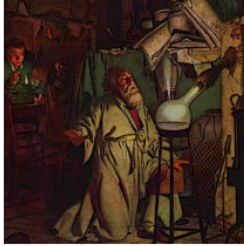




## Geochemical News #132 - July 2007



from *The Alchemist in Search of the...*

### In This Issue

[From the GS President \(#132\)](#)  
by Sue Brantley

[Computed Tomography Applied to Geosciences](#)  
by Allen H. Reed and Yoko Furukawa

[Disposal of Nuclear Waste: Innovative Fuels for Actinide Transmutation](#)  
by Geert-Jan L.M. de Haas, Frodo C. Klaassen and Ronald P.C. Schram

[Award News \(#132\)](#) Holly Stein receives Helmholtz-Humboldt Award

[Nick's Picks \(#132\)](#) Geo-relevant podcasts  
by Nicholas Wigginton

### Editors

Johnson R. Haas, Ph.D.  
Western Michigan University  
[johnson.haas@wmich.edu](mailto:johnson.haas@wmich.edu)

Carla Koretsky, Ph.D.  
Western Michigan University  
[carla.koretsky@wmich.edu](mailto:carla.koretsky@wmich.edu)

### Associate Editors

Thilo Behrends, Ph.D.  
Utrecht University  
[behrends@geo.uu.nl](mailto:behrends@geo.uu.nl)

Yoko Furukawa, Ph.D.  
Naval Research Laboratory  
[yoko.furukawa@nrlssc.navy.mil](mailto:yoko.furukawa@nrlssc.navy.mil)

Mitch Schulte, Ph.D.  
University of Missouri  
[schultemd@missouri.edu](mailto:schultemd@missouri.edu)

Angie Souren, M.Sc.  
SmarterScience  
[angelinasouren@smarterscience.com](mailto:angelinasouren@smarterscience.com)

Nicholas Wigginton, Ph.D. Candidate  
Virginia Tech  
[wigginto@vt.edu](mailto:wigginto@vt.edu)

Nathan Yee, Ph.D.  
Rutgers - The State University of New Jersey  
[nyee@envsci.rutgers.edu](mailto:nyee@envsci.rutgers.edu)

[Join or Renew](#)  
[Facebook](#)

[Geochemical News](#)  
[Elements Magazine](#)  
[Geochimica et Cosmochimica Acta](#)

[Goldschmidt Conference](#)  
[Follow GS on Twitter](#)



[home](#) > [publications](#) > [geochemical news](#) > [gn132 \(jul07\)](#) > [from the gs president \(#132\)](#)

## From the GS President (#132)



Prof. Susan L. Brantley

As I write this, I am looking forward to the Cologne Goldschmidt conference where more than 2000 geochemists will gather. These conferences are organized by the Geochemical Society when they are in North America and by the European Association of Geochemistry when they are in Europe.

Currently, we are still seeking draft proposals to host the 2010 and 2012 conferences in North America. If you have ever run a large meeting, you know that reserving conference and hotel facilities well in advance in North America is a requirement. For a number of months we have been soliciting self-nominations for conference teams and venues. We have one absolutely excellent proposal for 2010 but we would love to have at least one more for backup or for 2012.

Why aren't we seeing more volunteers to run the Goldschmidt? Of course, our meetings have become very large and this represents a significant responsibility and privilege for organizers. Even though Seth Davis, our GS business manager, is now in charge of helping to coordinate with the conference organizers, the job of organizing is still huge. I personally believe that the GS may need to retain a professional conference organizer to facilitate these meetings. However, the last time I floated this idea at a GS Executive meeting, it was not positively received. Several geochemists argued that part of the charm of the Goldschmidt meeting is that it changes 'flavor' every time.

Another option would be to hold the Goldschmidt meeting at the same venue every other year in the U.S. For example, given the extremely large numbers at the European Goldschmidt meetings, the EAG is discussing whether the Goldschmidt in Europe should always return to Davos Switzerland (the proposed site for 2009). Obviously, returning to a site every year would make organizational tasks easier. I would like to hear from the membership: *Would you like to see the Goldschmidt meeting held in the same place every other year in North America, and if so, where would you like it to be held? If you do not like the idea of holding the Goldschmidt in the same venue every other year in North America, then are you in favor of retention of a professional organizer by the GS?*

I would like to end on a personal note. I would like to express my personal sadness about the loss of Professor Hal Helgeson. Hal died of cancer on May 28, 2007 in Alta Bates Hospital, Berkeley, California. Born in 1931, he was internationally known for his insights and contributions with respect to all aspects of theoretical geochemistry from high- to low-temperature. The mark he has left on geochemistry is profound, ranging from significant theoretical advances, to his many excellent students, to the oft-told Helgeson stories. He will be missed greatly by all but especially by the many 'Friends of Prediction Central'. Hal served as a director of the Geochemical Society from 1973-1976. He also received the prestigious Goldschmidt medal from the Geochemical Society in 1988. It was well deserved and his dynamic presence will be well missed.

Susan L. Brantley, President of the Geochemical Society  
Earth and Environmental Sciences Institute  
Pennsylvania State University  
University Park, PA 16802  
[brantley@eesi.psu.edu](mailto:brantley@eesi.psu.edu)

## GN132 Content

[From the GS President \(#132\)](#)  
by Sue Brantley

[Computed Tomography Applied to Geosciences](#)  
by Allen H. Reed and Yoko Furukawa

[Disposal of Nuclear Waste: Innovative Fuels for Actinide Transmutation](#)  
by Geert-Jan L.M. de Haas, Frodo C. Klaassen and Ronald P.C. Schram

[Award News \(#132\)](#) Holly Stein receives Helmholtz-Humboldt Award

[Nick's Picks \(#132\)](#) Geo-relevant podcasts  
by Nicholas Wigginton

[Join or Renew](#)

[Facebook](#)

[Geochemical News](#)

[Elements Magazine](#)

[Geochimica et Cosmochimica Acta](#)

[Goldschmidt Conference](#)

[Follow GS on Twitter](#)



## Computed Tomography Applied to Geosciences

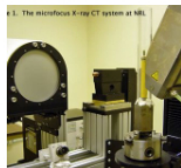


Figure 1

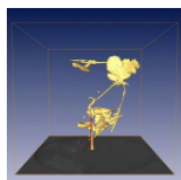


Figure 2

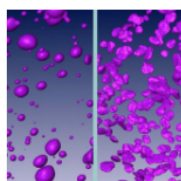


Figure 3

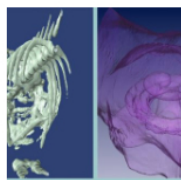


Figure 4

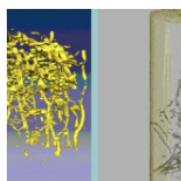


Figure 5

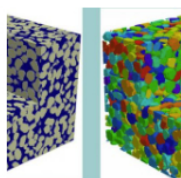


Figure 6

by Allen H. Reed and Yoko Furukawa

Computed tomography (CT) provides an exciting capability to geosciences by capturing the geometry of objects bounded by optically opaque materials. The geometric information provides guidance, constraints and boundaries within which to model processes, systems, transitions, and changes. The development of CT has progressed dramatically in the 30 years since it was first developed. In the beginning, CT was slow and cumbersome, but it demonstrated a clear ability to resolve a material embedded within another material. Hounsfield (1973) performed the first scans, one of which was of a pig's pancreas. The scan took the entire day and by the end of the day gas bubbles had developed as artifacts within the decaying tissues, yet the ability to resolve the pancreas in 3D had been achieved. Soon afterwards, the medical profession adapted this technology and pioneered many changes to increase speed and utility and to enhance image quality. Seeing an opportunity to advance understanding in fluid mechanics, groups at Shell and Chevron applied medical CT systems to evaluate multiphase flow in core samples (2-3). Currently, medical scanners have improved image-resolution (to ~250-400  $\mu\text{m}$  from 1-2  $\mu\text{m}$ ; Note: image resolution is equivalent to the size of the voxels, a 3D pixels). As industry and security needs have emerged, high-resolution industrial scanners (to <10  $\mu\text{m}$  resolution) have enabled scanning of smaller objects, such as pores and grains in a sediment or rock assemblage and the fluid and gas phases trapped in the pore space. With this advance, the ability to visualize and research multiphase fluid flow, grain interactions, pore clogging, gas formation, and a host of other geological processes is possible.

Computed tomography enables volumetric imaging of objects by emitting high-energy photons, (i.e., gamma rays or x-rays) through an object. These photons, produced either by gamma emitters, such as Ce-137 or by conventional electrical sources, are emitted from an x-ray tube through the sample material and to an x-ray detector. Once the x-rays reach the detector they are converted, through a series of processes, to an image. In the case of conventional systems, x-rays are produced electronically by illuminating a cathode to produce photons within an x-ray tube. The photons are accelerated through a magnetic field to an anode which produces x-rays when contacted by the photons. X-rays that pass through the focal spot, adjacent to the anode and at the front of the X-ray tube, are projected towards the sample material. The X-rays that pass through the sample and reach the X-ray detector, the image intensifier, are converted to an electric signal. This signal is then converted to an array of gridded gray-scale values or the voxels that comprise the image (2).

The CT system at the Naval Research Laboratory has a stationary source (x-ray tube) and receiver (image intensifier) and the sample is rotated and translated between the source and receiver. The distance between the source and receiver can be adjusted to accommodate large diameter samples (up to 11 cm) and small diameter samples (~3-5  $\mu\text{m}$ ). Sample diameter, as well as density and atomic number, play the key roles in determining sample resolution. See references 2 and 3 for more information on the specific determinants of x-ray attenuation and image resolution.

Samples scanned at NRL range from 6  $\mu\text{m}$  to 11 cm and it is common at NRL to scan small cores of sand to capture the pore- and grain-scale geometry and mud cores to capture features (e.g., gas bubbles, burrows) that persist in these cores. The small diameter sand cores, (6 to 8- $\mu\text{m}$ ) are imaged with 10  $\mu\text{m}$  resolution. The objects in mud cores, 6 to 11 cm diameter) are often larger. The 6-cm diameter cores of mud provide resolution of 120  $\mu\text{m}$ .

The capabilities of the microfocus x-ray CT system to resolve a variety of materials is presented in a series of figures and movies. The examples displayed below pertain to research in the geosciences, including geochemistry, marine geology, and geomechanics; they demonstrate the capabilities of this system. Specifically, these examples assist in the evaluation of structural changes in mud due to biogeochemical alteration of organic matter or gas bubble production. It turns out that assigning this problem to one sector of the geosciences would be too limited. As research in bubble production demonstrates, the mechanics of bubble growth differs in natural sediments from reconstituted sediments due to the difference in mechanical characteristics between natural and reconstituted mud (Boudreau et al., 2005; Figure 2, Figure 3; Movie 1: [Gas bubbles in natural sediments](#) (38.9MB MPEG Movie)).

Geosciences may require resolution of fine-scale geometrical details. The ability to resolve fine-scale details of small objects is demonstrated in the partial skeleton of the Mosquito fish and the inner ear of an extinct dolphin (Figure 4). The fish skeleton, although slightly off the topic, puts the resolution capabilities of NRL CT into perspective that may be easier to grasp. The image of the fish, a 1.5-cm long section scanned that was 0.5 cm thick, displays the ability to resolve small-scale features.

An important factor determining oxidation of organic matter and mineralogical changes in buried sediments is bottom water oxygen. In sediments, infauna rework sediments and create burrows, which serve as conduits for air in terrestrial environments or for oxygen rich bottom waters in aqueous environments. Burrow geometry is presented in bioturbated sediments (Briggs et al., 2005; Figure 5; Movie 2: [Bioturbation Example 1](#) (4.6MB MPEG Movie) and Movie 3: [Bioturbation Example 2](#) (4.3MB MPEG Movie)).

### GN132 Content

[From the GS President \(#132\)](#)

by Sue Brantley

[Computed Tomography Applied to Geosciences](#)

by Allen H. Reed and Yoko Furukawa

[Disposal of Nuclear Waste: Innovative Fuels for Actinide Transmutation](#)  
by Geert-Jan L.M. de Haas, Frodo C. Klaassen and Ronald P.C. Schram

[Award News \(#132\)](#) Holly Stein receives Helmholtz-Humboldt Award

[Nick's Picks \(#132\)](#) Geo-relevant podcasts  
by Nicholas Wigginton

Movie)) from which one can determine the magnitude of oxygen penetration.

The composition and interrelationship of sand grains in many settings determines fluid flow, diffusion, dispersion and chemical precipitation. Modeling these processes in marine systems, aquifers, and the vadose zone has been facilitated by the ability to resolve sand grains in high detail (Thompson et al., 2006; Figure 6; Movie 4: [Microfossils](#) (34.9MB MPEG Movie)).

As demonstrated in these examples, resolving the geometry of objects provides means to analyze geological problems, geochemical processes and geomechanical constraints. Numerous other objects and possibilities for research exist. It is our hope that this presentation will entice your imagination, foster new ideas and promote research options.

The wide array of materials that have been imaged at NRL demonstrate great potential for evaluating problems that exist within three-dimensional geometrical boundaries. These geometries provide the boundaries and constraints within which chemical and physicochemical processes can be analyzed. In addition to static three-dimensional problems we are currently evaluating dynamic processes in four-dimensions by including a time-component. These evaluations include rates of bioturbation, chemical transfer across burrow walls, single and multi-phase fluid flow, and bubble formation and ebullition; many additional possibilities exist.

Interested parties may contact the authors for further information on collaboration, research ideas and projects, and access to the CT facility. Post-doctoral candidates are sought to perform research on related projects.

Movies:

Movie 1: [Gas bubbles in natural sediments](#). Gas bubbles in natural sediments collected from the same area as the sample in figure 3.

Movie 2: [Bioturbation Example 1](#) and Movie 3: [Bioturbation Example 2](#). These movies depict sandy sediment systems which have inclusions of shells (white) and burrows (brown). The sand has been made transparent. Cores are 6-cm in diameter. See Briggs et al. (2006) for more information.

Movie 4: [Microfossils](#). An assemblage of sand-sized microfossils from Mihlos, Crete is presented in a slice progression movie. The sand sized materials have a median grain size of ~470 micrometers. The image resolution is 11 um.

Allen H. Reed  
Marine Geosciences Division  
Naval Research Laboratory  
Stennis Space Center, MS 39529  
Phone: 228-688-5473  
Email: [allen.reed@nrlssc.navy.mil](mailto:allen.reed@nrlssc.navy.mil)

We are grateful for the support of NRL, which provided the funds under a CPP to acquire the CT and to Drs. Eric Hartwig, Herbert Eppert, Philip Valent, Michael Richardson, and Kevin Briggs. Funding for these projects was provided by provided by NRL core funds and ONR's offices of Coastal Geosciences, Acoustics, and Optics. Periotic membrane provided courtesy of Calvert Marine Museum, Solomons MD. We thank Kevin Wilson, an undergraduate student, for producing the movies, Karsten Thompson for sand images and Chris Algar/Mark Barry for image of injected bubble.

#### Literature Cited

1. Hounsfield G. N., 'Computerized transverse axial scanning (tomography). 1. Description of system', Br. J. radiology, 46(552) 1016-1022, 1973.
2. Wellington, S.L., Vinegar, H.J., X-ray computerized tomography. Journal of Petroleum Technology 39 (8), 885-898, 1987.
3. Flannery, B.P., Deckman, H.W., Roberge, W.G., D'Amico, K.L., 'Three-dimensional X-ray microtomography', Science 237, 1439-1444, 1987.
4. Boudreau B. P. C., Algar C, B. D. Johnson , I. Croudace , A. H. Reed, Furukawa Y, Dorgan KM, Jumars PA, Grader AS, Gardiner BS, 'Bubble growth and rise in soft sediments,' Geology (6), 517-520, 2005.
5. Briggs, K. B., A. H. Reed, and M. D. Richardson, 'Using CT to image storm-generated stratigraphy in sandy sediment off Fort Walton Beach, Florida, USA', Proceedings of the International Conference 'Underwater Acoustic Measurements: Technologies & Results,' Crete, Greece, 2005.
6. Thompson, K. E., C.S. Willson, and W. Zhang, 'Quantitative computer reconstruction of particulate materials from microtomography images,' Powder Technology 163, 169-182, 2006.
7. Reed, A. H., K. E. Thompson, K. B. Briggs, and M. D. Richardson, 'Quantification of pore and grain properties of quartz sands from microfocus computed tomography images,' in prep for IEEE-Oceanic Engineering 2007.

Join or Renew  
Facebook

Geochemical News  
Elements Magazine  
Geochimica et Cosmochimica Acta

Goldschmidt Conference  
Follow GS on Twitter



## Disposal of Nuclear Waste

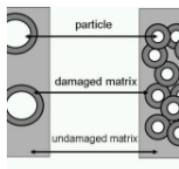


Figure 1



[Geert-Jan L.M. de Haas](#), Frodo C. Klaassen and Ronald P.C. Schram  
*Nuclear Research and consultancy Group (NRG), P.O. Box 25, 1755 ZG Petten, The Netherlands*

### Introduction

Discussions about the reduction of greenhouse-gas emissions have revitalized the discussion about the future role of nuclear power production. Uncertainties about the availability of conventional energy resources like oil and gas have also contributed to a renaissance of nuclear energy. Developments in the Middle East and, only recently, the conflict between Russia and Belarus and Ukraine have once again revealed the vulnerability of the industrialized world when it comes to a secure supply of oil and gas. In addition, the rise of new economic powers like China and India has led to increasing demands on the world markets for raw materials including oil and gas. The prospect of rapidly growing energy consumption rates in the next decades has fueled the debate about the role of other energy resources, including nuclear energy, in order to meet increasing demands.

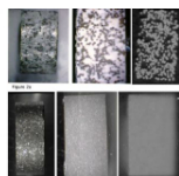


Figure 2a & 2b

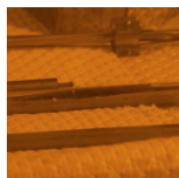


Figure 3

Over the last four decades nuclear power production has been proven to be a reliable technique, nowadays covering about 35% of the energy needs in the European Union and 17% of the needs world-wide. In contrast to oil and gas, a significant part of today's proven uranium resources are located in politically stable countries; about 50% of the uranium is currently produced in Canada (29%) and Australia (21%). These two factors and commitments to comply to the Kyoto protocol to reduce CO<sub>2</sub> emissions have resulted in the return of nuclear energy on the political agenda. The governments of France and Finland have already approved expansion of their nuclear infrastructure.

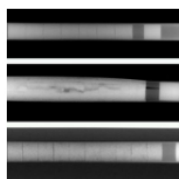


Figure 4

At the same time it is widely recognized that enlargement of the role of nuclear energy is directly related to the issue of nuclear waste. Enlargement of the nuclear energy's share in the world's energy portfolio requires a socially acceptable solution.

The waste consists of exhausted (spent) fuel rods, which remain radiotoxic for about 130,000 years due to the presence of long-lived isotopes of plutonium and minor actinides like neptunium (Np) and americium (Am), formed during reactor operation by neutron capture, and long-lived fission products like Cs-137, Tc-99 and I-129. For a detailed review on the composition of spent fuel the reader is referred to Bruno and Ewing (2006) in last year's issue of Elements on the nuclear fuel cycle.

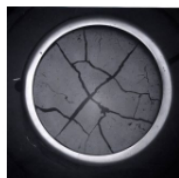


Figure 5

Over the last decades new concepts and initiatives for a more sustainable nuclear fuel cycle with emphasis on the management of radioactive waste have been launched. Storage of conditioned spent fuel, or high-level waste arising from reprocessing of spent fuel to recover uranium and plutonium, in underground repositories equipped with engineered barriers is one of the scenarios envisaged for future radioactive waste management. In several countries underground research facilities have become available (e.g. Belgium) or are currently under construction (e.g. Sweden).

Partitioning and transmutation (P&T) is another scenario under consideration. This scenario envisages reduction of the long-term environmental impact of radioactive waste by separation (partitioning) of the most radiotoxic or most long-lived components from the waste and re-irradiating them with neutrons, thereby converting the long-lived isotopes into more short-lived or stable isotopes. Realistic P&T scenarios indicate that the reduction in radiotoxicity equivalent to a period of 150,000 year of natural decay can be reached after 500 to 3000 years (Magill et al. 2003).

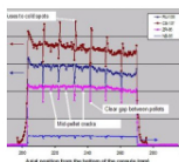


Figure 6

It is important to realize that the two scenarios do not only differ from a technical point of view. The final disposal route is an example of open cycle: the fuel is utilized once and then discarded as waste. The P&T route, in contrast, acknowledges the energy potential of irradiated fuel and processes are developed to exploit this potential: a closed cycle. The 'life cycle' of plutonium is a good example of the latter route.

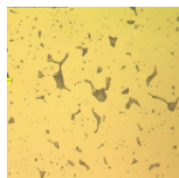


Figure 7

The bulk of the radiotoxic inventory of the waste, 90%, is made up by plutonium isotopes. The present global inventory of plutonium is over 1,400 metric tons next to about 250 metric tons of weapon-grade plutonium (Ewing 2004). Plutonium is therefore an obvious candidate for P&T studies. About 65% of the plutonium consists of fissile Pu-239 (half life 24,100 yr) and Pu-241 (half life 14,4 yr) (Gruppelaar et al. 1998). Part of the reprocessed, fissile plutonium is currently mixed with uranium oxide and used as MOX (Mixed OXide) fuel in some commercial nuclear power plants. During irradiation in existing light water reactors a maximum of about 50% MOX is applicable. During irradiation plutonium is fissioned (burnt) but at the same time new plutonium is generated by neutron capture in the non-fissile U-238. As a consequence, a net reduction of the worldwide plutonium stockpiles along this line is not feasible. New reactor designs are needed in order to increase the maximum loading of MOX.

Alternatively, effective reduction of the plutonium stockpiles may be achieved by using Pu-bearing fuels, which do not produce new waste. Over the last decade research has focussed on the incineration of plutonium - and

### GN132 Content

[From the GS President \(#132\)](#)  
by Sue Brantley

[Computed Tomography Applied to Geosciences](#)  
by Allen H. Reed and Yoko Furukawa

[Disposal of Nuclear Waste: Innovative Fuels for Actinide Transmutation](#)  
by Geert-Jan L.M. de Haas, Frodo C. Klaassen and Ronald P.C. Schram

[Award News \(#132\)](#) Holly Stein receives Helmholtz-Humboldt Award

[Nick's Picks \(#132\)](#) Geo-relevant podcasts  
by Nicholas Wigginton

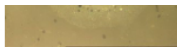


Figure 8

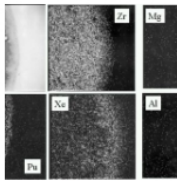



Figure 9

other actinides as well - in uranium-free matrices, more generally also referred to as IMF, inert matrix fuels. IMF are composed of a fissile-bearing phase, containing plutonium or a minor actinide like americium, embedded in an inert matrix, i.e. a matrix which does not interact with incident neutrons. The matrices may have a ceramic (e.g. Neeft et al. 2003; Schram et al. 2003) or a metallic composition (e.g. Fernandez et al. 2003). The results of recent irradiation experiments have demonstrated the potential of the concept of IMF (minor actinide incineration (Konings et al. 2000; Schram et al. 2003). In the next sections an outline on the concept of the IMF is presented, illustrated with the results of a successful irradiation experiment in the High Flux Reactor (HFR) in Petten, The Netherlands.


### Composition and fabrication of IMF


Important parameters in studies into IMF fabrication are the chemical composition of the matrix and the distribution of the fissile phase in the matrix. The chemical composition of the matrix is confined to materials (elements) that do not show (significant) interaction with neutrons during irradiation as activation of the matrix may lead to generation of new, extra radioactive waste and/or disturb the neutron balance required to generate a sustainable fission reaction. Examples of inert elements are silicon, aluminium, magnesium and zirconium. In addition, the inert matrix material should be resistant against the different types of radiation that may occur, and be able to accommodate fission products, amongst which are krypton and xenon, and, in some cases, large amounts of helium gas. Moreover, the material should have a sufficiently high melting point and thermal conductivity, and suitable elastic constants to provide mechanical stability. Studied matrix materials are MgO, spinel ( $MgAl_2O_4$ ),  $Al_2O_3$ , and  $ZrO_2$  (Shiratori et al. 1999; Klaassen et al. 2003; Neeft et al. 2003; Schram et al. 2003).


An important issue concerns the dispersion of the fissile material in the matrix material. A distinction can be made between composite and homogeneous types of IMF. The homogeneous type of IMF is a solid solution of the matrix and the fissile phase. A prominent example is the zirconia-based ( $ZrO_2$ ) fuel which forms a solid solution with  $PuO_{2-x}$ ;  $(Zr, Pu)O_{2-x}$ . Composite IMF consists of an inert matrix which contains a dispersion of either micro- (up to several tenths of a micrometer) or macro-sized (several hundred micrometers) particles of the fissile phase. The size of the particles exerts a significant influence on the behaviour of the fuel during, and after, irradiation. The production of fission products, alpha particles, recoil atoms and neutrons has a profound impact on the periphery of the individual particles. In micro-dispersive systems the damage will be more homogeneously distributed over the fuel (Fig. 1). On the other hand, damaged zones will overlap and result in more swelling than in a macro-dispersive IMF as recently observed (Schram et al. 2003). Clearly, excessive swelling is an important, safety-related, issue.


 HKS\_fig1\_sm.jpg: Figure 1  
Figure 1


Several routes for the fabrication of the fissile-phase bearing phase, and the IMF, are available. An important consideration concerns the amount of radioactive waste and dust generated during the fabrication process. This is especially important from a radiological point of view. Any handling of radioactive material is based upon the ALARA principle: As Low As Reasonably Achievable. Impregnation of porous yttria-stabilized zirconia sol-gel beads with an actinide (Ac)-bearing nitrate solution is a favoured route as the amount of radioactive waste and dust is minimized (Somers and Fernandez 2005). The impregnated beads are subsequently calcined during which the Ac-nitrate is converted into an oxide, forming a  $(Ac, Zr, Y)_2O_x$  solid solution.


 HKS\_fig2\_sm.jpg: Figure 2a & 2b  
Figure 2a & 2b


 HKS\_fig3\_sm.jpg: Figure 3  
Figure 3


 HKS\_fig4\_sm.jpg: Figure 4  
Figure 4

 HKS\_fig5\_sm.jpg: Figure 5  
Figure 5

 HKS\_fig6\_sm.jpg: Figure 6  
Figure 6

 HKS\_fig7\_sm.jpg: Figure 7  
Figure 7

 HKS\_fig8\_sm.jpg: Figure 8  
Figure 8

 HKS\_fig9\_sm.jpg: Figure 9  
Figure 9

During fabrication of the IMF samples (pellets) the fissile-bearing phase and the inert matrix are thoroughly mixed in the desired proportions and pressed to pellets. Pressures applied generally range between 500 and 600 MPa. The pressed pellets are then calcined at temperatures between 1400 and

1700°C, depending on the type of material selected, to achieve their final density. Typically, the calcined pellets have a diameter between 6 and 8 mm. A high density, i.e. between 90 and 95% of the theoretical density, is required to guarantee sufficient high transfer of the heat generated during irradiation. By far most of the ceramic inert matrices have a poor thermal conductivity properties which may result in temperatures well over 1500°C for  $ZrO_2$ -based IMF. Too high temperatures may result in melting and other unwanted interactions with the cladding material in which IMF pellets are stacked.

### The OTTO experiment

The OTTO experiment has been a joint research project between NRG (The Netherlands), PSI (Switzerland) and JAERI (nowadays JAEA, Japan) on the feasibility of plutonium transmutation using inert matrices. OTTO is an acronym for Once Through Then Out. The OTTO concept envisages burning of plutonium in IMF, designed such that the irradiated pellets are suitable for final storage, i.e. without additional post-treatment. For this purpose two geo-chemically stable matrices were selected: zirconia and spinel. The zirconia-based pellets were prepared from crushed  $(Zr, Y, Pu, U)O_2$  particles. The spinel-based pellets were prepared by mixing spinel powder with 20 vol.% micro (< 25 µm) and macro-sized (200-250 µm) spheres of  $(Zr, Y, Pu, U)O_2$ . Examination of the surface and the interior of the macro-dispersive type spinel made clear that homogeneous distribution of the spheres was only partly achieved (Fig. 2a), despite the fact that the spheres were mixed with a slurry of spinel (Schram et al. 2003). In contrast the micro-dispersive type pellets showed a massive, homogeneous appearance (Fig. 2b).

The samples were irradiated for 548 full power days in the 45 MW HFR in Petten, the Netherlands during which 30-35% of the plutonium was burnt. After irradiation part of the pellets was closely inspected. During this so-called post-irradiation examination (PIE) the physical and chemical properties of the irradiated pellets are recorded in order to assess their performance during irradiation. Non-destructive techniques include visual inspection, gamma spectrometry, X-ray imaging, tomography and fission gas analysis. Destructive PIE focuses on the microscale properties of the pellets using microscopy, SEM and microprobe analysis. It is good to recall that the samples are highly radioactive. The examinations, including the preparations of the samples, are therefore performed in cells shielded with concrete and lead (hot cells). For handling of the samples and the instruments use is made of manipulators.

Upon inspection of the capsules the cladding of capsule with the micro-dispersive type spinel pellets appeared to be damaged (Fig. 3). Failure of the thermocouple during irradiation had already hinted to (mechanical) problems (note that during irradiation the capsules were stacked in a closed containment so that failure did not result in dangerous situation). Failure of the pellets is clearly visible on X-ray images of the capsules (Fig. 4). The zirconia-based pellets and the spinel pellets with the macro-dispersion on the other hand have remained intact.

Microscopy provides a more detailed, external picture of the thermal cracking of the zirconia-based pellets (Fig. 5). These cracks are typically the result of the high thermal gradients in zirconia, which is a poor conductor. The cracking pattern resembles that of irradiated uranium pellets in common light water reactor fuel. Like zirconia uranium has a low thermal conductivity.

The effects of high temperatures in the core and the dissipation of the heat to the margins of the pellets, where temperatures are lower, is also reflected in the distribution of Cs-137 as recorded by gamma spectrometry. By moving the detector stepwise along the cladding an axial profile of the distribution of several fission products in the pellets, like Cs-137 and Ru-106 is obtained. The axial profile of the capsule with the zirconia-based pellets (Fig. 6) clearly reveals enhanced Cs-137 concentrations at the outer margins of the individual pellets as a result of diffusion away from the core. Less volatile fission products like Ru, Zr and Nb do not show such behaviour.

SEM studies revealed more detailed information with regard to the performance and behaviour of the different types of pellets. The irradiated zirconia-based pellets appeared to have retained a very homogeneous, dense structure. Fine-grained metallic fission products like palladium and ruthenium are evenly distributed over the pellet (Fig. 7). In the case of the macro-dispersive type spinel pellets most of the metallic fission products are concentrated inside the particles (Fig. 8). In contrast to the zirconia-based pellets inclusions of fission gasses (Kr, Xe) are clearly visible. Part of the gasses have apparently diffused outwards, the inclusions neatly aligned along the outline of the spheres. In addition an approximately 10 µm thick halo around the sphere has formed. In order to obtain more information about the composition of the halo microprobe analyses of a macro-sphere/halo/matrix section were executed. (Fig. 9). Plutonium shows a clear sharp transition, accentuating the outline of the macro-sphere. Apparently, plutonium does not show any redistribution: it remains within the pellet during irradiation. Fission product concentrations, on the other hand, are clearly enhanced in the immediate surroundings of the sphere, i.e. the halo. The fission products are injected into the matrix over a distance of 10 µm, which is the typical range of high energy (~70 - 90 MeV) fission products in matrix material.

## Future work

As mentioned before the basic thought behind the OTTO concept is the transmutation of plutonium using IMF suitable for direct disposal after irradiation. After having assessed the performance of the various types of pellets during irradiation - and the preliminary conclusion that the microdispersive type spinel IMF is less suitable than a macrodispersive type spinel IMF or homogeneous type zirconia-based IMF - the various pellets will therefore be subjected to a leaching test in NRG's hotcell laboratories. Over the last few years a leaching facility has been designed and constructed. In this facility the pellets will be leached for 6 months using distilled water as leachant at a temperature of 90 °C. The start of the leaching experiment is foreseen for the end of 2007. After completion the leached pellets will be characterized studied by light and scanning electron microscopy; the leachates will be analysed by ICPMS at PSI (Switzerland). The results of the leaching test are expected to provide crucial information about the feasibility of the OTTO concept and the properties of irradiated IMF in general.

## References

- Bruno J. and Ewing R.C., 2006. Spent nuclear fuel. *Elements* 2, 343-349.
- Ewing R.C., 2004. The environmental impact of the nuclear fuel cycle: climate change, nuclear waste and nuclear weapons. *Geochim. Cosmochim. Acta*, 2004, 68 (Supplement 1), A13.
- Fernandez A., Konings R.J.M. and Somers J., 2003. Design and fabrication of specific ceramic-metallic fuels and targets, *J. Nucl. Mat.* 319, 44-50.
- Gruppelaar H., Kloosterman J.L. and Konings R.J.M., 1998. Advanced technologies for the reduction of nuclear waste. Netherlands Energy Research Foundation, ECN report R-98-008.
- Klaassen F.C., Bakker K., Schram R.P.C., Klein-Meulenkamp R., Conrad R. Somers, J. and Konings, R.J.M., 2003. Post irradiation examination of irradiated americium oxide and uranium oxide in magnesium aluminate spinel. *J. Nucl. Mat.* 319, 108-117.
- Konings R.J.M., Conrad R., Dassel G., Pijlgroms B.J., Somers J. and Toscano E., 2000. The EFTTRA-T4 experiment on americium transmutation. *J. Nucl. Mat.* 282, 159-170.
- Magill J., Berthou V., Haas D., Galy, J., Schenkel R., Wiese H.-W., Heusener G., Tommasi J. and Youinou G., 2003. Impact limits of partitioning and transmutation scenarios on radiotoxicity of actinides in radioactive waste. *Nucl. Energy* 42, 1-15.

Neeft E.A.C., Bakker K., Belvroy R.L. Tams W.J., Schram, R.P.C., Conrad R. and van Veen, A., 2003. Mechanical behaviour of macro-dispersed inert matrix fuels. *J. Nucl. Mat.* 217-225.

Schram R.P.C., Laan R.R. van der, Klaassen F.C., Bakker K., Yamashita T. and Ingold F., 2003. The fabrication and irradiation of plutonium-containing inert matrix fuels for the 'Once Through Then Out' experiment. *J. Nucl. Mat.* 319, 118-125.

Shiratori T., Yamashita T., Ohmichi T. Yasuda A. and Watarumi K., 1999. Preparation of rock-like oxide fuels for the irradiation test in the Japan Research Reactor No. 3. *J. Nucl. Mat.* 274, 40-46.

Somers J. and Fernandez, A., 2005. Fabrication routes for yttria-stabilized zirconia suitable for the production of minor actinide transmutation targets. *J. Am. Ceram. Soc.* 88, 827-832.

[Join or Renew](#)

[Facebook](#)

[Geochemical News](#)

[Elements Magazine](#)

[Geochimica et Cosmochimica Acta](#)

[Goldschmidt Conference](#)

[Follow GS on Twitter](#)





[home](#) > [publications](#) > [geochemical news](#) > [gn132 \(jul07\)](#) > [award news \(#132\)](#)

## Award News (#132)

HOLLY STEIN of COLORADO STATE UNIVERSITY RECEIVES HELMHOLTZ-HUMBOLDT AWARD

FORT COLLINS - Colorado State University scientist Holly Stein received the prestigious Helmholtz-Humboldt Research Award for her groundbreaking scientific research in ore deposit geology and geochemistry.

Stein is the director of the AIRIE Program, Applied Isotope Research for Industry and the Environment, in the Department of Geosciences. AIRIE is a leading institution in producing state-of-the-art developmental and analytical work in Re-Os (rhenium-osmium) chronology. Re-Os dating allows scientists to better understand how metallic ore deposits form and their temporal relationship to regional geologic, metamorphic and tectonic processes.

In particular, AIRIE developed the technology to date the mineral molybdenite, well known to Coloradans through the Climax and Henderson mines. This relatively modest mineral occurs globally. Dating this mineral by Re-Os has unleashed a wealth of age information that reveals the primary histories of many otherwise complex rocks. The mineral industry now seeks this technology to aid exploration for new ore deposits.

The AIRIE Program recently acquired the expertise to date hydrocarbons, so that the source of oil and the time that the oil migrated into distant geologic traps can aid petroleum exploration. This work is backed by \$2.3 million from the Norwegian petroleum industry and the Norwegian Research Council.

The Helmholtz Association and the Alexander von Humboldt Foundation jointly grant up to six research awards annually to internationally recognized scientists for their research achievements to date. The award categories cover all disciplines of science including energy, earth and environment, health, technology, structure of matter, transport and space.

Nominations for the award are made by members of the Helmholtz Association National Research Center in Germany. Awardees are scientists whose discoveries, theories and findings have a strong influence on the immediate and broader disciplines over and beyond their specific research area.

As part of the award, Stein will receive a 50,000 Euro - or about \$70,000 - prize presented by the president of Germany in a formal reception in Berlin. She will also have a formal affiliation with the GeoForschungsZentrum and the University of Potsdam, and she will be an invited speaker at other universities throughout Germany.

In addition to her expertise in ore deposit geology and geochemistry, Stein has been active in several geologic societies and editorial boards. She received the 2005 Silver Medal from the Society of Economic Geologists for excellence and original work in the geology of mineral deposits. In 2000, she received a Fulbright Research Fellowship. She received a Gilbert Fellowship from the U.S. Geological Survey in 1992-1993 to develop the Re-Os dating tool, a means of determining the age of samples on the geologic time scale. In 1992, Stein received the Outstanding Woman Alumna Award at Western Illinois University.

Stein has been a senior research scientist in the Department of Geosciences at Colorado State since March 1998. After receiving her bachelor's degree from Western Illinois University, Stein obtained her master's of science degree and doctorate from the University of North Carolina at Chapel Hill. In addition to her research work, Stein is an accomplished cellist and soprano. She also enjoys writing poetry.

## GN132 Content

[From the GS President \(#132\)](#)  
by Sue Brantley

[Computed Tomography Applied to Geosciences](#)  
by Allen H. Reed and Yoko Furukawa

[Disposal of Nuclear Waste: Innovative Fuels for Actinide Transmutation](#)  
by Geert-Jan L.M. de Haas, Frodo C. Klaassen and Ronald P.C. Schram

[Award News \(#132\)](#) Holly Stein receives Helmholtz-Humboldt Award

[Nick's Picks \(#132\)](#) Geo-relevant podcasts  
by Nicholas Wigginton

[Join or Renew](#)

[Facebook](#)

[Geochemical News](#)

[Elements Magazine](#)

[Geochimica et Cosmochimica Acta](#)

[Goldschmidt Conference](#)

[Follow GS on Twitter](#)

## Nick's Picks (#132)

### Geo-relevant podcasts

As we are now fully immersed in the iPod culture, "podcasts," or regularly updated audio files on a particular topic or theme, have become increasingly popular.\* You can download podcasts on a broad spectrum of topics such as [how to learn a new language](#), [how to catch a fish](#), or even [how to appreciate wine](#). Fortunately, some podcasts have found their home in the sciences. Some of the more prominent science publications such as *Science* and *Nature* have weekly podcasts, and there are other podcasts directly related to the Earth Sciences (some are even specifically focused on geochemistry). Now you can stay up-to-date on exciting research wherever you go: on the bus, in the gym, or even out in the field! Here are some Geo-relevant podcasts I regularly listen to, but I'm sure I missed a few. Feel free to email me your favorites to the address at the right so I can post more up here. For your listening pleasure:

#### Science Magazine



This podcast, as well as a few others in this list, is great because it is a review of many fields in science. In one episode, you can learn about climate change, stem cells, governmental funding, and how bees swarm. Quite often they have features on Earth science, and it's nice to have that mixed in with other areas of science. The stories are presented at a high level, but are technical enough to get some real information out of them.

[Subscribe](#), [listen to the most recent show](#), or [check out an episode](#) from May 2006 focusing on the Earth's magnetic history, polio eradication, and viruses/nanotechnology.

#### Science Friday



This is the podcast edition of a great radio program that airs on many NPR stations across the country. The language is a little more simplified because it's meant for a general audience, but sometimes it's nice to hear something boiled down to the basics. Also, they have a page for [help with signing up](#) for podcasts, keeping them current, and actually listening to them.

[Subscribe](#), [listen to the most recent show](#), or [check out this episode](#) on the Arctic.

#### Living on Earth



This podcast is entirely Earth-related topics from environmental geochemistry and climate change to ecology and sustainability. Like *Science Friday*, it's also a public radio program available in podcast form. It features great interviews and coverage of the most current issues like energy and global warming. If you're at all involved with Earth and Environmental science, this is the podcast for you.

[Subscribe](#), [listen to the most recent show](#), or [check out this episode](#) from May 2007 remembering the legacy of Rachel Carson (author of *Silent Spring*), the discovery of bacteria in the La Brea Tar Pits, and the complex role that coal plays

in our communities.

#### Other podcasts

**Nature Magazine:** Similar to Science magazine, but with British accents!

**Pulse of the Planet:** 'Two-minute sound portraits of the Earth.' Bite-sized science clips that are easy to digest.

(Note: One of their ongoing features this summer is a Science Diary by Former Geochemical Society President Mike Hochella. Check out his [Science Diary](#) on their website and find links to all of his episodes.)

**American Mineralogist:** This mineralogy journal has a few really interesting audio files in which they interview authors of recent Am. min. papers to discuss the implications of their research. You can't subscribe to the podcast yet, but they have a page with all the audio files.

\*For help with downloading podcasts to your computer or iPod, check out [Apple's official podcast site](#).

### GN132 Content

[From the GS President \(#132\)](#)

by Sue Brantley

[Computed Tomography Applied to Geosciences](#)

by Allen H. Reed and Yoko Furukawa

[Disposal of Nuclear Waste: Innovative Fuels for Actinide Transmutation](#)  
by Geert-Jan L.M. de Haas, Frodo C. Klaassen and Ronald P.C. Schram

[Award News \(#132\)](#) Holly Stein

receives Helmholtz-Humboldt Award

[Nick's Picks \(#132\)](#) Geo-relevant podcasts

by Nicholas Wigginton

### Who's Nick?

Nick is Nicholas Wigginton.

He is a Ph.D Candidate in the Department of Geosciences at Virginia Tech. If you have a pick or a suggestion for Nick please contact him at [wigginto@vt.edu](mailto:wigginto@vt.edu).

Join or Renew

Facebook

Geochemical News

Elements Magazine

Geochimica et Cosmochimica Acta

Goldschmidt Conference

Follow GS on Twitter