

## 元素戦略プロジェクト<研究拠点形成型>

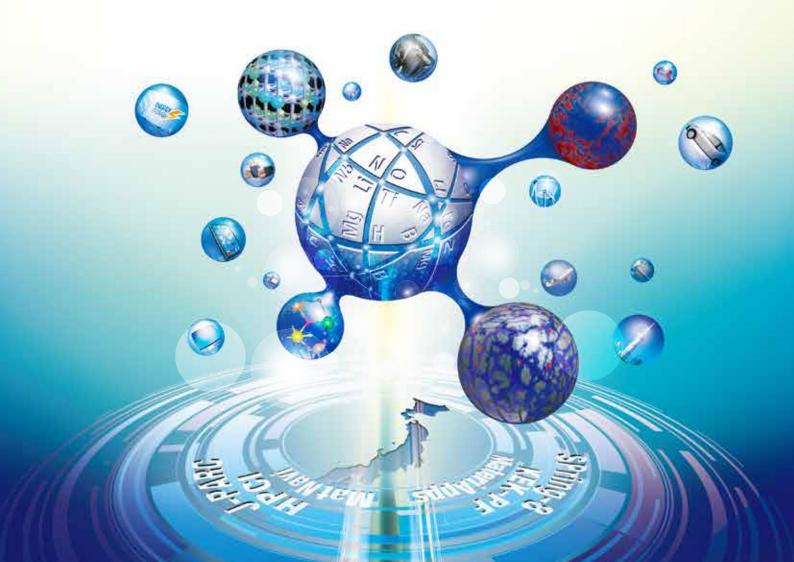
# HERE DIOTENASI

地球は多様な元素の集まりです。私たちの暮らしは元素からつくった無数の材料で成り立っています。持続可能な未来社会を構築するには、地球上の有限な元素資源から優れた材料をつくりだしていく「元素戦略」が必要です。

「元素戦略プロジェクト」は2012年に発足。材料の機能や特性を決めている元素のサイエンスを追究し、イノベーションの創出と産業応用を促進しています。

### Omotenashi (O Mo Te Na Si): Being Hospitable Hosts to the Earth

The Earth is a collection of diverse elements. Our existence is made possible by countless materials produced from these elements. In order to build a sustainable future, we need an "element strategy" for developing superior materials from the limited element resources on Earth. The Element Strategy Initiative was launched in 2012 to pursue the science of elements that determine material functions and properties and for encouraging innovation and industrial applications.



# 基礎から応用に直結する研究開発を 4拠点で進めています。

Working to develop practical applications directly from basic research at four research centers.



元素戦略プロジェクト<研究拠点形成型> プログラム・デイレクター(PD)

玉尾皓平

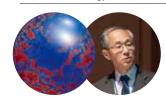
#### Kohei Tamao

Program Director, Element Strategy Initiative: To Form Core Research Centers

#### 磁性材料研究拠点

The Elements Strategy Initiative Center for Magnetic Materials (ESICMM)





設置機関:物質·材料研究機構(NIMS) Core: National Institute for Materials Science (NIMS)

代表研究者 : 広沢 哲 Satoshi Hirosawa, Director General

連 携 機 関 : 東北大学、産業技術総合研究所、東京大学、東京大学物性研究所、京都大学、 高エネルギー加速器研究機構、高輝度光科学研究センター、名古屋大学、

北陸先端科学技術大学院大学、東京工業大学、九州大学、東北学院大学、兵庫県立大学 Collaborating institutes: Tohoku University, the National Institute of Advanced Industrial Science and Technology, the University of Tokyo, the Institute for Solid State Physics at the University of Tokyo, Kyoto University, the High Energy Accelerator Research Organization (KEK), the Japan Synchrotron Radiation Research Institute, Nagoya University, the Japan Advanced Institute of Science and Technology, the Tokyo Institute of Technology, Kyushu University, Tohoku Gakuin University, and the University of Hyogo

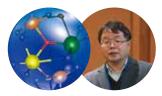
開発対象:電気エネルギーと機械エネルギーの変換に適用されているバルク永久磁石材料

開発目標: 希少元素フリーの新規高性能永久磁石材料の創製

## 電子材料研究拠点

Tokodai Institute for Element Strategy (TIES)





設置機関: 東京工業大学 Core: Tokyo Institute of Technology

代表研究者: 細野秀雄 Hideo Hosono, Representative 連携機関: 物質・材料研究機構、高エネルギー加速器研究機構、東京大学

で Mig 1947年 の 大阪神 向上イルイー 加速船が元成神 未ぶ入子 Collaborating institutes: National Institute for Materials Science, High Energy Accelerator

Research Organization (KEK), and the University of Tokyo

開発対象: 半導体、透明電極をはじめとする電子材料全般開発目標: 多存元素を使って革新的な機能を実現

### 触媒•電池材料研究拠点

 $\textbf{Elements Strategy Initiative for Catalysts and Batteries} \, (\textbf{ESICB})$ 





設置機関: 京都大学 Core: Department of Molecular Engineering, Kyoto University

代表研究者 : 田中庸裕 Tsunehiro Tanaka, Director

連携機関:東京大学、自然科学研究機構分子科学研究所、九州大学、熊本大学、東京理科大学

Collaborating institutes: The University of Tokyo, the Institute for Molecular Science of the National Institutes of Natural Sciences, Kyushu University, Kumamoto University, and the

Tokyo University of Science

開発対象: 固体触媒と二次電池材料

開発目標: 希少元素フリーの新規高性能触媒と二次電池を実現

### 構造材料研究拠点

Elements Strategy Initiative for Structural Materials (ESISM)





設置機関:京都大学 Core: Department of Materials Science and Engineering, Kyoto University

代表研究者: 田中 功 Isao Tanaka, Director

連携機関:東京大学、大阪大学、物質·材料研究機構、九州大学、経産省ISMA、内閣府SIP

Collaborating institutes: The University of Tokyo, Osaka University, the National Institute for Materials Science, Kyushu University, the Innovative Structural Materials Association

(METI), and the Strategic Innovation Promotion Program (the Cabinet Office) 開発対象: 実用材料として適用している金属材料、高靭性セラミックス材料、それらの複合材料

開発目標: 強度と靭性が両立する究極材料の実現

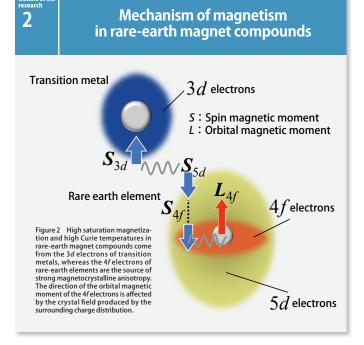




- · Growing market for high-performance permanent magnets
- Anticipation for novel materials beyond neodymium magnet
- · Anticipation for novel magnet materials that do not include critical elements

- Understanding high saturation magnetization and high magnetocrystalline anisotropy based on quantum theory
- Efficient materials discovery using a combination of computational, data-driven, and theoretical approaches
- · High magnetic properties of Fe-rich rare-earth magnet compound NdFe<sub>12</sub>N: prediction by first-principles calculations and visualization

#### **Requirements for** high-performance magnet materials Microstructure Main phase Nd<sub>2</sub>Fe<sub>14</sub>B NdFe<sub>12</sub>N Sub phases · Neodymium oxides Main phase Amorphous High saturation magnetization Sub phases High magnetocrystalline Optimized microstructures anisotropy for high coercivity High Curie temperature Figure 1 High-performance permanent magnet materials require a main phase having high saturation magnetization, high magneto crystallineanisotropy, high Curie temperature, and a sub phase that blocks the magnetic coupling among main phases. Coercivity has strong positive correlation with the microstructure.



#### Search for maximum performance of magnets through ab-initio data catalyst support

#### Procedure for materials design

Several hundreds of data points obtained by high-throughput ab-initio calculation are visualized

Materials located around the peak position in performance are searched and targeted as candidate materials after their structural stabilities are

Among candidates, those possibly synthesized are further investigated theoretically in detail .

Syntheses of thus predicted mate and their characterizations are tried out.

Ferromagnetism is unstable in the region to the left of the dashed line.

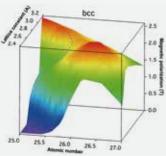


Figure 3 Saturation magnetic polarization JS of the system plotted against the lattice constant a and atomic number of a fictitious atom Z. The ferromagnetic state is unstable in the region to the left of the dashed line. Here a bcc structure is assumed, and the electron number corresponds to the atomic number of fictitious atoms. A dome-like structure appears around a = 2.65 A and Z = 26.4, where JS takes the maximum value of 2.66 T, which is much larger than that of  $Nd_2Fe_{14}B$ , is seen.

#### **Visualization in Mechanism** of magnetism

- Visualization of electron spin-density that may affect the magnetic
- properties.

   Development of 3D-Printing technology that enables us to depict the crystal structure and electron density in the transparent resin.

This helps us to get better understanding and deep insights, supporting the creation of new ideas.

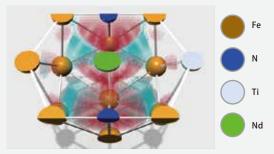


Figure 4 Difference in the electron spin-density is obtained by subtracting that of NdFe $_{11}$ Ti from that of NdFe $_{11}$ TiN. The difference near the N atoms is conspicuous, is seen.

- Discovery of novel magnet compounds and optimization of chemical composition by machine learning / AI
- Mechanism elucidation of coerciviry

- Electric motors for xEVs (hybrid vehicles, electric vehicles etc.)
- Wind turbines
- · Home electric appliances
- · Industrial robot
- · drone etc.

Contact us here The Elements Strategy Initiative Center for Magnetic Materials

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# Creation of Novel-Function & High-Performance Electronic Materials based on Novel Concepts for Materials Design





**GL: Hideo Hosono,** Representative,TIES

Backgroun to the research

- There have been numerous large-scale national projects on electronic materials to date.
- Materials design has become fixated on just a few concepts, such as diamond-structure semiconductors and perovskite dielectrics.
- There is a need for novel perspectives on abundant elements independent of obsolete approaches based on previous successes.

Points of the research

- Exploring novel material candidates making full use of condensed matter physics, electron theory, computational chemistry, materials informatics (M. I.), etc.
- Rapid syntheses of candidate materials utilizing such extreme experiments as super-high pressure and super-high vacuum.
- Rapid feedback of material property analyses using synchrotron radiation (KEK-PF and SPring-8), neutrons, and muons (J-PARC).

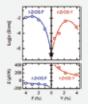
Outline of the research

Novel Direct Band-Gap, Bipolar Semiconductors derived from New Concepts of M. I. and Molecular Orbitals



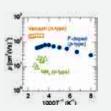
Red Luminescence as designed

Figure 1 Novel nitride semiconductor,  $CaZn_2N_2$ , discovered by M.I. and synthesized under high pressure



**Bipolar Doping** 

Figure 2 Oxysulfide semiconductor based on early-transition metals, ZrOS, designed under molecular orbital concepts



Hall Mobilities

Figure 3 A new nitride semiconductor, Cu<sub>3</sub>N, for solar-cell applications by p-type doping with interstitial fluorine

Outline of the research

Novel Semiconductor Materials for Next-Generation Light-Emission-Type Flat-Panel Displays



Green Electroluminescence with FWHM of 16 nm and Maximum Intensity of 180,000 cd m<sup>-2</sup>

Figure 4 Low-voltage, high-brightness LED based on halide perovskite semiconductors with the novel oxide

semiconductor Zn-Si-O as an electron-transport layer.

Figure 5 High efficiency blue-light emitting lead-free inorganic iodate semiconductor, Cs<sub>3</sub>Cu<sub>2</sub>I<sub>5</sub>.

Electroluminescence

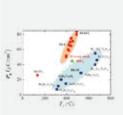
by electron injection from Zn-Si-O

Figure 6 Low-temperature, printable, high-mobility, p-type, transparent, amorphous semiconductor: Cs<sub>2</sub>Cu<sub>2</sub>I<sub>5</sub>.

**Hall Mobilities** 

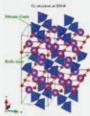
Outline of research

High-temperature stable, high dielectric constant, non-perovskite dielectrics for power-electronics applications



Higher  $T_{\rm C}$  and Higher  $P_{\rm S}$  than conventional materials

Figure 7 Fluorite ferroelectrics with high  $T_C \& P_S$ : HfO<sub>2</sub>: Y.



Crystal Structure

Figure 8 Silicate ferroelectrics satisfying requirements for vehicle applications: (Bi,La)<sub>2</sub>SiO<sub>5</sub>



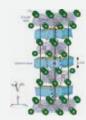


2-inch Single Crystal and Sensor Module

Figure 9 High-temperature stable piezoelectrics for vehicle pressure sensors: CTAS (Ca<sub>3</sub>TaAl<sub>3</sub>Si<sub>2</sub>O<sub>14</sub>).

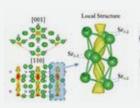
Outline of the research

Inorganic and Solid-State Electride Materials soon to be commercialized as catalysts for ammonia synthesis at low temperature and pressure



Crystal Structure of Ca<sub>2</sub>N

Figure 10 Two-dimensional electrides under consideration for the base of commercial material systems



Crystal Structure and Electron Density Mapping

Figure11 A novel, intermetallic, one-dimensional electride explored using genetic algorithms: Sr<sub>s</sub>P<sub>3</sub>



a 2D electride, Y₂C

Figure 12 A novel concept of "topological electrides" and their candidate materials.

Roadmap

- Stabilization of creation rates of novel materials and their design concepts
- Technical transfer to industries
- Concept proposal for post Elements Strategy

Applications

- High-efficiency & low-cost semiconductor materials for flat-panel displays
- High-temperature stable & high-dielectric constant dielectrics for vehicle power electronics
- Electride materials for catalysts

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### Unraveling the Roles, Functionalities, and States of Hydrogen in Condensed Matter and Establishing **Highly Sensitive Quantification Methods**





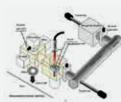


GL: Hideo Hosono,

- · The presence of hydrogen in oxides has often been ignored despite its high concentration ( $\geq 10^{20} \text{cm}^{-3}$ ).
- · Experimental and theoretical investigations have implied that the hydrogen strongly influences local and electronic structures of condensed matter.
- It is important to establish methods of quantification and state analysis for hydrogen and to unravel the roles and functionalities of hydrogen in order to utilize hydrogen as a key functional element.

- · Development of thin films with extremely low hydrogen content and a method and apparatus to quantify the hydrogen concentration with high sensitivity.
- Combined use of density functional theory calculations and analyses using hydrogen-sensitive probes, such as NMR, infrared absorption, neutrons, and muons.
- Investigations of relationships between the concentrations and states of hydrogen in semiconductor thin films and the stability of their electron devices.

#### Development of a High-Sensitivity Method & Apparatus to Quantify the Hydrogen Concentration in Thin Films



All-Vacuum System Setup from Thin-Film Deposition



Overall Configuration of

Thermal Desorption Spectra of an a-IGZO Thin Film with Extremely Low Hydrogen Content (no more than 10<sup>16</sup>cm<sup>-3</sup>)

Figure 1 High-Sensitivity method and apparatus for quantifying hydrogen concentration and their demonstration on a-IGZO thin films with extremely low hydrogen contents (under 10<sup>16</sup> cm<sup>-3</sup>).

Implemented using an extremely low-out-gassing, ultra-high vacuum system with

all-vacuum transfer from the thin-film deposition chamber, and by performing calibration using high-accuracy ion implan-tation and the introduction of trace amounts

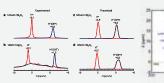
Used for preparing reference samples with

#### Hydride Ions in Oxide Hosts Hidden by Hydroxide ions





Crystal Structure of C12A7 Mayenite (12CaO·7Al<sub>2</sub>O<sub>3</sub>) Having a Cage Structure



<sup>1</sup>H NMR Spectra Derived from DFT Calculations of Hydrogen-Incorporated Structure Models, Which Reproduce the Experimental Observations

A reversal in Chemical Shifts of H and H<sup>-</sup> accompanying an Increase in O-H and M-H Distances

Figure 2 Demonstrating the presence of

hydride in solid-state oxide crystals
• Splits of <sup>1</sup>H NMR spectra were observed in nite and apatite crystals

 A reversal in chemical shifts of H<sup>+</sup> and H<sup>-</sup> is predicted by DFT calculations based on crystal structure models with hydrogen

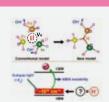
#### Hydrides (hydrogen anions) in Amorphous Oxide Semiconductors and the Origin of Instabilities in their Electron Devices



A Negative Shift of Threshold Voltage in a-IGZO TFT under Negative Bias Illumination Stress (NBIS)



Extra Peaks Originating from M-H Bonds (H<sup>-</sup>) in the Infrared Absorption Spectrum



Local Structure of Hydrogen Anions and Generation of Occupied Deep Subgap States

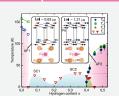
Figure 3 Hydrides (hydrogen anions) in a-IGZO thin films and the origin of instabilities

- Amorphous oxide semiconductor TFTs such as a-IGZO exhibit large negative shifts in threshold voltage under illumination and negative bias stresses.
- Hydrides composed of metal cation-hydro-gen anion bonds are observed in infrared

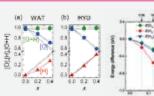
absorption spectra and identified by DFT calculations of hydrogen-incorporated

Hydrogen atoms incorporated into oxygen vacancies, V<sub>o</sub>, bond to adjacent metal cations to form hydrides (hydrogen anions, H<sup>-</sup>), which generate occupied deep subgap states above the valence-band maximum (VBM) and cause NBIS instabilities in a-IGZO TFTs.

Electron Doping by Hydrogen Incorporation in Iron Oxyprictides to Produce Superconducting Phase



Crystalline/Magnetic Phase Diagram of LaFeAsO<sub>1-X</sub>H<sub>X</sub> Unraveled by Quantum Beams of SRX-ray Scattering, Neutron Scattering, and Muons



x-Dependence of Hydrogen and Oxygen Contents in an LaFeAsO<sub>1-x</sub>H<sub>X</sub> Specimen Formed under  $H_2$ O (WAT),  $H_2$ (HYD) Ambient

Theoretical Prediction for x-Dependence of Stability by DFT Calculations

- Figure4 Superconducting phase generated by hydrogen doping in Iron Oxypnictide LaFeASO<sub>1-x</sub>H<sub>x</sub> and states of the non-substituted hydrogen.

   Iron oxypnictide LaFeASO<sub>1-x</sub>H<sub>x</sub> shows, depending on x, two superconducting phases (SC1, SC2) originating from two antiferromagnetic mother phases (AF1, AF2).
- Hydrogen and oxygen content were found experimentially to be complementary to each other.
- DFT calculations predict that stability is greatest when hydrogen anions H<sup>-</sup> occupy the sites of oxygen vacancies V<sub>0</sub>, which suggests the common image of V<sub>0</sub> stabiliza-

- Clarifying the relationships between material functionalities and device characteristics
- Extension to dielectric materials
- Exploration and creation of hydrogen-functionalized electronic materials

- Performance enhancement and stabilization of electronic materials and electron devices
- Novel hydrogen-functionalized materials including hydrides
- Analysis of low-hydrogen-content specimens

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# Novel Automotive Catalysts Using Less Noble Metals

- · Reducing the use of platinum-group metals (PGM) in automotive catalysts is of practical importance.
- Thermal sintering of metal nanoparticles poses a significant problem to catalyst life.
- · Catalytic active sites are thought to be the surface of noble metal particles.

- · Plasma preparation for Rh nanofilm-coated metal honeycomb
- The catalytic activity of the Rh nanofilm is markedly superior to that of Rh nanoparticles.
- · Improvement in catalytic performance using the function of a catalyst support.

#### Paradigm shift in catalyst preparation and structure

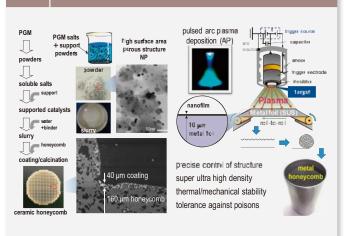


Figure 1 (Left) Conventional catalyst preparation via wet coating of powder containing metal

nanoparticles.
(Right) Novel catalyst preparation via pulsed arc plasma deposition of active metal nanofilms onto metal foil substrates.

#### Catalytic performance of Rh nanofilm and Rh nanoparticle catalysts

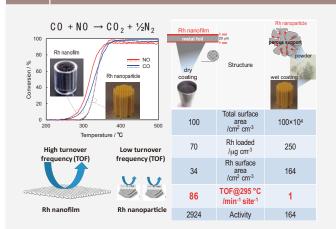
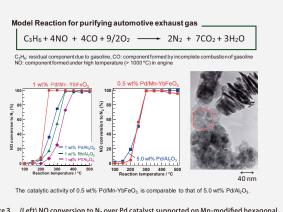


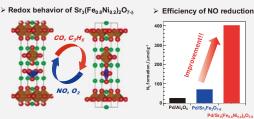
Figure 2 Catalytic performance of a Rh nanofilm-coated honeycomb catalyst compared with a conventional honeycomb prepared by wet coating. 0.1% NO, 0.1% CO,  $N_2$  balance. GHSV = 1.2×

#### Reduction of Pd usage by improving a function of catalyst support



(Left) NO conversion to N<sub>2</sub> over Pd catalyst supported on Mn-modified hexagonal (Right) TEM image of Pd/Mn-YbFeO<sub>3</sub> having hexagonal plate-like nanoparticles.

#### Development of novel catalyst support havingoxygen storage capacity



➤ Mechanism for NO reduction on Sr<sub>3</sub>(Fe<sub>0.8</sub>Ni<sub>0.2</sub>)<sub>2</sub>O<sub>7-8</sub>

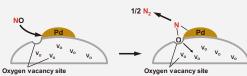


Figure 4 (Upper left) Structural change of  $Sr_3(Fe_{0,0}Ni_{0,2})_2O_{7-\epsilon}$  during the purification of automotive exhaust gas (Upper right) Efficiency of NO reduction at oxygen vacancy site in  $Pd/Sr_3(Fe_{0,0}Ni_{0,2})_2O_{7-\epsilon}$  (Below) Mechanism for NO reduction in  $Pd/Sr_3(Fe_{0,0}Ni_{0,2})_2O_{7-\epsilon}$ .

- Elucidation of the reaction mechanisms for nanofilm catalysts
- Development of nanofilm catalysts for various chemical
- · Development of novel catalyst materials using the crystal structure of catalyst supports

- Automotive catalysts
- Microreactors and smart reactors

#### Contact us here

**Elements Strategy Initiative for** Catalysts and Batteries, Kyoto University admin@esicb.kyoto-u.ac.jp



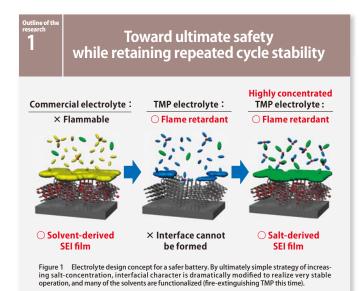


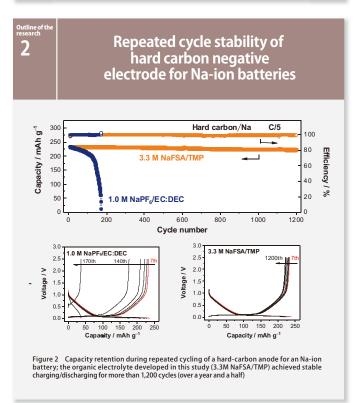




- Fire incidents with explosions occurring in lithium-ion batteries have been problematic.
- The simple addition of flame-retardant solvents significantly degrades the battery cycle.
- · How can we produce better batteries with a high level of

- · Highly concentrated fire-extinguishing TMP-based electrolyte can form a very robust passivation film on the anode surface.
- This enables an excellent battery life of more than 1,000 charge-discharge cycles with negligible degradation.
- Computational analysis using the K computer has revealed the formation mechanisms of stable passivation film.





#### Comparison of flammability

#### General electrolytes



#### New highly concentrated electrolytes





Figure 3 Flame tests on conventional 1.0 M NaPF $_6$ /EC:DEC (1:1 by vol) electrolyte (left) and laboratory-made 3.3 M NaFSA/TMP electrolyte (right)

#### **Electronic structures of** highly concentrated electrolytes

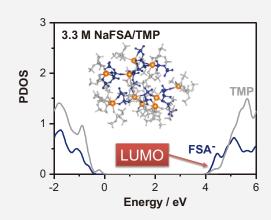


Figure 4 Electronic structures and coordination structures (insets) of 3.3 M NaFSA/TMP solutions obtained with DFT-MD simulations. Na $^{+}$  cations, FSA $^{-}$  anions and TMP solvents are shown in orange, blue and grey, respectively. The pDOS, projected density of states, shows that LUMO are located at FSA and that the FSA is attacked first upon reduction to form a passivation film.

- · Paradigm shift of electrolyte concept
- · Cost-cutting and industrial development
- · Optimization for mass production

- Consumer batteries
- · Batteries for vehicles
- · Stationary batteries

#### Contact us here

**Elements Strategy Initiative for Catalysts** and Batteries, Kyoto University admin@esicb.kyoto-u.ac.jp





# Concurrent enhancement of strength and ductility in bulk nanostructured hexagonal metals



- Concurrent enhancement of strength and ductility in structural materials is essential for guaranteeing a safe and secure society.
- Applications for strong, light-weight titanium and magnesium alloys have been limited by poor ductility owing to their hexagonal crystal structures.
- · Creation of innovative hexagonal metals through bulk nanostructuring rather than the addition of critical alloying elements is targeted.

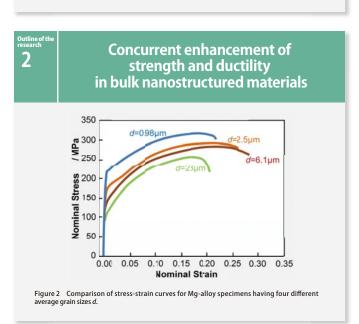
- · Fully recrystallized titanium and magnesium alloys with various average grain sizes down to the sub-micrometer scale were successfully fabricated.
- ·Concurrent enhancement of strength and ductility was successfully achieved in bulk nanostructured titanium and magnesium alloys.
- Atomistic mechanisms of the superior mechanical properties in bulk nanostructured materials were clarified through state-of-the-art experiments, such as transmission electron microscopy and neutron diffraction, as well as theoretical calculations.

Figure 3 Transmission electron micrographs of ultrafine grained magnesium alloy sample with average grain sizes of d=0.98  $\mu$ m after tensile deformation to a strain of 0.095. (a) Bright-field image observed along [10-10] zone axis. (b) Bright-field and (c) corresponding dark-field images observed under two-beam condition with diffraction vector g= (0002). (d) Weak-beam dark-field image of the rectangular area in (b) at a higher magnification. Highly dense, unusual dislocations with c-component can be observed. Such dislocations are presumed to be nucleated at grain boundaries when the stress level increases. boundaries when the stress level increases

Mechanism of enhanced ductility in

bulk nanostructured materials

# Fully recrystallized magnesium alloys with various average grain sizes 400 °C,30min 10 µm Magnesium-alloy samples having various average crystal grain sizes fabricated by high-pressure torsion (HPT) followed by recrystallization annealing treatments at the given temperatures and durations. (Top) Inverse pole figure (IPF maps by electron backscatter diffraction (ESSD). Colors correspond to crystallographic orientations of grains. (Bottom) Grain boundary (GS) maps in which the blue and grow lines correspond to high angle and low angle grain boundaries.



## Comprehensive understanding of ductility enhancement in bulk nanostructured metals under the new concept of *plaston*

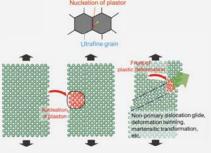


Figure 4 Unique plastic deformation mechanisms different from primary dislocation glide, such as non-primary dislocation glide, deformation twinning, and martensitic transformation, have been experimentally found to take place in bulk nanostructured materials. These mechanisms can be comprehensively explained under the new concept of plaston. When a large enough stress is applied, collective atomic motion occurs at lattice imperfections such as surfaces and grain boundaries, which eventually leads to the nucleation of plastic deformation. Similar phenomena are thought to take place at the front of the plastic deformation.

- Concurrent enhancement of strength and ductility in bulk nanostructured steels based on the new concept of plaston.
- Fundamental understanding of plaston in other materials having a variety of chemical bonds in order to overcome their brittleness.

- Vehicles and aircrafts
- Structures
- Biomaterials

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