Application of the ATOMKI-ECRIS for materials research and prospects of the medical utilization

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Collaborators:

• National Institute of Radiological Sciences (NIRS), Chiba, Japan
• Bio-Nano Electronics Research centre, Toyo University, Kawagoe, Japan
• Division of Electrical Engineering, Osaka University, Osaka, Japan
• Tateyama Machine Co., Ltd., Toyama, Japan
Outline

Introduction

1. New fullerene-based materials to be produced in ECR discharge

2. Bone cell growth on titanium coated with fullerene.

3. Optical changes in thin films by HCI irradiation

4. Guiding of HCI through nano- and micro-capillaries

Comparison, summary
The ATOMKI-ECRIS

- 14.3 GHz/1 kW
- Axial field: 1.2/0.4/1.0 T
- Radial field: 0.9 T
- Chamber: ø58x200
- Extraction: 0.2-30 KV
Two versions of the ECRIS

**ATOMKI-ECRIS-A**

- Klystron, 14 GHz, 1000 W
- Magnets: 1.2/0.4/1.0 and 0.9 T
- Plasma chamber: φ6x22 cm
- For HCl plasmas/beams

**ATOMKI-ECRIS-B**

- TWTAs, 6-18 GHz, 20 W
- Magnets: 0.9/0.3/0.9 and 0.7 T
- Plasma chamber: φ10x40 cm
- For LCI, large-size plasmas
1. Fullerene-based new materials ($C_{60}+X$) to be produced in ECR-discharge

Since 2000 fullerene plasmas and beams have been produced at the ATOMKI ECRIS

- $C_{60}$: just a working material (as Ne, Ar, Fe), no "$C_{60}$ research"
- Production of $C_{60}$ ion beam (intensity, charge)
- Fullerene derivatives (destroying-adding)
- Mixture plasmas ($C_{60}+X$)
- New materials (by $C_{60}+X$) to detect in beam
- New materials to produce in macroscopic quantity
- Applications (surface coating, etc.)
Basic C60 experiments

Typical fullerene derivative spectrum. The plasma was tuned for C_{60}^{2+}. The well-known shape of the derivative peaks is clearly observed.

Fullerenes can break into clusters: carbon chains or rings with atom numbers between 2 and 30 (both odd and even). Spectrum shows plasma tuned for C_{11}^{+}. Highest peaks: 3+, 7+, 11+, etc. The 4n+3 "magic" rule.
Next-Generation Information & Nano-Bio Technologies Devices

(energy / evaluation / semiconductor / storage / display / bio)

Creation of New-Functional Evolved-Nanomaterials

Nanoscopic Plasma Processes

Fullerene Nanotube Colloid

Nanocarbon Network

High-efficient organic solar cell
Small-sized electronic circuit
Quantum computing
Superconductivity Spintronics
High-density magnetic memory
Low-power flat panel display
High-accuracy gene diagnosis
High-sensitivity Bio sensor

Gaseous Plasmas
Solution Plasmas

Slide made by R. Hatakeyama, Tohoku University, Sendai
Ha \text{X}=\text{Fe} \rightarrow \text{Fe}[@\text{C}_60

Nanotechnology

- new SC materials
- quantum computing

Medical treatments

- ultra-contrast Magnetic Resonance Imaging (MRI) agent, magnetic nano-particle

In ECR ion sources:

- Evaporation of $\text{C}_60$ is solved (500 C)
- Production of fullerene derivatives is possible
- such as $(\text{C}60)+, (\text{C}58)+, (\text{C}56)+,\ldots, (\text{C}60)+++, (\text{C}58)+++, (\text{C}56)+++,\ldots$
- Calculations showed that fullerene derivatives are less stable
- Therefore we must make a two-components plasma
- 1. component: $\text{C}_60$ (mass and charge to be regulated)
- 2. component: \text{Fe: pure iron} „gas“, positive ions with optimal energy
- Synthesis: volume or surface?
Iron plasma in ECR ion sources (pre-experiments)

- Filament ovens: high temperature, large size, impurities
- Sputtering: high microwave power, high voltage,
- Sublimation: only in chemical compounds, e.g. ferrocene Fe(C₅H₅)₂: many non-wanted C, H, CₓHₓ
- External ion source (e.g. MEVVA): complicated to connect (?)
- Induction oven: only iron!
It was decided to build a new ECR ion source at Toyo University, Kawagoe, Japan. To produce new, fullerene-based materials, mainly metal-encapsulated fullerenes. It is called: Bio-Nano-ECRIS

**Collaborators:** Toyo Univ., NIRS, Osaka Univ., Tateyama Co. and ATOMKI

**1st step:** geometry and magnetic trap

Hexapole: modified AECR-U

ATOMKI-ECRIS-B
The Bio-Nano-ECRIS project at Toyo University

- **Geometry:** plasma chamber OD=14 cm, L=35 cm.
- **Microwave:** 8-10 GHz, optional 2.45 GHz 2nd frequency.
- **Mirror field:** 2 identical RT-coils, max. 0.64 Tesla.
- **Hexapole:** NdFeB, modified AECR-U design, 0.72/0.45 Tesla.
- **Fullerene gas:** using simple filament oven or evaporation boat.
- **Iron gas:** by induction oven (under development).
- **Beamline:** AM to transport upto 5 KV beams with M=800.
- **First gas plasma:** 2008 March
- **Fe+C60 mixing:** 2009-2010

Bio-Nano-ECRIS, Toyo University, 2008

Induction oven, under development
2. Bone cell growth on titanium coated with fullerene.

ATOMKI ECRIS Group and Unideb Faculty of Dentistry collaboration

• "Osseointegration": bone formation on Ti implants.
• Efforts: accelerate bone formation, improve lifetime and mechanical stability.
• Nanotechnology: new implant technics.
• Biological usefulness of Ti implants can be improved by (1) surface modification or (2) surface coating.

Our idea: fullerene coating as intermediate layer between metal and organic tissue.
• C60 ions can be shot to the metal surface with any required velocity: balls remain intact or damaged.
• Fullerene molecules are very reactive: hydroxyl and other groups can be connected.
• Such derivatised fullerenes are medically useful.
• Physical part: to coat titanium surfaces by fullerenes with various velocity and thickness.
• Biological tests: growing bone cells on Ti+C60.
• **Task:** irradiate simultaneously 10 Ti samples
• **Size:** 10x10x0.5 mm (d=50 mm beam is necessary)
• **ATOMKI-ECRIS-B configuration**
• **Primary beamline,** 50 cm distance from plasma
• 90% of the beam is single-charged
• 80% of the extracted beam is fullerene
• 90 deg beamline: just composition check
• **U=250 V and U=500 V** extraction voltages
• beam accuracy on 5 segments: less than 10% difference
• total intensity hit the samples: 300-800 enA
• **C60 thickness estimated:** 1 and 5 layers
Summary of the Ti coatings by C60

<table>
<thead>
<tr>
<th>Ti sample series</th>
<th>C$_{60}$ fraction in beam (%)</th>
<th>Beam energy (eV)</th>
<th>Number of C$_{60}$ molecular layers on the Ti surface</th>
<th>Time of irradiation (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>80</td>
<td>500</td>
<td>4.3</td>
<td>90</td>
</tr>
<tr>
<td>2</td>
<td>93</td>
<td>500</td>
<td>1.2</td>
<td>23</td>
</tr>
<tr>
<td>3</td>
<td>74</td>
<td>250</td>
<td>4.9</td>
<td>87</td>
</tr>
<tr>
<td>4</td>
<td>84</td>
<td>250</td>
<td>1.1</td>
<td>32</td>
</tr>
</tbody>
</table>
Human embryonic bone cells were cultured onto the Ti substrates for 48 hours (type: palatal mesenchymal pre-osteoblast, HEPM 1486, ATCC)

- Cells dual labeled with special markers (FITC-falloidin), actine and vinculine
- Confocal imaging: laser scanning microscope (LSM 510, Carl Zeiss).

Bone cells grown on glass

Bone cells grown on Ti+C60 (250 eV)
The morphology of the cells is different compared to the control substrate. The control cells on glass are quite spread showing an interconnected morphology. The cells grown on the Ti substrates are more spindle-like shape showing denser actine and vinculine structure. But there is no remarkable difference between the four Ti+C60 series, so far.

Glass:

Ti+C60:

This first experiment: successful
Proved: C60 coating does not prevent bone cells grows.
High number of bone cells grew on Ti+C60 (~5*10^5/cm3)
Further experiments: optimal beam properties (energy, rate, density, composition)
Goal: improve physical (Ti-C60) and biological (C60-cells) properties
3. Modification of thin films by multiply charged ions

2004-2006: individual slow ions hitting the surface
- Amorphous Se layers, 500 nm, on mica
- Needle-like SbSJ crystals

Xe gas natural
- Charge: 20+, 24+
- Ion flux: 1-100 ion/nm²
- Extraction voltage: U = 4...10 KV
One possible mechanism of nano-hillock formation consists in the process, similar to the radiation-stimulated creation and diffusion of defects to the drains at the surface.
2007-2008: volume effects

Precedents
- Laser induced structural changes
- Ion irradiation induced structural changes ($H^+, D^+$)
- $As_2S_3, AsSe, Se$

- Ions: $Ne^{q+}$ ($q=4...8$)
- $E_{kin} = 120$ keV fixed
- Sample: $AsSe$ layer on glass
- $d=700$ nm
- Penetration depth $R=200$ nm
- Measured parameter: darkening ($T/T_0$) depending on the charge

$$T / T_0 = \exp(-ad) \approx \exp(-a_1 R) \exp(-a(d-R)) \approx \exp(-a_1 R)\text{const}$$
Our experiment showed that $(T/T_0)_{\text{sat}}$ is larger for the sample irradiated with Ne8$^+$ ions than for the one irradiated with Ne4$^+$ ions.

Global optical property can be effected by the charge!
4. Guiding of slow, highly charged ions through nano-capillaries

- Ions with few keV energy have been transmitted through capillaries of thin PET and SiO$_2$ insulating foils.
- There was significant transmission even if the capillaries were tilted by large angles, i.e., when there is no geometrical transparency for straight line trajectories.
- The initial charge state may be or may be not changed.

Prospective applications:
- Ion guiding, directing, focusing slow ion beams in nanoscale devices.
- They might be used for irradiating single cells and writing on charge sensitive surfaces.
2004-2008: ATOMKI-ECRIS-A

- Target: membranes of nanochanneled \( \text{Al}_2\text{O}_3 \), thickness 15 \( \mu \text{m} \), honeycomb
- Capillary diameters: 140-260 nm
- To prevent charging up: 20 nm Nb layers
- Neon (6+, 7+), argon (8+, 9+)
- Extraction: 500 V
- Beam: two 1 mm diaphragms at 205 mm
- Beam current 300-500 pA
- Detectors: FC, ion-spectrometer (channeltron), multi-channel-plate (MCP)
Experimental setup
**Results**

The capillary guiding works!

UV photons were generated in the AM chamber

Deflector + MCP can separate them

Ion CSD after the capilleries

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Deflector plates

- +

Ion beam

\[ \sim 4 \text{ mm} \]

Sample

- 

Photons

\[ \sim 4 \text{ mm} \]

---

Angular distributions at different tilt angles

Target: 290 nm Al₂O₃

- 5°
- 2.5°
- 0°
- -2.5°
- -5°
- Primary Beam

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Ion energy/Charge (keV)

Sample: Al₂O₃, d=140 nm
Tilt angle: 0°

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Normalization counts/s

- 10
- 1
- 0.1
- 0.01
- 1E-3

Observation angle (degree)

- 5°
- 2.5°
- 0°
- -2.5°
- -5°
- Primary Beam

---

Sample: 290 nm Al₂O₃

---

Primary Beam

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Results

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## Comparison and summary

<table>
<thead>
<tr>
<th>Project short name:</th>
<th>Endohedral fullerenes</th>
<th>Ti implants coating</th>
<th>Thin layer modification</th>
<th>Capillary guiding</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Ion source:</strong></td>
<td>ATOMKI-ECRIS-B and Bio-Nano-ECRIS</td>
<td>ATOMKI-ECRIS-B</td>
<td>ATOMKI-ECRIS-A</td>
<td>ATOMKI-ECRIS-A</td>
</tr>
<tr>
<td><strong>Plasma/beam:</strong></td>
<td>Fe⁺, C₆₀⁺</td>
<td>C₆₀⁺</td>
<td>Ne⁴⁺...⁸⁺</td>
<td>Ne⁶⁺,⁷⁺, Ar⁸⁺,⁹⁺</td>
</tr>
<tr>
<td><strong>Beam diameter (mm)</strong></td>
<td>10-20</td>
<td>50</td>
<td>4</td>
<td>1</td>
</tr>
<tr>
<td><strong>Extraction voltage (V)</strong></td>
<td>500-5000</td>
<td>250-500</td>
<td>15000-30000</td>
<td>500-1000</td>
</tr>
<tr>
<td><strong>Microwave frequency (GHz)</strong></td>
<td>8-12</td>
<td>12-14</td>
<td>14.3</td>
<td>14.3</td>
</tr>
<tr>
<td><strong>Microwave power (W)</strong></td>
<td>1-50</td>
<td>4-20</td>
<td>200-400</td>
<td>200-600</td>
</tr>
<tr>
<td><strong>Specification:</strong></td>
<td>Synthesis in plasma or on surface</td>
<td>Irradiation in the zero-degree beamline</td>
<td>Beams with same total energy</td>
<td>Puller on high negative voltage</td>
</tr>
</tbody>
</table>

The tasks and beam requirements were different, but the ECR source proved to be versatile enough to fulfill all these jobs and serves as a very useful multi-purpose facility.
Thank you for your attention!