulfilling a multitude of requirements and to discover cathode materials enabling high-energy Mg batteries. The combination of Mg anode with a sulfur cathode is one of the promising electrochemical couples due to its advantages of safety, low costs, and a high theoretical energy density of over 3200 Wh/L. However, the research on magnesium–sulfur (Mg–S) battery is just at its beginning and the development of suitable electrolytes has been the key challenge for further improvement, and, thus, in the focus of recent research. In this review, we highlight the recent progress achieved in Mg electrolytes and Mg–S batteries and discuss the major technical issues, which must be resolved for the improvement of Mg–S batteries. DOI:10.1557/mrc.2017.101

Recent progress in the concurrent atomistic-continuum method and its application in phonon transport

Xiang Chen, Weixuan Li, Adrian Diaz, Yang Li, and Youping Chen, University of Florida, Gainesville, USA; and **David L. McDowell,** Georgia Institute of Technology, USA

This work presents the recent progress in the development of the concurrent atomistic-continuum (CAC) method for coarse-grained space- and time-resolved atomistic simulations of phonon transport. Application examples, including heat pulses propagating across grain boundaries and phase interfaces, as well as the interactions between phonons and moving dislocations, are provided to demonstrate the capabilities of CAC. The simulation results provide visual evidence and reveal the underlying physics of a variety of phenomena, including phonon focusing, wave interference, dislocation drag, interfacial Kapitza resistance caused by quasi-ballistic phonon transport, etc. A new method to quantify fluxes in transient transport processes is also introduced. DOI:10.1557/mrc.2017.116

Transmission electron microscopy with atomic resolution under atmospheric pressures

Sheng Dai and Wenpei Gao, University of California, Irvine, USA; **Shuyi Zhang and George W. Graham,** University of California, Irvine, USA, and University of Michigan, Ann Arbor, USA; and **Xiaoqing Pan,** University of California, Irvine, USA

Significant developments in micro-electrical-mechanical systems-based devices for use in transmission electron microscopy (TEM) sample holders have recently led to the commercialization of windowed gas cells that now enable the atomic-resolution visualization of phenomena occurring during gas–solid interactions at atmospheric pressure. *In situ* TEM study under atmospheric pressures provides unique information that is beneficial to correlating the structure–properties relationship of nanomaterials, particularly under real gaseous environments. We here provide a brief introduction of the advanced instrumentation of windowed gas cells and review recent progress of *in situ* atomic-resolution TEM study under atmospheric pressures, including some application examples of oxidation and reduction processes, dynamic growth of nanomaterials, catalytic reactions, and “operando” TEM. DOI:10.1557/mrc.2017.125

RESEARCH LETTERS

Influence of the injection temperature on the size of Ni–Pt polyhedral nanoparticles synthesized by the hot-injection method

J.L. Reyes-Rodríguez, A. Velázquez-Osorio, O. Solorza-Feria, D. Bahena-Uribe, and J. Roque, CINVESTAV, Mexico

Ni–Pt polyhedral nanoparticles were synthesized through a thermochemical route by the hot-injection method using Oleylamine (Oam) and Oleic acid (Oac) solvents as simultaneous stabilizing and reducing agents. Several syntheses were performed to study the effect of the hot-injection temperature on nanoparticle size distribution. Results revealed that the injection of precursors in a mixture of Oam and Oac at 180 °C produced paramagnetic nanoparticles with an approximate size of 27 nm; these particles have uniformly defined polyhedral structures and show greater Pt accumulation on the edges and corners. Ni–Pt

polyhedral nanoparticles with larger sizes and high polydispersity were obtained as the injection temperature was increased closer to the reduction temperature. DOI:10.1557/mrc.2017.122

Long-term stability of mechanically exfoliated MoS₂ flakes

Prachi Budania, Paul Baine, and John Montgomery, Queen's University, UK; **Conor McGeough and Tony Cafolla,** Dublin City University, Ireland; **Mircea Modreanu,** University College Cork, Ireland; **David McNeill and Neil Mitchell,** Queen's University, UK; **Greg Hughes,** Dublin City University, Ireland; and **Paul Hurley,** University College Cork, Ireland

The long-term stability of mechanically exfoliated MoS₂ flakes was compared for storage in the air and storage under vacuum. Significant changes in MoS₂ flakes were observed for samples stored in the air, whereas similar flakes on samples stored in vacuum underwent no change. Small speckles were observed to appear on the surface of flakes stored in the air, followed by thinning and eventual decomposition of MoS₂ flakes. The speckles are suspected to be formed by oxidation of MoS₂ in the presence of atmospheric oxygen and water molecules, resulting in the formation of hydrated MoO₃. DOI:10.1557/mrc.2017.105

A first-principles study of potassium insertion in crystalline vanadium oxide phases as possible potassium-ion battery cathode materials

Daniel Koch, Vadym V. Kulish, and Sergei Manzhos, National University of Singapore, Singapore

Four different vanadium oxide phases [α -vanadium pentoxide (V₂O₅), β -V₂O₅, bronze-type vanadium dioxide [VO₂(B)], and rutile-type VO₂ [VO₂(R)]] are investigated from first principles as potential electrode materials for potassium (K) ion batteries. Specifically, insertion energetics and diffusion barriers are computed. These phases are known as promising cathode materials for other types of metal ion batteries. Our results show that the metastable β -V₂O₅ provides the lowest (strongest) insertion energies for K and the lowest diffusion barriers compared with orthorhombic α -V₂O₅, VO₂(B), and VO₂(R). While three of these phases show energetically favorable potassiation and relatively small diffusion barriers, VO₂(R) is predicted to be incapable of electrochemical K incorporation. DOI:10.1557/mrc.2017.107

Effect of intermittent oxygen exposure on chemical vapor deposition of graphene

Selcuk Temiz, Zafer Mutlu, Sina Shahrezaei, Mihrimah Ozkan, and Cengiz S. Ozkan, University of California, Riverside, USA

Chemical vapor deposition is the most proficient method for growing graphene on copper foils due to its scalability, repeatability, and uniformity, etc. Herein, we systematically study the effect of oxygen (O₂) exposure on graphene growth. We introduced O₂ before and during the growth, and then studied its effects on the morphology, crystallinity, and nucleation density of graphene. We observe that introducing O₂ during growth significantly improves the graphene crystallinity while pre-dosing O₂ before growth reduces the graphene nucleation density. These studies suggest that intermittent O₂ exposure play a significant role in graphene growth, enabling scalable production of high-quality graphene. DOI:10.1557/mrc.2017.111

Dynamics of interacting interphases in polymer bilayer thin films

David D. Hsu, Wheaton College, USA; **Wenjie Xia**, National Institute of Standards and Technology, USA, and Northwestern University, USA; **Jake Song**, Massachusetts Institute of Technology, USA, and Northwestern University, USA; and **Sinan Keten**, Northwestern University, USA

We investigate how the local glass-transition temperature (T_g) depends on film thickness in monolayer and bilayer thin films with a polystyrene (PS) upper-layer and a poly(methyl methacrylate) (PMMA) lower-layer using coarse-grained simulations. Interactions between overlapping interphases demonstrate a superposition principle for describing their glass-transition behaviors. For supported bilayer films, the free surface effect on a PS film upper-layer is effectively eliminated due to an enhanced local T_g near the PS-PMMA interface, which cancels out depressed T_g near the free surface. However, at very low PMMA lower-layer thicknesses, the PMMA-substrate effect can penetrate through the polymer-polymer interface, leading to enhanced T_g in the PS upper-layer. DOI:10.1557/mrc.2017.113

Anomalous characteristics of pore formation in Graphene induced by Si-nanoparticle bombardment

Jae Hyun Park and **Ramki Murugesan**, Gyeongsang National University, South Korea; **Jaekwang Lee**, Pusan National University, South Korea; and **Narayana R. Aluru**, University of Illinois at Urbana-Champaign, USA

Graphene nanopores are utilized in various notable applications such as water desalination, molecular separation, and DNA sequencing. However, the creation of stable nanopores is still challenging due to the self-healing nature of graphene. In this study, using molecular dynamics simulations we explore the drilling of nanopores through graphene by bombardment with Si-nanoparticles. This enables the Si-passivation along the nanopore rim, which is known as an efficient way to stabilize graphene nanopores. The interplay between graphene and projectile causes the anomalous behaviors such as local maxima depending on particle size. The observations are thoroughly analyzed with interaction energy and shape changes. DOI:10.1557/mrc.2017.123

Dewetting of Co thin films obtained by atomic layer deposition due to the thermal reduction process

Samantha E. Holt, Texas A&M University, USA; **E. Sally Ward**, Texas A&M Health Science Center, USA; and **Raimund J. Ober** and **Daniel L. Alge**, Texas A&M University, USA

Cobalt oxide thin films with different thicknesses were synthesized by atomic layer deposition. After a thermal reduction process, under a controlled atmosphere of hydrogen, it was possible to convert cobalt oxide to metallic cobalt. The different thicknesses were obtained considering from 500 to 2000 cycles of CoCp_2/O_3 . The thin films were characterized by x-ray diffraction, scanning electron microscopy, energy-dispersive x-ray microanalysis, and by magneto-optical Kerr effect measurements. The indirect synthesis process allows us to obtain cobalt oxide and cobalt thin films with controlled thicknesses and extraordinary magnetic properties, with coercivities above 500 Oe. DOI:10.1557/mrc.2017.94

Birth of silicon nanowires covered with protective insulating blanket

Krishna Nama Manjunatha and **Shashi Paul**, De Montfort University, United Kingdom

Core-shell silicon-silicon oxide nanowires are synthesized at low temperatures using inorganic and organic compounds of a tin as a catalyst. *In situ* simultaneous one-dimensional growth of pristine silicon nanowires (SiNWs) using alloy catalyst is reported here. Such a development process generates a high-quality SiNW that is not determined by other atomic species in the plasma. A possible growth model is discussed to understand the synchronized precipitation of a SiNW core and an oxide shell. Nanowires grown here eliminate the additional fabrication steps to deposit anticipated oxide shell that is achieved by precipitation from the same catalyst that precipitates core nanowires. DOI:10.1557/mrc.2017.95

Rapid and scalable synthesis of crystalline tin oxide nanoparticles with superior photovoltaic properties by flame oxidation

Easwaramoorthi Ramasamy, International Advanced Research Centre for Powder Metallurgy and New Materials, India; **P. Kathirvel**, PSG College of Technology, India; and **S. Kumar**, **Koppoju Suresh**, and **Ganapathy Veerappan**, International Advanced Research Centre for Powder Metallurgy and New Materials, India

Tin oxide (SnO_2) nanoparticles in gram scale quantity were synthesized from inexpensive Sn feedstock by flame oxidation. Selection of optimal feedstock size based on computational fluid dynamics ensures complete conversion of Sn into SnO_2 nanoparticles. The rapid melting and oxidation of feedstock in high-temperature oxidative flame endow the crystalline and phase-pure SnO_2 nanoparticles, as evident from x-ray diffraction and transmission electron microscopy analysis. Dye-sensitized solar cells fabricated using flame- SnO_2 nanoparticles show higher efficiency ($\eta = 2.72\%$) than that of commercial SnO_2 nanoparticles ($\eta = 1.53\%$). The increased efficiency is attributed to suppression of electron recombination caused by passivation of sub-band-edge surface states. DOI:10.1557/mrc.2017.97

Electrode architecture of carbon-coated silicon nanowires through magnesiothermic reduction for lithium-ion batteries

Young Gyu Nam, **Mohammad Humood**, **Haejune Kim**, and **Andreas A. Polycarpou**, Texas A&M University, USA

Carbon-coated silicon nanowires (C-Si NWs) were prepared as anodes for lithium-ion batteries (LIBs). The C-Si NWs were synthesized using a simple and effective fabrication strategy via magnesiothermic reduction. The synthesis sequence of carbon coating before the chemical etching of the reduced Si NWs/MgO composite was found to be critical for improved battery performance. In addition, carbon coating was found to help to stabilize the solid electrolyte interphase layer during battery cycling, which is important to realize the benefits of Si-based LIBs. This synthesis method provides an efficient route to synthesizing high-performance Si electrodes via magnesiothermic reduction. DOI:10.1557/mrc.2017.106

Impact of enhanced oxide reducibility on rates of solar-driven thermochemical fuel production

Michael J. Ignatowich, California Institute of Technology, USA; **Alexander H. Bork**, ETH Zürich, Switzerland; **Timothy C. Davenport**, Northwestern University, USA; **Jennifer L.M. Rupp**, Massachusetts Institute of Technology, USA; **Chih-kai Yang**, California Institute of Technology, USA; **Yoshihiro Yamazaki**, Kyushu University, Japan; and **Sossina M. Haile**, Northwestern University and California Institute of Technology, USA

Two-step, solar-driven thermochemical fuel production offers the potential of efficient conversion of solar energy into dispatchable chemical fuel. Success relies on the availability of materials that readily undergo redox reactions in response to changes in environmental conditions. Those with a low enthalpy of reduction can typically be reduced at moderate temperatures, important for practical operation. However, easy reducibility has often been accompanied by surprisingly poor fuel production kinetics. Using the $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ series of perovskites as an example, we show that poor fuel production rates are a direct consequence of the diminished enthalpy. Thus, material development efforts will need to balance the countering thermodynamic influences of reduction enthalpy on fuel production capacity and fuel production rate. DOI:10.1557/mrc.2017.108

Formation of a hexagonal closed-packed phase in $\text{Al}_{0.5}\text{CoCrFeNi}$ high entropy alloy

J. Wang, **Y. Zhang**, **S.Z. Niu**, **W.Y. Wang**, **H.C. Kou**, and **J.S. Li**, Northwestern Polytechnical University, USA; **S.Q. Wang**, Xi'an Shiyou University, China; and **E. Beaunon**, University Grenoble Alps and CNRS, France

We report that a hexagonal closed-packed (HCP) phase with high cobalt content precipitates in $\text{Al}_{0.5}\text{CoCrFeNi}$ high entropy alloy (HEA) after 650 °C/8 h heat-treatment. The precipitate with the shape of plate is completely located at the interdendritic region. Results of electron diffraction and high resolution transmission electron microscopy show that the HCP phase was transformed from the body-centered cubic phase through a simple shear and the two phase obey an orientation relationship. The thermodynamic stability of $\text{Al}_{0.5}\text{CoCrFeNi}$ HEA should be carefully reevaluated, especially at the vulnerable temperature. DOI:10.1557/mrc.2017.109

Stable Pt clusters anchored to monovacancies on graphene sheets

Bharat K. Medasani, **Jun Liu**, and **Maria L. Sushko**, Pacific Northwest National Laboratory, USA

First principles simulations and global optimization predict new mode of binding of Pt clusters with defects on graphene that significantly enhances their stability. Pt clusters were found to firmly bind to monovacancies in configuration transacting the vacancy site, while retaining the integrity of the cluster. Diffusion calculations support tight anchoring of Pt cluster to monovacancy. Pt cluster adsorbed on pristine graphene or other common defects exhibit a different mode of adsorption and only decorate one side of graphene. This study reveals strong influence of defect chemistry on the structure and mobility of Pt nanoclusters adsorbed on graphene and have important implications for catalytic and gas sensing applications. DOI:10.1557/mrc.2017.112

The effects of hydrostatic pressure on the martensitic transition, magnetic, and magnetocaloric effects of $\text{Ni}_{45}\text{Mn}_{43}\text{CoSn}_{11}$

Sudip Pandey, Southern Illinois University, USA; **Ahmad Us Saleheen**, Louisiana State University, USA; **Abdiel Quetz**, Southern Illinois University, USA; **Jing-Han Chen**, Louisiana State University, USA; **Anil Aryal** and **Igor Dubenko**, Southern Illinois University, USA; **Philip W. Adams** and **Shane Stadler**, Louisiana State University, USA; and **Naushad Ali**, Southern Illinois University, USA

The magnetic and magnetocaloric properties of $\text{Ni}_{45}\text{Mn}_{43}\text{CoSn}_{11}$ have been investigated using heat capacity measurements and magnetization with hydrostatic pressure applications. A shift in the martensitic transition temperature by 40 K to higher temperatures was observed with application of pressure $P = 1.06$ GPa. The magnetic entropy changes significantly increases from 24 to 42 J/kgK at pressure of 0.73 GPa. A large adiabatic temperature change of 4 K was found from specific heat measurements. Also, the density of states and Debye temperature has been estimated from heat capacity measurements. The mixed effects of pressure and magnetic field on the transition temperature are discussed. DOI:10.1557/mrc.2017.110

Three-dimensional microscale flow of polymer coatings on glass during indentation

L.R. Bartell and **N.Y.C. Lin**, Cornell University, USA; **J.L. Lyon**, **M.L. Sorensen**, **D.A. Clark**, **M.J. Lockhart**, **J.R. Matthews**, **G.S. Glaesemann**, and **M.E. DeRosa**, Corning Research and Development Corporation, USA; and **I. Cohen**, Cornell University, USA

We present an indentation-scope that interfaces with confocal microscopy, enabling direct observation of the three-dimensional (3D) microstructural response of coatings on substrates. Using this method, we compared microns-thick polymer coatings on glass with and without silica nanoparticle filler. Bulk force data confirmed the >30% modulus difference, while microstructural data further revealed slip at the glass-coating interface. Filled coatings slipped more and about two times faster, as reflected in 3D displacement and von Mises strain fields. Overall, these data indicate that silica-doping of coatings can dramatically alter adhesion. Moreover, this method compliments existing theoretical and modeling approaches for studying indentation in layered systems. DOI:10.1557/mrc.2017.114

N-Phenyl naphthalene diimide pendant polymer as a charge storage material with high rate capability and cyclability

Subashani Maniam, Monash University, Australia; and **Kouki Oka** and **Hiroyuki Nishide**, Waseda University, Japan

Pendant-type polymers are attractive materials which allow the flexibility to introduce various redox active moieties that facilitate rapid ion/electron transport and enable charge storage. Here, we demonstrate naphthalene diimide polymers with polynorbornene backbone having *N*-phenyl, PNA $\mathbf{5}$ and *N*-(4-nitrophenyl), PNNO $\mathbf{6}$. Small changes in the molecular design have led to a significant difference in bulk material and device properties. PNNO $\mathbf{6}$ maintained 80% of its capacity at 1C after 10 cycles in a Li-ion coin cell. PNA $\mathbf{5}$ $\mathbf{5}$

displayed exceptionally high charge capacity and rate capability with excellent cyclability, maintaining almost its theoretical capacity at various C-rates throughout 500 cycles. DOI:10.1557/mrc.2017.127

Covalent immobilization of lysozyme in silicone rubber modified by easy chemical grafting

G.G. Flores-Rojas, Universidad Nacional Autónoma de México, Mexico, and Nano Medical Engineering Laboratory, Japan; F. López-Saucedo and E. Bucio, Universidad Nacional Autónoma de México, Mexico; and T. Isoshima, Nano Medical Engineering Laboratory, Japan

Functionalization of silicone rubber films with lysozyme was achieved by grafting copolymerization and its chemical activation allowing the covalent immobilization of the enzyme. The new materials were characterized by means of Fourier-transform infrared spectroscopy, differential scanning calorimetry, thermogravimetric analysis, contact angle, atomic force microscopy, and mechanical properties of films. The enzymatic activity of films was studied by a suspension of lyophilized *Micrococcus lysodeikticus*. The activity test was inquired at different pH and temperatures, exhibiting enzymatic activity 20 °C above the free lysozyme, and at pH = 5 where the free lysozyme did not show activity. DOI:10.1557/mrc.2017.115

Fabrication of Mn-CeO_x/polyphenylene sulfide functional composites by an *in situ* reaction for low-temperature NO reduction with NH₃

Weijie Zheng, Yuying Zheng, and Jian Chen, Fuzhou University, China; and Yanbing Zhang, Henan University of Urban Construction, China

Polyphenylene sulfide (PPS) has good corrosion resistance, chemical stability, and thermal stability, which was widely applied in precipitator field. In this paper, a novel *in situ* synthesis protocol was selected to fabricate the Mn-CeO_x/PPS functional composites with excellent low-temperature denitration activity. Results show that the as-obtained Mn-CeO_x/PPS filter possessed of superb denitration activity at 180 °C under a weight hourly space velocity of 210,000 mL/g_{cat}/h, which may be stemmed from the generation of amorphous and well-dispersed mixed metal oxide catalysts. DOI:10.1557/mrc.2017.120

Achieving antimicrobial activity through poly(*N*-methylvinylimidazolium) iodide brushes on binary-grafted polypropylene suture threads

F. López-Saucedo, G.G. Flores-Rojas, and E. Bucio, Universidad Nacional Autónoma de México, Mexico; C. Alvarez-Lorenzo and A. Concheiro, Universidade de Santiago de Compostela, Spain; and O. González-Antonio, Universidad Nacional Autónoma de México, Mexico

Harnessing the properties of imidazolium species, antimicrobial activity against Gram-negative and Gram-positive bacteria was attained by binary-grafting 2-hydroxyethyl methacrylate (HEMA) or *N*-isopropylacrylamide, followed by *N*-vinylimidazole onto polypropylene (PP) monofilaments (sutures) using ⁶⁰Co γ -rays. Ulterior functionalization with methyl iodide was carried out to endow brushes with antimicrobial activity on the PP surface. The PP-grafted sutures were characterized by means of Fourier-transform infrared spectroscopy attenuated total reflection, scanning electron microscopy, differential scanning calorimetry, and thermogravimetric analysis, and regarding the mechanical properties and the responsiveness to pH and temperature. Tests were performed on *Escherichia coli* and *Staphylococcus aureus* achieving large inhibition zones. DOI:10.1557/mrc.2017.121

Characteristics of nickel thin films prepared by electroless plating in foam of electrolyte

Takahiro Furuhashi and Yoshiyasu Yamada, Yamada Co. Ltd., Japan; and Masato Hayashi, Shoji Ichihara, and Hiroaki Usui, Tokyo University of Agriculture and Technology, Japan

Nickel thin films were prepared by electroless plating in a foam of electrolyte that was generated by bubbling nitrogen gas into a hypophosphite-based electroless plating solution to which was added a surfactant of sulfuric acid monododecyl ester sodium salt. Although the film growth rate in the foam was considerably lower than that in the conventional liquid, film growth was enhanced by inducing a flow in the foam. Compared with films deposited in liquid, the films deposited in foam had a smaller number of pinholes, smaller crystallite size, and superior corrosion resistance. The ferroxyl indicator test showed that the area of corrosion can be reduced to less than 1/20 by depositing the film in foam instead of liquid. DOI:10.1557/mrc.2017.124

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