EXPLORING HEAVY-ELEMENT SCIENCE USING PRESSURE*

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Scientific studies often employ variable experimental parameters to elucidate fundamental and technological aspects of materials. In this regard, pressure provides a very useful and powerful variable. Under laboratory conditions, pressure can alter interatomic distances solids far more than can temperature, and we are employing it to pursue the science of the heavy elements.

There have been significant advancements in the science of f-elements under pressure due in part to new designs of the diamond anvil cell (DAC), the use of synchrotron radiation and laser spectroscopy. These advancements permit one to study the actinides through californium under pressure in the megabar region. With such pressures, materials are compressed to fractions of their initial volumes. The significant reductions in interatomic distances that result can induce important changes in electronic interactions, energy levels and often alter the bonding.

One point of interest with actinides is whether pressure can bring about delocalization of f electrons and permit them to participate in bonding. The spatial extension of the 5f-electrons permits delocalization and/or hybridization processes to occur more readily with the transplutonium elements, as compared to their 4f-homologs. In the case of compounds, the behavior under pressure may be more complex, as electrons and orbitals from two or more different atoms must now be considered. Another facet of interest here is the effect of pressure on electronic transitions in f-electron materials, which may be altered significantly as the relative energies of the states are changed by pressure.

We have multiple interests for using pressure as an experimental variable for probing the heavy elements, and this includes altering temperature and pressure simultaneously. One emphasis has been to determine the effect of pressure by monitoring structures using X-ray diffraction techniques, where the effect of pressure on the electronic nature and the bonding is determined using established structure-bonding relationships. We have also employed spectroscopic techniques to monitor changes in materials brought about by applying pressure.

One recent thrust was to study americium metal up to one megabar using synchrotron radiation. It was found that the americium exhibited four different structures, where two transformations also involved "volume collapses". The latter are believed to reflect the pressure-induced involvement of americium's 5f-electrons in the metallic bonding. It was determined that pressure forces americium to adopt known structural forms of plutonium and uranium.

In the future, we plan to pursue systematic, comparative investigations that employ pressure as at least one of the experimental variables. These collaborative [1] efforts will employ synchrotron radiation at the ESRF, and hopefully also at the upcoming APS high-pressure beamline. An immediate goal is to investigate americium-curium alloys, curium and californium (the highest actinide currently feasible to study) under pressure to examine their electronic/structural behavior. We also expect to pursue the effects of pressure on compounds (e.g., actinide pnictides and chalcogenides) to determine the effects of pressure on materials with multiple atoms that may also have a potential for displaying a change in valence. Also planned are studies to examine, via spectroscopy, pressure induced changes in the energies of electronic states, the effect on energy up-conversion processes and energy transfers, and the effect on Raman vibrational spectra. Targeted materials will be single and multiple f elements in hosts (e.g., inorganic ceramic materials), as well as several pure materials.

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* Sponsored by the Division of Chemical Sciences, Geosciences and Biosciences, OBES, USDOE, under contract DE-ACO5-00OR22725 with ORNL, managed & operated by UT-Battelle, LLC.

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