Featured Highlight

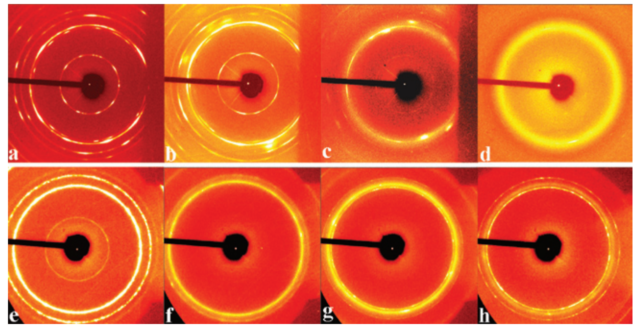
Response of Pyrochlore to Extreme Conditions

At the NSLS, a team of researchers from the University of Michigan and Rensselaer Polytechnic Institute has demonstrated the effect of composition on the stability and response of the pyrochlore structure under high pressure and in a high radiation field. This study lays the foundation for understanding how complex ceramics will respond to the extreme environments, such as those in advanced nuclear reactors.

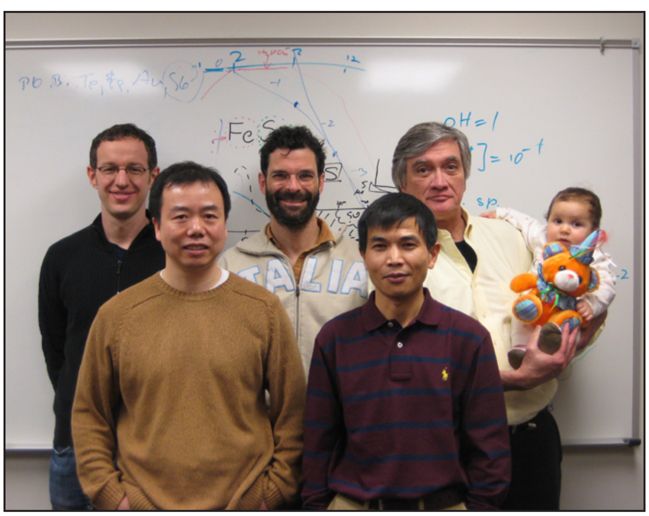
Pyrochlore has a flexible structure, which can exist in more than 500 compositions that have a wide variety of chemical and electronic properties with many technological applications. Some pyrochlores can also incorporate actinides, such as plutonium, so there is great interest in their use as nuclear waste forms or as inert matrix fuels.

In order to investigate the effects of high pressures and irradiation on pyrochlore, two closely related compositions, a gadolinium titanate oxide and a gadolinium zirconate oxide, were compressed with up to 45 gigapascals (GPa) of pressure, roughly more than six million pounds per square inch. That, by comparison, is almost 200,000 times the pressure in a car tire and more than the weight of a space shuttle on one's fingertip.

The structure of gadolinium titanate oxide remained relatively stable under the highest pressures tested (45 GPa), but the gadolinium zirconate oxide only maintained stability up to 23 GPa, above which it structurally distorted. When the pressure was released, the zirconate pyrochlore further disordered into a very stable defect-fluorite structure, which held up when irradiated to high doses.



X-ray diffraction images for $Gd_2Ti_2O_7$ at (a) 22.2 GPa, (b) 45.0 GPa, (c) 0 GPa (recovered sample and measured immediately), (d) 1 bar, 5 h after release of pressure; $Gd_2Ti_2O_7$ at (e) 16.0, (f) 44.0, (g) 0 GPa (recovered sample and measured immediately) and (h) 1 bar, 10 h after releasing pressure.



Authors (from left) Maik Lang, Jianwei Wang, Udo Becker, Fuxiang Zhang and Rod Ewing (holding Sofia). Not pictured: Jie Lian (faculty member at Rensselaer Polytechnic Institute).

Under ion-beam radiation, however, the behaviors of the two pyrochlores changed. The zirconium pyrochlore was more resistant to radiation damage, and therefore remained crystalline. In contrast, the titanate pyrochlore transformed into disordered structure at low doses and finally became stable at higher doses.

After using Raman scattering, calculations, and angle dispersive x-ray diffraction at NSLS beamline X17C, the researchers concluded that subtle changes in their structures significantly influence the response of pyrochlore to elevated pressure and irradiation fields. The performance of these materials in extreme environments is directly related to the energetics of the disordering process. Their results were reported in the February 1, 2008 edition of *Physical Review Letters*.

Now, Ewing's team is further studying titanium and zirconium composition under very high-energy irradiations, finding that the materials have consistent susceptibility to radiation damage. A single uranium atom fired through pyrochlore material leaves a damaged zone, which exists as a linear track. Using high-resolution electron microscopy, the researchers have found every type of structural stage within this trail of damage.

"The research team is now actively involved in studies that investigate the simultaneous effects of high pressure and a high radiation field using very high-energy, swift, and heavy ions passing through a diamond anvil pressure cell," said research group leader and University of Michigan geological sciences professor Rod Ewing. "The next year should provide new and exciting results under a pressure and irradiation regime that have never been investigated," Ewing said.

Researchers involved included Fuxiang Zhang, Jianwei Wang, Maik Lang, Udo Becker, and Rod Ewing of the University of Michigan, and Jie Lian of Rensselaer Polytechnic Institute.

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For more information, please see: F.X. Zhang, J. W. Wang, J. Lian, M. K. Lang, U. Becker, and R.C. Ewing, "Phase Stability and Pressure Dependence of Defect Formation in $Gd_2Ti_2O_7$ and $Gd_2Zr_2O_7$ Pyrochlore," Phys. Rev. Lett., ***100****(4): 045503 (2008).*

– Satya Shanmugham