Gaining novel insight into Fukushima Daiichi Nuclear Power Plant derived fallout.

Peter Martin

Thomas Scott, David Richards & Yosuke Yamashiki
The Fukushima Accident

Crisis at Fukushima nuclear power plant

Tens of thousands evacuated from 20km radius of the plant, people living within a further 10km of the zone urged to stay indoors

Fukushima 1 atomic plant

Source: ANPRO/USGS
Release Monitoring

Source: US DoE / NNSA / MEXT

03/05/2011

Source: US DoE / NNSA / MEXT
Release Monitoring

Table 1. Concentration of fission and neutron capture products (in ppm) in a low burn-up spent nuclear fuel

<table>
<thead>
<tr>
<th>Element</th>
<th>ppm</th>
<th>Element</th>
<th>ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Xe</td>
<td>5656.5</td>
<td>La</td>
<td>1269</td>
</tr>
<tr>
<td>I</td>
<td>258.7</td>
<td>Ce</td>
<td>2469.3</td>
</tr>
<tr>
<td>Cs</td>
<td>2605.3</td>
<td>Pr</td>
<td>1161</td>
</tr>
<tr>
<td>Sr</td>
<td>794.2</td>
<td>Nd</td>
<td>4189.8</td>
</tr>
<tr>
<td>Ba</td>
<td>1740.6</td>
<td>Sm</td>
<td>815.2</td>
</tr>
<tr>
<td>Se</td>
<td>57.7</td>
<td>Eu</td>
<td>154.5</td>
</tr>
<tr>
<td>Te</td>
<td>528.6</td>
<td>Gd</td>
<td>141.5</td>
</tr>
<tr>
<td>Zr</td>
<td>3639.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Mo</td>
<td>3497.5</td>
<td>Np-237</td>
<td>468</td>
</tr>
<tr>
<td>Tc</td>
<td>798.8</td>
<td>Pu (total)</td>
<td>9459</td>
</tr>
<tr>
<td>Ru</td>
<td>2404.9</td>
<td>Am (total)</td>
<td>484</td>
</tr>
<tr>
<td>Rh</td>
<td>483.5</td>
<td>Cm (total)</td>
<td>39</td>
</tr>
<tr>
<td>Pd</td>
<td>1684</td>
<td></td>
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</tr>
<tr>
<td>Ag</td>
<td>91.8</td>
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</tr>
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</table>

*Buck et al 2004*
# Release Analysis To Date

<table>
<thead>
<tr>
<th>Element</th>
<th>Value</th>
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<tbody>
<tr>
<td>Cesium</td>
<td>4865</td>
</tr>
<tr>
<td>Iodine</td>
<td>1308</td>
</tr>
<tr>
<td>Strontium</td>
<td>441</td>
</tr>
<tr>
<td>Xenon</td>
<td>729</td>
</tr>
<tr>
<td>Silver</td>
<td>372</td>
</tr>
<tr>
<td>Technetium</td>
<td>290</td>
</tr>
<tr>
<td>Cerium</td>
<td>164</td>
</tr>
<tr>
<td>Zirconium</td>
<td>128</td>
</tr>
<tr>
<td>Barium</td>
<td>81</td>
</tr>
<tr>
<td>Uranium</td>
<td>122</td>
</tr>
<tr>
<td>Plutonium</td>
<td>145</td>
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</table>

<table>
<thead>
<tr>
<th>Region</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Japan</td>
<td>4147</td>
</tr>
<tr>
<td>Asia (excl. Japan)</td>
<td>640</td>
</tr>
<tr>
<td>Americas</td>
<td>105</td>
</tr>
<tr>
<td>Europe</td>
<td>92</td>
</tr>
<tr>
<td>Africa</td>
<td>36</td>
</tr>
</tbody>
</table>

As of 4th January 2016
Particle Analysis To Date

Emission of spherical cesium-bearing particles from an early stage of the Fukushima nuclear accident

Kouji Adachi, Mizuo Kajino, Yuji Zaizen & Yasuhiro Igashira

Meteorological Research Institute, 1-1 Magono, Tsukuba, Ibaraki, Japan 305-0002.

Detection of Uranium and Chemical State Analysis of Individual Radioactive Microparticles Emitted from the Fukushima Nuclear Accident Using Multiple Synchrotron Radiation X-ray Analyses

Yoshinari Abe,1,6 Yushin Izawa,7 Yasuko Terada,7 Kouji Adachi,8 Yasuhiro Igashira,8 and Izumi Nakai9,7

1Department of Applied Chemistry, Faculty of Science, Tokyo University of Science, 1-3 Kagurazaka, Shinjuku, Tokyo 162-8601, Japan
2Japan Synchrotron Radiation Research Institute (JASRI), SPring-8, 1-1-1 Uehara, Sayo-cho, Sayo-gun, Hyogo 679-5148, Japan
3Meteorological Research Institute, 1-1 Magono, Tsukuba, Ibaraki 305-0002, Japan

Supporting Information

Adachi et al 2011 and Abe et al 2014
Difficulties in “Bulk-Analysis”

Isotope Ratio Analysis of $^{235}$U and $^{238}$U Nuclide
Using a Microwave Digestion Associated with ICP-MS and
the Large Areal Soil Survey Related to Fukushima Daiichi Nuclear Disaster

Yoshitaka Takaya$^1$, Makoto Furukawa$^2$, Yoshitaka Nagahashi$^3$,
Tsuneko Takase$^4$, Osamu Shikino$^5$ and Yutaka Kameo$^6$

$^1$ Cluster of Science and Technology, Fukushima University, 1, Kanayagawa, Fukushima-shi, Fukushima 960-1296
$^2$ PerkinElmer Japan Co., Ltd., 134, Gocho-cho, Hodogaya-ku, Yokohama-shi, Kanagawa 240-0005
$^3$ Nuclear Cycle Backend Directorate, Japan Atomic Energy Agency, 2-4, Shirane, Shirakata, Tokai-mura, Ibaraki 319-1195

(Received 12 September 2011, Accepted 19 October 2011)
Fukushima Regional Sampling: May 2014 & Oct 2015

- 1 - Iitate
- 12 - Emataira
- 14 - Katsuro
- 19 - Minamisoma
  Exclusion Zone (20 km)
- 25 - Okuma
- FDNPP

20 km
Particle Capture
Particle Identification

34 – 62 wt% Uranium
Particle Extraction
Particle Extraction

Spectrochimica Acta Part B: Atomic Spectroscopy
Available online 31 December 2015
In Press, Accepted Manuscript — Note to users

In-situ removal and characterisation of uranium-containing particles from sediments surrounding the Fukushima Daiichi Nuclear Power Plant

P.G. Martin\textsuperscript{a} \textsuperscript{*}, I. Griffiths\textsuperscript{b}, C.P. Jones\textsuperscript{a}, C.A. Stitt\textsuperscript{a}, M. Davies-Milner\textsuperscript{c}, J.F.W. Moolmans\textsuperscript{d}, Y. Yamashikii\textsuperscript{f}, D.A. Richards\textsuperscript{i}, T.B. Scott\textsuperscript{a}
SR-μ-XANES

Strong compositional similarity to the centres of micron-scale spherical particles analysed by Abe et al. 2014
Particle Sectioning: Slice and View & TEM
Additional Fallout Material

- **Ag**
- **Cd**
- **Ce & Pr**
- **Ba**

Also:
- Pb
- Zr
- I
- Bi
- Au
- Nd
- Nb
- La
- Y
- S. Steel

500 nm
Classifying & Quantifying Material

- ACICULAR
- ANGULAR
- CUBIC
- CYLINDRICAL
- ROUNDED
- FLAKEY
- SPHERICAL
- SPONGY
Classifying & Quantifying Material

*Uranium Particles – All localities*

- Irregular rounded: 19.18%
- Angular Fragment: 20.55%
- Cubic: 23.29%
- Quadrilateral: 4.11%
- Diamond: 6.85%
- Rod: 5.48%
- Rounded: 10.96%
- Elliptical: 1.37%
- Sub Rounded: 8.22%
Classifying & Quantifying Material

Uranium Particles

![Graph showing volume (μm³) against distance (11.45 km, 40.05 km, 65.67 km)]

- Max
- 75%
- 50%
- 25%
- Min
Classifying & Quantifying Material

**Iitate Village**

![Graph showing volume (µm³) for different materials: Uranium, Lead, Lanthinides, Tin. The legend shows Uranium in white, Lead in red, Lanthinides in blue, and Tin in pink.](image-url)
Classifying & Quantifying Material

_Lanthanide Particles_

- Iitate
- Minamisoma
- Emataira
- Yamakiya Junior High School
U Particle Solubility

<table>
<thead>
<tr>
<th>Species</th>
<th>Result</th>
</tr>
</thead>
<tbody>
<tr>
<td>UK Rain Water</td>
<td>✗</td>
</tr>
<tr>
<td>Simulated Fukushima Groundwater</td>
<td>✗</td>
</tr>
<tr>
<td>Abukama River Water</td>
<td>✗</td>
</tr>
<tr>
<td>DI Water</td>
<td>✗</td>
</tr>
<tr>
<td>10% HNO₃</td>
<td>✓</td>
</tr>
</tbody>
</table>

- Particles adhered to borosilicate glass capillaries.
- SEMGlu used as non-reactive when “cured”.
  - 4 week duration.
- Examination: prior to commencement, weekly and at the conclusion of the study.
- Natural fluids failed to dissolve material, supporting uranium existing as insoluble UO₂ (+4) state.
Future Work

- Further dissolution experiments
- DLS I14 Nano-focus XAS beamline
  - LA-ICP-MS
  - Transport modelling
- Analysis of further particles, from additional localities
  - Fingerprint individual plume contributors
Thanks for listening

Peter G. Martin

Thomas B. Scott, David A. Richards & Yosuke Yamashiki