Some bacteria appear capable of altering isotopic ratios of Uranium --- Is it the result of prosaic chemical fractionation processes and/or LENRs?

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December 7, 2010
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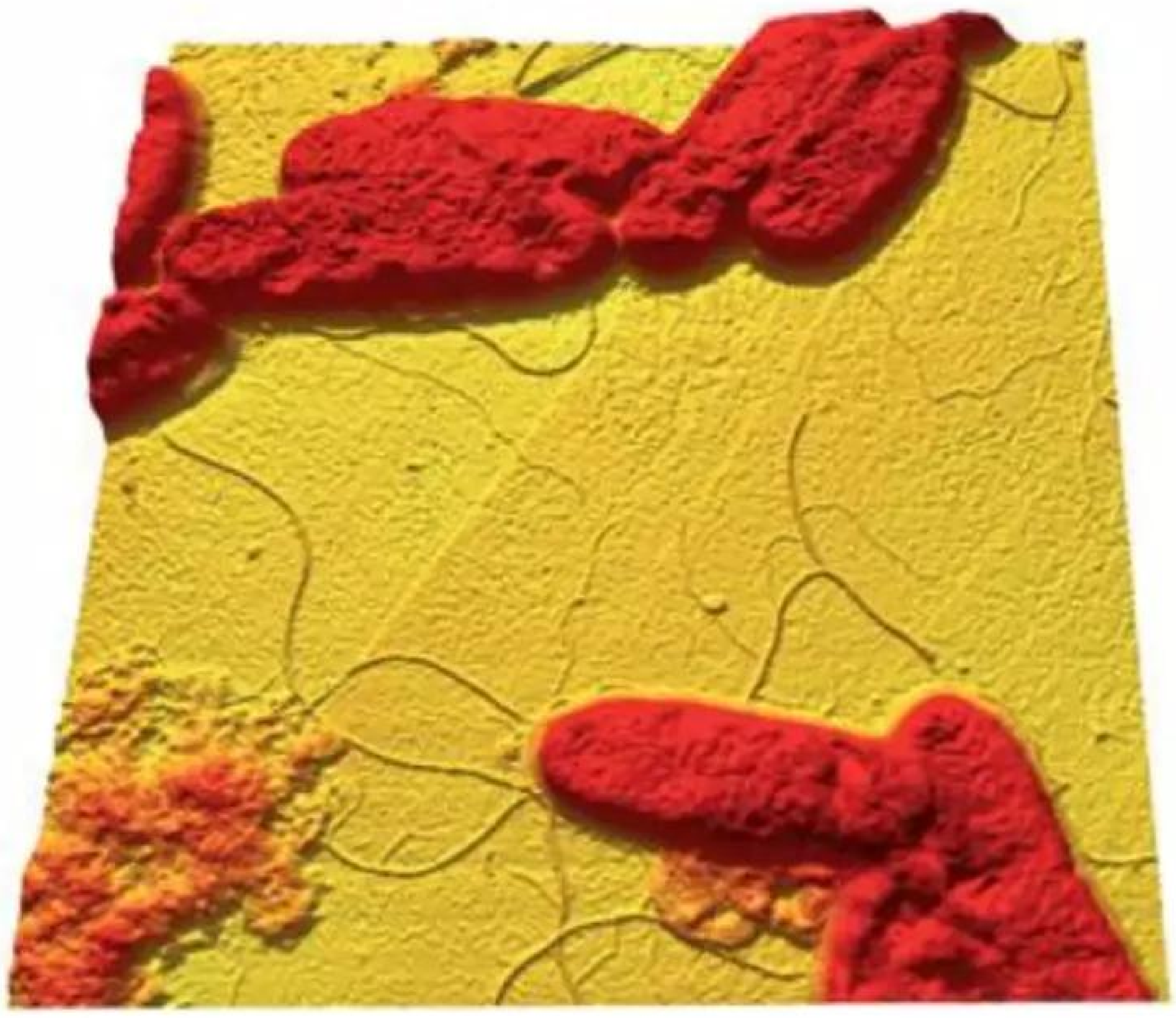
Perhaps to readers' amazement, in examples of published, peer-reviewed papers (citations are provided in this document) you will see that certain bacteria appear to be capable of significantly altering measured ratios of Uranium isotopes. If this data is correct, then what is the mechanism?



Shewanella oneidensis strain MR-1 bacteria showing electrically conductive bacterial nanowires, 5 μm scan

Credit: M. El-Naggar, USC and
Y. Gorby, J. Craig Venter Institute

Conceptual outline of contents

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Old paradigm - *Earth's Uranium isotope ratios are essentially invariant*:

Up until relatively recently, it was widely presumed that present-era $^{238}U/^{235}U$ ratios were, within some characteristic range of natural variance, essentially identical throughout the planet earth and its environs. This long-held belief was based on the underlying assumption that no significant amounts of nucleosynthesis have taken place locally since solid materials condensed out of the presolar nebula and aggregated under the influence of gravity to form planets. Under the old paradigm, observations of anomalous variations in Uranium isotope ratios were assumed to be strictly the result of spontaneous, time-dependent nuclear fission and alpha/beta-decay processes.

Obvious exceptions to the above 'rule' were ancient Oklo-type natural fission reactors, man-made stable and unstable isotopes produced in fission reactors and detonations of nuclear weapons, as well as captured interstellar debris produced during 'nearby' supernova explosions.

New experimental evidence - *challenges the old paradigm*:

Recently, greatly increased use of various types of mass spectroscopy by geochemists, microbiologists, and environmental scientists has revealed that the longstanding assumption of effective natural uniformity of $^{238}U/^{235}U$ ratios across the earth is clearly erroneous; importantly, present-era abiological and/or biologically mediated processes appear to be responsible for such anomalous variances.

Key question **-** *how might significant changes in $^{238}U/^{235}U$ ratios actually occur?*

Are anomalous variances in such isotopic ratios the result of purely chemical "fractionation" process or processes of some sort, and/or could they be caused by low energy nuclear reactions (LENRs), either abiologically or somehow induced by the actions of bacteria through some yet to be clarified mechanism?

Currently popular chemical fractionation paradigm - *besides exceptions noted above, it is widely believed that purely chemical "fractionation" processes correctly explain most if not all observations of significant changes in terrestrial Uranium isotope ratios, including those associated with bacterial activity*:

The chemical fractionation paradigm and the Widom-Larsen theory of LENRs (insofar as W-L may do a better job explaining a presently indeterminate subset of data now thought previously to be strictly the result of chemical "fractionation") are extensively discussed and compared in a public online SlideShare PowerPoint presentation as follows:

"*Lattice Energy LLC - LENRs in catalytic converters: are 'green' LENRs occurring in common devices?*" – June 25, 2010 - [76 slides] at source URL =

http://www.slideshare.net/lewisglarsen/lattice-energy-llc-len-rs-in-catalytic-convertersjune-25-2010

Quoting selected text/topics from the June 25, 2010, SlideShare document:

"*Please be aware that the following discussion involves a radical (some might say heretical) paradigm-shift in thinking about the types of environments where nuclear reactions may occur. LENRs may well be relatively common ---hidden in plain sight*" [Slide #4]

"Nuclear reactions in few places; chemistry explains everything else" [chart subtitle: Slides #13 - 22]

"*Today's fractionation theories include equilibrium and kinetic effects and mass-independent: nuclear field shift, photochemical, and Q-M symmetry effects that attempt to extend such concepts to higher-mass elements/isotopes in the periodic table*." [Slide #14]

"*Nuclear field shift effect in mass independent isotope fractionation*" [explained on Slides # 17 and 21]

"*Although not explicitly acknowledged by fractionation theorists, an intrinsic fundamental assumption underlying all of this theory and interpretation of data is that no nucleosynthetic processes are occurring anywhere in any of these systems, at any time, that are capable of altering isotope ratios and/or producing new mixtures of different elements over time; ergo, chemistry explains everything … However, if the Widom-Larsen theory is correct, for some of this data the above fundamental assumption may be wrong*." [Slide #23]

"*In systems with LENRs, some of chemistry's key fundamental assumptions break down and are violated on small length scales. Thus, while 'pure chemistry' may reign supreme on 98 -99% of a given sample surface, there can be many tiny sub-regions comprising just a few % of the total surface area in which W-L weak interaction-dominated nuclear processes can also occur*." [Slide #26]

"*Key point:* [LENR] *transmutation processes can mimic chemical fractionation*" [Slide #28]

My opinion - *there are problems with chemical fractionation theories in the case of Uranium isotopes:*

Prior to publication of seminal work on bacterial transmutations in Russia (Vysotskii et al. 2003, 2008), prosaic explanations for many observed Fe and other isotope anomalies observed in many studies assumed that biological systems (mainly bacteria) are able to effectively "fractionate" heavier isotopes through various types of biochemical pathways that utilize purely chemical, non-nuclear mechanisms. *Unfortunately, very specific, believable biophysical details of precisely how such processes are supposed to operate are lacking in my opinion, especially for significantly heavier elements, e.g., Iron and beyond.*

I can readily accept that chemical kinetic processes in biological systems are able to discriminate between isotopes of comparatively light elements (e.g., hydrogen, carbon, nitrogen, and perhaps up to oxygen) and can somehow physically separate them from each other; that is, via purely chemical "fractionation." However, I have difficulty believing that ordinary, prosaic chemical processes alone are capable of easily and efficiently discriminating and significantly enriching various isotopes of substantially heavier elements, especially beyond the atomic masses of Fe isotopes and/or transition metals.

In particular, peer-reviewed papers have recently been published in which *Geobacter* species are seemingly observed to "fractionate" Uranium isotopes (Bopp et al. 2010). In such papers authors attempt to explain Uranium isotope changes with the so-called "nuclear field shift effect" and nuclear volume/shape effects (Bigeleisen 1996). In my view, such explanations for isotopic data could potentially be erroneous in the case of Uranium.

Why? Because the reason giant high-speed gas centrifuges are used to "fractionate", i.e., enrich, uranium isotopes for use in nuclear weapons and fission reactor fuel is that 'unassisted' wet chemical kinetic fractionation/separation processes by themselves have been found to be inadequate in terms of practical efficacy. The only other known-to-be practical method for sufficiently enriching Uranium isotopes employs powerful lasers and the same type Uranium hexafluoride gas used in high-speed centrifuges.

If the recently hypothesized chemical fractionation mechanisms truly worked as well on Uranium isotopes in non-gas-phase systems as their proponents claim, one would surely think that such effects would have been utilized long ago in one or more of the government nuclear weapons and/or commercial Uranium enrichment programs scattered around the world (e.g., Iran today). To my knowledge, during the past 60 years they have never been used for such a purpose anywhere. That well-established fact is the basis for my strong skepticism about "nuclear field "or "nuclear volume" shifts being correct explanations for the underlying mechanism(s) responsible for creating observed changes in Uranium isotope ratios.

Summarizing: while these new types of chemical fractionation theories may well be logically consistent, do not violate accepted physics, and are capable of supporting calculations that do agree with some data, in the case of Uranium isotope shifts *such agreement may be phenomenological rather than fundamental*.

Until too many planets were discovered, Ptolemaic epicycles permitted calculations that more-or-less agreed with astronomers' observations up to that point. However, ever-growing numbers of epicycles required to explain volumes of (then) newly collected astronomical data were ultimately replaced by Newtonian mechanics --- a much simpler, fundamental theory of planetary orbits that enabled even better calculations to be performed that explained a vastly broader range of natural phenomena.

New Widom-Larsen theory of LENRs explains nuclear transmutations of isotopes *- combinations of chemical "fractionation" processes and/or LENRS may better explain some anomalous isotopic data*:

I am not claiming that the existing chemical fractionation paradigm fails to adequately explain most reported isotope anomalies with respect to statistically significant deviations from natural abundances --- indeed it may well explain the vast majority of them. That said, present literature appears to contain a significant subset of cases in which the chemical fractionation paradigm must be pushed very hard (which includes use of various ad hoc constructs) to explain certain data, i.e. it is being overly stretched to be able to comfortably accommodate some present isotope anomalies. *What is proposed is that in Nature a variety of prosaic chemical fractionation processes operate in parallel with abiotic and biological low energy nuclear reaction (LENR) transmutations, producing the vast range of different isotopic shifts and anomalies that are observed in a rapidly increasing body of high quality experimental data.*

With fissile isotopes, Incident neutron energies determine induced fission versus 'simple' capture – in fissile isotopes of Actinide elements (e.g., Uranium-235, Plutonium-239, etc.) neutron energy is key:

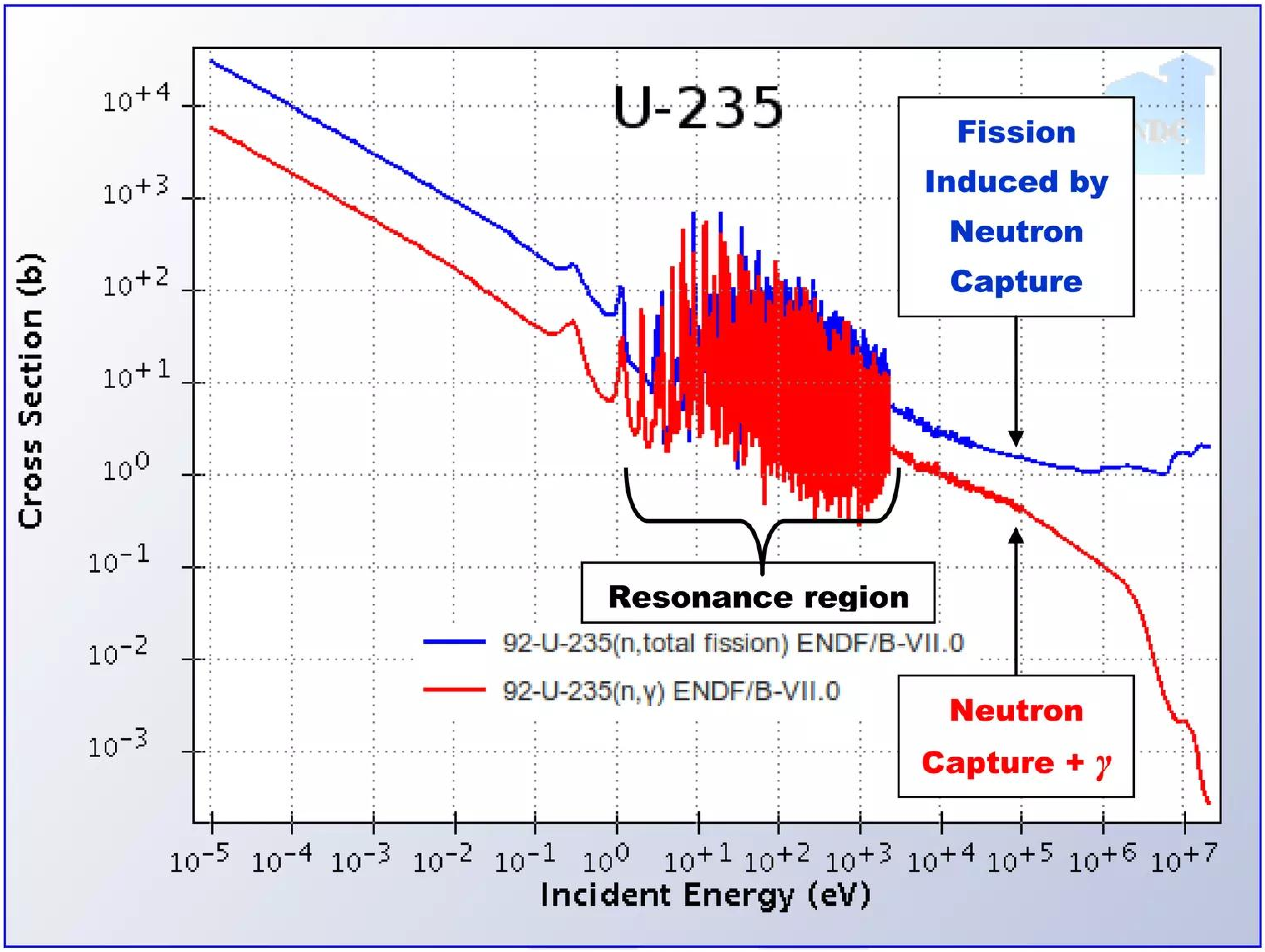
As we have noted in our many public technical documents, quantum-mechanical DeBroglie wavelengths of ULM neutrons in condensed matter LENR systems can range from as little as ~0.28 nm (2.8 Angstroms) on benzene rings all the way up to perhaps as large as 100+ microns (> ~1 million Angstroms) on fully 'loaded' metallic hydride surfaces . By comparison, a free neutron at thermal energies passes though condensed matter at a velocity of ~2,200 m/sec (v) with directly related kinetic energy of ~0.025 eV and Q-M DeBroglie wavelength of ~2 Angstroms.

Now, please note that viruses typically range from ~0.004 – 0.100 microns in size (i.e., 40 to 1,000 Angstroms); bacteria from ~0.2 – ~2.0 microns (2,000 to 20,000 Angstroms). *Incredibly, for brief periods of attoseconds to picoseconds, ULM neutrons (normally a subatomic particle of femtometer dimensions when inside a nucleus) created collectively via the weak interaction in condensed matter systems have quantum mechanical wavelengths that can sometimes be as large as the dimensions of microorganisms.*

In condensed matter, the wavelength, i.e., kinetic energy (and thus capture cross-section on 'target' atoms situated within the envelope of the wave function) of a given ULM neutron is determined by the physical dimensions of the many-body entangled, collectively oscillating 'patch' of protons in which it is created via the weak reaction $e^* + p \rightarrow n$ + neutrino. That being the case, if the *dimensions* of LENR-active patches of protons can somehow be 'controlled,' then *kinetic energies* of produced neutrons can also be 'controlled.' This sort of capability could be important because with fissionable isotopes (e.g., Actinides like ^{235}U, ^{239}Pu, etc.) the energy of the incident neutron being absorbed determines whether the result will be a capture process that simply produces a heavier unstable isotope along with a prompt capture gamma, or whether it induces a much more energetically violent fission of the 'target' nucleus.

With certain exceptions, most known isotopes generally follow the so-called 1/*v* rule: that is, neutron capture cross-sections (measured in barns - *b*) for different 'target' isotopes are roughly a linear function of the incident neutron kinetic energy (directly related to *v*) --- the lower the energy, the higher the capture cross-section. However, in the course of such measurements on many different isotopes, in some cases certain ranges of incident neutron energies were discovered in which both neutron capture and induced fission cross-sections varied enormously in parallel, e.g., 10^3, and non-linearly with very small changes in incident neutron energy; these are referred to as "resonance regions;" an example with ^{235}U follows.

An example of a measured resonance region for Uranium-235 is shown in Chart 1 below:



Fissioning ^{235}U (U-235) nucleus releases ~181 MeV of prompt energy in complex mixture of energetic fission fragments and gammas **-** by contrast, 'simple' ULMN capture (n, γ) on U-235 releases ~6.6 MeV

Please note that the blue line in Chart 1 above is the mostly measured neutron capture cross-section for fission; the red line is for 'simple' capture and prompt gamma emission. At most energies, fission is a significantly more likely event than neutron capture for ^{235}U. However, in the resonance region that is not the case; depending on a neutron's exact energy, capture can sometimes be as or *more likely* than fission. So, if one somehow had extremely precise control over produced neutron energy one could in principle create a situation in which neutron capture events were substantially more likely than fission.

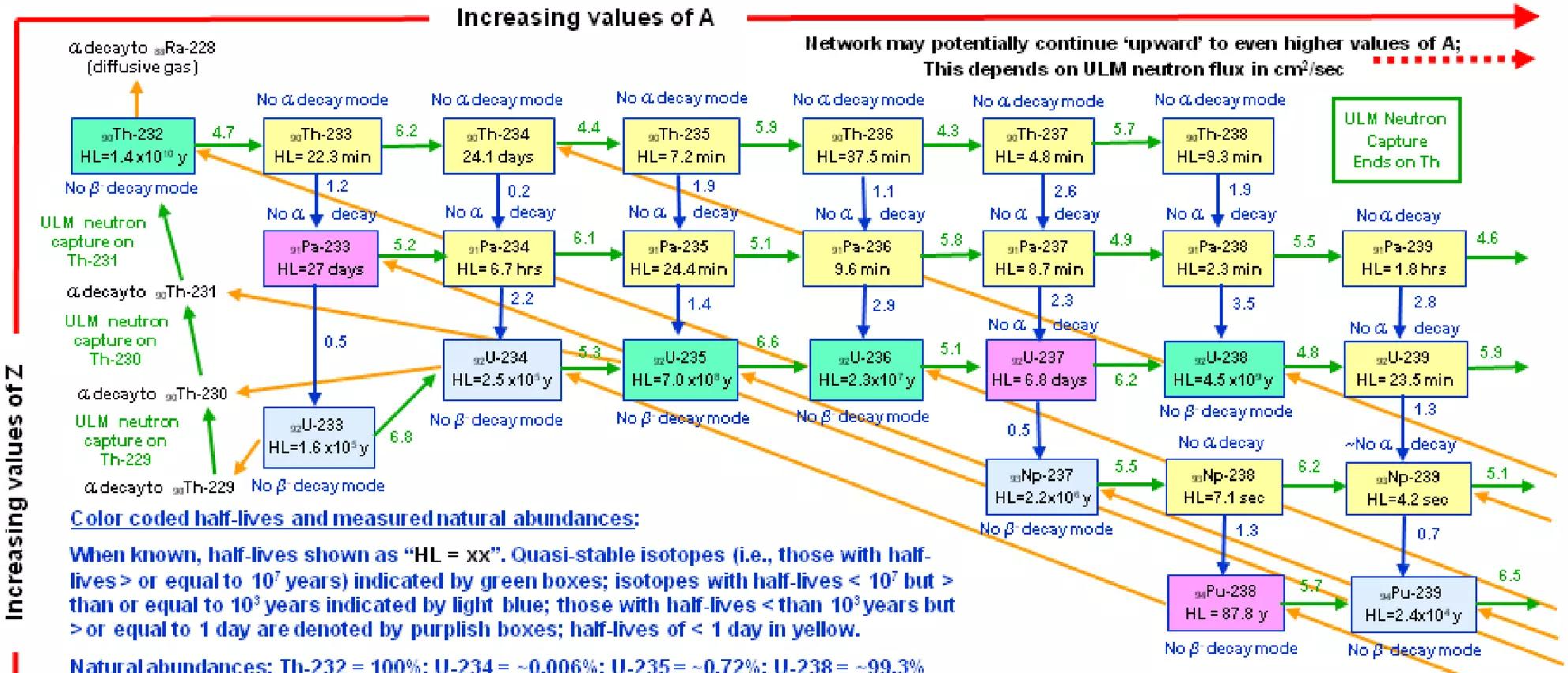
For ^{235}U and other fissile Actinides, comparative cross-sections for neutron captures vs. fission have never been measured in the region of ultra low momentum energies (which are vastly lower than the 10^{-5} eV shown in Chart 1 above). That said, there is nothing in the laws of nuclear physics that would a priori prevent the existence of additional, analogous resonance regions in the ultra low momentum (ULM) neutron regime. *We will now hypothesize that such low-energy resonance regions exist in Nature and explore potential consequences of that possibility, recognizing that direct evidence for such resonances has not yet been observed experimentally; it is presently a speculative conjecture.*

For any organism assumed capable of producing ULM neutrons via the weak interaction that then uses such neutrons to create LENR-based nucleosynthetic networks (for whatever purpose), there would be strong selective pressures to evolve an ability to minimize the number of neutron-induced fissions versus 'simple' neutron captures (i.e., make heavier isotope + gamma conversion to IR by heavy electrons): *it is the difference between dealing with very damaging, essentially uncontrolled release of large amounts of energetic fission fragments and gamma radiation versus other, vastly less damaging nuclear processes.*

Now let us examine a specific example of a hypothetical terrestrial nucleosynthetic network based upon Widom-Larsen's new nuclear paradigm – a ~fissionless ULM neutron-catalyzed LENR network beginning with neutron-capture on Thorium-232 (natural abundance = 100%; half-life = 1.4 x 10^{10} years):

Fig. 1 - ULMN catalyzed LENR network starting from $_{90}Th^{232}$

ULMN capture on Thorium, neutron-rich isotope production, and related decays



Color coded half-lives and measured natural abundances:

When known, half-lives shown as "HL = xx". Quasi-stable isotopes (i.e., those with half-lives > or equal to 10^7 years) indicated by green boxes; isotopes with half-lives < 10^7 but > than or equal to 10^3 years indicated by light blue; those with half-lives < than 10^3 years but > or equal to 1 day are denoted by purplish boxes; half-lives of < 1 day in yellow.

Natural abundances: Th-232 = 100%; U-234 = ~0.006%; U-235 = ~0.72%; U-238 = ~99.3%

Legend:

Please note: once created, the process of capturing an ULM neutron on a nearby atom happens very quickly --- on the order of picoseconds (0.000000000001 seconds).

ULM neutron captures proceed from left to right except for upper-left corner; Q-value of capture reaction (MeV) in green either above or below horizontal arrow. ⟶

Beta decays proceed from top to bottom; denoted with blue vertical arrow with Q-value (MeV) in blue either to left or right. ⟶

Alpha decays, where shown with orange arrows, proceed mostly from right to left with Q-value (MeV) in black on either side of the arrow. ⟵

Electron captures (e.c.) indicated by purple vertical arrow; Q-value (MeV) to left or right. ↑

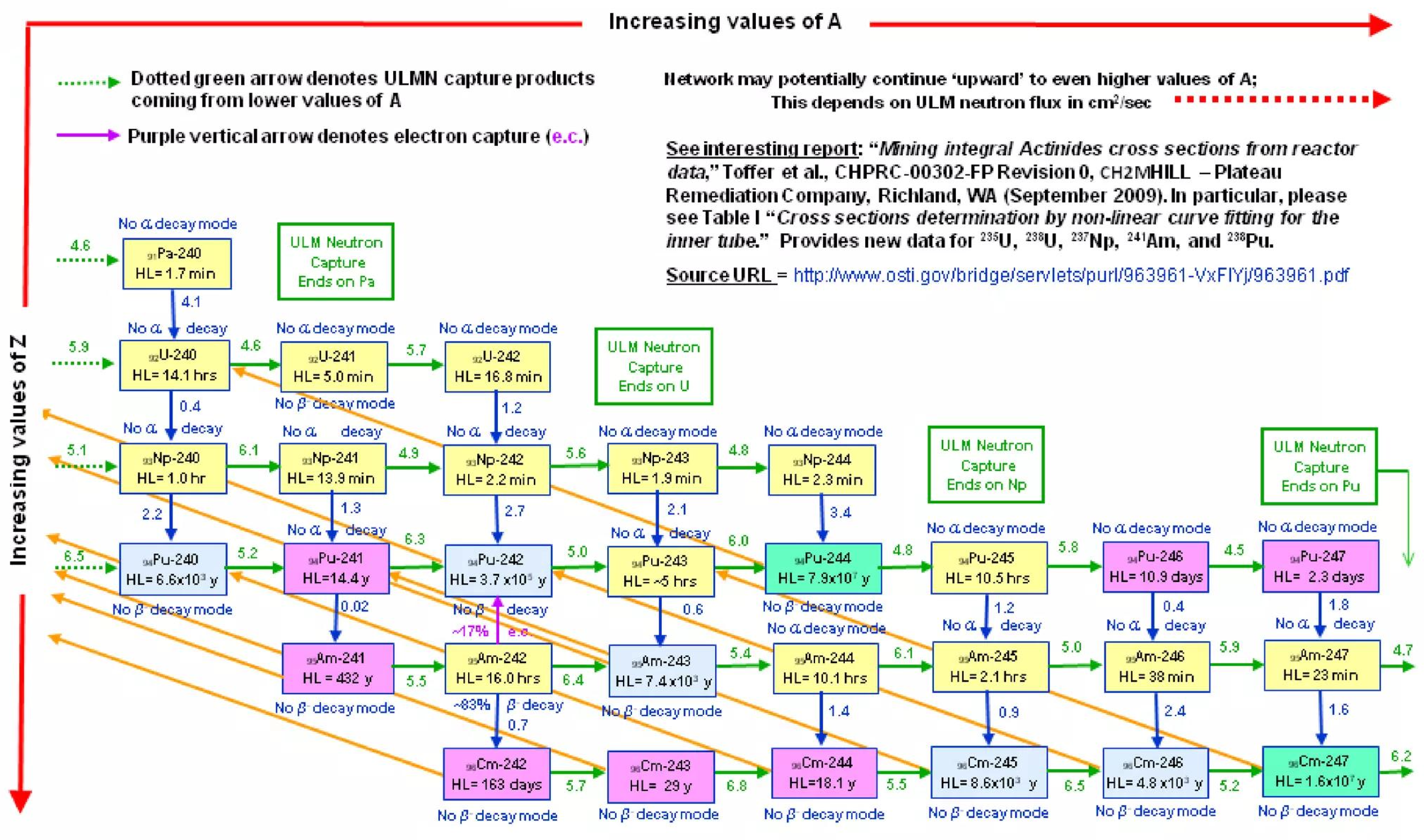
Fissionless LENR nucleosynthetic network:

Conjecture: this LENR network 'avoids' 'unwanted' neutron-induced fission by tightly controlling ULM neutron energies in capture resonance regions so that neutron capture and gamma emission (n, γ) and any subsequent beta and/or alpha decays vastly dominate

Any gamma emissions are automatically converted directly to infrared photons by heavy e^* electrons always present in the same nanoscale regions as LENR ULMN production (> ~0.5 MeV to ~10 MeV suppressed)

Fig. 2 - ULMN catalyzed LENR network starting from $_{90}Th^{232}$

ULMN capture on Thorium, neutron-rich isotope production, and related decays



See interesting report: "*Mining integral Actinides cross sections from reactor data,*" Toffer et al., CHPRC-00302-FP Revision 0, CH2MHILL – Plateau Remediation Company, Richland, WA (September 2009). In particular, please see Table I "*Cross sections determination by non-linear curve fitting for the inner tube.*" Provides new data for ^{235}U, ^{238}U, ^{237}Np, ^{241}Am, and ^{238}Pu.

Source URL = http://www.osti.gov/bridge/servlets/purl/963961-VxFIYj/963961.pdf

Roughly ~fissionless (except for spontaneous non-neutron-induced fissions) neutron-catalyzed LENR network beginning with ULMN capture on Thorium-232 – discussion of Figures 1 and 2 above:

Somewhat analogous to a prosaic chemical or biochemical reaction network, the above two Figures describe a hypothetical ~fissionless, Widom-Larsen ULM neutron catalyzed, LENR-based nucleosynthetic network characterized by an interlocking series of neutron captures beginning on Thorium-232 (^{232}Th) 'seed' nuclei (100% natural abundance) and subsequent beta decays, alpha decays, electron capture (one instance), neutrino emissions, and direct conversion of prompt and delayed gamma photons to infrared (heat) by heavy *e** electrons involved in weak interaction ULM neutron production via the [$e^* + p \rightarrow n$ + neutrino] reaction. Where known, neutron capture and beta decay Q-values are shown in MeV; where applicable, neutrino and gamma emissions are implicit and not shown to reduce visual clutter.

What is shown above is a 'full network' of nucleosynthetic *possibilities*; in any given situation in Nature, only a portion of this network may be operating at any given point time and/or at any particular geospatial location. *Furthermore, some portions of the network may operate abiologically, whereas some or all of it may also operate biologically, albeit perhaps spread across a wide variety of different ecologically interacting species of microorganisms found in natural environments*, e.g., *Geobacter*, *Shewanella*, etc.

In my view, isotopic 'nodes' in the above Actinide network where neutron-induced fission is 'avoided' after ULM neutron capture (e.g., ^{235}*U,* ^{233}*U,* ^{237}*U,* ^{239}*Pu,* ^{241}*Pu, etc.) are likely to be mediated most effectively by biological systems. I believe this could be the case because exquisitely fine-grained, highly reproducible control over produced neutron energies would appear to be necessary to achieve such a result. Since abiological processes cannot 'learn' and in essence store information over time (i.e., 'embed' it in physical structures of evolved proteins specified by a genetic code), it would seem to be more difficult to abiologically 'optimize' an effectively spontaneous, multistep, many-body collective LENR process.*

If truly present, nucleosynthetic abilities could have been acquired early in the history of life – microorganisms here on Earth may have been altering and synthesizing isotopes for billions of years:

If certain microorganisms can truly engage in isotopic nucleosynthesis (not decisively proven yet), one can imagine that various species of bacteria now on Earth could have easily spent the last 3 – 4 billion years of natural selection and trial-and-error evolution 'learning' how to accomplish such an amazing feat.

Frankly, it should be unsurprising that certain bacteria, other microorganisms, and some fungi exhibit extreme resistance to radiation (e.g., *Deinococcus radiodurans*, which can thrive under huge radiation fluxes present inside operating light water ^{235}U fission reactors; *Chroococcidiopsis*, *Thermococcus gammatolerans,* and some species of *Rubrobacter*) and/or are apparently able to alter isotopic ratios of heavy elements. Roughly 3.5+ billion years ago, at a much early point in the evolution of life on Earth, there is no doubt whatsoever that there were vastly larger quantities of radioisotopes and various forms of hard radiation present in the natural terrestrial environment. Given such a history, we shouldn't be shocked that early life adapted and evolved to deal with high-radiation regimes; evidence of such early adaptation has been preserved through eons of time and remains today in some modern microorganisms.

For a moment, let us speculatively travel even further back in time to the era of the presolar nebula from which our solar system's planets condensed. If in fact some sort of primitive Ur-microbial life was then living on isolated dust grains embedded in the radiation-laced, hydrogen-rich dusty presolar cloud, the ability to create needed elements/isotopes when necessary, utilize heavy-mass *e** electrons as gamma radiation shields, and perhaps even generate 'raw' heat via nucleosynthesis to help accelerate Ur-microbes' kinetically-driven *chemical* reactions, would have provided valuable competitive advantages for any living organisms possessing such capabilities that were living in such a harsh environment.

Who knows, perhaps water in some form was also present somewhere in the dusty presolar cloud. This possibility is suggested by the recent, very surprising discovery of a team of astronomers led by L. Decin ("*Warm water vapour in the sooty outflow from a luminous carbon star*," *Nature* **467** pp. 64-67 September 2010). Using infrared observations from the ESA's Herschel Space Observatory, Decin et al. concluded that water vapor was definitely present at ~700° C in the outer atmosphere of an aging carbon-rich star named IRC+100216 located 500 light-years from earth. Prior to this new discovery, the presence of large amounts of water vapor as such in and around hot stellar atmospheres was thought to be impossible.

Objectives herein – generate interest and stimulate further inquiry into LENR transmutations:

My purpose in bringing selected papers and new ideas to readers' attention is a hope that other researchers will find the controversial concept of LENR transmutations intriguing enough to acquaint themselves with the W-L theory and then use it to help them unravel what I believe is a deep mystery surrounding Uranium isotopic shifts and other anomalous isotopic "fractionations" observed in Nature.

Preliminary remarks and caveats – readers encouraged to directly examine experimental evidence:

I will now cite, extensively quote from, and make brief comments about selected reports concerning various types of shifts in Uranium and Plutonium isotope ratios that have been observed in different terrestrial environments. Rather than my discussing each paper *ad nauseum*, readers are encouraged to focus directly on the experimental data presented in these publications in the light of the Widom-Larsen theory of LENRs and the hypothetical fissionless LENR-based Actinide nucleosynthetic network shown in Figs. 1-2 herein. After studying the data, readers can decide for themselves whether LENR transmutation processes might be a new area of inquiry that is worthy of their further consideration and investigation.

Please note I am not suggesting that this collection of papers constitutes proof-positive that biological and/or abiological LENR-based nucleosynthesis is occurring in Nature. That said, I believe that the available experimental data suggests it is very likely in some instances, albeit a controversial interpretation in the light of the widely accepted chemical fractionation paradigm. In my opinion, further investigation by experimentalists is warranted to map-out the boundaries in parameter space where the chemical fractionation paradigm ends and a nuclear paradigm is needed to fully understand data.

Early seminal experimental work on microbial transmutations in Russia by Vysotskii & Kornilova – their work has generally not been peer-reviewed simply because mainstream journals have steadfastly refused to send any of their manuscripts out to referees; nonetheless, while this work has some shortcomings, I believe they have reported important early data that merits attention.

General comment about their publications: in my opinion, Vysotskii et al.'s "cold fusion" speculations about the mechanisms underlying their experimentally observed isotopic data are simply physics nonsense; please focus on examining their reported data, as opposed to how they try to explain it:

Nuclear Fusion and Transmutation of Isotopes in Biological Systems [hardcover book]
V. Vysotskii and A. Kornilova
Especially Chapter 4, **"*Experimental study of transmutation of isotopes and chemical elements in microbiological systems*,"** from pp. 179 - 212
Mir Press - Moscow 2003 [now out of business --- I can e-mail a scanned copy of Chap. 4 if interested]

Comment on Chapter 4: these experimental observations suggest that cultures of D. radiodurans M-1 and B. subtilis GSY-228 may be able to directly synthesize elemental iron (Fe - four stable isotopes) from Manganese (Mn - has only one stable isotope). In the context of W-L theory of LENRs, we would explain such data with the hypothesized capture of one or more ULM neutrons on stable ^{55}Mn, followed by subsequent beta decays into various stable isotopes of Iron (^{56}Fe, ^{57}Fe, etc.). *Very interesting data.*

"*Experimental observation and modeling of Cs-137 isotope deactivation and stable isotopes transmutation in biological cells*"
V. Vysotskii, A. Tashyrev, and A. Kornilova [peer-reviewed book chapter]
Low-Energy Nuclear Reactions Sourcebook
Chapter 13, pp. 295 – 309 Chapter DOI: 10.1021/bk-2008-0998.ch013
ACS Symposium Series, Vol. 998 ISBN 13: 9780841269668 eISBN: 9780841221413
American Chemical Society - 2008

An updated version of the now out-of-print 2003 book by Mir Press was recently republished:

Nuclear Transmutation of Stable and Radioactive Isotopes in Biological Systems
V. Vysotskii and A. Kornilova
ISBN: 978-81-8274-430-1 Hardcover 192 pages (presently ~US$25 + shipping on Amazon.com)
Pentagon Press (India) – 2010

Examples of peer-reviewed publications reporting various types of shifts in terrestrial Uranium isotope ratios – they are no longer regarded as being essentially invariant within the Earth's crust and environs (source URLs to free online copies of cited papers will be provided where possible):

"*Natural fractionation of $^{238}U/^{235}U$*"
S. Weyer et al.
Geochimica et Cosmochimica Acta **72** pp. 345 – 359 (2008)
Source URL = http://boyle.mit.edu/~ed/PDFs/Weyer(2008)GCA72_345.pdf

Direct quote: "The isotopic composition of U in nature is generally assumed to be invariant. Here, we report variations of the $^{238}U/^{235}U$ isotope ratio in natural samples (basalts, granites, seawater, corals, black shales, suboxic sediments, ferromanganese crusts/nodules and BIFs) of ~1.3‰, exceeding by far the analytical precision of our method (~0.06‰, 2SD) … The largest isotope variations found in our survey are between oxidized and reduced depositional environments, with seawater and suboxic sediments falling in between … Our findings imply that U isotope fractionation occurs in both oxic (manganese crusts) and suboxic to euxinic environments with opposite directions. In the first case, we hypothesize that this fractionation results from adsorption of U to ferromanganese oxides, as is the case for Mo and possibly Tl isotopes. In the second case, reduction of soluble U^{VI} to insoluble U^{IV} probably results in fractionation toward heavy U isotope compositions relative to seawater … The discovery that $^{238}U/^{235}U$ varies in nature also has implications for the precision and accuracy of U - Pb dating."

Comment: establishes that Uranium isotope ratios are not essentially invariant in the Earth's crust

"*Low-temperature isotopic fractionation of Uranium*"
C. Stirling et al.
Earth and Planetary Sciences Letters **264** pp. 208 - 225 (2007)
Source URL = http://www.jcsservices.in/pdf/Low-Temperature-Isotopic.pdf

Direct quote: "Uranium is the heaviest naturally occurring element and isotope fractionation between ^{235}U and ^{238}U is not normally considered significant given the small ~1% difference in mass. It is therefore usual to assume that $^{238}U/^{235}U$ is constant in the terrestrial environment and equal to 137.88. We have developed experimental protocols for the precise measurement of $^{235}U/^{238}U$ by multiple-collector ICPMS (MC-ICPMS) and have analyzed a suite of samples formed in a range of low-temperature environments … Here we demonstrate sizeable (13 epsilon units) natural variability in $^{235}U/^{238}U$, exceeding the analytical reproducibility by more than an order of magnitude. Compositions that are both isotopically heavier and lighter than our terrestrial standard, by 4 and 9 epsilon units respectively, are observed. The largest excursions are found in speleothem samples. Furthermore, $^{235}U/^{238}U$ appears broadly correlated with $^{234}U/^{238}U$ in samples showing the most extreme isotopic compositions. The present study investigates the role of abiotic processes in fractionating ^{235}U from ^{238}U … Our observations have a direct impact on the U-series and U–Th–Pb chronometers, when applied to samples formed in low-temperature environments, as these chronometers currently assume an invariant $^{238}U/^{235}U$ equal to 137.88."

Comment: generally reinforces conclusions of S. Weyer et al. (2008) noted above

"*Experimentally determined Uranium isotope fractionation during reduction of hexavalent U by bacteria and zero valent iron*"
L. Rademacher et al.
Environmental Science & Technology **40** pp. 6943 - 6948 (2006)
Source URL = free copy unavailable

Direct quotes: "Presented here are measurements of mass-dependent U isotope fractionation induced by U(VI) reduction by zerovalent iron (Fe^0) and bacteria under controlled pH and HCO_3^- conditions. In abiotic experiments, Fe^0 reduced U(VI), but the reaction failed to induce an analytically significant isotopic fractionation. Bacterial reduction experiments using *Geobacter sulfurreducens* and *Anaeromyxobacter dehalogenans* reduced dissolved U(VI) and caused enrichment of ^{238}U relative to ^{235}Uin the remaining U(VI). Enrichment factors (ε) calculated using a Rayleigh distillation model are -0.31‰ and -0.34‰ for *G. sulfurreducens* and *A. dehalogenans*, respectively, under identical experimental conditions."

"Mass-dependent isotope fractionations are useful for detecting and potentially quantifying reduction reactions. In general, bonds involving lighter isotopes are easier to break than bonds with heavier isotopes. Thus, bonds involving lighter isotopes react more rapidly and are concentrated in solid products (*8*). These differing reaction rates result in kinetic isotope fractionation between reactants and products if back-reaction rates are small. The remaining unreacted pool becomes progressively enriched in heavier isotopes as reduction proceeds. Thus, the degree of isotopic enrichment is a useful means for detecting and quantifying the extent of reduction."

"Few studies have examined mass-dependent fractionation of ^{238}U and ^{235}U … The present contribution reports results from controlled laboratory experiments designed to determine the extent of ^{238}U/^{235}Ufractionation during chemical reduction by bacteria and Fe^0. We observed systematic changes in ^{238}U/^{235}U with progressing bacterial reduction but no isotopic fractionation during Fe^0 reduction."

[As the Uranium reference source for their experiments] "…we used an enriched U standard, SRM U-500 from the National Institute of Standards and Technology (NIST) (*25*) as the U source for the experiments. The NBS SRM U-500 contains nearly equal quantities of ^{238}U and ^{235}U and has a ^{238}U/^{235}U [ratio] of 1.0003 +/- 0.001. **[Readers, note especially: the ^{238}U/^{235}U (ratio) of natural U is 137.85 +/- 0.4]** …subsequent bacterial experiments were performed using a UO_2Cl_2 solution, which does not appear to interfere with biotic reduction processes … Results are only reported for uranyl chloride solutions used in the biotic experiments. Uranyl nitrate was used during abiotic (Fe^0) experiments … *G. sulfurreducens* was selected for its success at anaerobically reducing metals (e.g., ref *29*). *G. sulfurreducens* and other *Geobacteriacea* are currently being evaluated for their ability to reduce U in situ in contaminated groundwater … Cultures (100 mL) of *A. dehalogenans* and *G. sulfurreducens* were grown with 4 mM acetate and 5 mM fumarate in a bicarbonate-buffered (10 mM), low phosphate basal salts medium (pH 7-7.5) … no free oxygen was detected during the experiments … Approximately 0.25 mL (+/-25%) of 1360 mgL^{-1} U (as UO_2Cl_2) was added to each 25 mL culture tube to make a final solution strength of approximately 14 mgL^{-1} total U in the serum bottle … Control experiments, without the addition of cells, were included during each experiment set. All experiments were performed in triplicate. ^{235}U/^{238}U, ^{233}U/^{238}U, and ^{236}U/^{238}U isotope ratios were measured on a Micromass Isoprobe MC-ICP-MS located at the Field Museum of Natural History in Chicago, Illinois."

"Wu et al. (*32*) established that U(VI) reduction by *A. dehalogenans* is biologically mediated and does not proceed in the absence of cells … [our] Fe^0 Reduction Experiments = Despite decreasing concentration and apparent U reduction in these experiments, no measurable changes in isotope ratios were detected, with ^{238}U/^{235}U remaining constant … Bacterial Reduction Experiments = Both *A. dehalogenans* and *G. sulfurreducens* successfully reduced U during experiments when acetate was added as an electron donor. Bacterial experiments were terminated after 96 hours … Control experiments, which contained no cells, indicated that U reduction did not occur in the absence of either bacterium."

"Anomalous Concentrations Early in Bacterial Reduction Experiments**.** Anomalous changes in concentration occurred in the early stages of all of the bacterial reduction experiments (Figure 2A). Samples extracted at time zero (within 30 min of the experiment start) had U(VI) concentrations lower than expected initial values based on the amount of U added during experimental setup. During the first 8 h after the addition of U to the reaction vessels, U concentrations increased to approximately 14 mgL^{-1} +/- 25% followed by a steady, gradual decrease through the remainder of the experiment. The initial increase in U(VI) concentration during bacterial experiments was unexpected, as we anticipated a monotonic decrease in dissolved U as U(VI) was reduced to U(IV) and precipitated from solution. The increase in U(VI) in solution during the first 8 h indicates that U(VI) was removed from solution via very rapid adsorption or precipitation and then re-entered the solution over the first 8 h of the experiments … Adsorption is a likely explanation … Regardless of the process responsible for the early drop and subsequent recovery of U concentration, our determinations of reduction are valid as long as this process was complete 8 h into the experiments. The evolution of concentrations starting at 8 h follows a smooth progression of first-order reaction kinetics (R^2 = 0.99), so the assumption that the sorption processes are complete by 8 h appears to be valid. The 8-hour samples are the effective starting points of the experiments, and fractionation factors are calculated for reduction beginning at that time."

"In the absence of a back-reaction, a closed-system Rayleigh distillation model can be used to extract the magnitude of the kinetic isotope effect from the experimental data. When the fractionation factor, α, is constant during isotopic evolution in a closed system (such as the experiments in this study), the δ^{238}U value of the remaining U(VI) evolves according to the Rayleigh relationship … R is the fractionation factor expressed as the ratio of the isotope ratios for the reactant and product $\alpha = R_{reactant}/R_{product}$ where $R_{reactant}$ and $R_{product}$ are the ratios of ^{238}U to ^{235}U **[below R = measured isotopic ratio = $^{238}U/^{235}U$ for samples vs. standard]**… enrichment factor, ε, is used to express R in ‰ notation … Best-fit values (excluding samples prior to 8 h) of ε for the *G. sulfurreducens* and *A. dehalogenans* are -0.31‰ and -0.34‰."

[Table 1 in the paper clearly shows an increase in $^{238}U/^{235}U$ ratio with time in bacterial experiments and lack of comparable change in Fe^0 experiments; reproduced below for readers' convenience]

$$\delta = \left(\frac{R_{sample} - R_{standard}}{R_{standard}}\right) x1000$$

TABLE 1. Summary of Isotopic results from Abiotic and Bacterial Reduction Experiments

Experiment	Time (min)	$\Delta\delta^{238}U$ (‰)[a]	error[b]
Fe^0	0	-0.03	0.07
	41	-0.08	0.09
	64	-0.02	0.10
A. dehalogenans	0	-0.09	0.09
	120		
	240		
	480	-0.10	0.06
	2160	-0.03	0.05
	4320	0.19	0.06
	5760	0.26	0.04
G. sulfurreducens	0	-0.02	0.10
	480	-0.02	0.06
	2160	0.04	0.05
	4320	0.19	0.05

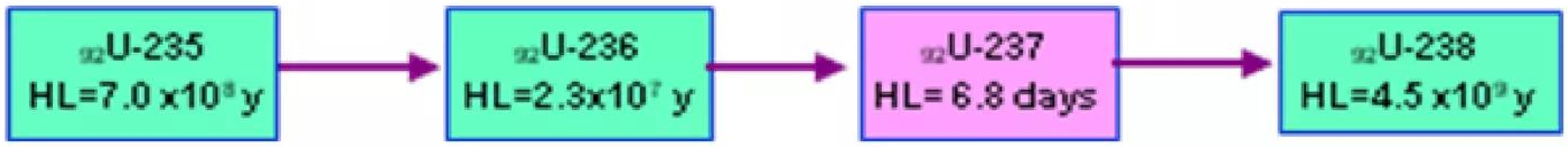
[a] **$\Delta\delta^{238}U$ represent the offset of the data point relative to the standard [NIST SRM U-500]**

[b] **Standard analytical error**

"Considerable research on isotopic fractionation during redox reactions involving Cr, Se, and more recently, Fe … suggests that ε tends to depend on reaction mechanism and environmental conditions … amount of isotopic fractionation for a given reaction tends to vary both with the reaction mechanism and with the rate of the reaction … In reaction steps that involve breakage of chemical bonds, reaction rates for lighter isotopes tend to be greater than those for heavier isotopes … single isotopic fractionation factor cannot be determined for a given reaction … range of likely values may be defined for the reaction or for a given reaction pathway under a particular set of conditions."

"Little is known about the exact reaction mechanisms for U reduction and potential U(VI)-U(IV) isotopic equilibration … Complex redox pathways occur in nature and can affect isotopic shifts. At this stage, U disproportionation cannot be ruled out and only a general understanding of the driver(s) of isotopic fractionation during U(VI) reduction is possible based on prior research of isotopic fractionation during reduction of other elements … while the magnitude of fractionation produced in nature remains unknown, the results of our bacterial reduction experiments indicate that reduction can induce small but significant shifts in U isotope composition."

Comment: while the chemical fractionation explanation advocated for this data by Rademacher et al. is not illogical, the W-L LENR Actinide nucleosynthetic network shown in Figs. 1 and 2 can also explain these experimental results alternatively as follows (please refer back to Figures on pp. 5-6 for details):



Please note that this theorized series of three sequential ULM neutron captures from ^{235}U to ^{238}U would be very fast (picoseconds/capture) and that neutron captures in this sequence of LENRs are fissionless. If the W-L mechanism were in fact operating during these experiments, sufficient production of ^{238}U from ^{235}U via ULMN captures could have occurred and been detected with mass spectroscopy. Also, when measured at *thermal energies* ^{235}U has a substantially *higher* 'simple' neutron capture cross-section than ^{238}U (~95 b vs. ~2.7 b); so all other things being equal, ULM neutron capture on ^{235}U would be expected to occur at substantially higher rates than capture on ^{238}U, decreasing ^{235}U relative to ^{238}U, and thus increasing δ^{238}U values, which is observed in Rademacher et al.'s controlled laboratory experiments.

"*Uranium ^{238}U/^{235}U isotope ratios as indicators of reduction: results from an in situ biostimulation experiment at Rifle, Colorado, U.S.A.*"
C. Bopp et al.
Environmental Science & Technology **44** pp. 5927 - 5933 (2010)
Source URL = free copy unavailable

Direct quotes: "We apply measurements of ^{238}U/^{235}U to a U bioremediation field experiment at the Rifle Integrated Field Research Challenge Site in Rifle, Colorado. An array of monitoring and injection wells was installed on a 100 m^2 plot where U(VI) contamination was present in the groundwater. Acetate-amended groundwater was injected along an up-gradient gallery to encourage the growth of dissimilatory metal reducing bacteria (e.g., *Geobacter* species). During amendment, U concentration dropped by an order of magnitude in the experiment plot. We measured ^{238}U/^{235}U in samples from one monitoring well by MC-ICP-MS using a double isotope tracer method. A significant ~1.00‰ decrease in ^{238}U/^{235}U occurred in the groundwater as U(VI) concentration decreased. The relationship between ^{238}U/^{235}U and concentration corresponds approximately to a Rayleigh distillation curve with an effective fractionation factor (R) of 1.00046. We attribute the observed U isotope fractionation to a nuclear field shift effect during enzymatic reduction of U(VI)$_{(aq)}$ to U(IV)$_{(s)}$."

"Recent research demonstrates that on … short time scales, changes in ^{238}U/^{235}U are induced by certain geochemical processes, such as chemical reduction. Several authors have shown ^{238}U/^{235}U to vary depending on environment and mode of U deposition (i.e., high vs. low temperature or igneous vs. epigenetic sedimentary deposition). These include studies showing broad changes in ^{238}U/^{235}U in U-bearing solid materials across several environments, with higher ^{238}U/^{235}U occurring in chemically reduced black shales (*11, 12*) and in epigenetic U ores produced by reductive entrapment of U(VI)$_{(aq)}$ from groundwater (*13*); this latter study found that tabular sandstone ore deposits were consistently depleted in ^{235}U relative to magmatic deposits."

"Because different isotopes may have slightly different bonding behavior in a given chemical reaction, isotope ratios may be fractionated during chemical reaction. The fractionation of ^{235}U from ^{238}U as a result of reduction is analogous to the behavior observed in other redox-active elements, such as S (*15, 16*) and Cr (*17*). Here, we apply the method of using variations in ^{238}U/^{235}U to monitor U reduction in a contaminated aquifer undergoing stimulated bioremediation through organic carbon amendment."

"To test the feasibility of in situ methods of remediating U contamination in groundwater, an experimental plot was built on the former site of a U mill at Rifle, Colorado. The experiments at this site (the Rifle Integrated Field Research Challenge (IFRC) site) have succeeded in reducing the previously elevated U concentrations by an order of magnitude via stimulation of the native subsurface bacteria (i.e., *18, 19)*. This site is designated the Old Rifle site and was part of the Uranium Mill Tailings Remedial Action (UMTRA) program under the U.S. Department of Energy."

"The 'Winchester' field experiment involved the injection of groundwater amended with sodium acetate (5 mM) and potassium bromide (2 mM) into the subsurface over 31 days during August and September 2007 to stimulate the bioreduction of U in a manner similar to previous experiments at the site (i.e., *19*). Acetate is a common electron donor, and so amending the groundwater with acetate stimulates the activity of [many different species of] dissimilatory metal-reducing microbes."

"Background samples were taken from every monitoring well before the injection experiment began. Once acetate injection began, the monitoring wells were sampled periodically (Table 1); this study focuses on samples taken over a period of 45 days from the start of the experiment … samples were first analyzed by inductively-coupled plasma mass spectrometry for cation content at Lawrence Berkeley National Laboratory before being shipped to the Isotope Geochemistry Laboratory at the University of Illinois at Urbana-Champaign. The samples were stored at 4 °C continuously, and were at room temperature only when aliquots were removed for processing and analysis."

"The double-isotope tracer (double spike) is a mixture of ^{233}U and ^{236}U having a known ratio (^{233}U/^{236}U ≈ 0.146001). The double spike was admixed into each sample or standard prior to chemical separation to correct for instrumental mass fractionation, (i.e., *20, 21)*. Correcting for these effects yields a highly precise determination of the original ^{238}U/^{235}U. The double spike has a ^{235}U/^{236}U ratio of ~0.00202 and a ^{236}U/^{238}U ratio of ~691, such that the contribution to the total ^{235}U and ^{238}U from the spike is small and can be corrected for. Samples were spiked so as to produce a ^{238}U/^{236}U of ~3-4. The double spike methods are similar to those in the literature."

"Although there were small differences in the pre-experiment δ^{238}U of groundwater within the experimental plot, pronounced changes were observed only after biostimulation occurred. Background samples (Figure 2, Table 1) have a mean $\delta^{238}U_{IRMM\ U\text{-}A}$ of 0.26‰ and standard deviation of 0.12‰ (2σ) excluding two outlying data points. We attribute these anomalous points to the presence of a naturally occurring reducing zone defined by the presence of reduced minerals and organic material (*25*). In essence, there is a zone of slow natural reduction in the area of the outlier wells, and the observed lower $\delta^{238}U_{IRMM\ U\text{-}A}$ values reflect a combination of partial U reduction in that zone and mixing of partially reduced groundwaters with unaffected groundwaters, as a result of either in situ advection or sampling."

"The down-gradient well D-07 showed a significant change in δ^{238}U values as the experiment progressed (Figure 3) and groundwater U(VI) concentrations decreased from 0.8 to 0.09 μM. Well D-07 showed a total δ^{238}U change of ~1.0‰, decreasing from preinjection values of $\delta^{238}U_{IRMM\ U\text{-}A}$ ≈ 0.40‰ to postinjection values of $\delta^{238}U_{IRMM\ U\text{-}A}$ ≈ -0.60‰. This decrease was well outside both analytical uncertainty and the background variability of the site."

"We interpreted these results using a mathematical framework developed in earlier work on isotopic fractionation observed during reduction reactions involving sulfur, selenium, iron, and chromium (*17, 26, 27*). The Rayleigh distillation model (*28*) is often used to describe the relationship between isotope ratios and the extent of reaction. The model describes the isotope ratios of reactants and products in a closed system where a reaction showing isotopic preference proceeds in the absence of back-reaction. To a rough approximation, groundwater masses moving through the bioreduction zone can be described using a Rayleigh model. Each groundwater mass is not a closed system because of dispersive mixing, but this effect leads to relatively small errors in interpretations of isotopic data (*29*). We assume U(IV) does not undergo back-reaction or otherwise interact substantially with U(VI)."

" … The resulting value of R (see eq 2) is 1.00046, indicating a preference for heavier isotopes in the products. The best fit Rayleigh distillation curve is plotted with our results in Figure 4 … These results indicate that ^{238}U has a greater reaction rate than ^{235}U. This sense of fractionation is opposite that observed for the reduction of lighter elements (i.e., Se, Cr (*17, 33*), S (*34*)), in which lighter isotopes have greater reaction rates. However, our findings are consistent with results from U isotopic analyses of solid earth materials (*11-13*) and theoretical work (*35*). Recent work has shown that U ore deposits created by entrapment of U via U(VI) reduction in aquifer settings (e.g., roll-front or tabular sandstone deposits) have a distinctly greater ^{238}U/^{235}U compared to higher temperature "magmatic" deposits (*13, 14*)."

"Generally, the ZPE [zero point energy] effect on reduction reactions is for lighter isotopes to have greater reaction rates (*26, 28, 34, 37*). Our results show the opposite effect, in which ^{238}U reacts faster than ^{235}U, and thus they do not fit this general pattern. However, kinetic isotope effects are known to vary with reaction mechanism and changes in the relative rates of individual steps within multistep reactions, so at present we cannot definitively rule out an unusual ZPE as the cause of the observed fractionation."

"Conversely, a mass-independent effect known as the nuclear field shift (NFS; *38-40)* appears to provide a mechanism by which ^{238}U could react faster than ^{235}U. The NFS has been shown to be important for heavier elements such as mercury, and it has been applied to explain U isotope fractionation in recent studies (*11-13*). Unlike mass dependent effects, the NFS is controlled by the shape and volume of the atomic nuclei independently of atomic mass. Theoretical estimates of equilibrium isotopic fractionation between U(VI) and U(IV) suggest that the NFS-driven fractionation will be 3 times greater and in the opposite sense to that of the ZPE effect (*38*). We suggest that the energy differences driving this equilibrium fractionation also drive a kinetic fractionation in the same direction, i.e., with ^{238}U enrichment in the U(IV) reaction product, that produces the isotopic shifts observed in the present study, U ores (*13, 14*), and sediments … the NFS interpretation seems consistent with the results of this experiment."

"Although our results are consistent with a NFS-dominated scenario, contradictory results from bacterial reduction experiments do exist. Rademacher, et al. (*22*) found that U reduction in laboratory experiments by *Geobacter sulfurreducens* and *Anaeromyxobacter dehalogenans* resulted in the remaining unreduced U(VI) in solution having progressively higher ^{238}U/^{235}U; while this is in agreement with a mass dependent fractionation effect, it is opposite to what is observed in every naturally reduced sample thus far measured (both solids and, in the case here, pore fluids/groundwater). The reason for the difference between the fractionation in the laboratory experiments and that observed during natural U reduction is not understood at present. However, the laboratory microcosm experiments were very different from a natural groundwater system. The results of Rademacher, et al. (*22*) **[cited and discussed above in this document]** suggest that additional experiments are needed to understand the mechanistic controls on biological fractionation of U isotopes in both natural and laboratory environments."

Comment: A contradiction of sorts is apparent here. On one hand, a comparatively well-controlled and characterized microbiological laboratory study by Rademacher et al. (2006) reported a significant reduction in the measured concentration of dissolved U(VI)$_{(aq)}$ Uranium in conjunction with a parallel *increase* in the U(VI)$_{(aq)}$'s ^{238}U/^{235}U ratio over time after the addition of acetate (as an electron donor) to the experimental culture medium. On the other, a bioremediation field experiment by Bopp et al. (2010) reported similar reduction in measured concentration of soluble U(VI)$_{(aq)}$ Uranium but instead observed a parallel *decrease* in the U(VI)$_{(aq)}$ ^{238}U/^{235}U ratio over time after analogous injection of acetate-amended groundwater into tailings (soil) of an old Uranium mill in Rifle, Colorado. *Comparing these two published reports, at first glance we seem to see an opposite effect on the U(VI)$_{(aq)}$ ^{238}U/^{235}U ratio after roughly the same causative action (adding acetate to stimulate the growth of "dissimilatory metal-reducing bacteria")*.

In both papers, authors presume a chemical process paradigm; i.e., Uranium isotopes must somehow be "fractionating" between soluble U(VI)$_{(aq)}$ compounds (measured w. mass spectroscopy in withdrawn aliquots of aqueous solution) and U(IV)$_{(s)}$ compounds assumed to be precipitated out of solution as insoluble particulates during bacterial reduction of U(VI)$_{(aq)}$ to U(IV)$_{(s)}$. Note that ^{238}U/^{235}U ratios are not measured in uncollected insoluble U(IV)$_{(s)}$ residues, so the true mass balance of elemental Uranium is not known in such cases. However, under a purely chemical paradigm, there would be no reason to suspect that total mass balance of Uranium could potentially change and/or neutron captures might be occurring.

Effects of the underlying chemical paradigm on researchers' thinking about their experiments can sometimes be subtly pernicious. For example, in the case of Rademacher at al., [quoting directly from pp. 6946] *"…we assume the produced U(IV)$_{(aq)}$ was bound within a precipitate (even though it was not clearly visible in the experiments used for isotopic analysis) and was unable to interact with the dissolved U(VI)$_{(aq)}$. Accordingly, the Rayleigh model is appropriate*." So in effect, they assumed a solid precipitate that they did not clearly observe experimentally because of an underlying subconscious belief in a chemical conceptual paradigm. Interestingly, *explanations* for the observed isotopic shifts also differ somewhat between the two papers. While Rademacher et al. did not come to a hard-and-fast conclusion about the likely physical mechanism behind the observed shifts in ^{238}U/^{235}U ratios, they leaned more-or-less toward the idea that they were the result of *mass-dependent* (i.e., purely kinetic) processes related to the idea that [quoting directly] *"…In reaction steps that involve breakage of chemical bonds, reaction rates for lighter isotopes tend to be greater than those for heavier isotopes*."

By contrast, to explain U(VI)$_{(aq)}$ ^{238}U/^{235}U shifts in the *opposite direction* in somewhat similar experiments, Bopp et al. invoked a [quoting directly], *"…nuclear field shift effect during enzymatic reduction of U(VI)$_{(aq)}$ to U(IV)$_{(s)}$.*" They go on to say that, *"…Conversely, a mass-independent effect known as the nuclear field shift (NFS; 38-40) appears to provide a mechanism by which U(VI)$_{(aq)}$ ^{238}U could react faster than U(VI)$_{(aq)}$ ^{235}U. The NFS has been shown to be important for heavier elements such as mercury, and it has been applied to explain U isotope fractionation in recent studies (11-13). Unlike mass dependent effects, the NFS is controlled by the shape and volume of the atomic nuclei independently of atomic mass*."

So we have one group (Rademacher et al.) suggesting that observed ^{238}U/^{235}U shifts during bacterial reduction of U(VI)$_{(aq)}$ to U(IV)$_{(s)}$ are the result of *mass-dependent* chemical kinetic effects, and the other group (Bopp et al.) saying that their observed ^{238}U/^{235}U shifts *in the opposite direction* are the result of theoretically very different *mass-independent* chemical fractionation processes based on nuclear field-shift and volume effects that were first proposed by Bigeleisen (1996) and further elaborated by Schauble (2007). *From the standpoint of underlying mechanisms, these alternative explanations are contradictory.*

Before attempting to reconcile these two sets of apparently conflicting experimental observations, a few comments about important differences between the two types of experimental systems are in order:

First, only two relatively well-characterized species of bacteria were involved in Rademacher et al.'s laboratory experiments; by contrast, Bopp et al.'s Rifle, Colorado, field site undoubtedly involved a very large array of many different types of soil microorganisms (only some of which are identified and well-characterized) embedded in a complex, dynamically interacting, rapidly changing natural micro-ecology.

Second, the input Uranium U(VI)$_{(aq)}$ compounds being reduced by specified bacteria in Rademacher at al.'s controlled laboratory experiments were created from extraordinarily well-characterized, NBS-certified material, NIST's Standard Reference Material U-500 (established in 1981 --- has an isotopic composition guaranteed to be ^{234}U = 0.5181 atom percent +/- 0.0008; ^{235}U = 49.696 a.p. +/- 0.050; ^{236}U = 0.0755 a.p +/- 0.0003; ^{238}U = 49.711 a.p +/- 0.050). *Note that NIST SRM U-500 does not contain any Thorium*.

By contrast, in Bopp et al.'s field experiments, the 'input' U(VI)$_{(aq)}$ Uranium isotopes actually being reduced by bacteria are whatever isotopic mixture happens to be naturally present at the particular time acetate is injected into the mine tailings to stimulate growth of whatever "*reducing dissimilatory bacteria*" may then be present.

Also note that in Bopp et al.'s Rifle bioremediation site, significant quantities of customary natural Uranium decay products are invariably present in tailings, including Thorium-232 as well as probably some Plutonium isotopes derived from atmospheric fallout contamination.

Importantly, the presence of significant amounts of ^{232}Th has been unequivocally verified in the Rifle test site tailings in multiple test pits. Please see "*Radiological survey of the inactive Uranium-mill tailings at Rifle, Colorado,*" F. Hayward et al., Report #ORNL-5455, Oak Ridge National Laboratory, Health and Safety Research Division, Contract # W-7405-eng-26, published June 1980 at:

Source URL = http://www.osti.gov/bridge/servlets/purl/5345200-AR4QVh/5345200.pdf

The potential significance of ^{232}Th being present at the natural Rifle test site soils versus its very likely absence in Rademacher at al.'s controlled laboratory experiments will be explained momentarily.

Please be aware that some or all of this theorized LENR transmutation network, if present in Nature, is likely to: be distributed across multiple species of microorganisms; vary spatially on sub-meter length-scales; and change dynamically over short time-scales in response a variety of environmental factors.

Let us now apply some of the ideas embodied in the fissionless W-L LENR Actinide network outlined in Figures 1 and 2 herein and see whether a few simple examples can provide us with some useful insights that can help reconcile some differences between the apparently contradictory ^{238}U/^{235}U data of Rademacher at al. versus that of Bopp et al. These arbitrarily constructed examples are provided to illustrate how identical isotopic shift data can be interpreted very differently when it is viewed through the conceptual lens of a widely accepted chemical, versus a still controversial nuclear, process paradigm.

Example 1 - Measured ^{238}U/^{235}U U(VI)$_{(aq)}$ ratio can INCREASE for very different reasons under different paradigms:

- **Chemical:** via a "fractionation" mechanism, *soluble* ^{235}U U(VI)$_{(aq)}$ is *reduced and converted to an insoluble* U(IV)$_{(s)}$ *form at a faster rate* than ^{238}U (VI) $_{(aq)}$ *is reduced and converted into insoluble* U(IV)$_{(s)}$ compounds

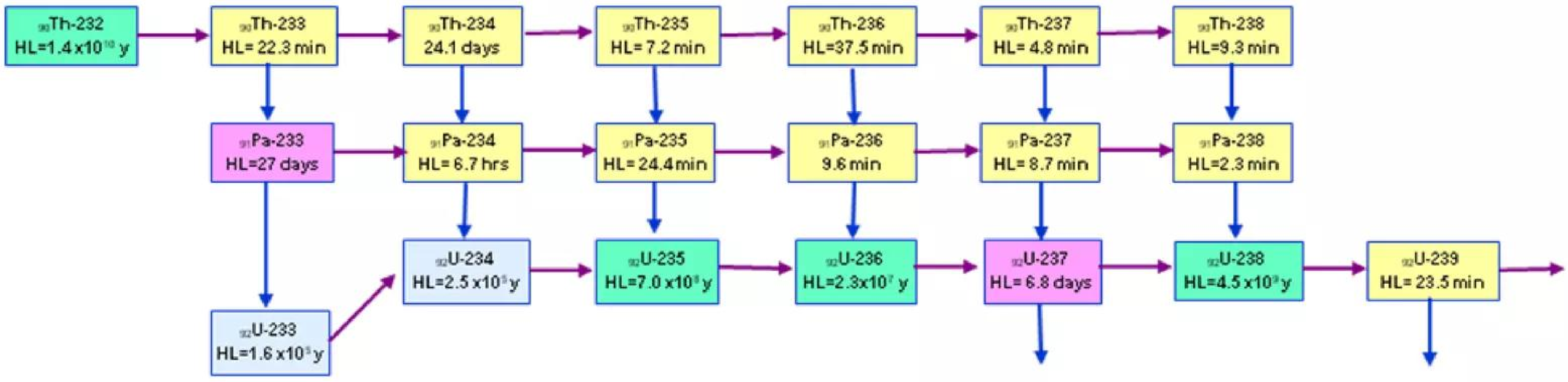
- **LENR:** in a given experimental system, the network subset of spatially- and species-distributed *nucleosynthetic pathways producing* ^{238}U from ^{235}U and ^{232}Th *may be operating at much faster rates than ULM neutron captures are transmuting* ^{238}U into stable or unstable isotopes of another element besides Uranium or Thorium

Example 2 - Measured ^{238}U/^{235}U U(VI)$_{(aq)}$ ratio can DECREASE for very different reasons under different paradigms:

- **Chemical:** via a "fractionation" mechanism, *soluble* ^{238}U U(VI)$_{(aq)}$ is *reduced and converted to an insoluble* U(IV)$_{(s)}$ *form at a faster rate* than ^{235}U U(VI)$_{(aq)}$ *is reduced and converted into insoluble* U(IV)$_{(s)}$ *compounds*

- **LENR:** in a given experimental system, the network subset of spatially- and species-distributed *nucleosynthetic pathways producing* ^{238}U from ^{235}U and ^{232}Th *may be operating at much slower rates than ULM neutron captures are transmuting* ^{238}U into stable or unstable isotopes of another element besides Uranium or Thorium

Thorium 'seed' nuclei present - in a given experimental system that has a functioning W-L LENR Actinide nucleosynthetic network, any ^{232}Th atoms initially present therein can potentially be transmuted via ULM neutron capture and decays into Uranium isotopes via the following subset of network pathways:



Possible reconciliation of differences between the results of Rademacher at al. versus Bopp et al.:

The segment of the hypothetical LENR Actinide nucleosynthetic network shown above illustrates how Thorium-232 (^{232}Th) can be transmuted into ^{235}U and/or ^{238}U by a combination of fissionless ULM neutron captures and subsequent radioactive decays.

Please recall that ^{232}Th was almost assuredly NOT present in significant amounts at the beginning of Rademacher et al.'s controlled laboratory experiments, but it undoubtedly was present in significant quantities in the tailings at the beginning of Bopp et al.'s field experiments in Rifle, Colorado.

If one looks at the differences in their results through the lens of a chemical paradigm, they look somewhat contradictory and confusing. That lack of clarity is probably what impelled the two groups to invoke diametrically opposite chemical fractionation mechanisms to explain their ^{238}U/^{235}U isotopic data (i.e., mass-dependent versus mass-independent processes). *By contrast, if one applies the new LENR paradigm, the observed differences are simply the result of a combination of: (1) different initial conditions (^{232}Th was initially present in one set of experiments but not the other); and (2) different segments of the LENR Actinide nucleosynthetic network operating at different relative rates during the two sets of experiments. Under an LENR paradigm, these results are readily comprehensible rather than confusing.*

"Sulfur isotopes as indicators of amended bacterial sulfate reduction processes influencing field scale Uranium bioremediation"
J. Druhan et al.
Environmental Science & Technology **42** pp. 7842 - 7849 (2008)
Source URL = http://www.osti.gov/bridge/servlets/purl/961529-3R6Vg7/961529.pdf

Abstract: "Aqueous uranium U(VI) concentrations in a contaminated aquifer in Rifle CO, have been successfully lowered through electron donor amended bioreduction. Samples collected during the acetate amendment experiment were analyzed for aqueous concentrations of Fe(II), sulfate, sulfide, acetate, U(VI), and δ^{34}S of sulfate and sulfide to explore the utility of sulfur isotopes as indicators of in situ acetate amended sulfate and uranium bioreduction processes. Enrichment of up to 7‰ in δ^{34}S of sulfate in down-gradient monitoring wells indicates a transition to elevated bacterial sulfate reduction. A depletion in Fe(II), sulfate, and sulfide concentrations at the height of sulfate reduction, along with an increase in the δ^{34}S of sulfide to levels approaching the δ^{34}S values of sulfate, indicates sulfate-limited conditions concurrent with a rebound in U(VI) concentrations. Upon cessation of acetate amendment, sulfate and sulfide concentrations increased, while δ^{34}S values of sulfide returned to less than -20‰ and sulfate δ^{34}S decreased to near-background values, indicating lower levels of sulfate reduction accompanied by a corresponding drop in U(VI). Results indicate a transition between electron donor and sulfate limited conditions at the height of sulfate reduction and suggest stability of biogenic FeS precipitates following the end of acetate amendment."

Direct quotes: "Briefly, during the course of the experiment a solution of up-gradient groundwater with 10 mM sodium acetate and 1 mM potassium bromide was continuously injected at a rate of 0.1 to 0.3 L/min for 69 consecutive days to shift the system through iron and sulfate reducing conditions."

"These experiments demonstrated aqueous uranium removal in association with iron reduction attributed to increased activity of *Geobacter* species. As acetate amendment continued, the system shifted from iron reduction to BSR, at which point U(VI) removal rates decreased. Prolonged U(VI) removal following cessation of acetate amendments was also noted in areas where sulfate reduction had occurred, though the mechanisms of post amendment U(VI) removal have not been characterized (*3, 34, 35*). Following the success of these experiments, two additional "minigalleries" were constructed south of the larger gallery to further explore the relationships between iron and sulfate reduction and uranium removal."

"Subsurface microbial activity is often electron-donor limited, and thus addition of organic compounds such as lactate, acetate, ethanol, and glucose offer a straightforward means of uranium sequestration … Enhanced in situ uranium bioreduction has been successfully demonstrated at … the Old Rifle Site in western Colorado (3–8). At the Old Rifle Site, acetate has been utilized as the electron donor, and the efficiency of the associated bioremediation processes [of Uranium] has been shown to depend directly on the redox conditions generated in the aquifer. During previous acetate amendments, the highest rates of uranium removal were observed at redox levels optimal for microbial iron reduction (3). U(VI) bioreduction continued at lower redox conditions favoring microbial sulfate reduction but at decreased rates."

"Bacterial sulfate reduction (BSR) in the subsurface can be identified through characteristic fractionations in the stable isotope compositions of sulfate and sulfide. BSR preferentially utilizes sulfate containing ^{32}S, the lighter isotope of sulfur. This preference results in enrichment of ^{34}S, the heavier isotope of sulfur, in the residual sulfate**.**"

"U(VI) reduction by sulfate reducers has also been shown to depend on the type and variety of organic substrate, but, in general, sulfur isotope fractionation during BSR is principally dependent on the magnitude of the preference for reducing sulfate with the lighter sulfur isotope … Under most conditions, the difference between the δ^{34}S of sulfate minus the δ^{34}S of the sulfide produced is ≥ 10‰; however, under sulfate-limited conditions the fractionation factor reduces to almost zero (25). Precipitation of sulfide minerals produces an isotopic fractionation of <1‰ (*27, 30*) and will, therefore, not cause a significant change in the δ^{34}S values of the remaining aqueous sulfide even when a large fraction of the sulfide has precipitated out of solution. For these reasons, the δ^{34}S of sulfide is an excellent indicator of whether or not a system has become sulfate limited."

Figure 4 in Druhan et al. is quite interesting and is reproduced below for readers' convenience:

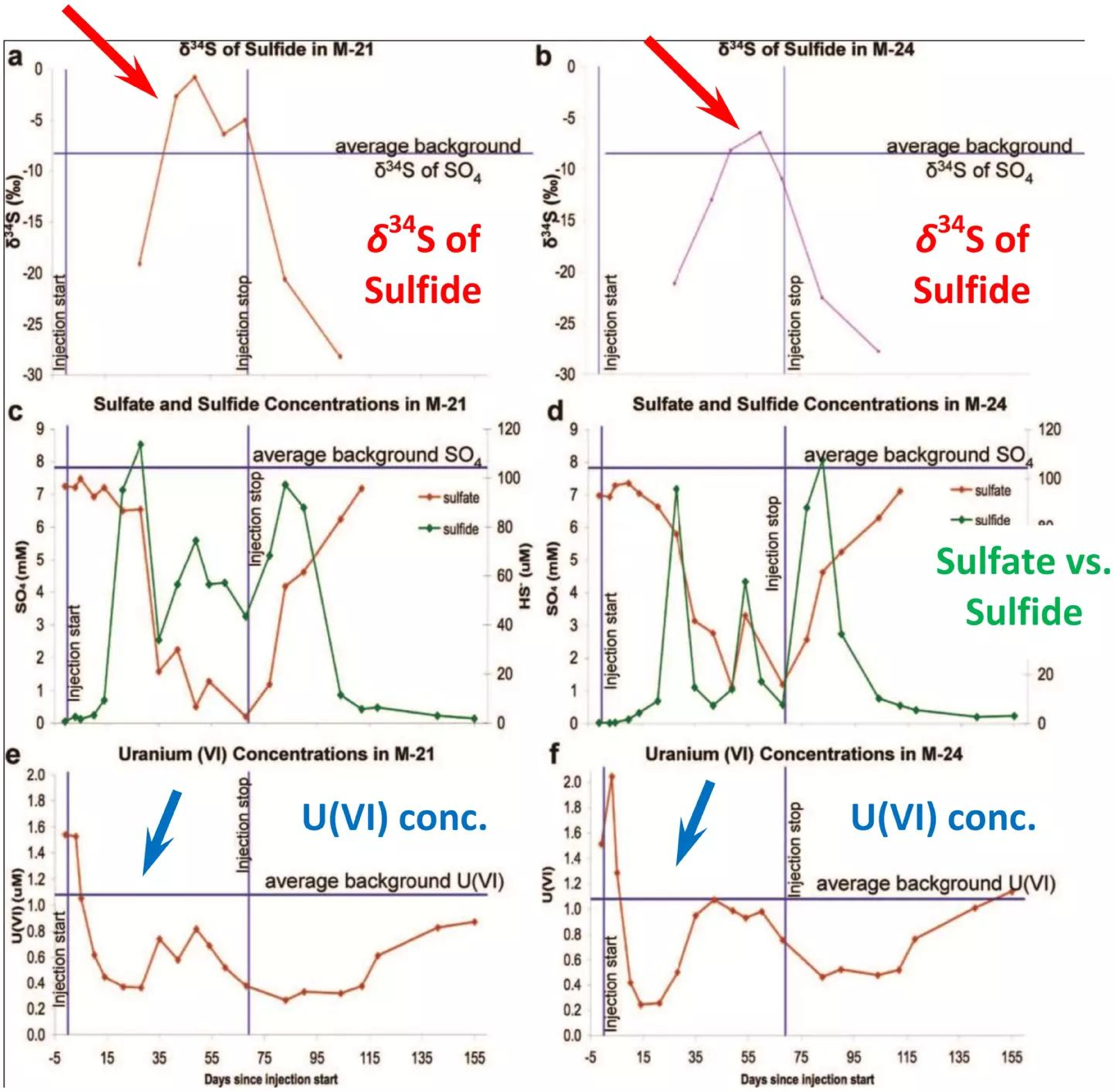


FIGURE 4. Stable sulfur isotopes of sulfide vs. time for a) well M-21 and b) well M-24. Sulfate and sulfide concentrations for c) well M-21 and d) well M-24 and Uranium (VI) concentrations for e) well M-21 and f) well M-24 are also given for comparison. Background δ^{34}S of SO_4, sulfate, sulfide, and uranium (VI) concentrations are given as averages of all measurements in well B-05 throughout the course of the experiment. The background value for sulfide concentration (not labeled) is 1.25 μM.

"In wells M-21 and M-24, the highest levels of U(VI) removal occurred in association with concomitant Fe(II) and sulfide production. Microbial community analysis during this period of the amendment showed that the groundwater microbial population was still dominated by Fe-reducing bacteria (i.e., *Geobacter* spp.) though sulfide concentrations and sulfur isotopes indicate increasing BSR activity. This suggests that maximum uranium removal may have been achieved in association with FeS precipitation (*44*), a connection supported by the sulfur isotope data …"

Comment: it is interesting to note that while Uranium ^{238}U/^{235}U U(VI)$_{(aq)}$ isotope ratios were changing during reduction of U(VI)$_{(aq)}$ to U(IV)$_{(s)}$ at the Rifle site (Bopp et al. 2010 --- discussed earlier), roughly in parallel, Sulfur was being reduced from Sulfate to Sulfide and undergoing "fractionation" which (for a period of time) significantly increased the δ^{34}S of Sulfide reduction products (see a. and b. in Fig. 4 above) relative to the average measured background δ^{34}S of SO_4. *Alternatively, a sequence of LENR ULM neutron captures such as ^{32}S -> ^{33}S -> ^{34}S would also be consistent with this experimental data.*

"Variations in $^{238}U/^{235}U$ in Uranium ore deposits: isotopic signatures of the U reduction process?"
C. Bopp et al.
Geology **37** pp. 611 - 614 (2009)
Source URL = (partial extract) ftp://rock.geosociety.org/pub/reposit/2009/2009143.pdf

Abstract: "The ability to measure $^{238}U/^{235}U$ to high precision presents an important new opportunity to study the fate and transport of uranium in the environment. The ratio of $^{238}U/^{235}U$ was determined by multicollector–inductively coupled plasma–mass spectrometer in six uranium ore samples representing two different classes of deposits. Significant offsets in $^{238}U/^{235}U$ are observed between uranium ores precipitated from groundwaters at low temperature versus hydrothermal deposits precipitated at high temperatures, reinforcing an observation made previously but lacking the needed precision. Specifically, tabular sandstone-type uranium deposits were found to be depleted in ^{235}U, with a total offset between low-temperature deposits and higher temperature deposits of ≈1.0‰. We attribute this offset to reflect a temperature-dependent fractionation related to the nuclear field shift effect during chemical reduction of uranium in ambient temperature groundwaters."

Comment: in this publication, Bopp et al. report significant differences in $^{238}U/^{235}U$ ratios between natural uranium ores located in sandstone deposits formed at low temperatures versus those found in hydrothermal magmatic deposits formed at much higher temperatures.

Patent activity - interestingly, the United States Patent and Trademark Office has recently issued a patent concerning Uranium enrichment utilizing microorganisms as follows:

US 7,452,703 B1
URANIUM ENRICHMENT USING MICROORGANISMS
Date of Patent: November 18, 2008
Inventors: K. Czerwinski and M. Polz
Assignee: Massachusetts Institute of Technology
Source URL = http://www.freepatentsonline.com/7452703.pdf

Abstract: "The present invention provides methods for separating isotopes of actinide elements such as uranium using microorganisms, e.g., metal or sulfate reducing bacteria. The microorganisms reduce the actinide element to form a precipitate, which contains a greater proportion of the lighter isotope relative to the heavier isotope than the starting material. The precipitate may be collected, reoxidized, and subjected to multiple rounds of enrichment. Alternately, separation processes not requiring formation of a precipitate may be used. The invention also features cell-free systems for isotope separation. The invention further provides compositions produced according to the foregoing methods, including compositions comprising enriched uraninite."

"*The ecological behavior of Plutonium and Americium in a freshwater ecosystem*: *Phase II – Implications of differences in Transuranic isotopic ratios"*
R. Emery and T. Garland
Report: BNWL-1879 UC-48 December 1974
Special Distribution – 30 pages of text and figures
Battelle - Ecosystems Department
Pacific Northwest Laboratories
Richland, WA 99352
Source URL = http://www.osti.gov/bridge/servlets/purl/4218018-vthkUT/4218018.pdf

Abstract: "The ecological behavior of Pu and Am in a freshwater processing waste pond has been studied since July 1973 to characterize the pond's limnology and define the isotopic distributions in this ecosystem (Emery et al. 1974). Results from this study show that the history of Transuranics discharged to the pond has created a complex combination of source terms and isotopic ratios. Since these source terms and chemical forms entering the pond have not been clearly defined, a material balance for Pu and Am in this ecosystem cannot be developed. Relatively large quantities of ^{238}U have been released to the pond, and these, along with unidentified levels of ^{232}Th and ^{228}Th create a problem of interpreting the ecological behavior of Pu and Am in this ecosystem. Ratios of ^{238}Pu to $^{239,240}Pu$ found in the pond are much higher than those in the processing wastewaters released to the pond. Ratios of ^{238}Pu to ^{239,240}Pu and ^{241}Am to ^{239,240}Pu are significantly higher in the pond biota than in the sediments, suggesting a common source of availability to the biota. This contention is supported by the rapid establishment of these ratios in goldfish (*Carassius*) which were experimentally introduced in to the pond. Ratios of Am to Pu in the pond biota are also significantly different from those of the sediments but to a lesser degree than ratios of ^{238}Pu to ^{239,240}Pu and ^{241}Am to ^{239,240}Pu. A wide range of particle sizes of ^{238}Pu and ^{239,240}Pu may exist in the pond sediments. The pond sediments, however, may not be the major source of 'available' Pu and Am because results of this study suggest that sediments in a trench carrying the processing wastes to the pond may be the primary source." **[A variety of different nuclear wastes arising from Plutonium and Uranium processing operations on the Hanford Reservation have been discharged into this so-called "U-pond" since it was dug for the Manhattan Project in 1944]**

Direct quotes: "CHEMICAL FORMS OF ISOTOPES IN AQUEOUS WASTES - The only information concerning the chemical forms of Pu and Am in aqueous Pu processing wastes are extrapolations of forms generated in the processing plants. These include Pu and Am oxides, metals, nitrates, polymers, hydroxides, and organic complexes. The U present in the pond was probably discharged primarily of UO_3, U_30_8, or $U0_2(N0_3)_2$."

"Because the source terms and chemical forms of the actinides entering and present in the pond cannot be clearly defined, it i s important to be realistic about the limitations to which the data generated by this study may be used**.** It is now clear that a material balance for Pu or Am between levels found in the pond and those entering and leaving the pond cannot be developed."

"Adding further complexity to the problem of making ecological interpretations of the Pu and Am values in U-Pond is the existence of comparatively high levels of U in the pond sediments … A source of other radionuclides in U-Pond was a uranium processing plant which operated from 1953 to 1957. This plant added very large quantities of ^{238}U, about 1400 kg (3000 Ibs, Anderson 1973), in addition to unknown quantities of ^{232}Th and ^{228}Th during its operation. Processing and purification of several batches of Th fuel elements, used for the production of ^{233}U, could have released both ^{232}Th and ^{228}Th. An alternative source of ^{228}Th is from the decay of ^{232}U, which is found in the ^{233}U product as an impurity. Levels of these isotopes in the pond have not been determined."

[Here comes one of the fascinating anomalies contained in this report] "If the 1400 kg of ^{238}U, the recorded discharged quantity through 1966, were evenly distributed across the pond's bottom to a depth of 10 cm, the average concentration in the pond sediment would be about 170 μg of ^{238}U/g of sediment. Levels of ^{238}U measured in 51 samples of U-Pond sediment (down to 10 cm) show a mean concentration of 647 μg /g (range of 70 to 2300 μg/g) or about 216 pCi of ^{238}U/g dry weight of sediment. This ^{238}U content in the sediments amounts to about four times more ^{238}U than the recorded discharge into the pond system would allow. At present there are no explanations for this difference. Whichever the case, this level of U in the pond becomes a matter of concern when attempting to interpret the ecological behavior of Pu or Am in this ecosystem. Since U is more mobile than Pu in aquatic systems and is also toxic to many organisms (Bowen 1966), determinations of the behavior of Pu or Am in this pond could be impaired by superimposed chemical and possibly radiological effects of ^{238}U on biota."

"Concerns over the presence of Th in the pond, particularly ^{228}Th is primarily due to the fact that ^{228}Th plus its daughters can bias the analytical determination of ^{238}Pu in a way to cause the levels to appear higher than they actually are. However, procedures used for Pu analysis have taken the levels of Th into account, and values of Pu have been corrected for the presence of Th."

"ISOTOPIC RATIOS OCCURRING IN U-POND - If ratios of ^{238}Pu to ^{239,240}Pu and ^{241}Am to ^{239,240}Pu which occur in U-Pond biota are compared to those occurring in the sediments, it is obvious that there are significant differences (Figs. 5 and 6). In both cases the ratios of these isotopes are greater in the biota than in the sediments … Another possible explanation is that the sampling and processing of the sediment caused a fractionation of the ^{238}Pu and ^{239,240}Pu isotopes."

"When the pond sediments are sampled repeatedly in the same location over a short time interval, a substantial variation in the concentrations of ^{238}Pu and ^{239,240}Pu is observed**."**

"Since the input of Pu and Am to U-Pond has been intermittent over a long period, it is not possible to calculate precisely what the ^{241}Am to ^{239,240}Pu ratio should be. However, most of the U-Pond sediments sampled have ratios that are greater than the expected maximum for weapons grade Pu (i.e., about 0.26), thus providing evidence that there are other sources of ^{241}Am."

"Although weapons grade Pu processing operations should have discharged wastes containing ratios of ^{238}Pu to ^{239,240}Pu of about 0.02, these ratios in the pond sediments are much higher, about 1.0 **[roughly 50x larger than would be expected]**, indicating the contribution of ^{238}Pu production processing after the early 1960's."

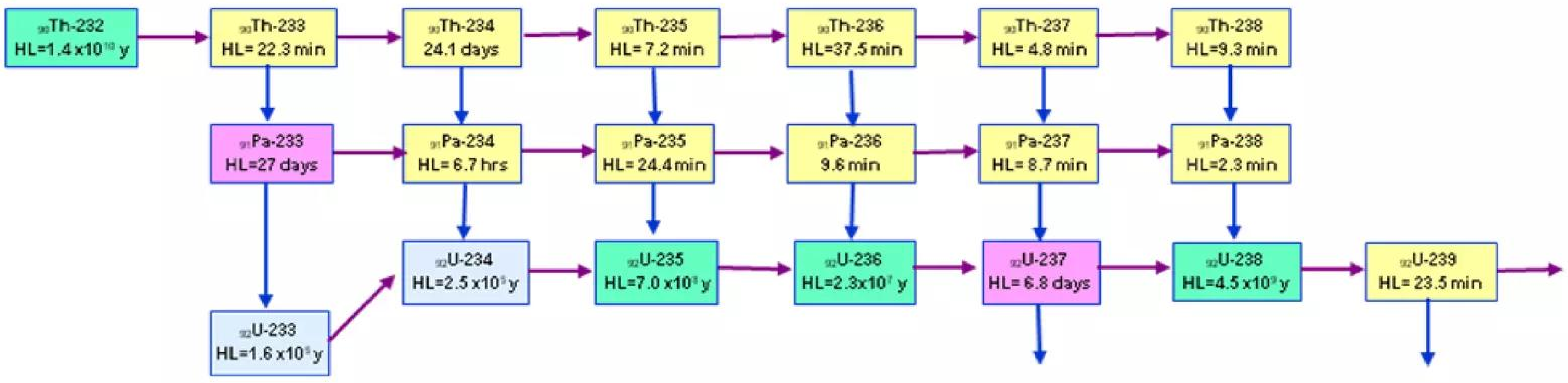
Comment: Battelle report BNWL-1879 contains some extremely interesting isotopic data as well as some fascinating mysteries. It includes a very honest discussion of results in which Emery and Garland were quite candid about inherent underlying limitations in their data; they also readily admitted that some of their observed results were poorly understood if not inexplicable as of 1974.

Perhaps the observed very large Uranium-238 anomaly was simply a result of very bad recordkeeping on exactly what types and quantities of nuclear wastes were dumped into the Hanford U-pond over the years (although the authors would presumably have carefully investigated this possibility); or maybe the wide variability in Plutonium and Americium isotope ratios were produced by particle-size sorting and/or unremarkable chemical fractionation processes of one type or another. However, what if those were not the case? Might there be another alternative explanation that is also consistent with such data?

In particular, if one were to shift conceptually from a strictly chemical paradigm to the W-L LENR paradigm can it provide some useful insights that can help make better sense out of this enigmatic data? The answer is yes; I believe it can.

First, please refer back to the LENR Actinide nucleosynthetic network shown in Figs. 1 and 2; also recall our previous discussion of differences in Uranium isotope shifts between the laboratory results of Rademacher et al. (2006) and the Rifle, Colorado, field study of Bopp et al. (2010) in which ^{232}Th 'seed nuclei' were initially present in the Rifle mine tailings but not in the controlled laboratory environment.

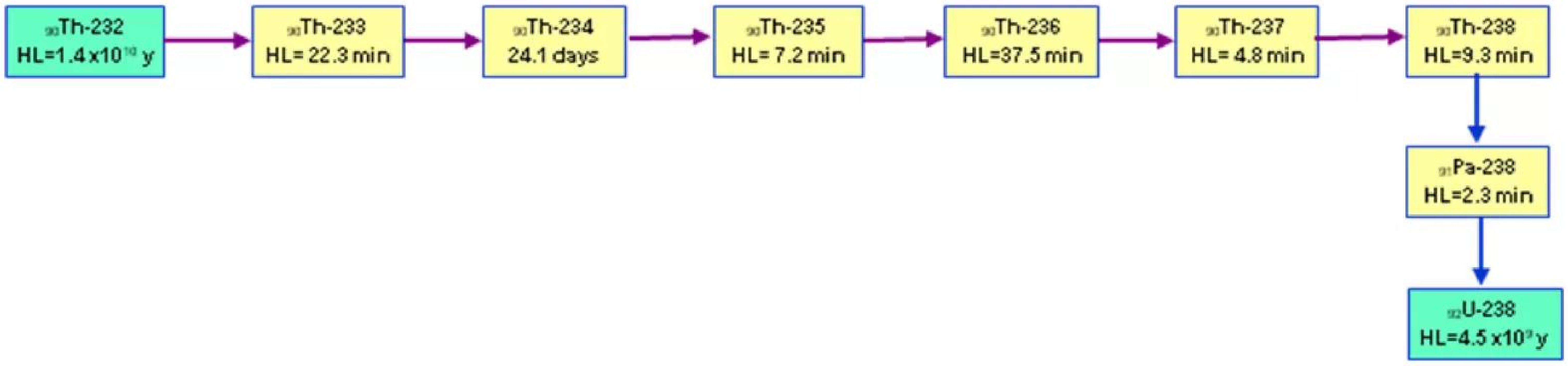
Now, based on the availability of substantial (albeit indeterminate) quantities of ^{232}Th to microorganisms assuredly present in the U-pond wastes at Hanford, if the LENR Actinide network was in fact operating within that pond, it is clear from the network subset shown below that ^{238}U can be produced from ^{232}Th:



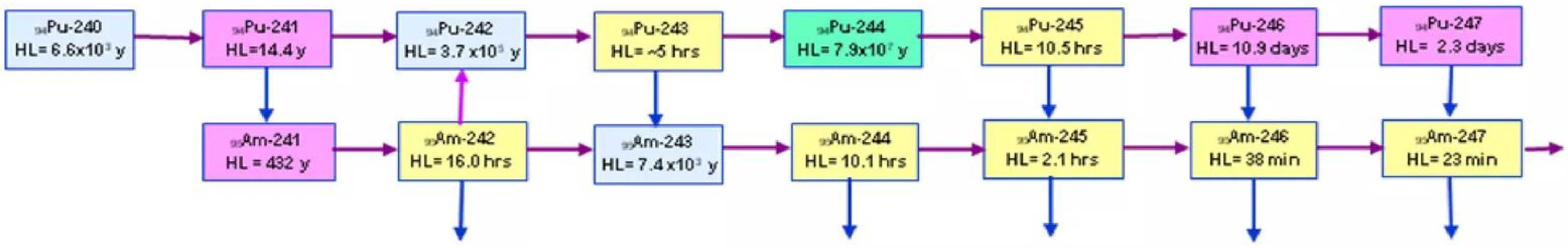
Given the presence of such an LENR network, these hypothetical nucleosynthetic pathways could easily have produced substantial amounts of ^{238}U over the period of ~66 years since waste dumping in the U-pond began in 1944, even at relatively low fluxes of LENR ULM neutrons.

On the other hand, since neutron captures on 'seed' nuclei such as ^{232}Th are lightning fast processes (on the order of picoseconds) such LENR networks can potentially operate at amazingly rapid rates of nucleosynthesis if the ULM neutron fluxes are high enough (for whatever reason).

For example, in the theoretical high-neutron-flux network subset shown below, readily measurable ^{238}U production from ^{232}Th can occur in as little as an hour or two. Under such conditions (which may well be relatively infrequent in Nature) LENR network branching, isotopic products and isotope ratios can vary substantially over short time scales:



An array of Americium isotopes can also be produced from ^{240}Pu by the LENR network as follows:



Thus in theory, over time an LENR ULM neutron-catalyzed Actinide nucleosynthetic network is capable of explaining the elemental anomalies (too much ^{238}U) and various isotope shifts (unusual ratios, substantial changes over short time-scales) observed by Emery and Garland in the U-pond at Hanford. As in the case of Rademacher et al.'s (2006) results versus those of Bopp et al. (2010), by applying a nuclear paradigm seemingly confusing and hard-to-explain data can suddenly become readily comprehensible.

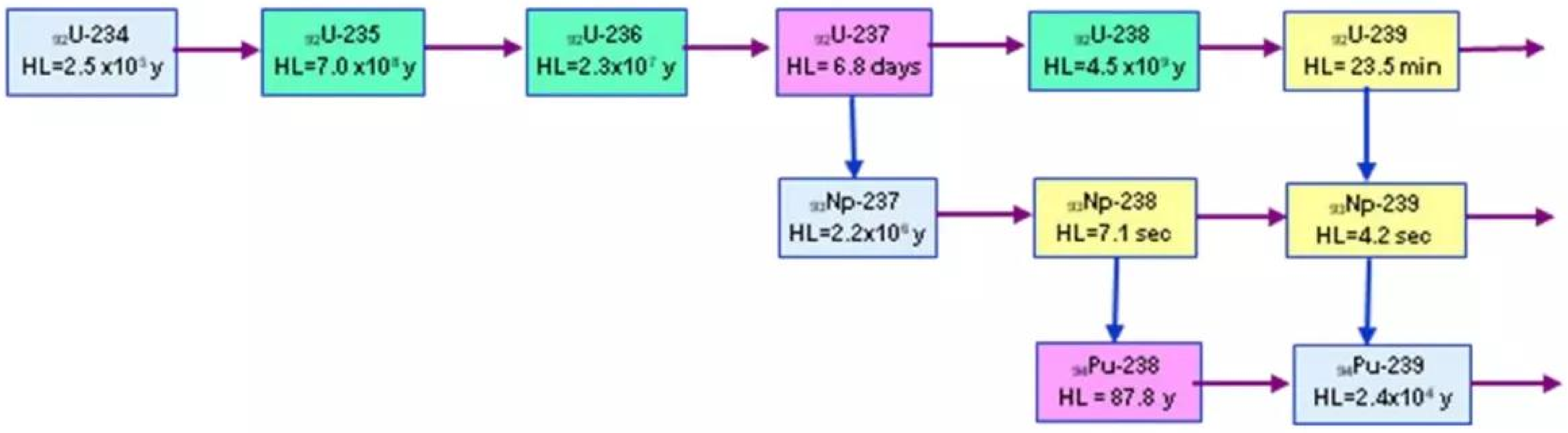
Earthly Plutonium (Pu) isotopes – is all Plutonium on earth strictly man-made (except in the case of Pu-244, from supernova debris entering the earth's atmosphere) or could biological or abiological LENRs also be responsible for creating some indeterminate portion of this element via transmutation?

Plutonium (Pu) is inherently unstable and has no isotopes with extremely long half-lives. Thus, until now it has been near universally assumed that virtually any Plutonium found on earth, at whatever oxidation state in whatever chemical form, is anthropogenic in origin. There are sound reasons for this longstanding belief: with the exception of somewhat less unstable Pu-244 (half-life = 7.9 x 10^7 years), all other known Plutonium isotopes have comparatively short half-lives (see Figs. 1 and 2 herein) and simply would not survive in any measurable quantities over long geological time periods, unlike Uranium and Thorium.

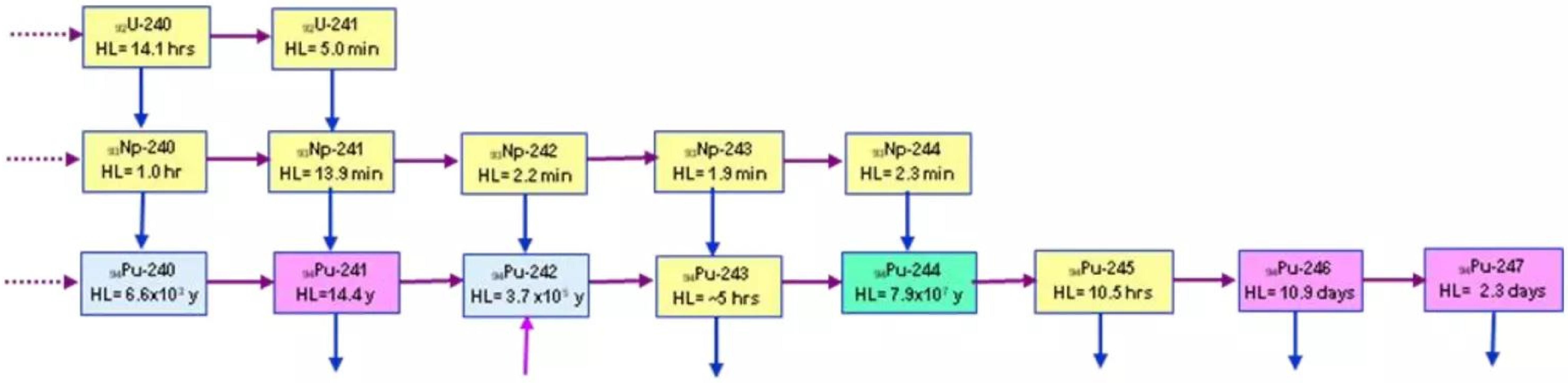
Interestingly, as shown in Figs. 1 and 2, Plutonium isotopes can theoretically be produced by the W-L LENR Actinide nucleosynthetic network via a series of fissionless ULM neutron captures and radioactive decays. If such a transmutation network were in fact operating biologically or abiologically somewhere out in Nature, *one would expect to observe examples of anomalous Plutonium that are questionably man-made in origin, albeit perhaps comprising a minuscule percentage of total Pu found in the environment.*

Specific segments of the LENR Actinide network that could potentially be involved in natural Plutonium production are as follows:

From Fig. 1 - Uranium LENR network segment leading to Neptunium and then to Plutonium:



From Fig. 2 - LENR network continues through region of increasingly heavy Plutonium isotopes:



Experimental evidence – are there any published, peer-reviewed experimental reports which suggest that anomalous Plutonium of unclear origin might be present in Nature?

The answer to this question is a qualified yes, with a strong caveat that data published to date is definitely suggestive, but not conclusive. That said, I think the question is sufficiently interesting and important for experimentalists to consider trying to determine whether anomalously produced Plutonium really exists somewhere out in the terrestrial environment, simply waiting to be discovered and published.

Before further discussing the controversial possibility that non-anthropogenic, non-supernova Plutonium could potentially be present at different geospatial locations in the terrestrial environment, let us ask an important preliminary question: are there any microorganisms that are known to reduce and/or otherwise interact with Plutonium? The answer is definitely yes as shown in two papers cited below:

"***Plutonium speciation affected by environmental bacteria***"
M. Neu et al.
Radiochimica Acta **93** pp. 705 – 714 (2005)
Source URL = free copy unavailable

Abstract:"Plutonium has no known biological utility, yet it has the potential to interact with bacterial cellular and extracellular structures that contain metal-binding groups, to interfere with the uptake and utilization of essential elements, and to alter cell metabolism. These interactions can transform plutonium from its most common forms, solid, mineral-adsorbed, or colloidal Pu(IV), to a variety of biogeochemical species that have much different physico-chemical properties. Organic acids that are extruded products of cell metabolism can solubilize plutonium and then enhance its environmental mobility, or in some cases facilitate plutonium transfer into cells. Phosphate- and carboxylate-rich polymers associated with cell walls can bind plutonium to form mobile biocolloids or Pu-laden biofilm/mineral solids. Bacterial membranes, proteins or redox agents can produce strongly reducing electrochemical zones and generate molecular Pu(III/IV) species or oxide particles. Alternatively, they can oxidize plutonium to form soluble Pu(V) or Pu(VI) complexes. This paper reviews research on plutonium-bacteria interactions and closely related studies on the biotransformation of uranium and other metals.

"***Plutonium(IV) reduction by the metal-reducing bacteria Geobacter metallireducens and Shewanella oneidensis MR1***"
H. Boukhalfa et al.
Applied and Environmental Microbiology **73** pp. 5897 – 5903 (2007)
Source URL = http://aem.asm.org/cgi/reprint/73/18/5897

Abstract: "The bacterial reduction of Actinides has been suggested as a possible remedial strategy for Actinide-contaminated environments, and the bacterial reduction of Pu(VI/V) has the potential to produce highly insoluble Pu(IV) solid phases. However, the behavior of Plutonium with regard to bacterial reduction is more complex than for other Actinides because it is possible for Pu(IV) to be further reduced to Pu(III), which is relatively more soluble than Pu(IV). This work investigates the ability of the metal-reducing bacteria *Geobacter metallireducens* GS15 and *Shewanella oneidensis* MR1 to enzymatically reduce freshly precipitated amorphous Pu(IV) (OH)4 [Pu(IV)(OH)4(am)] and soluble Pu(IV)(EDTA). In cell suspensions without added complexing ligands, minor Pu(III) production was observed in cultures containing *S. oneidensis*, but little or no Pu(III) production was observed in cultures containing *G. metallireducens*. In the presence of EDTA, most of the Pu(IV)(OH)4(am)present was reduced to Pu(III) and remained soluble in cell suspensions of both *S. oneidensis* and *G. metallireducens*. When soluble Pu(IV)(EDTA) was provided as the terminal electron acceptor, cell suspensions of both *S. oneidensis* and *G. metallireducens* rapidly reduced Pu(IV)(EDTA) to Pu(III)(EDTA) with nearly complete reduction within 20 to 40 min, depending on the initial concentration. Neither bacterium was able to use Pu(IV) (in any of the forms used) as a terminal electron acceptor to support growth. These results have significant implications for the potential remediation of plutonium and suggest that strongly reducing environments where complexing ligands are present may produce soluble forms of reduced Pu species."

Comment: thus, existing published data shows that at least one type of common, widely-distributed soil microorganism, *Geobacter*, is definitely known to reduce Uranium, Sulfur, and Plutonium. Also, *Geobacter* was present in the controlled laboratory experiments of Rademacher at al. (2006), Bopp et al.'s Rifle Uranium isotope field study (2010), Druhan et al.'s Rifle field study on Sulfur isotopes (2008), and was almost undoubtedly present in the Hanford U-pond environment studied by Emery and Garland (1974). *Noting that I have not fully surveyed all the relevant literature yet, I would hazard a guess that Shewanella and a number of other types of common microorganisms probably possess the same capabilities.*

Question of whether non-anthropogenic terrestrial Plutonium truly exists – dates back to the 1960s:

Questions about whether non-man-made, non-supernova Plutonium really does exist somewhere out in the terrestrial environment began in the USSR in the 1960s and continued through the 1970s and mid-1980s. Still unresolved, from circa 1990 onward this possibility has been pretty much dismissed by geochemists; it is now near-universally assumed that, for the most part, natural Plutonium does not exist.

Apart from a lack of conclusive experimental data, a major underlying, unstated part of the problem with accepting this possibility is that there has been no plausible abiological or biological mechanism capable of producing Plutonium isotopes from other elements under conditions that prevail on the earth of today.

Importantly, the theoretical dimension of this longstanding controversy has recently changed: thanks to development of the Widom-Larsen theory of LENRs (which is based on known, well-accepted physics --- in effect, it integrates many-body collective effects with electroweak theory under the Standard Model) a ULM neutron-catalyzed mechanism now exists that could potentially produce Plutonium from other elements and operate in terrestrial environments under comparatively 'mild' physical conditions. In other words, W-L theory suggests that stars, supernovae, natural Oklo-type reactors, and man-made fission reactors are not necessarily required to perform significant and detectible amounts of nucleosynthesis.

The history of this old controversy about reports of natural Plutonium has been chronicled in the following selected papers that were published in peer-reviewed journals in Russia and elsewhere:

"*Has element 108 been discovered?"*
V. Kulakov
Atomic Energy **29** pp. 1166 - 1168 (1971)
Translated from *Atomnaya Energia* **29** pp. 401 – 402 (1970)

Source URL = http://www.springerlink.com/content/p042g7671h185055/fulltext.pdf

Direct quotes: “Sergenium is genetically related to Pu^{239}. In some preparations of uranium isolated from molybdenite, and in some isolated rocks, including material from the upper mantle of the earth, V.V. Cherdyntsev has discovered an excess of that plutonium isotope. Clearly, Pu^{239} itself could not have been retained in nature up to the present day. But it could be formed in uraniferous ores in negligible quantities (~10^{-10} fraction of the U^{235} present) because of the presence of neutrons deriving from spontaneous fission of the uranium. Any excess over that fraction means, in Cherdyntsev's view, that the Pu^{239} discovered in the minerals has accumulated as a result of the decay of its parent, which would have to be some more remote transuranium element (i.e., sergenium).”

“From the foregoing discussion, only two facts emerge which reflect direct, and consequently more or less reliable, observations. This applies, first and foremost, to establishing the excess amount of Pu^{239} in some rocks. Here, however, as Prof. Cherdyntsev himself has noted, it is not possible to exclude from consideration Pu^{239} scattered over the earth's surface as a result of nuclear weapons tests. The amount of plutonium scattered over the globe in that manner does not yield to any exact estimate. Secondly, there is the problem of detecting α-emitters of ~4.6 MeV particle energy. The presence of such an emitter in nature appears probable. Many α-emitters with a similar, or slightly different, α-particle energy have been discovered in rocks since the Twenties. Some of these have been identified subsequently as rare earths, some have defied a full explanation up until recently, and at least in some cases the problem has been resolved as simply one of experimental errors. We should note that the energy of α-particles from some of the emitters discovered by V.V. Cherdyntsev has varied over a wide range (4.4 to 4.7 MeV) in his several articles published on the topic. A more precise determination of the energy is of the utmost importance, since α-emission peaks of U^{235} (E_α = 4.35 MeV) and of Np^{237} (E_α = 4.76 MeV) are to be found in the immediate vicinity of the spectral region. Either of these two nuclides would be capable of simulating the α-emitter, if the errors in the energy determination were appreciable. Note also that, in experiments conducted with the same specimens at Dubna (at JINR), there were no α-emitters of 4.6 MeV particle energy detected. These negative results are also reported by the renowned American physicist A. Ghiorso. At the same time, J. Adams has confirmed the existence of the emitter in fractions isolated from ancient granites.”

Comment: Prof. Viktor Cherdyntsev was a famous Russian geochemist with an excellent reputation and long experience who spent many years collecting experimental field data on Actinides found in Nature. He served on the staff of the Radium Institute of the Academy of Sciences of the USSR from 1930 to 1944 and on the staff of the Institute of Astronomy and Physics of the Academy of Sciences of the Kazakh SSR from 1944 to 1960. From 1946 to 1960, he was also the head of the subdepartment of experimental physics at the Kazakh University in Alma-Ata; he became a professor at the university in 1948. In 1960 he became the head of the laboratory of absolute age determination at the Geological Institute of the Academy of Sciences of the USSR in Moscow.

Cherdyntsev’s main works dealt with the geochemistry of isotopes, nuclear geophysics, and radiogeology. Cherdyntsev was the first to substantiate the origin of heavy elements in massive stars. In 1954, together with P. I. Chalov, he discovered the spontaneous fission of the uranium isotopes ^{234}U and ^{238}U; the spontaneous fission of the two uranium isotopes is called the Cherdyntsev-Chalov effect. Cherdyntsev demonstrated the importance of radioactive and radiogenic isotopes, such as ^{4}He and ^{40}Ar, in the study of the earth’s upper mantle.

By 1970, Cherdyntsev had been publishing field studies reporting anomalous ‘natural’ Plutonium in Russian language journals for years. Well aware of the comparatively short half-lives of Plutonium isotopes and worldwide bomb testing contamination issues, he was groping for a mechanism that could explain his data; at that time, no processes were known that could possibly create neutrons in terrestrial environments (natural fission reactors in were not discovered in Oklo, Gabon, until 1972). In April 1970, to explain his Pu^{239} data Cherdyntsev proposed the existence of a long-lived superheavy element 108, that he named “Sergenium,” from which ^{239}Pu could hypothetically be produced over time via a decay chain.

Unfortunately, Cherdyntsev died in August 1971 at 59 years old, and was unable to continue in the debate with Academician G. B. Flerov and others who dismissed his ideas and data. *While proposed “Sergenium” was later proven wrong, most of his reported ^{239}Pu anomalies have never been explained.*

"***Geochemical isotopic anomalies and the hypothesis of natural nuclear reactors***"
R. Prasolov
Atomic Energy **36** pp. 61 - 83 (1974)
Translated from *Atomnaya Energia* **36** pp. 57 – 59 (1974)
Source URL = http://www.springerlink.com/content/m5q0624066217t33/fulltext.pdf

Direct quotes: "Some experimental data have by now been accumulated concerning variations in the abundance of isotopes of a number of elements found in natural substances. For example, deviations of 3-20% from the Clarke isotope ratios have been observed in the case of ^{2}H, ^{13}C, ^{18}0, and other isotopes. These differences may be attributable to isotopic fractionation in gravitational and temperature fields, to diffusive and biogeochemical separation, and also to phenomena involved in evaporation and radioactive recoil following alpha disintegration [1-4]."

"However, in addition to the minor differences mentioned above, observations reveal some very substantial isotopic anomalies which are difficult or impossible to explain on the basis of the above-mentioned mechanisms. For example, isotopic variations of the order of 10^{3} - 10^{10}% [1, 3-9] have been observed in the case of isotopes of helium, neon, xenon, samarium, plutonium, and other elements. One of the most probable reasons for such large deviations may be the occurrence of various nuclear reactions, including reactions of the (n, γ) type, in natural neutron fields, as the result of cosmic radiation, spontaneous fission of heavy nuclei, and other neutron-generating processes, which have been discussed in [3, 5]. It must be noted that some studies for determining the value of the natural neutron background (in particular, on the basis of the accumulation of fission-produced xenon in minerals) have shown a discrepancy between the calculated and measured values of the background, with the calculated neutron flux sometimes greater by several orders of magnitude than the measured value."

"In the light of these facts, an explanation of the observed anomalies should be sought on the basis of the hypothesis of natural nuclear reactors (NNR) [3, 5, 8, 10, 11]; the anomalies conform to the laws deduced from this hypothesis. In particular, if other conditions are equal, there should exist a correlation between the effective neutron cross section and the value of the isotopic deviation for each particular isotope. We give below the results of a study bringing out such a correlation in the form of a single-parameter function φ = f(σ); this is only a crude first approximation and does not take into account the length of time the neutron field has been acting, the probable migration of isotopes under geophysicochemical influences, and other factors."

"The most probable kinds of NNR are those which involve thermal neutrons produced by the fission of ^{235}U; for this reason, the effective neutron cross section, σ, for (n, γ) reactions, or the partial cross section of the (n, f) reaction for fragment products, was taken on the basis of known data used in reactor design for thermal reactors [12, 13], although in theory a number of anomalies may be caused by fluxes of intermediate and fast neutrons, resonance-absorption phenomena, etc."

"The numerical value of the isotopic deviation φ was found from the ratio $\varphi = (x_{max} - x_{min})/x_{min}$, where x_{max} and x_{min} are the maximum and minimum experimental values of the percentage of the isotope under consideration in the natural mixture of isotopes of a particular specimen, as given in a number of studies [1-9], or else the extremum values of ratios of the type ^{234}U/^{238}U."

"Since the generation of a particular isotope from different original isotopes may have different values of σ, we first found the value of the Clarke factor of the element -- the abundance of the isotope -- the cross section of the nuclear reaction, on the basis of which we can make a rough choice of the cross-section value σ corresponding to a particular value of the parameter φ. We assumed an (n, γ) reaction for all of the isotopes analyzed; an exception was made in the case of ^{140}Ce for which the NNR [8] was found to be more probable with an (n, f) reaction, whose partial cross section (taking into account the yield of fission fragments) is about 30 - 60 b. Despite the relative abundance of experimental data for inert gases, we did not analyze these data in the present study, since the values of φ are very poorly defined, owing to the difficulty of preserving these gases [5]."

Figure 1 from Prasolov (1974) reproduced below reveals a remarkable relationship:

What Fig. 1 shows is that, for the specific array of isotopes selected by Prasolov, there appears to be a reasonably well-behaved relationship between increased effective neutron cross-sections σ of the (n, γ) 'simple' capture reaction versus increased relative isotopic deviations φ as calculated by Prasolov.

This type of causal relationship would be expected if W-L LENR ULM-neutron catalyzed transmutation processes, not just chemical fractionation, were occurring to some indeterminate degree out in Nature.

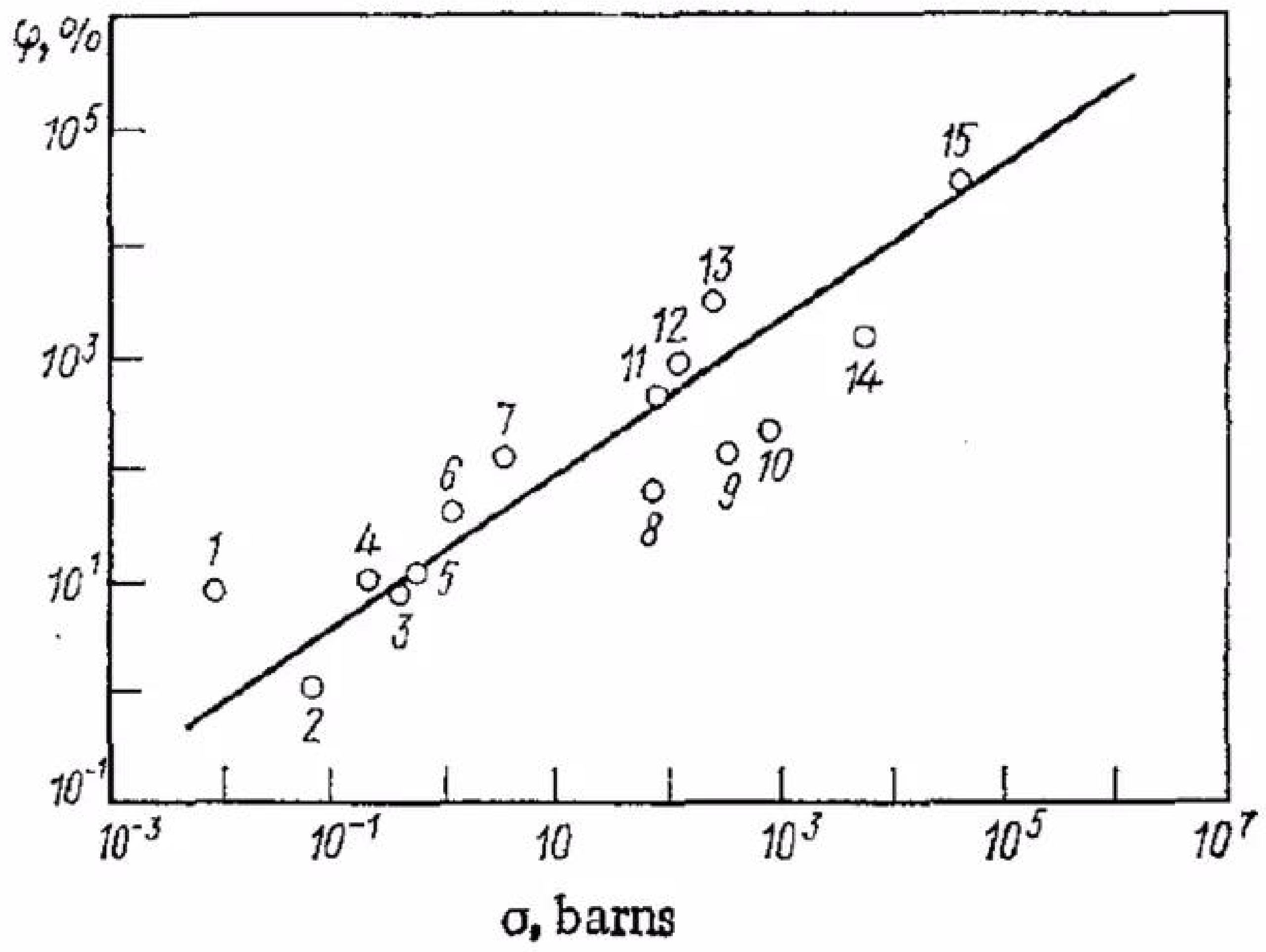


Fig. 1. Relative isotopic deviations as a function of effective neutron cross sections of the (n, γ) reaction: 1) ^{13}C; 2) ^{15}N; 3) ^{18}O; 4) ^{34}S; 5) $^{24}Mg/^{26}Mg$; 6) $^{87}Sr/^{86}Sr$; 7) $^{148}Nd/^{150}Nd$; 8) $^{144}Nd/^{145}Nd$; 9) ^{235}U; 10) $^{6}Li/^{7}Li$; 11) $^{140}Ce/^{142}Ce$; 12) $^{234}U/^{238}U$; 13) $^{143}Nd/^{142}Nd$; 14) $^{151}Eu/^{153}Eu$; 15) $^{149}Sm/^{147}Sm$.

Incredibly, to my knowledge Prasolov's intriguing ideas about the likely existence of "natural reactors" were never really picked-up and embraced by many other researchers; thought to be very rare a la Oklo.

There was another spate of discussion on this topic during the 1980s but that thread appears to have more-or-less died-out after about 1990.

We will now cite several papers from the 1980s in which Paul Kuroda was a coauthor; importantly, he had (correctly) theoretically predicted the existence of natural Oklo-type fission reactors back in 1956.

Tide turns in the mid-1980s; many researchers begin to doubt evidence for 'natural' Plutonium - based mainly on extrapolation of elemental/isotopic data from rain and snow samples collected at one location in Fayetteville, Arkansas, as well as other selected data, Prof. Paul Kuroda et al., then of the Chemistry Dept. at the University of Arkansas, published two papers summarily dismissing the reported anomalous data of Cherdyntsev and others, concluding with sweeping assertions that essentially *all* Plutonium found on earth was simply the result of man-made atmospheric fallout contamination:

"*Plutonium anomaly in nature*"
I. Essien and P. Kuroda
Geochemical Journal **18** pp. 101 – 108 (1984)
Source URL = http://www.terrapub.co.jp/journals/GJ/pdf/1803/18030101.PDF

Abstract: "The $^{239}Pu/^{238}U$ ratios are reexamined in volcanic ashes and rock, uranium mineral and other contemporary terrestrial samples and also in the atmospheric samples. This ratio in rain samples collected at Fayetteville (36'N, 94'W), Arkansas, reaches up to 10^{-5} (atom/atom), which is about 2 orders Of magnitude higher than those of volcanic ashes and rock and about five or six orders of magnitude higher than the $^{239}Pu/^{238}U$ ratio of 10^{-12} (atom/atom) typical of uranium minerals. The data obtained from our measurements indicate that the high $^{239}Pu/^{238}U$ ratios observed in this contemporary terrestrial samples are due to fallout contaminations."

Direct quotes: "This means that ^{239}Pu once existed in nature in far greater quantities than the amounts found in uranium-bearing minerals and rocks today."

[Essien and Kuroda then cite Cherdyntsev and Prasolov]

"It has been reported by a number of investigators, however, that plutonium is highly enriched relative to Uranium in volcanic rocks, tufas and thermal waters. CHERDYNTSEV et al. (1968) have reported that the $^{239}Pu/^{238}U$ ratios reached 1.7 x 10^{-5} (atom /atom) for a sample of limonite from the fumarole field in Karnchatka and 1.8 x 10^{-5} (atom/atom) in a sample of volcanic bomb from the volcanic region of Caucasus. They also found that the $^{239}Pu/^{238}U$ ratio in a sample of water from the fumarole field of Kamchatka was also as high as 1.3 x 10^{-6} (atom/atom) and suggested that these abnormal enrichments of ^{239}Pu relative to ^{238}U were probably due to existence of an unknown transuranium element in nature, which decays into ^{239}Pu. PRASOLOV (1974) then put forward the hypothesis that these isotopic anomalies may be attributable to the Oklo phenomenon, which is still occurring inside the earth's crust today."

"The fact that ^{239}Pu is highly enriched relative to ^{238}U in volcanic samples was confirmed by MEIER et al. (1974), who found that the $^{239}Pu/^{238}U$ ratios in various samples from different locations and eruptions reached values up to 10^{-7} (atom/atom), several orders of magnitude higher than the values of 10^{-11} (atom/atom), which is typical for uranium minerals. They reported that a contamination by ^{239}Pu from fallout was out of the question and suggested that either the Oklo phenomenon or the existence of an unknown superheavy element was the likely cause of the isotopic anomalies observed in plutonium."

[After which Essien and Kuroda state their intention to dismiss ideas of anomalous Plutonium]

"In this paper, the authors intend to report the set of data for the $^{239,240}Pu/^{238}U$ (atom/atom) ratios obtained from the study of these two radionuclides in the atmosphere and contemporary terrestrial samples, in order to show that the anornaly of plutonium isotope in nature is due to fallout contamination."

Comment: I find Essein and Kouda's analysis and attempted extrapolations from their own data faulty. They were, in my view, comparing 'apples and oranges' and for whatever reason appear to have had some sort of an axe to grind. Also, in some ways this paper is scientifically schizophrenic --- on one hand they assert that all Plutonium observed on Nature must be man-made contamination, and on the other they appear to concede that Plutonium *seems to* be emanating from volcanic eruptions (which is, in part, what Cherdyntsev was implicitly also saying about some of his data). Oddly, they then seem to hedge their bets on that notion by concluding that, *"… If one does not assume that the Oklo phenomenon is still occurring inside the earth's crust today, it appears then that the observed plutonium anomaly in nature is a result of the input of man-made plutonium into the earth's environment and by volcanic eruptions."*

What Essein and Kouda do not explain in any qualitative or *quantitative* detail is the precise mechanism(s) by which atmospheric Plutonium fallout from nuclear weapons detonations is somehow *later reinjected into the atmosphere by volcanoes*. On a simplistic basis one would think that 99.99999999+% of the megatons of gaseous and solid materials ejected from a volcano during a massive eruption would come from INSIDE the volcano (where presumably no man-made Plutonium fallout is present). In any case, readers are encouraged to download this (free) paper and decide for themselves.

"***The origin of Plutonium in the atmosphere***"
S. Lee et al.
Geochemical Journal **19** pp. 283 – 288 (1986)
Source URL = http://www.terrapub.co.jp/journals/GJ/pdf/1905/19050283.PDF

Abstract: "Radiochemical measurements of $^{239,240}Pu$ were carried out for a total of 288 individual rain and snow samples collected at Fayetteville (36'N, 94'W), Arkansas, during the period between February 1976 and October 1984. The results indicate that the plutonium inventory in the northern stratosphere has been decreasing during the past several years at a rate corresponding to about 1.2 years for the mean stratospheric residence time of plutonium. However, the fallout of plutonium may not be attributable solely to the atmospheric injections of nuclear debris from the weapons tests. It appears that the recent volcanic eruptions may have played an important role in altering the stratospheric inventory of plutonium."

Direct quotes: "… plutonium data obtained at Fayetteville during the past few years indicated that the fallout of ^{239,240}Pu may not be attributable solely to the atmospheric injections of debris from the nuclear weapons tests. It appeared as if the recent volcanic eruptions may have played an important role in altering the stratospheric inventory of plutonium during the past few years. Meanwhile, a number of reports on the occurrence of 'natural' plutonium in volcanic samples have appeared in the literature. Meier et al. (1974) … reported Pu/U ratios of up to 10^{-7} atom/atom in samples from Kilauea, several orders of magnitude higher than the value of about 10^{-11} to 10^{-12} atom/atom, which is typical for uranium minerals. Fried et al. (1985) have recently reported, however, that they found the ratio in the volcanic samples collected before 1930 and stored in the vaults of the Field Museum of Natural History in Chicago to be about 10^{-10} or less, that is, only about one order of magnitude more than the value observed for uranium minerals. These results indicate that the abnormally high Pu/U ratios reported for the volcanic samples are most likely due to radioactive fallout contamination (Fried et al., 1985; Sandoval et al., 1985; Essien and Kuroda, 1985). This means that the surface of the entire earth is contaminated with 'man-made' plutonium and, whenever a major volcanic eruption occurs, a large quantity of plutonium together with uranium is blown up into the troposphere and the stratosphere resulting in world-wide fallout of plutonium. In an attempt to find an answer to the question concerning the origin of plutonium in the atmosphere, we have decided to re-examine a set of plutonium data obtained in our laboratories prior to 1980 and also after April 1983 together with those reported by Sakuragi et al. (1983) and Sandoval et al. (1985)."

Comment: Although skeptical about their thinking, I concede that Kuroda and his collaborators have a plausible (albeit still dubious and ad hoc) prosaic 'contamination' explanation for the Plutonium and Uranium anomalies that seem to be associated with large volcanic eruptions. That said, is there any other plausible mechanism that might be able to produce nucleosynthesis and isotopic shifts during volcanism?

Answer is yes: neutron-catalyzed LENRs occurring on the surfaces of tiny charged dust grains exposed to ultra-high-current lightning discharges that are well-known to occur within and around huge clouds of hot, dusty gases, ash, and larger ejecta produced during large, violent volcanic eruptions. For example, here are typical images of huge lightning bolts coursing through dusty volcanic clouds during an eruption:

Close-up images of lightning during the Eyjafjallajökull, Iceland Eruptions - 2010

In the context of our published theoretical work on the Widom-Larsen theory of LENRs, lightning discharges inside hot volcanic clouds are roughly analogous to a cross between 'classic' exploding wires (which create hot, dusty plasmas filled with metallic nanoparticles) and high-current electric discharge experiments that have been conducted for years at Proton-21's laboratory facility in Kiev, Ukraine.

Are there examples of similar or otherwise comparable terrestrial environments/situations that are theoretically relatable to lightning discharges in hot volcanic plumes and in which anomalous shifts in Uranium and/or Plutonium isotopes have been observed? – Again, the answer is yes:

First, please examine some relevant background information:

“***Generation of electric fields by dust grains during the explosions of condensed explosive charges in air***”
M. Erokhin et al.
Technical Physics **47** pp. 121 – 124 (2002)
Translated from *Zhurnal Tekhnichesko I Fiziki* **72** pp. 125 – 128 (2002)
Source URL = free copy unavailable

Abstract: “Results are presented from two-dimensional numerical simulations of the generation of electric signals during the explosions of chemical explosive charges in air. The calculated results are compared with the data from experiments carried out under the auspices of the International Science and Technology Center (project no. 835) at the Institute of Geosphere Dynamics (Russian Academy of Sciences), the Research Institute of Pulsed Technique (the Ministry of Atomic Energy of the Russian Federation), and the Zababakhin All-Russia Research Institute of Technical Physics, Russian Federal Nuclear Center (the Ministry of Atomic Energy of the Russian Federation). The electric field is assumed to be generated by charged dust grains produced during the explosion of a condensed chemical explosive charge. Numerical predictions based on the dusty plasma model for describing the parameters of dust grains are found to agree satisfactorily with experiment.”

Comment: it is thus established that dust grains of varied compositions can accumulate charge and build-up very high nano-scale electric fields on their surfaces during chemical explosions. Indeed, this is the likely source of intense lightning discharges often observed in hot, dusty volcanic eruption clouds.

*“**Fractionation of Plutonium isotopes in aerosol fallout at locations of accidental nonnuclear explosions**”*
L. Evterev et al.
Atomic Energy **89** pp. 1004 – 1010 (2000)
Source URL = http://www.springerlink.com/content/j1r65t664325u88n/fulltext.pdf

Abstract: “It is concluded on the basis of a systematization of experimental data that fractionation of plutonium isotopes occurs in nonnuclear explosions with plutonium-containing components. An attempt is made to give a qualitative interpretation of this effect.”

Direct quotes: “The present work is based on a systematization of experimental data, taken from various sources, showing that fractionation of plutonium isotopes occurs during *nonnuclear* explosions with plutonium-containing components and pointing to a correct interpretation of the effect … The data taken together indicate that the effect under study occurs during the explosion and the development of a gas-dust cloud … It is obvious that isotope separation should occur at the stage of homogeneous condensation of the vapor, when very small base particles, highly enriched with ^{238}Pu, ^{240}Pu, and ^{241}Pu, are formed. We believe that this result is a simple consequence of the systematization of the experimental data. However, then the following complicated question arises: How can the plutonium isotopes, having close evaporation and melting temperatures, be separated?”

“The basic event is an explosion of a chemical substance inside an apparatus containing weapons-grade plutonium. The energy released does not exceed an energy equivalent to 22 tonnes of TNT. The center of mass of the apparatus is located at a small height above the surface of soil, concrete, or ice (accident in Thule, Greenland). The gas-dust cloud formed during the explosion rises to an altitude of not more than 1000 m, it is carried in the direction of the wind, and it becomes a source of radioactive fallout, in the form of small particles, onto the Earth’s surface.”

“Most experimental data concern the content of $^{238–240}$Pu and ^{241}Am in samples. The samples are primarily of two types – samples from a 5-cm thick surface layer of soil and samples along the soil profile. There are also samples of the vegetation cover and bottom deposits (Thule, Greenland). The content and activity of the isotopes was determined using standard procedures of radiochemical analysis and radiometry of samples [1]. On the basis of these data, and taking account of the sampling time, the number of nuclei n^e_i of the ith isotope at the moment of the explosion was reconstructed after the event. The procedure is more complicated for ^{241}Am, since on the one hand this nuclide is a daughter product of the decay of ^{241}Pu after the explosion ($T_{1/2}$ = 14.4 yr) while on the other it is initially present in the exploded device …"

[Here they discuss a key piece of data that I think is just one more tip-off suggesting LENRs]

"The observed dependence of the fractionation factor $r_{241,239}$ in close surface samples on the reduced height h^* of the explosion center (straight line in Fig. 4) is also helpful for understanding the possible mechanism of the phenomenon under consideration. The tendency for the average fractionation factor to decrease with increasing height and decreasing amount of dust in the initial cloud of the explosion is clearly seen. The variance of the data is due to the appreciable difference of the situations occurring during the indicated events. Thus, an accidental inactive explosion of a nuclear aerial bomb occurred in Palomars; in the series of experiments in projects 56, 57 and on the area GMX primarily inactive detonations of nuclear devices were conducted; in addition, on area GMX the plutonium additives were dispersed using chemical explosives, and in the experiment Little Feller 2 a nuclear device with an incomplete fission chain reaction was detonated."

[An annotated version of Evterev et al.'s Fig. 4 is reproduced below]

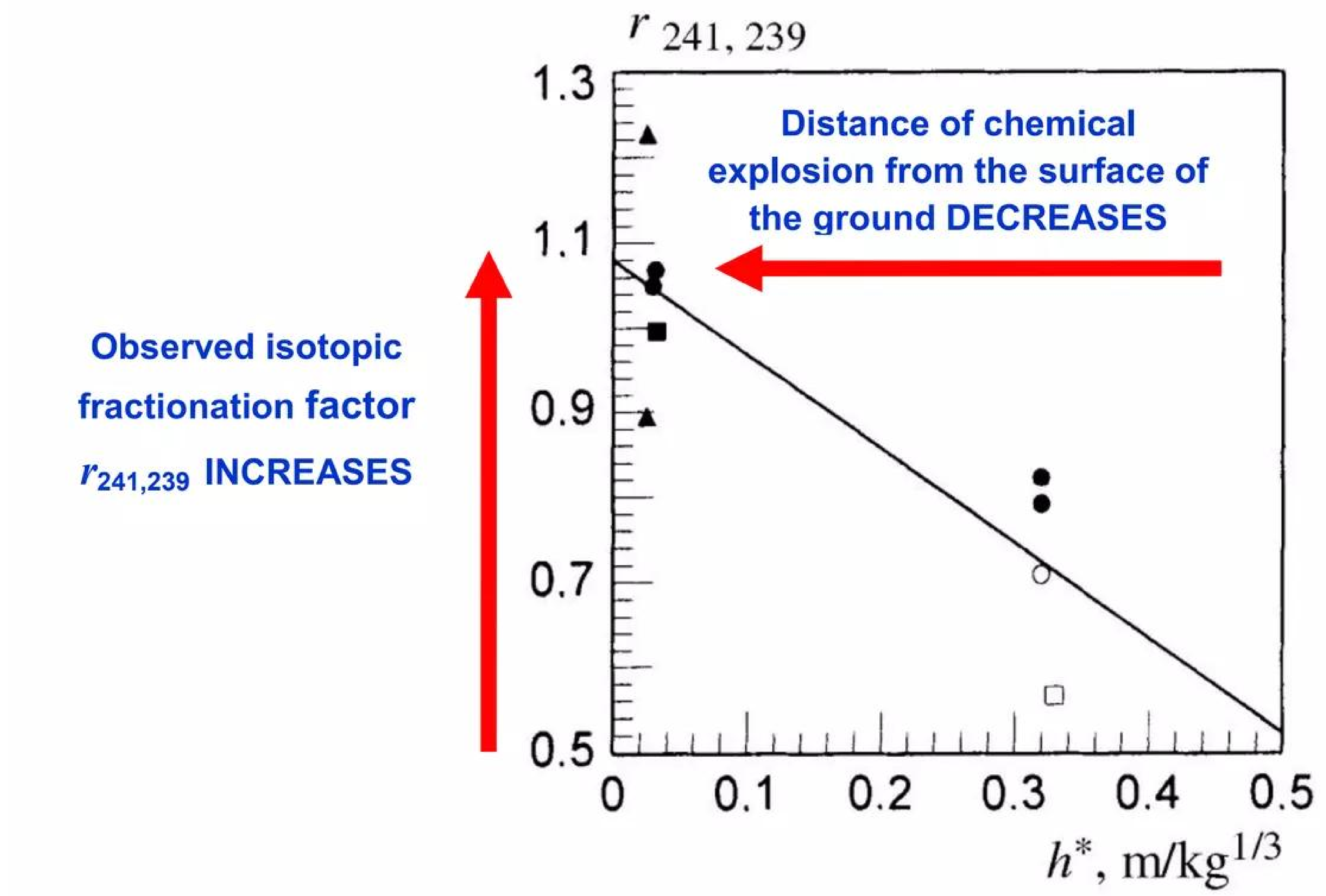


Fig. 4. Fractionation factor $r_{241,239}$ versus the reduced height of the explosion: ●, ❍) projects 56, 57, respectively; ■) Little Feller 2; ❑) GMX; ▲) Palomars.

<u>Comment</u>: this is fascinating data. What happened in the selected cases examined by Evterev et al. was that weapons-grade Uranium and Plutonium materials were subjected to unremarkable chemical explosions for one reason or another, accidental or planned. The startling anomaly that prompted these Russian nuclear weapons researchers to write a paper about these cases was that Plutonium isotope ratios appeared to have *changed* during otherwise ordinary, decidedly nonnuclear chemical explosions. For obvious reasons, isotopic compositions of fissile materials in nuclear weapons are carefully controlled within specified manufacturing tolerances; unexpectedly large variances would be noticed and of concern.

Since these particular explosions were nonnuclear, no significant isotope shifts would have been expected because purely chemical processes simply do not alter isotopes --- they also 'conserve' mass-balances of elements. Under presently accepted nuclear science paradigms, nonnuclear chemical explosions would not be expected to generate fluxes of neutrons large-enough to detectably transmute isotopes. Under that paradigm, if significant amounts of fission aren't occurring, then little or no neutron production/multiplication should occur. Unexpected, anomalous neutron fluxes could be a major problem when dealing with large amounts of 1/*v* fissile materials found in nuclear weapons; unpredictable neutron emissions occurring inside a weapon would raise the specter of catastrophic unplanned detonations.

There are only three likely alternatives with respect to the isotopic data collected on these chemical explosions involving weapons-grade special nuclear materials: (1) Plutonium and Americium isotopes were simply measured incorrectly, no significant isotopic shifts really occurred, and nothing remarkable happened; (2) the isotopes were measured correctly, significant isotopic shifts did in fact occur and were the result of some sort of prosaic, purely chemical isotope fractionation process that is interesting but not paradigm-changing; and (3) the isotopes were measured correctly, significant isotopic shifts did in fact occur and were the result of some sort of unexpected neutron captures on special nuclear materials, in which case something really remarkable happened during these explosions.

Implicitly assuming a strictly chemical conceptual paradigm, Evterev et al. chose alternative 2 and proceeded to develop an artful (if somewhat contrived) new type of chemical fractionation theory to explain this dataset. New theories of fractionation seem to be multiplying like Ptolemaic epicycles.

While Evterev et al.'s ideas have logic, and do fit the facts as observed, one might ask whether there is an alternative explanation that is also consistent with such observations. There is, and it is alternative 3.

Applying the W-L theory of LENRs to very high nanoscale electric fields created on the surfaces of tiny dust grains with appropriate initial compositions found inside the exploding chemical 'fireballs', one might expect that neutrons could be produced via the weak interaction that would be captured on some nearby materials, transmuting them and detectably altering isotopic ratios. The apparent relationship between height of the explosion above the surface of the ground and observed degree of isotopic "fractionation" (more fractionation occurs when the blast occurs CLOSER to the ground's surface) can be explained by presuming that increasing admixtures of natural 'ground dust' somehow promote electric charging of small particles inside a chemical explosion's fireball; it could also create admixtures of soil microorganisms that may have subsequently engaged in transmutations during the time period between the explosion and whenever the samples of fallout particulates were collected and bagged for analysis.

Again, applying the LENR paradigm can help provide new insights into otherwise perplexing data.

(a.) "***The case for the Younger Dryas extraterrestrial impact event: Mammoth, megafauna, and Clovis extinction, 12,900 years ago***"
R. Firestone
Journal of Cosmology **2** pp. 256 – 285 (2009)
Source URL = http://journalofcosmology.com/Extinction105.html

(b.) "***Terrestrial evidence of a nuclear catastrophe in Paleoindian times***"
R. Firestone and W. Topping
Mammoth Trumpet **16** pp. 9 – 16 (2001)
Source URL = http://www.bibliotecapleyades.net/ancientatomicwar/esp_ancient_atomic_11.htm

Abstract (from a. - 2009): "The onset of >1000 years of Younger Dryas cooling, broad-scale extinctions, and the disappearance of the Clovis culture in North America simultaneously occurred 12,900 years ago followed immediately by the appearance of a carbon-rich black layer at many locations. *In situ* bones of extinct megafauna and Clovis tools occur only beneath this black layer and not within or above it. At the base of the black mat at 9 Clovis-age sites in North America and a site in Belgium numerous extraterrestrial impact markers were found including magnetic grains highly enriched in iridium, magnetic microspherules, vesicular carbon spherules enriched in cubic, hexagonal, and n-type nanodiamonds, glass-like carbon containing Fullerenes and nanodiamonds, charcoal, soot, and polycyclic aromatic hydrocarbons. The same impact markers were found mixed throughout the sediments of 15 Carolina Bays, elliptical depressions along the Atlantic coast, whose parallel major axes point towards either the Great Lakes or Hudson Bay. The magnetic grains and spherules have an unusual Fe/Ti composition similar to lunar Procellarum KREEP Terrane and the organic constituents are enriched in ^{14}C leading to radiocarbon dates often well into the future. These characteristics are inconsistent with known meteorites and suggest that the impact was by a previous unobserved, possibly extrasolar body. The concentration of impact markers peaks near the Great Lakes and their unusually high water content suggests that a 4.6 km-wide comet fragmented and exploded over the Laurentide Ice Sheet creating numerous craters that now persist at the bottom of the Great Lakes. The coincidence of this impact, the onset of Younger Dryas cooling, extinction of the megafauna, and the appearance of a black mat strongly suggests that all these events are directly related. These results have unleashed an avalanche of controversy which I will address in this paper."

Abstract (from b. - 2001): "Our research indicates … entire Great Lakes region (and beyond) was subjected to particle bombardment and a catastrophic nuclear irradiation that produced secondary thermal neutrons from cosmic ray interactions … neutrons produced unusually large quantities of ^{239}Pu and substantially altered … natural uranium abundance ratios (^{235}U/^{238}U) in artifacts … other exposed materials including cherts, sediments, and … entire landscape … These neutrons necessarily transmuted residual nitrogen (^{14}N) in the dated charcoals to radiocarbon, thus explaining anomalous dates."

Direct quotes (from b. – 2001): "The evidence from uranium and plutonium **-** Natural uranium, which is ubiquitous in cherts, has a ^{235}U/^{238}U isotopic ratio of 0.72 percent, which varies by less than 0.1 percent in natural sources … Significant variations in the isotopic ratio do not occur because of chemical processes; however, a thermal neutron bombardment depletes ^{235}U and thus alters the ratio. Solar or galactic cosmic rays interacting with matter produce fast secondary neutrons that become thermalized by scattering from surrounding materials."

"Thermal neutrons see a target of large cross section (681 barns) for destroying ^{235}U, compared with a target of only 2.68 barns for neutron capture on ^{238}U. Therefore, despite the low abundance of ^{235}U, about 1.8 times as many ^{235}U atoms are destroyed as ^{238}U atoms by thermal neutrons."

"If a large cosmic-ray bombardment impacted the earth and irradiated the prehistoric landscape with thermal neutrons, the ^{235}U/^{238}U ratio would be changed; ^{239}Pu would be produced from neutron capture on ^{238}U, followed by the decay of ^{239}U. Neutrons colliding with nitrogen (1.83 barns) would create ^{14}C in exactly the same way ^{14}C is normally produced in the upper atmosphere, necessarily resetting the radiocarbon dates of any organic materials lying near the surface on the North American prehistoric landscape--including charcoals at Paleoindian sites--to younger values."

"^{239}Pu produced during the bombardment will also be partly destroyed by thermal neutrons with 10^{17} barn cross section … Assuming ^{239}Pu doesn't mobilize, it will decay back to ^{235}U (half-life 24,110 yr), partially restoring the normal abundance."

"Paleoindian artifacts from Gainey, Leavitt, and Butler, and two later-period artifacts from the same geographic area of Michigan were analyzed for ^{235}U content by gamma-ray counting at the *Phoenix Memorial Laboratory*, University of Michigan … They were compared with identical chert types representative of the source materials for the artifacts. Control samples were extracted from the inner core of the purest chert known to be utilized by prehistoric people."

"The Paleoindian artifacts contained about 78 percent as much ^{235}U as the controls and later-period artifacts, suggesting substantial depletion. Depletion of ^{235}U necessarily indicates that thermal neutrons impacted these artifacts and the surrounding prehistoric landscape."

"Various artifacts, cherts, sediments, and a control sample containing about 0.2 percent uranium obtained from uraninite were sent to the McMaster University Centre for Neutron Activation Analysis to determine ^{235}U concentration by delayed neutron counting and ^{238}U concentration by activation analysis. These results are shown in Table 2. The ^{235}U/^{238}U ratios for all samples except the control deviated substantially from the expected ratio … McMaster ran additional calibration standards and has considerable expertise analyzing low-level uranium."

"Samples of unaltered flakes from Taylor and sediment originally adjacent to Gainey artifacts showed ^{235}U enriched by 30 percent … Both samples were closely associated with the particles described above. The position of these samples appears to be related to the enrichment, which cannot be explained by thermal neutrons from the bombardment. To test this, we bathed another Taylor flake in 48-percent HF at 60°F for ten minutes to remove the outer 70 percent of the sample and the attached particles. Analysis showed the "inner" flake depleted in ^{235}U by 20 percent, consistent with the other depleted cherts."

"Chert is a glass-like material highly impervious to penetration by any nuclear fallout that might also contribute ^{239}Pu. We analyzed a long-exposed piece of Bayport chert by gamma-ray counting at the LBNL low-background facility for the presence of cesium-137 (^{137}Cs), a key indicator of fallout (from nuclear testing), and found none. The B-C interface typically lies sufficiently deep that contamination by fallout is improbable. It is important to note that fallout cannot explain the depletion of ^{235}U."

"Since the depletion of ^{235}U must have resulted from bombardment by thermal neutrons, the presence of ^{239}Pu from irradiation of ^{238}U is expected. The total thermal neutron flux required to produce the observed ^{239}Pu concentration can be calculated from the relative concentrations of ^{239}Pu (corrected for the decay) and ^{238}U, and the thermal neutron-capture cross section for ^{238}U … This neutron flux can then be used to estimate the amount of additional ^{14}C that would have been produced in charcoal by neutrons colliding with ^{14}N (^{14}N cross section = 1.83 barns)."

"We derive a thermal neutron flux of c. 10^{17} n/cm^2 …. These calculations necessarily neglect differences in the neutron flux experienced by the dated charcoal and the artifacts, the effects of residual ^{239}Pu from previous bombardments, and loss of ^{239}Pu due to leaching from chert over time … The neutron flux calculated from the ^{235}U/^{238}U ratio is more than 1000 times that implied by the level of ^{239}Pu. Since ^{239}Pu decays to ^{235}U, partly restoring the natural abundance, it appears that substantial quantities of ^{239}Pu have migrated out of the chert."

Comment: Dr. Richard Firestone is a highly respected nuclear scientist employed at Lawrence Berkeley National Laboratory who has published extensively on the subject of isotopes for many years.

In isotopic studies of terrestrial materials apparently affected by a hypothesized comet impact event that is thought to have occurred ~12,900 years ago, Firestone et al. unambiguously observed the presence of anomalous ^{239}Pu which clearly was not the result of recent anthropogenic contamination. A number of other anomalous isotopic shifts involving other elements were also observed and reported.

A 4.6 km-wide comet impacting the North American icecap would probably have created a very 'dirty' fireball. It would have been comprised of primeval cometary materials intermixed with vaporized terrestrial water ice (a copious source of protons/hydrogen 'fuel' for LENRs), any dust particles previously residing inside the struck icecap, and megatons of pulverized metals and oxide materials derived from the underlying terrestrial sedimentary and Precambrian shield rocks blasted-out of the underlying crust in the moments after the immediately overlying icecap was vaporized by the comet strike.

From a theoretical LENR perspective, volcanic eruption clouds and terrestrial impact events would be very similar to dusty chemical explosions. A comet strike back in the Pleistocene would undoubtedly have created a gigantic, very dense, extremely hot, dusty plasma fireball interlaced with huge lightning discharges and many nanoparticles (potential substrates for LENRs containing atoms that can serve as neutron capture 'targets') having a wide variety of initial chemical/isotopic compositions.

Viewing Firestone et al.'s data through an LENR paradigm, some degree of isotopic shifts in Uranium, Plutonium and other 'seed' elements exposed to an impactor's fireball and hot dust clouds would not be surprising. If proper initial conditions for initiating LENRs existed in various locations therein (i.e., metals and/or PAHs and/or graphene/fullerenes + hydrogen, i.e. protons + electrons + large non-equilibrium electric currents), some indeterminate tiny fraction of materials inside the fireball could have been exposed to varying fluxes of LENR neutrons at energies ranging from ULM up to well-above thermal.

According to the Widom-Larsen theory, LENR neutron fluxes produced in nanoscale, nuclear-active regions on various types of surfaces can range from zero all the way up to very large values. For example, such neutron fluxes could in principle go as high as 10^{20} cm^2/sec; in fact, such rates may have actually been achieved during the course of high-current, pulsed-power LENR experiments conducted at Proton-21 in Kiev, Ukraine. *That said, it is unclear exactly how high they might have gotten inside the fireball created by Firestone et al.'s impactor; nonetheless, total LENR neutron fluxes on the surfaces of nanoparticles at billions of scattered sites located within it could easily have been considerable.*

Are there other examples of experimental data in which anthropogenic origin of the Plutonium might potentially be questioned? - the answer is yes --- one illustrative publication follows:

"***Distribution of Plutonium isotopes in sediments of Melanesian Basin, Central Pacific***"
W. Dong et al.
Journal of Radioanalytical and Nuclear Chemistry
DOI: 10.1007/s10967-010-0884-y ISSN (online) 1588-2780 (September 2010)
Source URL = http://www.akademiai.com/content/32m8560r02588568/fulltext.pdf

Abstract: "The anthropogenic plutonium isotopes are important geochemical tracers for environmental studies. The distributions and sources of the Pu isotopes in water column or sediments of the North Pacific have been intensively studied. However, knowledge on the distribution of Pu isotopes in sediments of the Central Pacific, when available, is limited. To study the composition of Pu isotopes in the ocean, thus to identify the sources of radioactive pollution, sediment core samples were collected in the Central Pacific by R/V Hakuho Maru in the KH-04-5 cruise. The activity concentrations of $^{239+240}$Pu and the ^{240}Pu/^{239}Pu atom ratio were determined using a sector-field inductively coupled mass spectrometry (SF-ICP-MS) combined with a high efficiency sample introduction system (APEX-Q). Possible sources and sedimentation behavior of Pu isotopes are discussed."

Direct quotes: "*It was found that $^{239+240}$Pu penetrated to the top 10 cm in SX-02 site … The sedimentation rates in the deep sea ranged from 0.12 to 1.8 cm kyr-1 using U–Th dating method [10]. A depth of 10-cm sediments may represent a sedimentation of thousands or tens of thousands years. The release of Pu isotopes into the environment, however, began in ca. 60 years ago.* **[Italics for emphasis are mine]** Pu isotopes detected in this sediment core were not solely resulted from the sedimentation process, but also, and more importantly from the post-deposition processes, such as physical mixing of bottom currents and the bioturbation effect. The plutonium may be associated with surface derived large particles, of which food values are high, so that they are segregated and transported by benthic organisms to deeper depths in the sediment column."

"The results of ^{240}Pu/^{239}Pu atom ratio are presented in Fig. 3, which ranged from 0.186 to 0.238 in the Melanesian Basin of SX-02 with a mean value of 0.205, which is typically higher than that of global fallout (0.174 ± 0.014). It is reasonable to suggest that the global fallout is not the solely Pu source in this area."

"Buesseler [26] reported ^{240}Pu/^{239}Pu atom ratio in surface sediments and seawater collected during 1974–1982 in the North Pacific, and concluded that the Pacific Proving Ground (PPG) was a major source for the North Pacific, which was characterized by an elevated ^{240}Pu/^{239}Pu atom ratio ([0.2) relative to global stratospheric fallout (0.18–0.19). Norisuye et al. [27] observed high ^{240}Pu/^{239}Pu atom ratios (0.220–0.269) in seawaters in the NW Pacific. The range of contamination originated from Bikini and Enewetak nuclear weapon tests in the 1950s was evaluated by Zheng and Yamada [20] and by Kim et al. [28], which was believed to be extended to Japanese Coast and the Coast of the Korean Peninsula by the transport of the North Equatorial Current. Our results of ^{240}Pu/^{239}Pu atom ratio in SX-02 (0.205, inventory-weighted mean) are comparable to above results."

Comment: Dong et al.'s prosaic 'contamination' explanations for certain variances in their Plutonium data may very well be entirely correct. That said, I would like to specifically note their explanation for the observed presence of Plutonium at unexpectedly deep levels in the sediment column (given the likely local sedimentation rates) relies on an underlying assumption that, *"… post-deposition processes, such as physical mixing of bottom currents and the bioturbation effect ..,"* are processes that are in fact operating at the specific locations of the sampling sites and furthermore that activities of "*benthic organisms*" are responsible for effectively transporting Plutonium-rich particulate matter from the upper layers to much deeper levels in sediments found at the bottom of the Pacific ocean. While their assumption sounds reasonable, I am not aware that the phenomenon of 'well-mixed upper sediments' down 10 cm has been demonstrated quantitatively in any field experiments in the deep Central Pacific.

Lastly, biologically mediated LENR ULM neutron captures (n,γ) on ^{239}Pu to produce ^{240}Pu will result in enrichment of the heavier Pu isotope, exactly as observed in their reported data. *Thus in terms of isotope effects, in this instance biological transmutation processes can potentially mimic the effects of higher-than-normal neutron fluxes that occasionally occur during detonation of nuclear weapons.*

SPECIAL COMMENTS: as we are concluding our discussion of LENRs and Plutonium, I will reiterate that the vast majority of Pu present in the terrestrial environment today is probably anthropogenic; I fully concur with prevailing opinion on that point. What is proposed to readers is that based on the Widom-Larsen theory of LENRs, there may be a very modest, yet-to-be-determined percentage of existing Plutonium and Uranium isotopic shifts that are the result of biological or abiological LENR transmutations.

So far, readers have been provided with a description of a theoretical W-L LENR Actinide nucleosynthetic network and selected examples of published mainstream, peer-reviewed experimental data that provide intriguing evidence that this network might be operating in a variety of different environments on earth. We have seen how some or all of the Actinide network pathways could potentially be present in soils, ocean sediments, dusty chemical explosions, volcanic eruptions, and extraterrestrial impact events.

In the final section, we will speculatively explore how theoretical concepts developed to understand enigmatic LENR effects in laboratory experiments involving electrochemical cells might provide useful insights into LENR transmutation networks operating in aqueous natural environments.

Before going to the final section, here is some pertinent historical and technical information – while different types of LENR effects have been unknowingly observed and reported as anomalies in various scientific literature for more than 100 years, the subject came to light in the present era as a result of the furor over “cold fusion” that erupted into media and mainstream science communities back in 1989:

Since the inception of modern nuclear science in the 1940s, it has been widely believed that the only nuclear processes capable of triggering certain nuclear effects were strong interaction fission or fusion. It was widely held that nuclear transmutation reactions could only take place within special environments: e.g., fission reactors, nuclear weapons, or stars. Pons & Fleischmann’s 1989 discovery of what appeared to be nuclear processes operating inside what would otherwise be ordinary D_2O electrolytic chemical cells challenged long-established conceptual paradigms about nuclear science. Sadly, P&F rashly speculated that their observed radiation-free “excess heat” resulted from a hypothetical D-D “cold fusion” process. That totally erroneous theoretical idea, coupled with irreproducible experimental results, resulted in huge controversy and attacks on such work by some mainstream scientists that have continued to the present.

Starting with the release of our first arXiv preprint in 2005, the Widom-Larsen theory of LENRs has shown, using known physics, how energetic nuclear reactions can readily take place in ordinary chemical cells. According to W-L, key aspects of LENRs involve weak interactions that can occur in a variety of different laboratory and natural environments under relatively ‘mild’ physical conditions. Our theory posits that in condensed matter systems, many-body collective effects allow the otherwise disparate chemical and nuclear energy realms to interconnect with each other in special, nano-scale regions on surfaces.

A 5-page, highly technical description of the physics 'bedrock' of our theoretical work was published in:

"***Ultra low momentum neutron catalyzed nuclear reactions on metallic hydride surfaces***"
A. Widom and L. Larsen
European Physical Journal C - Particles and Fields **46** pp. 107 – 112 (2006)
Source URL = http://www.newenergytimes.com/v2/library/2006/2006Widom-UltraLowMomentumNeutronCatalyzed.pdf

More recently, a 21-page “*Primer*” paper, which provides readers with a high-level overview of the entire expanse of the Widom-Larsen theory of low energy nuclear reactions (LENRs) from the microcosm (very high local E-fields on nm-to-micron length-scales on condensed matter surfaces) to the macrocosm (dusty plasmas and magnetic-regime astrophysical phenomena), just published in the Indian Academy of Science’s peer-reviewed journal as follows (much less mathematically intensive than *EPJC* in 2006) :

“***A primer for electroweak induced low-energy nuclear reactions***”
Y. N. Srivastava, A. Widom, and L. Larsen
Pramana – Journal of Physics **75** pp. 617 – 637 (2010)
Source URL = http://www.ias.ac.in/pramana/v75/p617/fulltext.pdf

Now, fascinating new facts and speculation about 'electric bacteria' - recent exciting discoveries by microbiologists have revealed that electric potentials, currents, and 'nanowires' are associated with the activities of various species of bacteria; might this possibly have anything to do with biological LENRs?

Rather than waiting until various facts are better clarified and more experimental work specifically concerning the Widom-Larsen theory of LENRs has been conducted, I am trying to engage readers and otherwise interested scientists as early as possible in the creative discovery process by providing a briefly-sketched 'conceptual bridge' between the areas of W-L theory, earlier experimental work in LENRs, electrochemistry, publications cited and discussed in this working paper, and exciting new discoveries by microbiologists concerning, for lack of a better descriptive term, 'electric bacteria'.

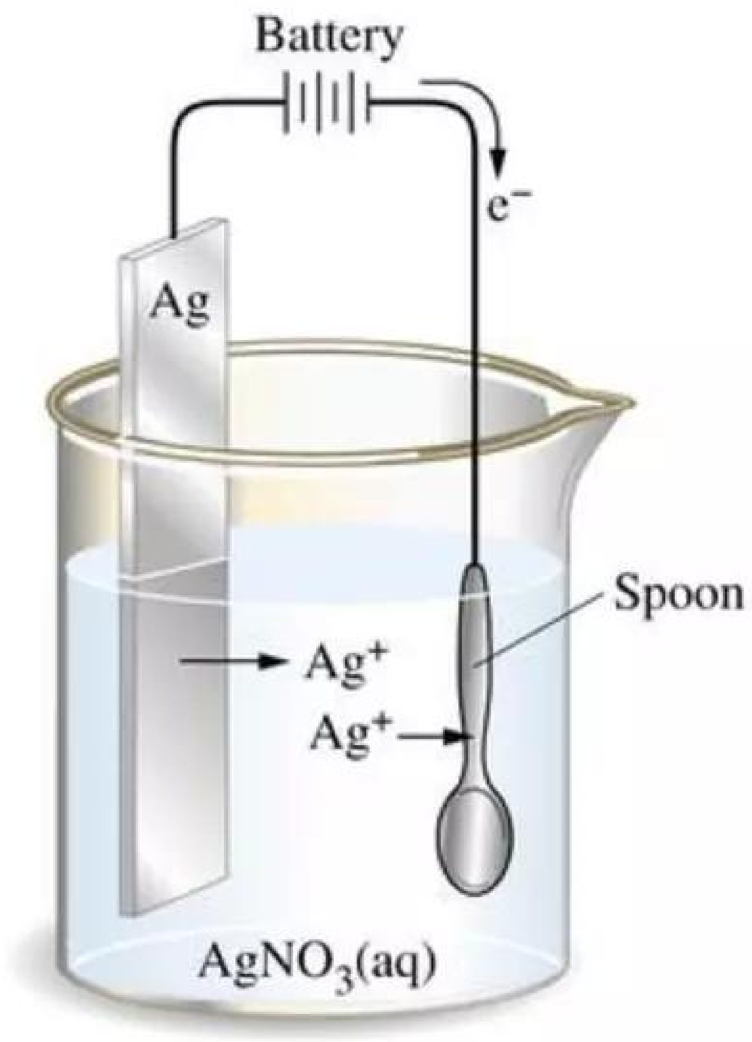
If my high-level conceptual distillation and intellectual proselytizing are successful, it will hopefully help interested researchers in a variety of disciplines leverage and extrapolate their present knowledge about some or all of the above-noted areas into participation in what I believe is an exciting, paradigm-shifting area of inquiry: biological and abiological LENR transmutations in natural and laboratory environments.

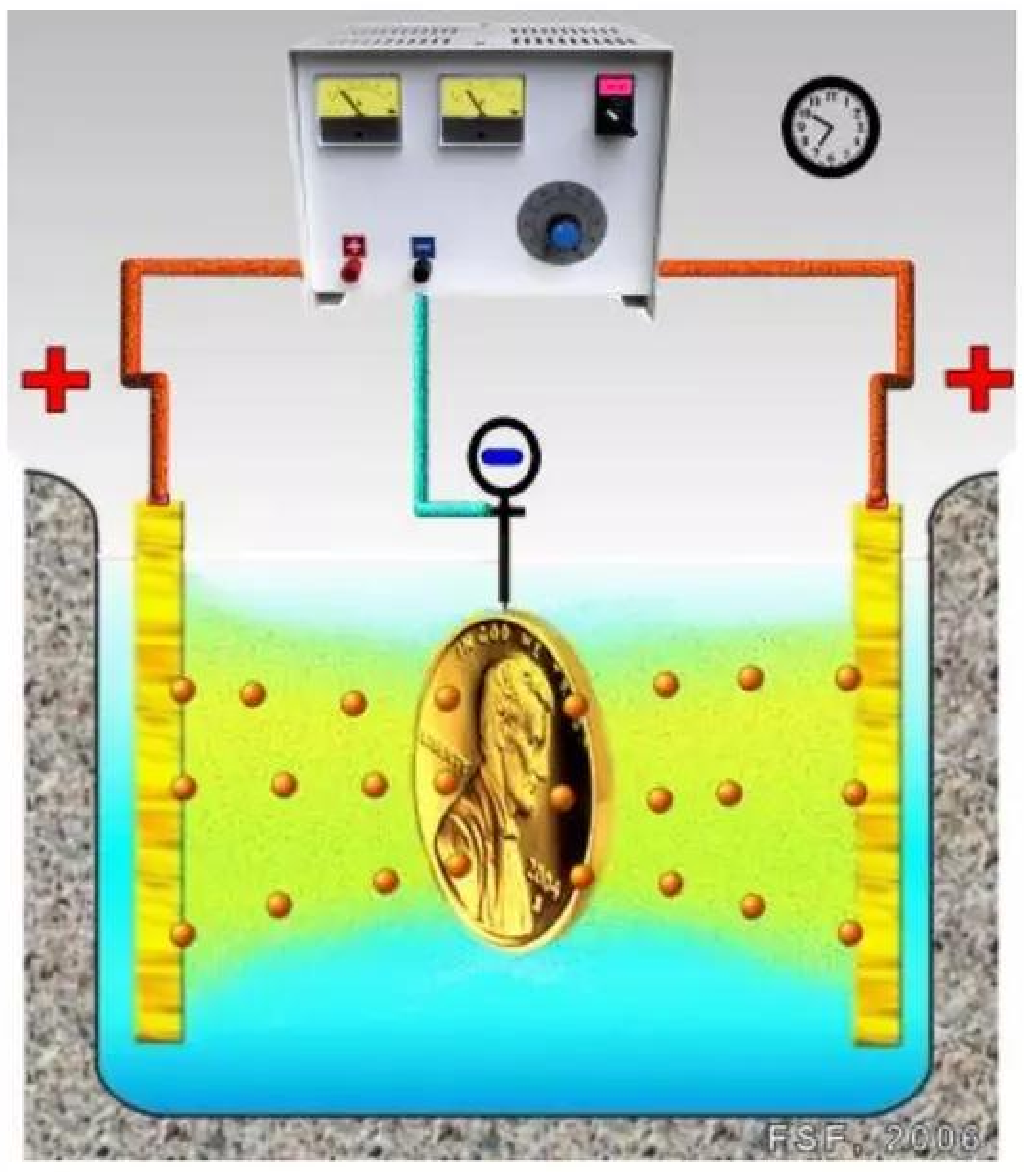
For 20 years, a small subset of LENR researchers periodically reported reasonably careful observations of nuclear transmutations in electrolytic chemical cells. For the most part, these were not published in traditional peer-reviewed scientific journals simply because mainstream science steadfastly refused to believe such experimental results. Until the recent development of the Widom-Larsen theory, there was no plausible theoretical framework that could make sense of these seemingly inexplicable observations.

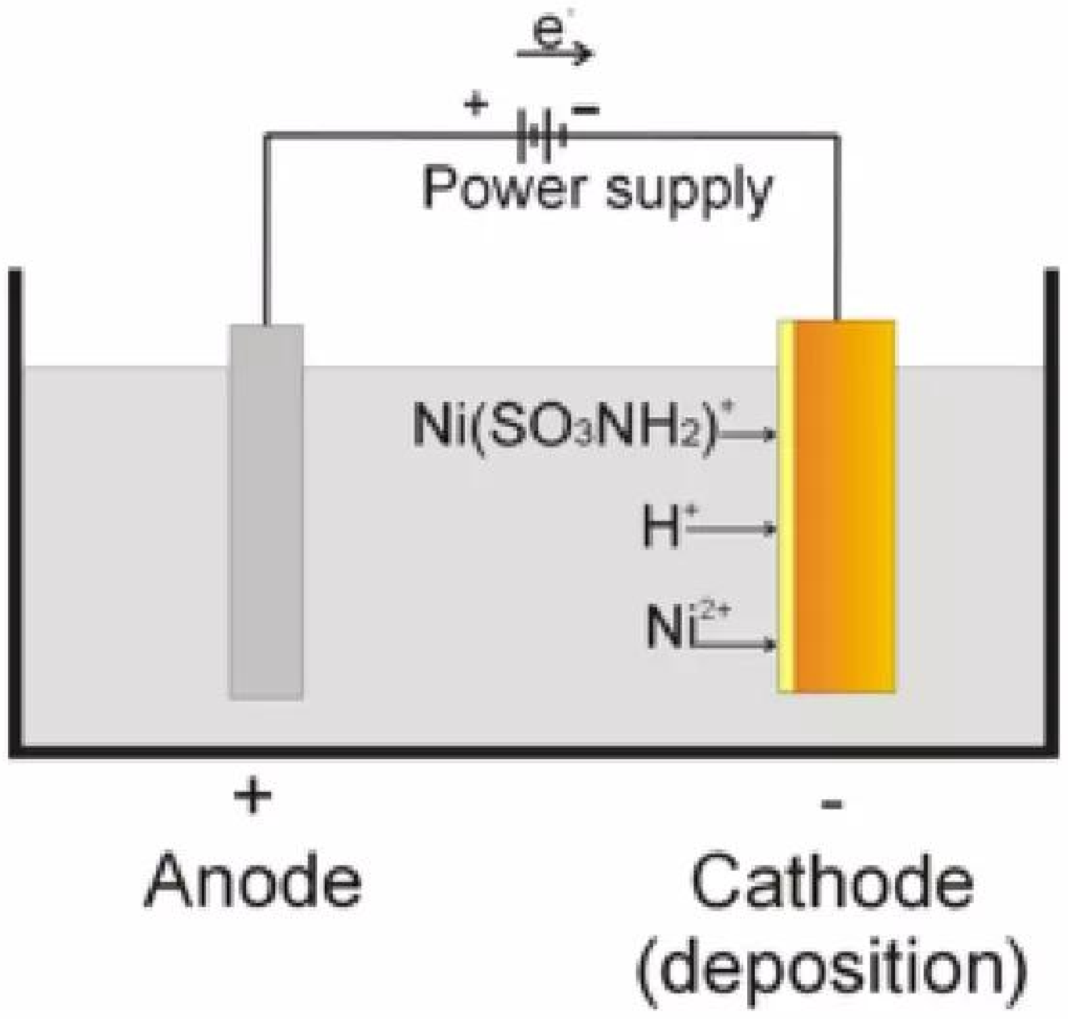
Some illustrative papers concerning LENR transmutations in electrochemical cells can be found on the free public website http://www.lenr-canr.org. However, *caveat emptor* --- this site contains a diverse range of non-peer-reviewed electronic publications: quality ranges from many low-quality totally nonsensical theories and/or very sloppy experimental work up to a few high-quality publications that could probably survive rigorous peer-review at a mainstream journal as long as referees selected by Editors were reasonably open-minded and fair (today, with few exceptions most are decidedly unreasonable).

In the simplest terms, abiological laboratory electrochemical cells conceptually look roughly like this:

Three examples showing schematic diagrams of electrolytic chemical cells

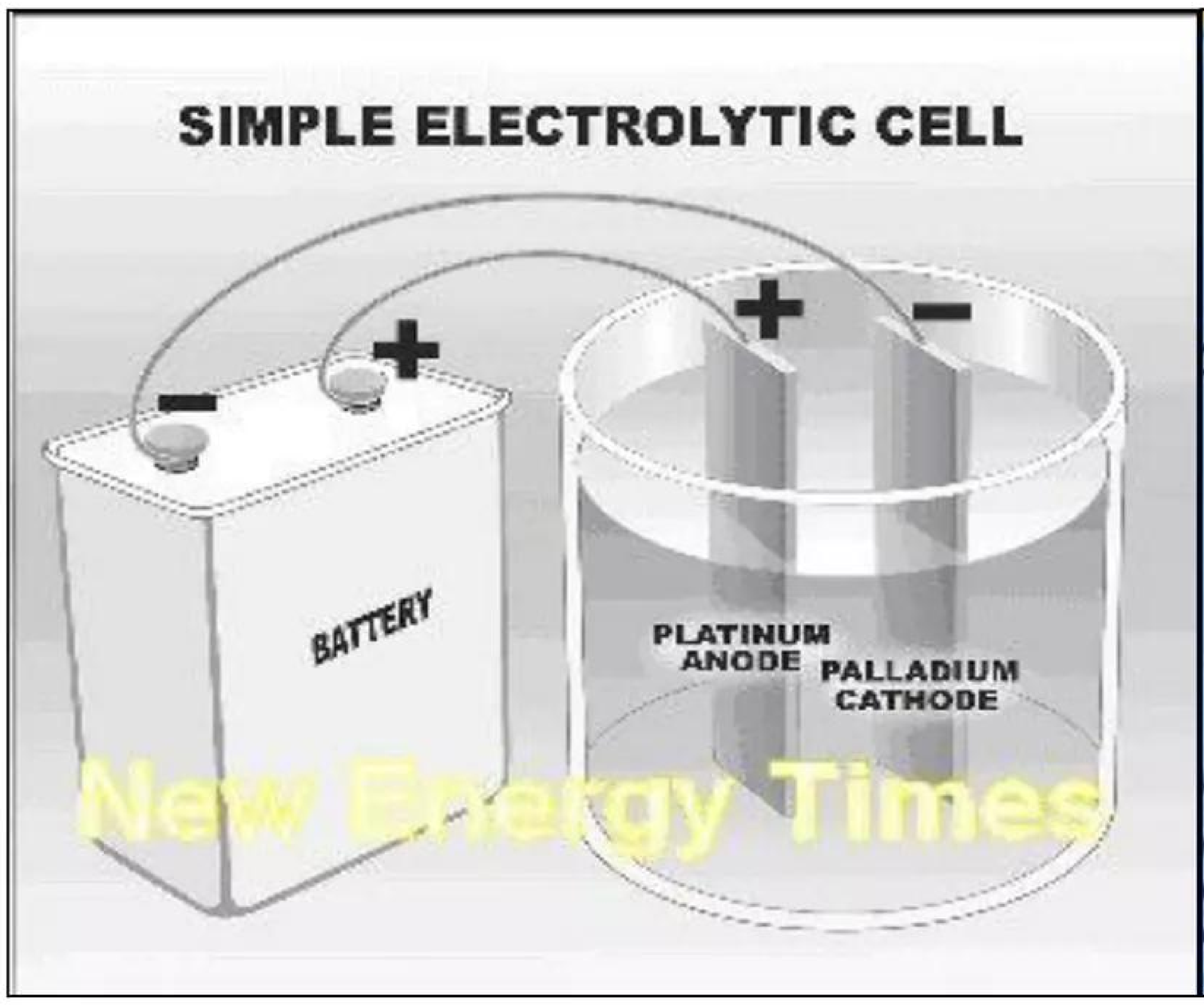






LENR electrolytic chemical cells

To the left blow below is a conceptual schematic of a type of electrolytic cell used in many LENR experiments (typically would use DC power supply instead of a battery; image credit is *New Energy Times*); at right is an image of an actual working version of a laboratory LENR electrolytic cell apparatus:



Now let us 'take a view of the situation from 50,000 feet,' looking simply and abstractly at some of the basic conceptual elements that comprise an LENR electrolytic cell in a laboratory; they are as follows:

1. Aqueous electrolyte containing a mixture of various types of mobile ions <u>and</u> hydrogen isotopes (H_2O and/or D_2O - when 'loaded' in/onto lattices of cathode metals via currents they can exist in ionic form as protons $p+$ or deuterons $d+$)
2. Source of energy in the form of a disequilibrium electrical (electron) current (e.g., provided by a battery or a DC power supply in a laboratory experiment)
3. Conductive metallic anode and cathode (many different types of metals will work) immersed in the aqueous ionic electrolyte and connected to wires
4. Connecting metallic wires conduct electric current (electrons) between components of electrolytic system; parallel ionic currents occur in the electrolyte
5. During disequilibrium current flow between the anode and cathode, electrically driven oxidation-reduction (Redox) chemical reactions take place on the surfaces of both the anode and cathode immersed in the aqueous electrolyte

Please again note that certain LENR researchers have carefully documented the production (via transmutation) of <u>minute</u> amounts of different elements and stable isotopes thereof on the surfaces of cathodes in a large number of laboratory experiments with aqueous electrolytic cells.

In the Widom-Larsen theory of LENRs in condensed matter, production of ultra low momentum (ULM) neutrons takes place via the weak interaction: $e^- + p^+ \rightarrow$ neutron + neutrino. Mass-renormalized heavy electrons that can react directly with protons are created by high nanoscale local electric fields enabled by surface many-body collective effects. An external energy source is required to drive this reaction forward (about 0.78 MeV per e^- to react with a proton; ~=2.6 million ATP molecules) which comes from an input disequilibrium electrical current. Transmutations occur when ULM neutrons are absorbed by nearby 'target' nuclei; heavy-mass electrons also present locally convert neutron capture gammas to much lower- energy infrared photons (heat).

How do key attributes of a typical laboratory LENR electrolytic cell compare with what is present inside bacterial cells and/or in their immediate environment? – please see Table below:

Present in electrolytic cells used in laboratory experiments that produced LENR transmutation products	Analogously present somewhere either inside microorganisms or located in their immediate physical environments?	Comments
Aqueous electrolyte containing mobile ions as well as sources of protons or deuterons (i.e., H_2O or D_2O)	Yes	Obvious
Disequilibrium electrical (electron) current	Yes	See papers cited down below --- recent discoveries re microbial currents
Conductive metallic anode and cathode structures that support redox reactions on their surfaces; also can absorb hydrogen (form hydrides) which enters metallic lattice as $p+$ protons	Yes	Various metals are commonly present inside bacteria; in some cases tightly integrated with macromolecules (e.g., enzyme cofactors); not clear to me exactly what biological systems' nanoscale structural equivalents of metallic laboratory cathodes/anodes are really like
'Wires' that can conduct electrical currents	Yes	Bacteria are now known to have electrically conductive nonmetallic nanowires
Oxidation-reduction (Redox) chemical reactions occur at the anodes and cathodes immersed in electrolyte	Yes	Redox reactions occur in metal-reducing bacteria --- observed isotopic shifts are now known to be closely associated with bacterial reduction processes; e.g., Uranium, Sulfur and perhaps Plutonium isotopes as we have discussed herein

Mapping this rough sketch of very high-level theoretical analogies into nanoscale details of exactly what might be going on in microbial systems that could enable LENRs to occur in them is a major unfinished task that will require the effort of many researchers; such work would intrinsically be very interdisciplinary.

For example, *on a nanoscale* in what specific macromolecular biological structures might the high electric fields occur that are necessary for ULM neutron production take place? Recall that transient E-fields > 10^{11} V/m in regions with nanometer to micron-scale dimensions are needed to renormalize electron masses so that heavy electrons can react with protons, creating ULM neutrons via the weak interaction.

Per W-L theory, collective many-body effects involve many thousands of quantum-mechanically entangled protons coupled (via breakdown of Born-Oppenheimer) to many mutually entangled metallic surface plasmon or ring π electrons; this temporarily creates huge local electric fields on 'surfaces'. Luckily for bacteria, such fields only have to exist for a number of attoseconds to 'get the job done.' How are these fields turned on/off; how is field-strength is controlled and finely modulated on a nanoscale? What types of macromolecular structural/conformational dynamics are involved in this process? Etc.

Another example of an intriguing open question for fellow researchers is the mechanism(s) by which microorganisms are able to ‘deliver’ and ‘control’ exactly which ‘target’ atoms/elements are ‘allowed’ to capture (i.e., absorb) ULM neutrons as they are produced inside a cell? If this process were not tightly controlled by microorganisms, it could potentially create a form of intracellular chemical chaos with one element unexpectedly changing into another (please recall that neutron captures occur in picoseconds).

Active sites of enzymes and membrane surfaces/interfaces would be logical possibilities where one might expect key aspects of biological LENRs could be occurring. How does this new possibility relate to our present knowledge of biophysical processes already thought to be taking place at such locations?

With regard to theoretical energetics of biological transmutations, it ‘costs’ the energy equivalent of ~2.6 million ATP molecules to produce a single ULM neutron from a proton. On the Internet, I discovered an estimate that a single *E. coli* cell can supposedly sustain an ATP production rate of ~0.8 to 4.0 x 10^{10} ATP molecules/cell/hr. That being the case, the ‘cost’ of ULM neutron production would be fairly ‘expensive’ for a bacterium but maybe not too onerous if the end result was important enough for growth and/or survival.

In that regard, let’s say some metal ion (say Fe) is not present inside a given cell or available in its external environment. Let us further suppose that this metal ion is required as a key cofactor in the active site of a particular metalloprotein enzyme that is crucial to proper functioning of a bacterium’s key metabolic pathways. Given that metal cofactors tend to persist and be ‘reused’ in cells, it might be ‘well worth it’ for an organism to invest the amount of energy needed to synthesize the required number of Fe cofactor ions from some other readily available element.

In fact, if readers examine the early work of Vysotskii and Kornilova (2003), they will find fascinating experimental evidence (laboratory studies) suggesting that, when Fe is unavailable in the growth medium, certain species of bacteria appear to be able to synthesize needed ^{56,57}Fe from ^{55}Mn (which has only one stable isotope) present in the medium via a W-L LENR neutron-capture transmutation process as follows:

1. **Mn-55 + n -> Mn-56 (unstable isotope half-life = 2.6 hrs); then**
2. **Mn-56 (beta decays) -> Fe-56 (stable isotope) + neutrino photon**
3. **Fe-56 + n -> Fe-57 (stable isotope)**

In the above example, only one ULM neutron is required to transmute Mn into Fe; not an excessive ‘cost’ per Fe if a bacterium must satisfy a critical requirement to synthesize certain metalloproteins. In such cases, it would seem that the ability to perform transmutations, i.e., nucleosynthesis, albeit energetically costly, could potentially provide a valuable competitive and survival advantage for a microorganism.

And so forth … the list of very interesting and important questions that still need to be answered to fully explore the intriguing possibility of LENRs in biological systems is filled with major R&D opportunities.

Readers are encouraged to examine the cited papers that follow and familiarize themselves with our theoretical work. It is hoped that the broad-brush conceptual analogies that I have sketched-out here provide some useful new insights that inspire participation and facilitate further experimentation to better understand enigmatic isotope-shifting processes that are occurring in abiological and biological systems. *Unequivocal experimental confirmation of LENR biological transmutations would be paradigm-shifting.*

Selected publications concerning bacterial nanowires, observations of electrodic potential anomalies and related electron currents, and bacterial electrokinesis – exciting recent information:

“***Extracellular electron transfer via microbial nanowires***”
G. Reguera et al.
Nature **435** pp. 1098 – 1101 (2005) includes another document w. additional supplementary material
Source URL = http://www.geobacter.org/publications/Nature_2005_Jun.pdf

Abstract: “Microbes that can transfer electrons to extracellular electron acceptors, such as Fe(III) oxides, are important in organic matter degradation and nutrient cycling in soils and sediments. Previous investigations on electron transfer to Fe(III) have focused on the role of outer-membrane *c*-type cytochromes. However, some Fe(III) reducers lack *c*-cytochromes. *Geobacter* species, which are the predominant Fe(III) reducers in many environments, must directly contact Fe(III) oxides to reduce them, and produce monolateral pili that were proposed, on the basis of the role of pili in other organisms, to aid in establishing contact with the Fe(III) oxides. Here we report that a pilus-deficient mutant of *Geobacter sulfurreducens* could not reduce Fe(III) oxides but could attach to them. Conducting-probe atomic force microscopy revealed that the pili were highly conductive. These results indicate that the pili of *G. sulfurreducens* might serve as biological nanowires, transferring electrons from the cell surface to the surface of Fe(III) oxides. Electron transfer through pili indicates possibilities for other unique cell-surface and cell–cell interactions, and for bioengineering of novel conductive materials."

Direct quotes: "As expected from previous studies, *G. sulfurreducens* produced pili during growth on Fe(III) oxide (Fig. 1a) but not on soluble Fe(III) (Fig. 1b), and the pili were localized to one side of the cell ... One known function of type IV pili in other microorganisms is establishing contact with surfaces … These results indicated that the pili might have a more direct role in electron transfer to Fe(III) oxides. To evaluate this we measured the electrical conductivity through the pili … When a voltage was applied to the tip there was a strong current response along the pilus filament, which was positive when a positive voltage was applied and negative with a negative voltage (Fig. 4b, c). In contrast, the non-pilin proteins had no detectable conductivity and in instances in which the non-pilin proteins covered the pili filaments they insulated the pili from the conductive tip."

"These results show that the pili of *G. sulfurreducens* are highly conductive. This indicates that *G. sulfurreducens* requires pili in order to reduce Fe(III) oxides because pili are the electrical connection between the cell and the surface of the Fe(III) oxides. This contrasts with the nearly universal concept that outer-membrane cytochromes are the proteins that transfer electrons to Fe(III) oxide in Fe(III) reducers. However, the outer-membrane cytochrome model for Fe(III) reduction has serious limitations."

"The conductive pili provide the opportunity to extend electron transfer capabilities well beyond the outer surface of the cells, which might be especially important in soils in which Fe(III) oxides exist as heterogeneously dispersed coatings on clays and other particulate matter. The pilus apparatus is anchored in the periplasm and outer membrane of Gram-negative cells, thus offering the possibility that pili accept electrons from periplasmic and/or outer membrane electron transfer proteins. These intermediary electron transfer proteins need not be the same in all organisms, which is consistent with the differences in cytochrome content and/or composition in different Fe(III) reducers1. The likely function of the pili is to complete the circuit between these various intermediary electron carriers and the Fe(III) oxide … In addition to serving as a conduit for electron transfer to Fe(III) oxides, pili could conceivably be involved in other electron transfer reactions. For example, pili of individual Geobacter cells are often intertwined, raising the possibility of cell-to-cell electron transfer through pili."

"***Electrical transport along bacterial nanowires from Shewanella oneidensis MR-1***"
M. El-Naggar et al.
Proceedings of the National Academy of Sciences (PNAS) DOI: 10.1073/pnas.1004880107 (2010)
Source URL = http://foundry.lbl.gov/assets/docs/pubs/MFNF_101001_UserElectrical.pdf
Free supplementary material =
http://www.pnas.org/content/suppl/2010/09/28/1004880107.DCSupplemental/pnas.201004880SI.pdf

Abstract: "Bacterial nanowires are extracellular appendages that have been suggested as pathways for electron transport in phylogenetically diverse microorganisms, including dissimilatory metal-reducing bacteria and photosynthetic cyanobacteria. However, there has been no evidence presented to demonstrate electron transport along the length of bacterial nanowires. Here we report electron transport measurements along individually addressed bacterial nanowires derived from electron-acceptor–limited cultures of the dissimilatory metal-reducing bacterium *Shewanella oneidensis* MR-1. Transport along the bacterial nanowires was independently evaluated by two techniques: (*i*) nanofabricated electrodes patterned on top of individual nanowires, and (*ii*) conducting probe atomic force microscopy at various points along a single nanowire bridging a metallic electrode and the conductive atomic force microscopy tip. The *S. oneidensis* MR-1 nanowires were found to be electrically conductive along micrometer-length scales with electron transport rates up to 10^9/s at 100 mV of applied bias and a measured resistivity on the order of 1 Ω·cm. Mutants deficient in genes for *c*-type decaheme cytochromes MtrC and OmcA produce appendages that are morphologically consistent with bacterial nanowires, but were found to be nonconductive. The measurements reported here allow for bacterial nanowires to serve as a viable microbial strategy for extracellular electron transport."

Direct quotes: "To date, several biological assays have demonstrated results consistent with electron transport along bacterial nanowires, including measurements of improved electricity generation in microbial fuel cells and enhanced microbial reduction of solid phase iron oxides. However, our direct knowledge of nanowire conductivity has been limited to local measurements of transport only across the thickness of the nanowires. Thus far, there has been no evidence presented to verify electron transport along the length of bacterial nanowires, which can extend many microns, well beyond a typical cell's length. Here we report electron transport measurements along individually addressed bacterial nanowires derived from electron-acceptor limited cultures of the DMRB *S. oneidensis* MR-1."

"Recent measurements by McLean et al. of the rate of electron transfer per cell from *S. oneidensis* MR-1 to fuel cell anodes were on the order of 10^6 electrons per cell per second. These measurements are consistent with the specific respiration rate estimated under the cultivation conditions used here (2.6×10^6 electrons per cell per second) (Materials and Methods). A comparison with our transport measurements demonstrates that a single bacterial nanowire could discharge this entire supply of respiratory electrons to a terminal acceptor … We found that these mutants produce nonconductive filaments, indicating that, in the case of *S. oneidensis* MR-1, cytochromes are necessary for conduction along nanowires. However, this finding does not preclude other mechanisms for long-range electron transport along bacterial nanowires from other organisms"

"In conclusion, our data demonstrate electrical transport along bacterial nanowires from *S. oneidensis* MR-1, with transport rates that allow for bacterial nanowires to serve as a viable microbial strategy for extracellular electron transport. The measurements reported here motivate further investigations into the molecular composition and physical transport mechanism of bacterial nanowires, both to understand and realize the broad implications for natural microbial systems and biotechnological applications such as microbial fuel cells."

Comment: in my opinion, this new publication is a very interesting and important paper --- a must read.

"***Electrokinesis is a microbial behavior that requires extracellular electron transport***"
H. Harris et al.
Proceedings of the National Academy of Sciences (PNAS) **107** pp. 326-331 (2010)
Source URL = http://nanobio.usc.edu/publications/assets/Harris2009.pdf
Free supplementary material =
http://www.pnas.org/content/suppl/2009/12/14/0907468107.DCSupplemental/pnas.200907468SI.pdf

Abstract: "We report a previously undescribed bacterial behavior termed electrokinesis. This behavior was initially observed as a dramatic increase in cell swimming speed during reduction of solid MnO_2 particles by the dissimilatory metal-reducing bacterium *Shewanella oneidensis* MR-1. The same behavioral response was observed when cells were exposed to small positive applied potentials at the working electrode of a microelectrochemical cell and could be tuned by adjusting the potential on the working electrode. Electrokinesis was found to be different from both chemotaxis and galvanotaxis but was absent in mutants defective in electron transport to solid metal oxides. Using in situ video microscopy and cell tracking algorithms, we have quantified the response for different strains of *Shewanella* and shown that the response correlates with current generating capacity in microbial fuel cells. The electrokinetic response was only exhibited by a subpopulation of cells closest to the MnO_2 particles or electrodes. In contrast, the addition of 1 mM 9,10-anthraquinone-2,6-disulfonic acid, a soluble electron shuttle, led to increases in motility in the entire population. Electrokinesis is defined as a behavioral response that requires functional extracellular electron transport and that is observed as an increase in cell swimming speeds and lengthened paths of motion that occur in the proximity of a redox active mineral surface or the working electrode of an electrochemical cell."

Direct quotes:

"*Shewanella* species are widespread in nature, enjoying a cosmopolitan distribution in marine, freshwater, sedimentary and soil environments. They have attracted considerable attention in recent years because of their ability to reduce an extensive number of different electron acceptors, including the solid (oxy)hydroxides of iron and manganese, such as $Fe(OH)_3$ and MnO_2, using one or more proposed mechanisms of extracellular electron transport (EET). The EET ability of *Shewanella* species is consistent with their ability to generate electric current in microbial fuel cells (MFCs) in the absence of exogenous electron shuttles. Various strategies of extracellular electron transfer have been proposed in metal-reducing microbes, including naturally occurring or biogenic soluble mediators that 'shuttle' electrons from cells to acceptors, as well as direct transfer using multiheme cytochromes located on the cell exterior and transfer via conductive nanowires."

"Here we present data that suggest that the shewanellae exhibit a unique motility response: we call it *electrokinesis*. This response, which involves increased swimming speeds and prolonged runs, was observed when cells were in close proximity to a redox active surface, such as an MnO_2 particle, or the working electrode of an electrochemical cell. Cells in the presence of MnO_2 particles were frequently observed to touch the mineral surface and sometimes pause for up to 1 sec, before swimming away. Video microscopy showed a concomitant reduction of the MnO_2 particles, suggesting that electrokinesis could be associated with the use of insoluble surfaces as electron acceptors. Electrokinesis is not a uniform response that can be observed in all cells, although if an electron shuttle is added the proportion of cells swimming and the average swimming speed of these cells rapidly increased."

"The motility response, which we call *electrokinesis*, was localized to the areas surrounding the metal oxide particles and was coincident with the reduction and subsequent dissolution of the particles (the highly motile cells are shown in Movie S1 and Movie S2; reduction of an MnO_2 particle, during a 24-h experiment, is shown in Fig. 1A and Movie. S2). The swimming activity was quantified within the first 2 h of each experiment using video-microscopic observations coupled with cellular tracking algorithms that yielded cell positions, trajectories, and 2D maps of swimming speed (Materials and Methods)."

"Cells were also frequently observed to touch the MnO_2 particles, sometimes pause for up to 1 sec, then swim away, suggesting that the motility response could be associated with the direct transfer of electrons to the mineral surface. When soluble electron acceptors (10 mM fumarate, 5 mM nitrate, or 10 mM thiosulfate) were added individually to anaerobic capillary tubes containing *S. oneidensis* MR-1 cells and MnO_2 particles, the fast-swimming motility response became uniform, rather than being localized around the MnO_2 particles."

[What H. Harris et al. state here in their conclusions is fascinating and frankly, astonishing]

"… Many of the motile cells in both the MnO_2 and electrode experiments were, however, observed to transiently touch the surface of the electron acceptor but not to attach to the mineral or graphite surfaces (Movie S1 and Movie S6). Cells displaying this behavior could hypothetically be storing electrons on some biologic 'capacitor,' as suggested by Esteve-Núñez et al., then rapidly discharging these electrons to the electron acceptor during these brief interaction events, and thus energizing the cell. If there is a kind of biologic capacitor, this might account for the different relaxation times observed for the different *Shewanella* strains. To this end, electrokinesis may require a combination of specialized electron storage/donating mechanisms by cells and efficient electron acceptor capacity by minerals. Our future research will explore whether electrokinesis has a thermodynamic or kinetic basis and will perform additional studies to determine whether the cells are indeed acting as biologic 'capacitors'."

Comment: this is truly incredible data; in my opinion this is another important 'must read' paper.

"***Analyses of current-generating mechanisms of Shewanella loihica PV-4 and Shewanella oneidensis MR-1 in microbial fuel cells***"
G. Newton et al.
Applied and Environmental Microbiology **75** pp. 7674 – 7681 (2009)
Source URL = http://aem.asm.org/cgi/reprint/75/24/7674

Abstract: "Although members of the genus *Shewanella* have common features (e.g., the presence of decaheme c-type cytochromes [c-cyts]), they are widely variable in genetic and physiological features. The present study compared the current-generating ability of *S. loihica* PV-4 in microbial fuel cells (MFCs) with that of well-characterized *S. oneidensis* MR-1 and examined the roles of c-cyts in extracellular electron transfer. We found that strains PV-4 and MR-1 exhibited notable differences in current-generating mechanisms. While the MR-1 MFCs maintained a constant current density over time, the PV-4 MFCs continued to increase in current density and finally surpassed the MR-1 MFCs. Coulombic efficiencies reached 26% in the PV-4 MFC but 16% in the MR-1 MFCs. Although both organisms produced quinone-like compounds, anode exchange experiments showed that anode-attached cells of PV-4 produced sevenfold more current than planktonic cells in the same chamber, while planktonic cells of MR-1 produced twice the current of the anode-attached cells. Examination of the genome sequence indicated that PV-4 has more c-cyt genes in the metal reductase-containing locus than MR-1. Mutational analysis revealed that PV-4 relied predominantly on a homologue of the decaheme c-cyt MtrC in MR-1 for current generation, even though it also possesses two homologues of the decaheme c-cyt OmcA in MR-1. These results suggest that current generation in a PV-4 MFC is in large part accomplished by anode-attached cells, in which the MtrC homologue constitutes the main path of electrons toward the anode."

Direct quote: "In conclusion, the present study shows that even though MR-1 and PV-4 are affiliated with the same genus, *Shewanella*, and possess similar genes for metal-reducing *c*-cyts, they employ substantially different extracellular electron transfer mechanisms (e.g., mediated versus direct electron transfer, amounts of excreted mediators, and roles of OM *c*-cyts). At present, the genomes of nearly 20 strains in the genus *Shewanella* have been completed, and the diversity in the genetic organization of metal-reducing *c*-cyts among these strains has been analyzed (6). We suggest that further genetic and physiological studies of these diverse *Shewanella* strains are needed to comprehend the consensus features and diversity in the metal-reducing and current-generating mechanisms in this environmentally and biotechnologically important genus."

"***Harnessing self-organizing bacterial networks for microbial fuel cells***"
R. Nakamura and K. Hashimoto
SPIE DOI: 10.1117/2.1200901.1477 (2009)
Source URL = http://spie.org/documents/Newsroom/Imported/1477/1477_5515_0_2009-01-16.pdf

Abstract: "Metal-reducing bacteria can construct electrically conductive networks using semiconductive nanominerals"

Direct quotes: "Recent reports demonstrated that metal-reducing bacteria can also use various artificial solid materials such as graphite, gold, and indium-tin-oxide (ITO) electrodes as a terminal-electron acceptor. This important result highlights the potential to harvest electricity by collecting waste electrons from bacteria through the electrode in microbial fuel cells (MFCs), a type of electrochemical fuel cell. A great deal of research has focused on identifying the OM proteins responsible for extracellular electron transfer and improving the MFC power density. However, the specific mechanisms of extracellular electron transfer have not been fully evaluated in a living system. In addition, the low power density of MFCs—approximately 500 μW/cm^2 in small-scale reactors— prevents widespread practical use."

"Direct evidence for the formation of an electrically conductive bacterial network was obtained by whole-cell cyclic voltammetry, a type of potentiodynamic-electrochemical measurement. We cultivated cells on the ITO electrode and confirmed that they exhibited a clear reduction-oxidation (redox) wave, having a midpoint potential (Em) of 20 *m*V: see Figure 1(b), trace 1. This value is in accordance with the E_m of OM c-Cyts isolated from *Shewanella* and thus demonstrates the c-Cyt-mediated electron exchange process at the cell-electrode interface. Also note that when we inoculated cells in the presence of α-Fe_2O_3 colloids the peak current showed a more than 300-fold enhancement: see Figure 1(b), trace 2."

"… Therefore, the radical enhancement of the redox current demonstrates the formation of a long-distance electron-transfer conduit in the colloidal network, which enables many cells located a considerable distance from the electrode to participate in current generation … We could thus describe long distance electron transfer on the basis of a semiconductor mediated electron-hopping process in which the OM c-Cyts serve as electrical glue in the α-Fe_2O_3 colloidal network: see Figure 1(c). The proposed model of long-distance electron transfer is analogous to the operational principle of dye-sensitized photovoltaic cells, where photogenerated charges are transported over a long distance by exploiting the properties of semiconductive nanomaterials … Semiconductive nanocolloids are abundant in natural minerals and it is not unlikely that bacteria have already taken advantage of semiconductive nanominerals in anaerobic environments to transport and share electrons in the bacterial network."

"***Electrodic voltages accompanying stimulated bioremediation of a Uranium-contaminated aquifer***"
K. Williams et al.
Journal of Geophysical Research **115** DOI:10.1029/2009JG001142 (2010)
Source URL = http://www.geobacter.org/publications/2009JG001142.pdf

Abstract: "The inability to track the products of subsurface microbial activity during stimulated bioremediation has limited its implementation. We used spatiotemporal changes in *electrodic potentials* (EP) to track the onset and persistence of stimulated sulfate-reducing bacteria in a uranium-contaminated aquifer undergoing acetate amendment. Following acetate injection, anomalous voltages approaching −900 mV were measured between copper electrodes within the aquifer sediments and a single reference electrode at the ground surface. Onset of EP anomalies correlated in time with both the accumulation of dissolved sulfide and the removal of uranium from groundwater. The anomalies persisted for 45 days after halting acetate injection. Current-voltage and current-power relationships between measurement and reference electrodes exhibited a galvanic response, with a maximum power density of 10 mW/m^2 during sulfate reduction. We infer that the EP anomalies resulted from electrochemical differences between geochemically reduced regions and areas having higher oxidation potential. Following the period of sulfate reduction, EP values ranged from −500 to −600 mV and were associated with elevated concentrations of ferrous iron. Within 10 days of the voltage decrease, uranium concentrations rebounded from 0.2 to 0.8 *μ*M, a level still below the background value of 1.5 *μ*M. These findings demonstrate that EP measurements provide an inexpensive and minimally invasive means for monitoring the products of stimulated microbial activity within aquifer sediments and are capable of verifying maintenance of redox conditions favorable for the stability of bioreduced contaminants, such as uranium."

Direct quotes: "Research efforts are currently underway to promote the in situ removal of soluble U(VI) species via stimulated bioremediation at the Rifle Integrated Field Research Challenge (IFRC) site near Rifle, Colorado (Figure 1), the site of a former mill tailings facility. Repeated biostimulation experiments at the site have shown that acetate injection into the aquifer promotes the growth of dissimilatory metal-reducing bacteria … *Geobacteraceae* and a decline in the U(VI) content of groundwater downgradient from the injection site. The reproducible, stimulated growth of *Geobacteraceae* along with the correlated loss of U(VI) suggests that these organisms are responsible for the in situ reduction of U(VI) …"

Prolonged addition of acetate to the aquifer results in a shift in the dominant terminal electron- accepting pathway from iron to sulfate reduction. This is evidenced by groundwater and sediment samples enriched in members of the order *Desulfobacterales* and a decrease in ferrous iron and sulfate within the aquifer. An unintended consequence of this shift in metabolism is an initial decrease in the rate of removal of soluble U(VI) from groundwater. At longer time points, however, and upon cessation of acetate injection, sustained removal of U(VI) under conditions of sulfate reduction does occur, ultimately reaching levels comparable to those achieved during the initial period of iron reduction."

"Here we show that the appearance and persistence of dissolved sulfide correlates with the generation of significant EP voltage anomalies indicative of subsurface microbial sulfate reduction. Furthermore, spatiotemporal variations in the onset and sustenance of the EP anomalies appear to track the location of active metabolism within the aquifer, offering an indirect means for verifying geochemically reduced conditions conducive to the removal of aqueous uranium."

"Following the injection of acetate, a pronounced increase in voltage magnitude was observed for both wells at all electrode locations (Figures 3 and 4). As a result of the measurement convention, the anomalous EP effect is reported as an increase in voltage magnitude, with values becoming more negative with time (e.g., −400 mV to −900 mV)."

"Upon reaching values between −850 and −900 mV, the EP values fluctuated within the range from −740 to −900 mV throughout the period of acetate injection. [20] Although injection of acetate into the aquifer was halted after 68 days, EP values more negative than −800 mV persisted for an additional 50 days (Figure 3). The length of time that such voltages persisted generally increased with depth. After a total elapsed time of 115 days, or 47 days after acetate injection was halted, EP values across the entire measurement interval approached relatively uniform levels of −550 ± 40 mV."

"Current flow from the deepest electrode location in SP-1 (6.5 m bgs) to the surface electrode increased approximately 4 orders of magnitude with a concomitant fortyfold decrease in voltage over the resistance range measured (Figure 5). When normalized to the surface area of the measurement electrode (50 cm^2), the resulting current density over this resistance range increased from 0.004 to 46 mA/cm^2. Power densities between the electrodes showed a parabolic trend reaching a maximum value of 9.7 mW/cm2 at 400 W. Similar values were observed for the same electrode pair 85 days after acetate injection began. This was not the case following the cessation of acetate injection. By day 150, both current and power density decreased dramatically, reaching maximum values of 2.4 mA/cm^2 and 0.305 mW/cm^2, respectively. Identical measurements made between SP-2 and the surface electrode revealed a similar trend, with anomalously high current flow and power production during acetate injection."

"Overall an excellent temporal correlation was found to exist between the downgradient EP response and the sulfide and ferrous iron concentrations, E_h, and pH values observed in M-21 … Dissolved uranium concentrations began to decrease downgradient of the injection gallery within 5 days after starting acetate injection (Figure S6). Uranium concentrations decreased to their lowest level after 27 days, after which time they rebounded somewhat for the duration of the injection. The initial decrease coincided with the accumulation of Fe^{2+} and extended into the period of sulfide accumulation. The rebound in uranium closely tracked the rapid increase in pH (Figure 6c) and the near complete removal of sulfate (data not shown). Following the end of acetate injection, uranium concentrations fell to their lowest levels, with a high degree of removal occurring for another 50 days. EP values in excess of −800 mV correlated generally with high rates of uranium removal over the entire experimental period and strongly in the period following the cessation of acetate injection. Once EP values fell below −650 mV, significant removal of uranium continued for a period of less than 15 days."

"Laboratory EP measurements were made using a galvanic cell to explore the mechanism underlying the anomalous voltages observed during sulfate reduction … Current-voltage and current-power relationships using the 1 mM concentration confirmed the transfer of electrons from anode to cathode upon completion of the circuit, with the results (data not shown) closely matching those obtained on day 58 of the field experiment."

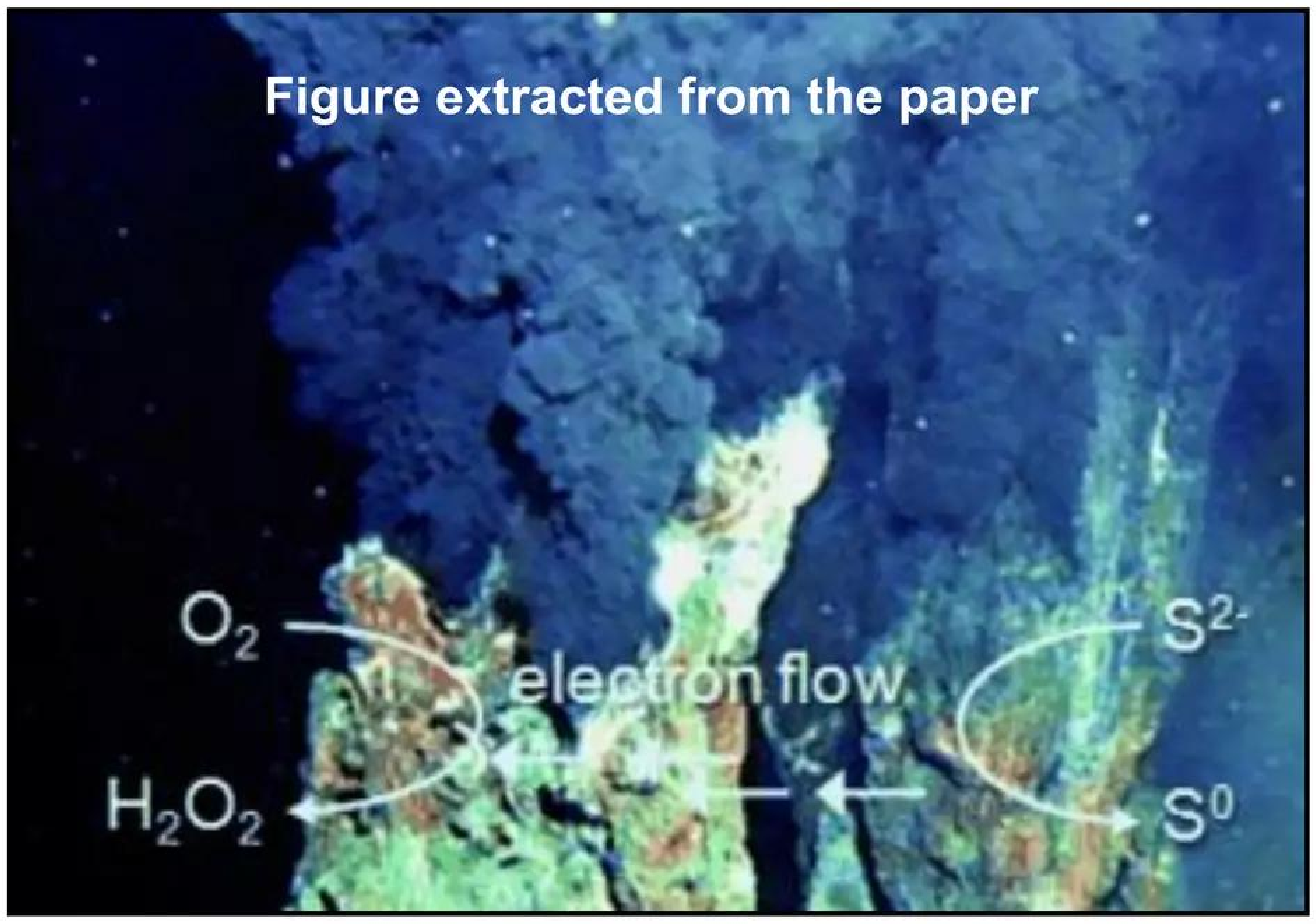
"Our results confirm that electrochemical interactions between an electrode surface and the surrounding environment over spatial scales exceeding 10 m can generate significant EP anomalies. We attribute the anomalies to electrochemical concentration gradients that developed between both sulfide- and Fe^{2+} -rich regions and areas having higher oxidation potential (e.g., the ground surface). Temporal voltage variations were diagnostic of the dominant metabolic process during and after acetate amendment and exhibited a general correlation with enhanced rates of uranium removal."

"Evidence in support of a galvanic mechanism underlying the EP response comes from the power production results. We observed significant electron flow through the circuit created by connection of the measurement and reference electrodes. Current and power densities are in excellent agreement with those reported for sulfide dependent galvanic fuel cells. If the transfer of electrons from reduced to oxidized locations via an endogenous conductor had been the dominant source of the measured potentials, current flow through the circuit would have been negligible. The EP signals are therefore unlikely to have resulted from electron transfer through biogenic materials or mineral phases and anomalies of the magnitude reported here cannot be generated via electrochemical diffusion driven by chemical concentration gradients."

Comment: these well-documented anomalous electrodic potentials were observed at the Rifle, Colorado, Uranium bioremediation site. They were clearly occurring in parallel with bacterial reduction of Uranium and Sulfur --- this is another 'must read' publication in the context of the subject matter of this document.

"***Electrical current generation across a Black Smoker chimney***"
R. Nakamura et al
Angewandte Chemie International Edition **49** pp. 7692–7694 (2010)
Source URL = free copy unavailable

Abstract: “Some like it hot: metal-like electrical conduction and electrocatalytic function of a black smoker chimney point to a possible new form of energy transfer from hot, reductive hydrothermal fluid to cold, oxygenated seawater by electrical current generation in the sulfide chimney wall.”



Brief commentary about the above paper found on the website of The Sugar Project:
Source URL = http://www.jamstec.go.jp/biogeos/e/xbr/sugar/achievements.html

Direct quote: “The SUGAR Program has worked in collaboration with Prof. Kazuhito Hashimoto group of Tokyo University, and carried out an electrochemical analysis with the sample collected from the chimney from the deep sea hydrothermal vent. From this experiment, it made clear of the chimney composed by Black Smoker, made of sulfide minerals possessing a high electrical conductivity. In addition, the presence of electrical current across the wall of the Chimneys was proved. This electrical current occurs due to the chemical reactions similar to the battery between inside and outside of the Chimney in natural environment. These experimental results are the world-first proof of the Earth’s internal energy being converted into electrical energy, when released at the seafloor. This Hydrothermal Chimney Battery phenomenon will surely explain more about the origin- of-life mechanisms, as well as deep sea biological activity and material circulation.”

“***Source of sustained voltage difference between the xylem of a potted Ficus benjamina tree and its soil***”
C. Love et al.
PLoS One **3** pp. 1 – 5 (2003)
Source URL =
http://www.plosone.org/article/fetchObjectAttachment.action;jsessionid=8CAFB45093865EDF6F620DA4B16D6D8A.ambra02?uri=info%3Adoi%2F10.1371%2Fjournal.pone.0002963&representation=PDF

Abstract: “It has long been known that there is a sustained electrical potential (voltage) difference between the xylem of many plants and their surrounding soil, but the mechanism behind this voltage has remained controversial. After eliminating any extraneous capacitive or inductive couplings and ground-mediated electric current flows, we have measured sustained differences of 50–200 mV between the xylem region of a Faraday-caged, intact, potted *Ficus benjamina* tree and its soil, as well as between its cut branches and soils and ionic solutions standardized to various pH values. Using identical platinum electrodes, no correlation between the voltage and time of day, illumination, sap flow, electrode elevation, or ionic composition of soil was found, suggesting no direct connection to simple dissimilar-metal redox reactions or transpirational activity. Instead, a clear relationship between the voltage polarity and magnitude and the pH difference between xylem and soil was observed. We attribute these sustained voltages to a biological concentration cell likely set up by the homeostatic mechanisms of the tree. Potential applications of this finding are briefly explored.”

Additional important questions for experimentalists - surface of this area has barely been scratched:

Presuming bacteria can engage in nucleosynthesis, why pray tell would earthly microorganisms still be manipulating isotopes of Actinide elements? Is it simply an ancient vestigial capability coded in organisms' genomes, or does it serve some useful biological purpose in the present era? Specifically, what network pathways are typically traversed, what organisms are involved in each segment of those pathways, and in what natural environments does/can such processes occur?

Are the observed biologically-induced *decreases* in measured U(VI) Uranium concentration and changes $^{238}U/^{235}U$ ratios over time simply a reflection of the conversion of Uranium isotopes into different chemical forms and concomitant chemical "fractionation" between those forms, e.g., from mobile and bioavailable U(VI) compounds (measured by Rademacher et al. and Bopp et al.) to fractionated, highly insoluble reduced U(IV) compounds (not presently measured); or are some, at the moment, indeterminate percentages of U(VI) and/or U(IV) Uranium atoms actually being transmuted to other elements entirely? If so, which elements/isotopes are produced and where do they go in the natural environment?

Is Plutonium produced by the Actinide LENR network in Nature? If so, is it further transmuted from there?

To answer such questions, an exhaustive isotopic 'inventory' and total mass balance of elements/isotopes comprising the theorized Actinide LENR transmutation network would have to be measured by experimentalists in different types of laboratory experiments and natural environments.

Speculation - imagine what might be possible in the future with commercial applications of LENRs:

In the near future, as suggested by recent work of Vysotskii et al. (2008), it is theoretically possible that collections of carefully selected bacterial species situated in a man-made 'transmutation network ecology' could be utilized to vastly speed-up remediation and 'destruction' (i.e., LENR-based transmutation of long-lived Actinides to less environmentally dangerous, faster-decaying isotopes) of accumulated high-level wastes from nuclear fission reactors located all over the world. While substantial amounts of time, money, and man-hours would be needed to achieve such a goal, the experimental data discussed herein strongly suggests that it may be possible to do so. If this were achieved, it could revolutionize the economics and dramatically reduce long-term environmental hazards of present-day fission technologies.

In the more distant future, maybe 'nuclear-capable' bacteria could be genetically altered to harness their LENR-based nucleosynthetic capabilities to produce a range of different valuable isotopes that specifically suit Man's purposes? If that were ultimately accomplished, it would appear that almost any extremely expensive (stable) element in the periodic table such as Gold and Platinum could potentially be produced by genetically altered bacteria, perhaps even at economic costs that are competitive with traditional mining and refining technologies. If achieved, this would be a 'black swan' technological shift.

First international workshop on LENR biological transmutations will be held early in 2011:

The first-ever one day workshop on the subject of biological transmutations, "*Biological Nuclear Transmutations: Historical Perspective and Applications*" will be held on February 15, 2011, at the SRM University (SRMU), Kattankulathur campus near Chennai, India. The main lectures will be given by Prof. Vladimir Vysotskii of the Kiev National Shevchenko University, Ukraine. This workshop is being jointly organized by the Kalpakkam Chapter of the Indian Institute of Metals and the Biotechnology Department of SRMU. For further information see:

Source URL = http://www.iscmns.org/iccf16/course_on_transmutation.htm

Final comments:

Thanks to new insights provided by the Widom-Larsen theory of LENRs, I believe that the interactions between microorganisms and their co-evolving geochemical environment could be more interesting and dynamic than anyone could have ever imagined. If the LENR-based Actinide nucleosynthetic network occurs somewhere in Nature, as I think it probably does, there will be outstanding opportunities for researchers to discover new and important details of LENR-driven biological and abiological nuclear processes and to develop many valuable, breakthrough commercial technologies from such discoveries.