

# Interaction of Radiation with Matter

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II Letnia Szkoła Energetyki i Chemii Jądrowej



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**UNIA EUROPEJSKA**  
EUROPEJSKI  
FUNDUSZ SPOŁECZNY



# **Interaction of Radiation with Matter**

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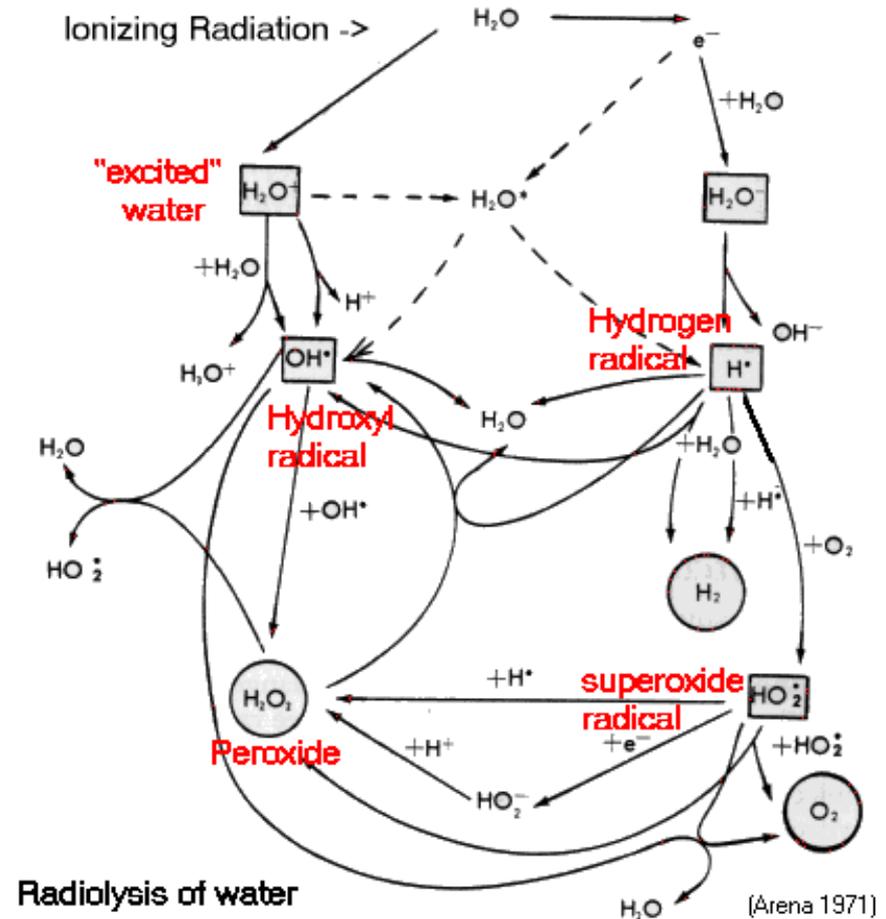
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# Outline

- Interaction of radiation with matter
- Dosimetry
  - Units
  - Measurements
- Hot Atom Chemistry
- Examples with radionuclides
  - Chemical behavior in nuclear fuel
  - Radiolysis studies
  - Decay and separations



# Interaction with matter overview

- **Neutrons**
  - **Very little interaction with electron, primary ionization is negligible**
  - **Interaction confined to nuclear effect**
    - scattering (elastic and inelastic)
    - reactions (n, $\gamma$ ), (n,p), (n, $\alpha$ ), (n,2n)
    - fission
- **Positive Ions**
- **Electrons**
- **Electromagnetic**

# Interaction of Radiation with Matter

- Interaction of radiation with matter leads to
  - dissociation of molecules
  - excitation of atoms or molecules
  - ionization of atoms or molecules
- Ionization is easily measured and often used for detection
- In air about 35 eV of energy are dissipated for each ion pair formed Value varies with gas
- Xe: 21.9 eV, He: 43 eV, NH<sub>3</sub>: 39 eV (IP = 10.8 eV), Ge: 2.9 eV

Radiation detected by interaction with matter

Interactions ultimately have the same effect (35 ev/ion pair formation)

Measuring total number of ions produced as a means to determine energy

## Energy Loss Overview

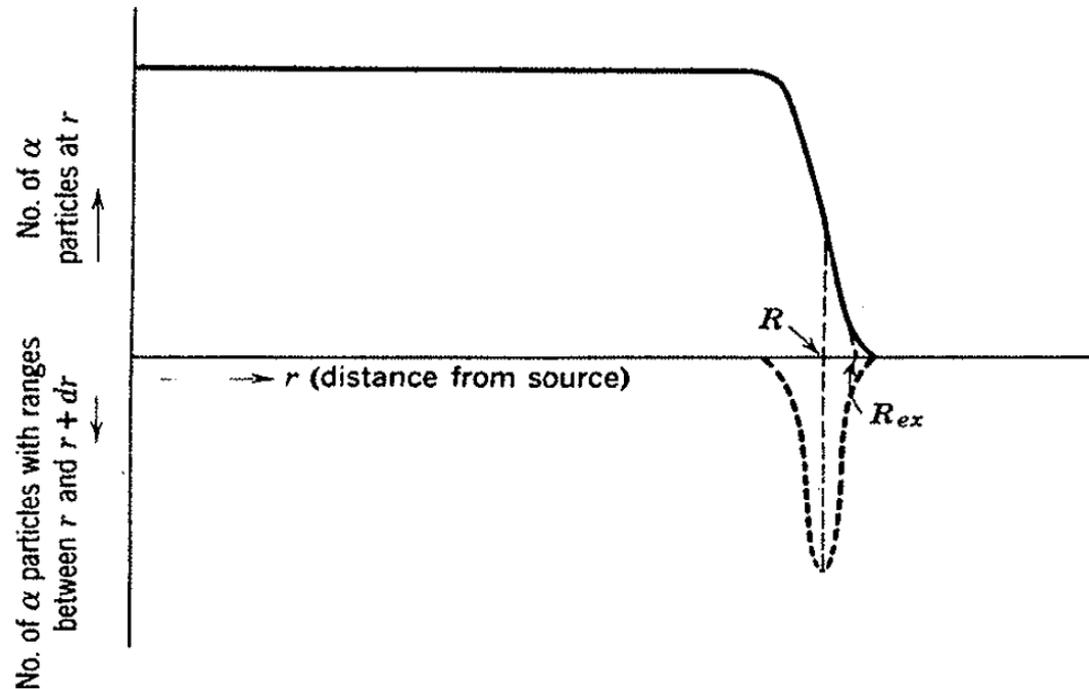
1. At sufficiently high energy ion is bare and energy loss is through electronic excitation and ionization of stopping material
  2. At velocities comparable to the K-shell electron ion begins to pick up electrons, stopping is still electronic
  3. At velocities comparable to valence electrons elastic collisions account for energy loss
    - nuclear stopping
- No sharp difference point of loss by method 2 and 3 between elastic and inelastic collisions

# Positive Ions

- **Processes for energy loss**
  - **Chiefly by interactions with electrons**
- **Maximum velocity (v) imparted to electron is 2v**
  - **$K_E = 0.5mv^2$ ;  $v = (K_E/0.5m)^{1/2}$** 
    - **Consider maximum energy from 6 MeV alpha to electron**
    - **Average energy from ions to electrons is 100-200 eV**
  - \* **Secondary ionization**
- **Electronic stopping**
  - **inelastic collisions between bound electrons and ion**
    - **Excitation of atomic electrons**
- **Nuclear stopping**
  - **velocity of ion close to velocity of valence electrons**
  - **elastic collisions dominate**
- **Velocity of the ion comparable to K shell electron, ion begins to pick up electrons**
  - **Ions passing through matter**
    - **stripped of all orbital electrons whose orbital velocity is less than ion velocity**

# Range

- Due to large mass of positive ion compared to electron
- Distances that positive ions travel in matter are in narrow limits
  - Particle and energy dependent
  - Defined as range
- Large mass drives behavior
  - Fractional energy loss per collision is small
    - large number of collisions required to stop ion
  - Deflection of ion in each collision is small
- Straggling is fluctuations in average energy loss and projected path
  - order of a few percent



Number of ions from a point source  $f_n(\text{distance})$

# Stopping Power

Relationship between energy of a positive ion and the range is seen as  $dE/dx$

- Defined as stopping power
  - Function of the energy, charge, and ion mass
- Maximum rate of energy loss at low energies

- Energy loss per distance travelled of charged particles
- Relativistic and non-relativistic approach
- Momentum transfer through Coulomb interaction

$\gamma$ -average fraction of electrons stripped off ion

$z$ -ion charge

$N$ -absorber atoms per unit volume

$Z$ -absorber  $Z$

$m$ - electron mass

$v$ -ion velocity (not electron velocity)

$I$ -effective ionization potential

- Rate of energy loss for charged particles moving with the same velocity in a given absorber proportional to squares of charges

- $^1\text{H}$  energy  $E$ ,  $^2\text{H}$   $2E$ , and  $^3\text{H}$   $3E$  all same  $dE/dx$
- 25 % of alpha  $4E$

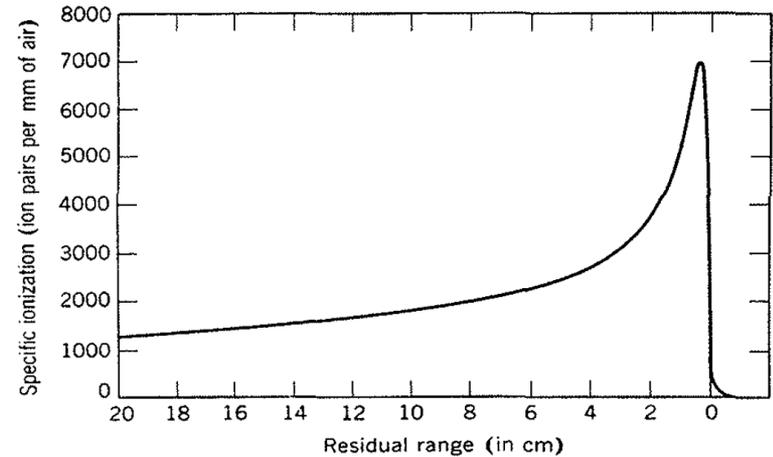


Fig. 6-2 Bragg curve for initially homogeneous  $\alpha$  particles.

$$-\frac{dE}{dx} = \frac{4\pi\gamma^2 z^2 e^4 NZ}{mv^2} \left[ \ln \frac{2mv^2}{I} - \ln(1 - \beta^2) - \beta^2 \right]$$

- $\beta = v/c$

$$-\frac{dE}{dx} = \frac{4\pi\gamma^2 z^2 e^4 NZ}{mv^2} \left[ \ln \frac{2mv^2}{I} \right]$$

- When  $\beta \ll 1$

# Stopping power of ions in Al

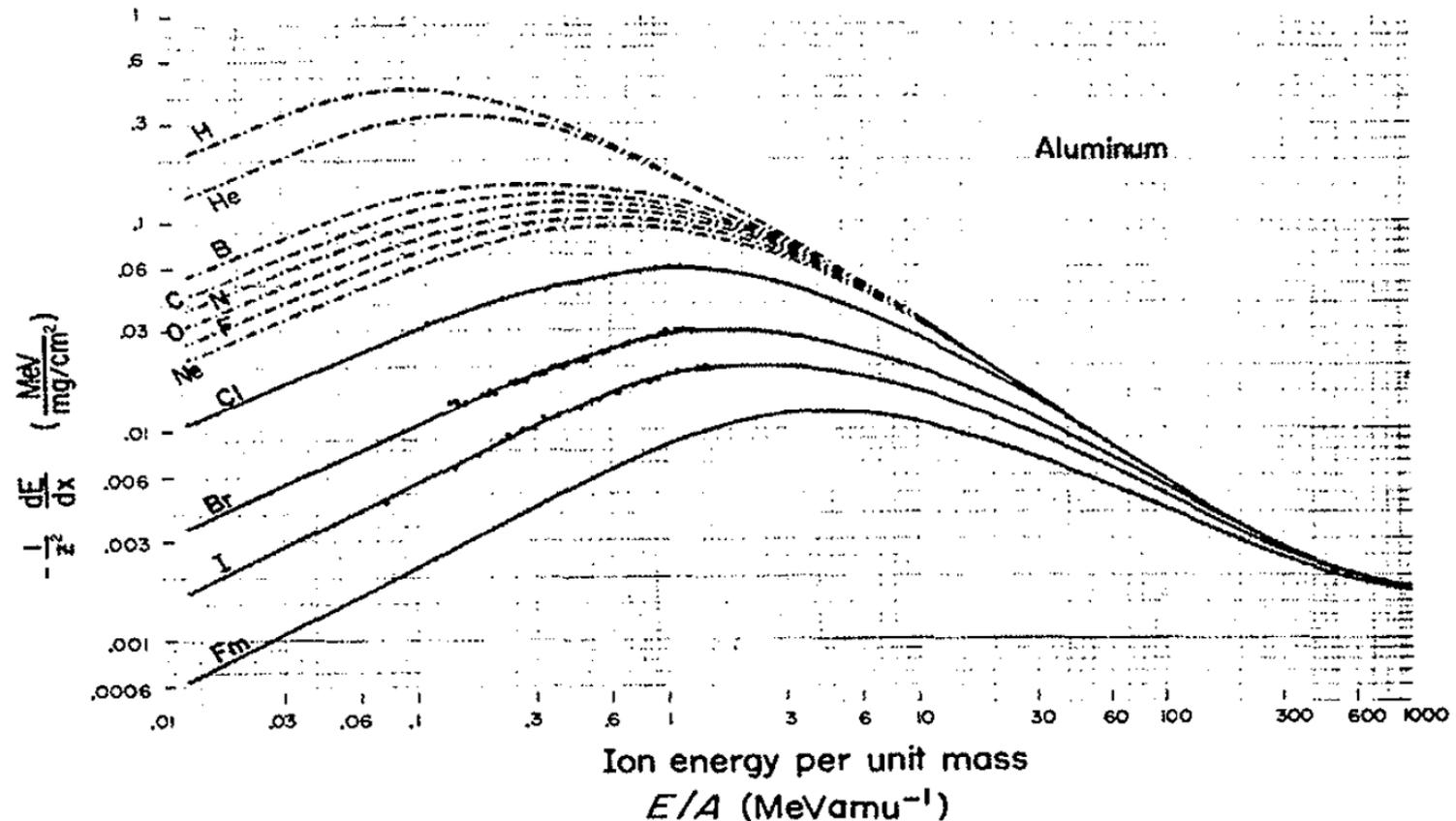


Fig. 6-4 Stopping power curves for various ions in aluminum. (From reference N1.)

- Rate of energy loss is not the same for different ions with the same energy
  - $^{16}\text{O}$ ,  $^{15}\text{O}$ ,  $^{14}\text{N}$  at 80 MeV in Al
    - Different isotopes examined with  $E/A$
  - 3.46, 3.32, and 2.49  $\text{MeV mg}^{-1}\text{cm}^2$

# Proton stopping in C and U

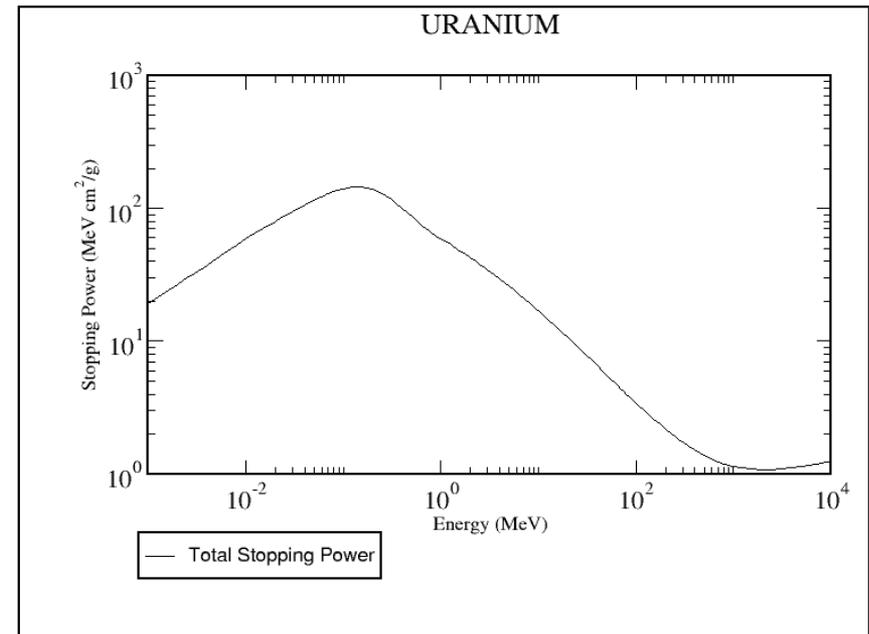
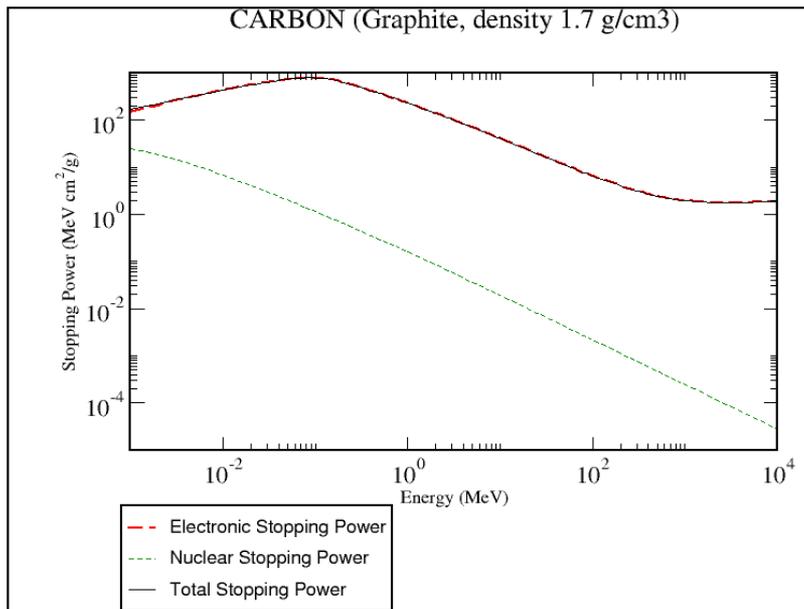
- Calculate thickness to decrease energy
- Determine stopping power from particle energy

## • C thickness to stop 10 MeV proton

- $4.084\text{E}+01 \text{ MeV cm}^2\text{g}^{-1}$
- $10 \text{ MeV} / 4.084\text{E}+01 \text{ MeV cm}^2\text{g}^{-1} = 0.244 \text{ g cm}^{-2}$
- Use density of material ( $1.7 \text{ g cm}^{-3}$ )
- $0.244 \text{ g cm}^{-2} / 1.7 \text{ g cm}^{-3} = 0.144 \text{ cm}$  of graphite

## • U thickness to stop 10 MeV proton

- $1.690\text{E}1 \text{ MeV cm}^2\text{g}^{-1}$
- $10 \text{ MeV} / 1.690\text{E}1 \text{ MeV cm}^2\text{g}^{-1} = 0.592 \text{ g cm}^{-2}$
- Use density of material ( $19.1 \text{ g cm}^{-3}$ )
- $0.592 \text{ g cm}^{-2} / 19.1 \text{ g cm}^{-3} = 0.031 \text{ cm}$  of U

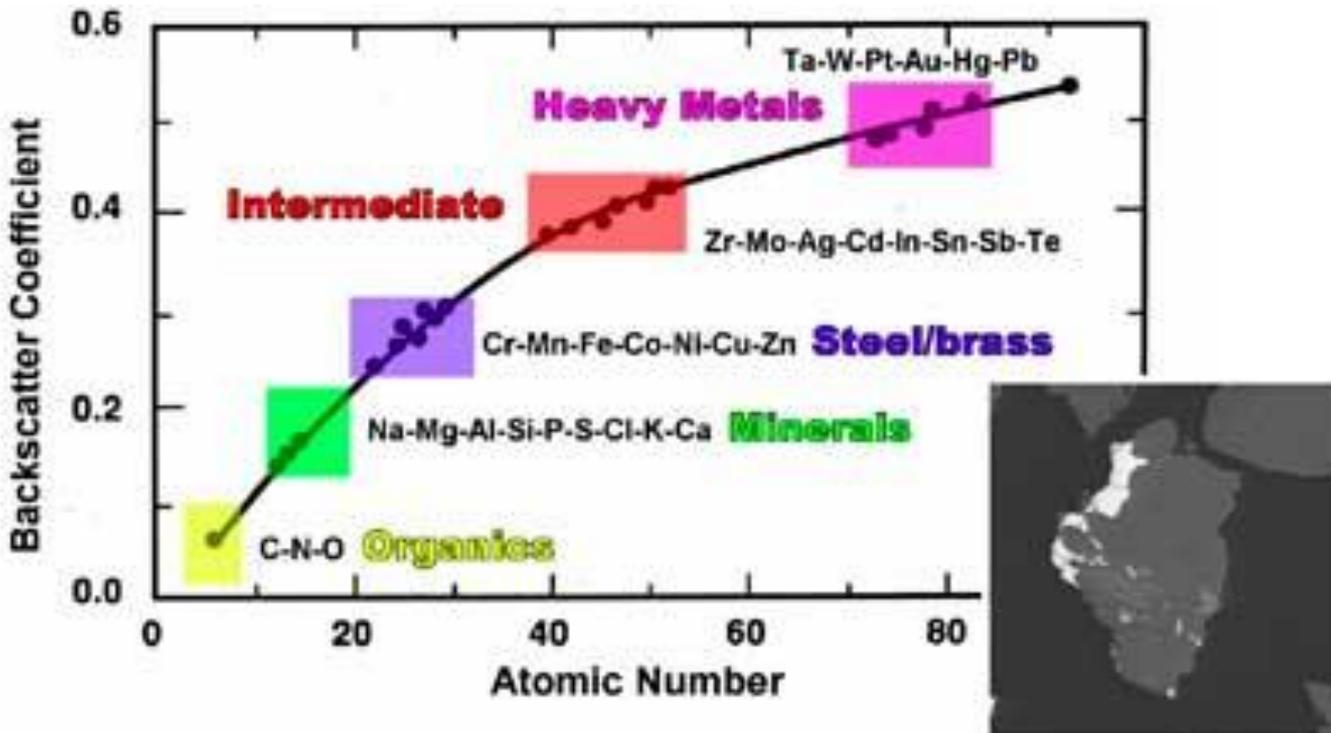


# Electrons

- **Energy loss**
  - **similar to that of positive ions**
  - **average ion pair formation about 35 eV in air**
  - **70-80% of ionization is secondary**
- **Electron has less mass than positive ions**
  - **For the same energy, higher velocity**
  - **Lower stopping power**
- **Maximum at 146 eV (5950 ion pairs per mg/cm<sup>2</sup>)**
- **In air ionization stops around 12.5 eV**
- **electron can lose a large fraction of energy in one collision**
- **Straggling is more pronounced**
- **Energy loss through electron interaction, nuclear scattering**

# Backscattering

- Significant fraction of electrons may be reflected from scattering
- Reflected intensity increases with increasing thickness of reflector
  - Saturation can be achieved
- Ratio of measured activity beta source with reflector to that without reflector is back-scattering factor
- Factor varies with material
  - Used to determine Z of material

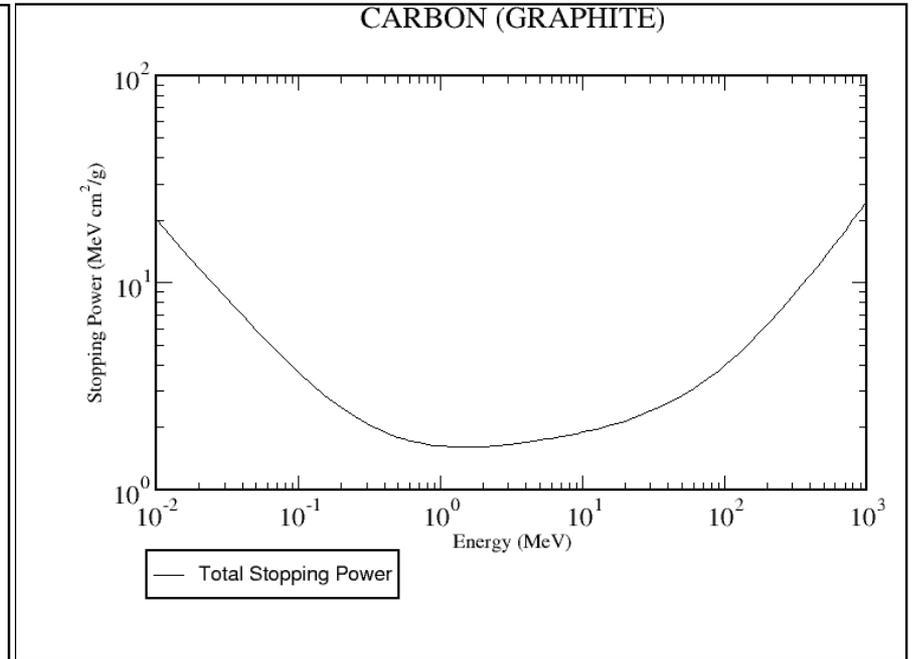
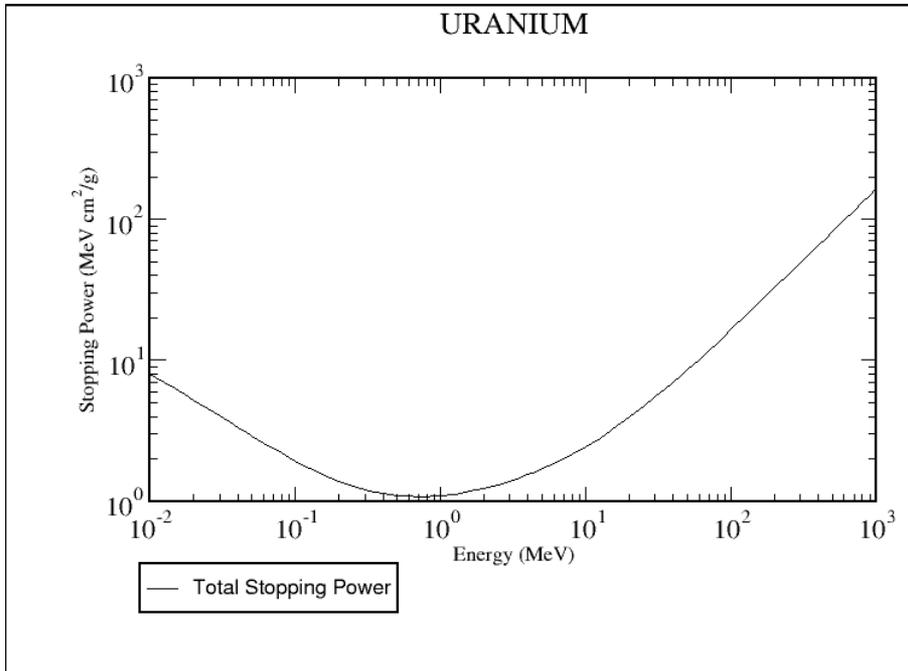


**Table 6-1 Specific Ionization and Velocity for Electrons of Various Energies in Air**

Velocity (in Units of the Velocity of Light, $c$ )	Energy (MeV)	Ion Pairs per 1.00 mg $\text{cm}^{-2}$
0.001979	$10^{-6}$	0
0.006257	$10^{-5}$	0
0.0240	$1.46 \times 10^{-4}$	5950 (maximum)
0.1950	$10^{-2}$	~850
0.4127	0.05	154
0.5483	0.10	116
0.8629	0.50	50
0.9068	0.70	47
0.9411	1.0	46
0.9791	2.0	46
0.9893	3.0	47
0.9934	4.0	48
0.9957	5.0	49
0.9988	10	53
0.99969	20	57
0.999949	50	63
0.9999871	100	66

# Electron Stopping Power

- Distance to stop 1 MeV electron in U metal
  - 1.1 MeV  $\text{cm}^2\text{g}^{-1}$
  - 1 MeV/1.1 MeV  $\text{cm}^2\text{g}^{-1} = 0.909 \text{ g cm}^{-2}$
  - Use density of material (18.95  $\text{g cm}^{-3}$ )
  - $0.909 \text{ g cm}^{-2} / 18.95 \text{ g cm}^{-3} = 0.048 \text{ cm}$  of U metal



# Photons

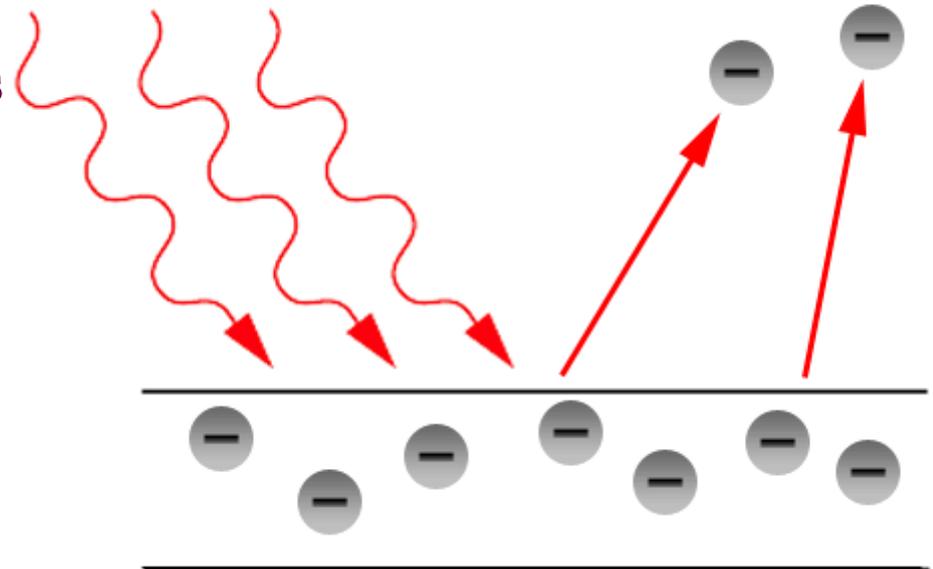
- Lose most energy in a few interaction or a single interaction
- Need more material for interaction than electron
- Average specific ionization is less than electron (10%)
- Average energy loss per ion pair formation in air is 35 eV

## Photoelectric effect (For low energies)

- photon with energy  $h\nu$  ejects a bound electron and imparts energy  $h\nu - \epsilon_b$  to electron where  $\epsilon_b$  is electron binding energy
- Mostly K-shell, some L-shell (about 20%)
- Proportional to  $Z^5$  of absorber

For 5% photoelectric effect,  $\gamma$  energy needed for different  $Z$

Al- 0.15 MeV, Cu-0.4 MeV, Sn- 1.2 MeV, Pb-4.7 MeV



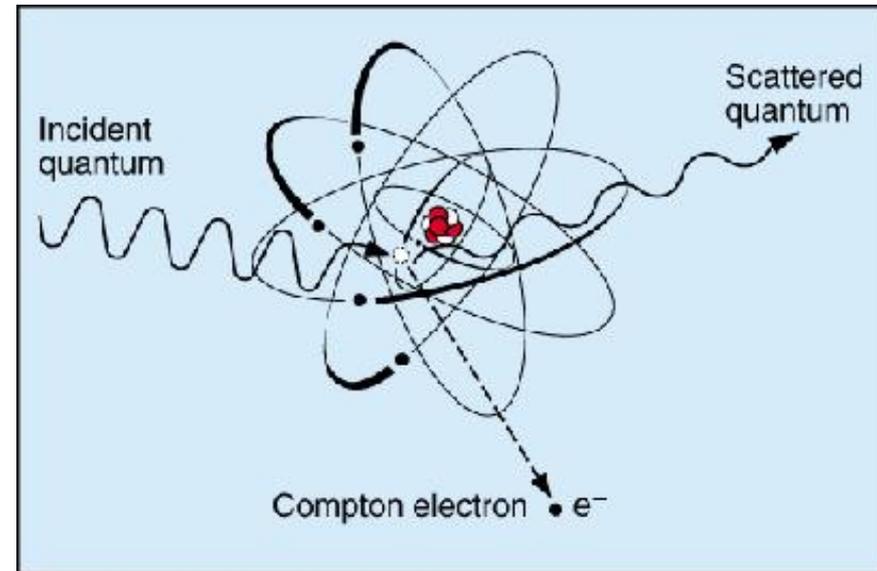
# Compton Effect

- Photon loss part of energy to electron
- Photon is scattered
- Minimum for scattered photon is

$$(E'_\gamma)_{\min} = \frac{E_0}{2} \frac{1}{1 + \frac{E_0}{2E_\gamma}}$$

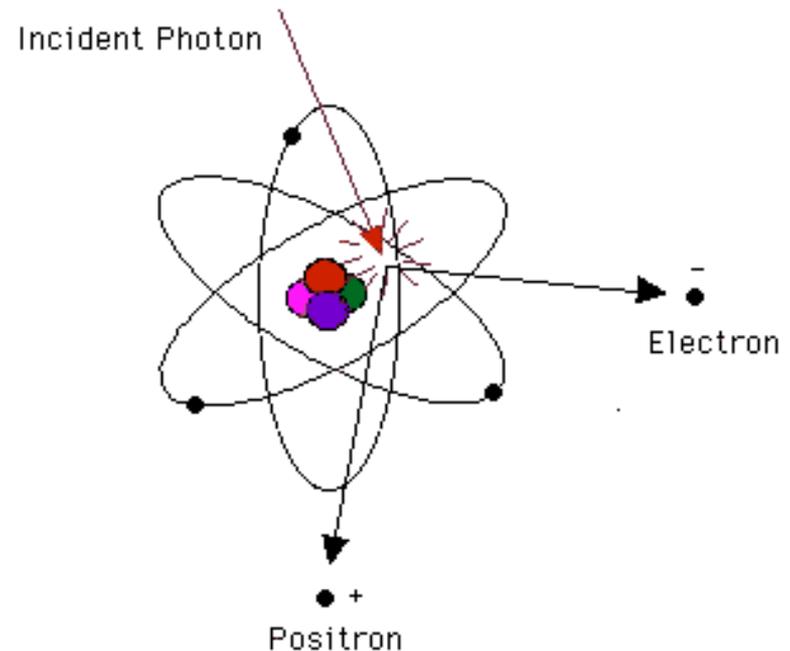
$E_0$  = electron rest energy

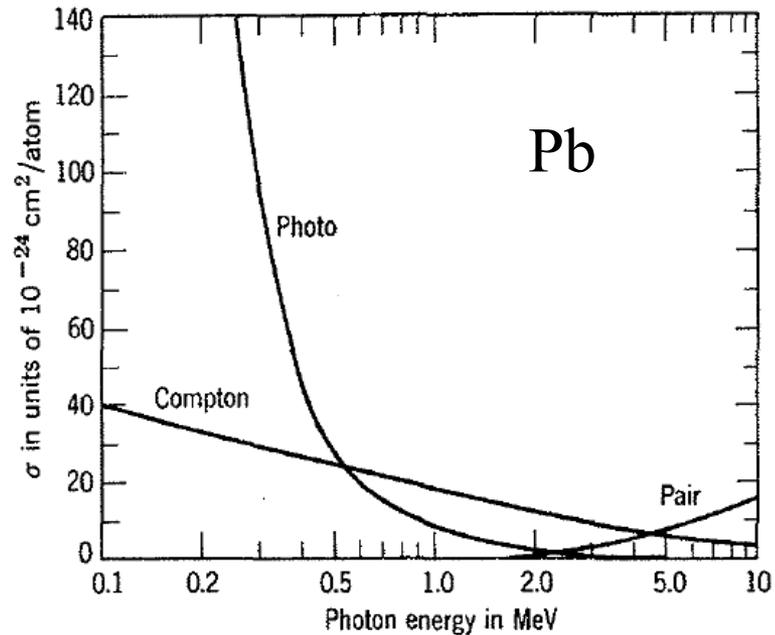
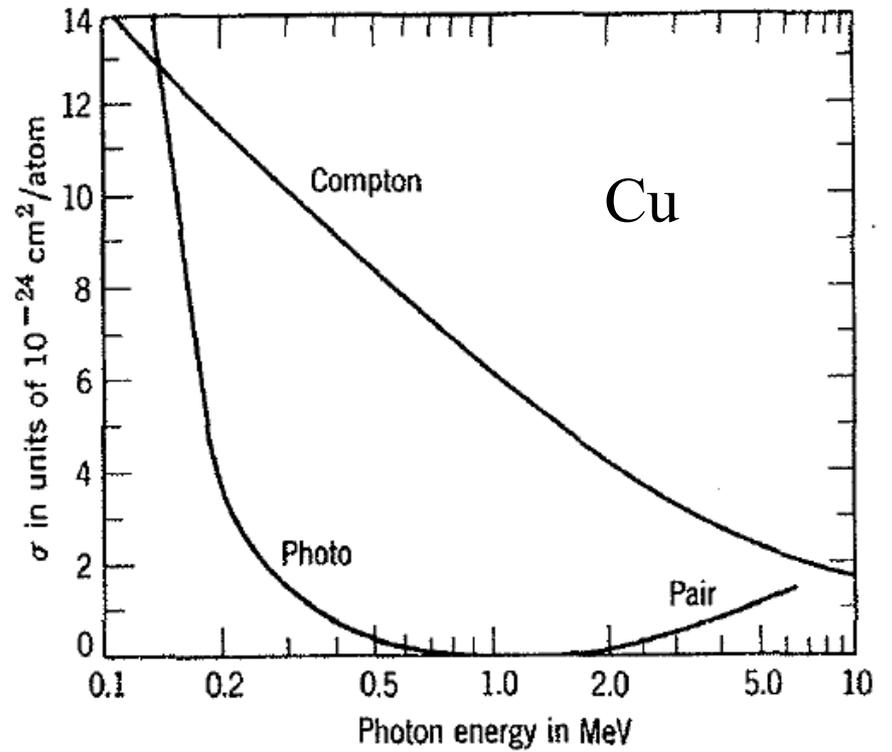
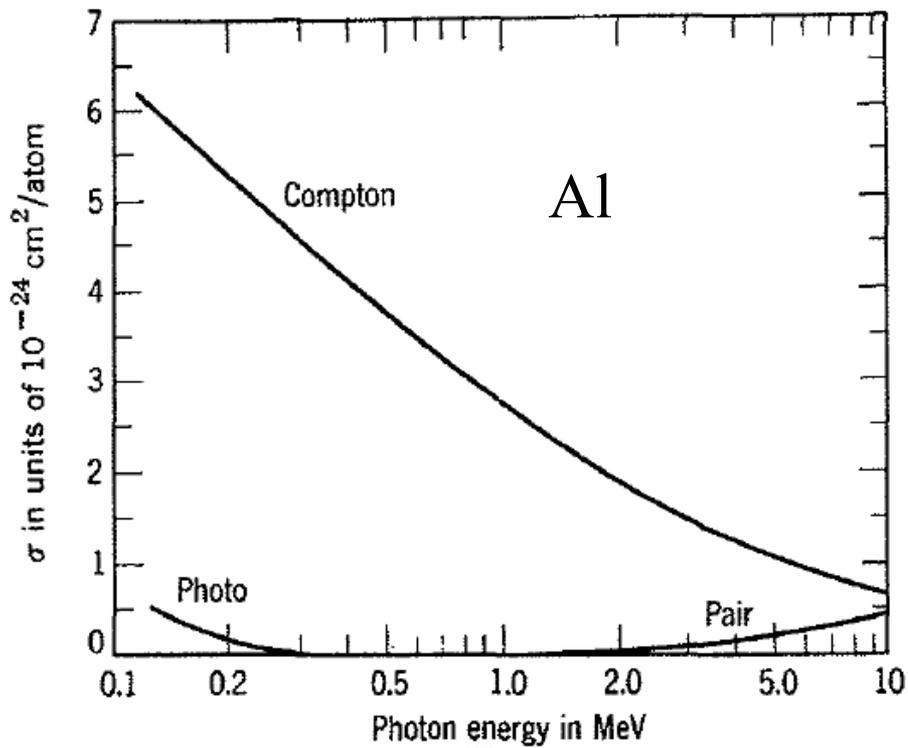
Back scattering peak can be seen on spectra



# Pair Production

- Production of  $\beta^+$  and  $\beta^-$
- Proportional to energy ( $\log E$  about 4 MeV) and  $Z^2$
- More common at high energy
- 511 keV from positron-electron annihilation





**Energy Dependence for photon Effects with different materials**

# Dose Units

Radicals are formed by the interaction of radiation with water

Radicals drive reactions

## Dosimetry

- Quantitative relation between specific measurement in a radiation field and chemical and/or biological changes
  - dose effect relationship
  - caused by production of ionized molecules, atoms, secondary electrons
  - chemical changes, biological effects

## • Absorbed Dose

- energy absorbed per unit mass of target for any kind of ionizing energy
  - Gray (Gy) = 1J/kg
  - in US; rad = 100 erg/g
  - $1\text{J/kg} = 10^7 \text{ erg}/10^3 \text{ g} = 10^4 \text{ erg/g} = 10^2 \text{ rad}$
- Absorbed dose is referred to as dose
  - Treated as point function, having a value at every position in an irradiated object

$$1 \text{ eV} = 1.60\text{E-}19 \text{ J}$$

$$1 \text{ charge pair separation} = 1.60\text{E-}19 \text{ C}$$

# Dose Equivalent

Absorbed dose needed to achieve biological effect is different for different types of radiation

Radiation	QF
X and γrays	1
Electrons and Positrons	1
Neutrons, E < 10 keV	3
Neutrons, E > 10 keV	10
Protons	1-10
Alpha Particles	1-20
Heavy Ions	20

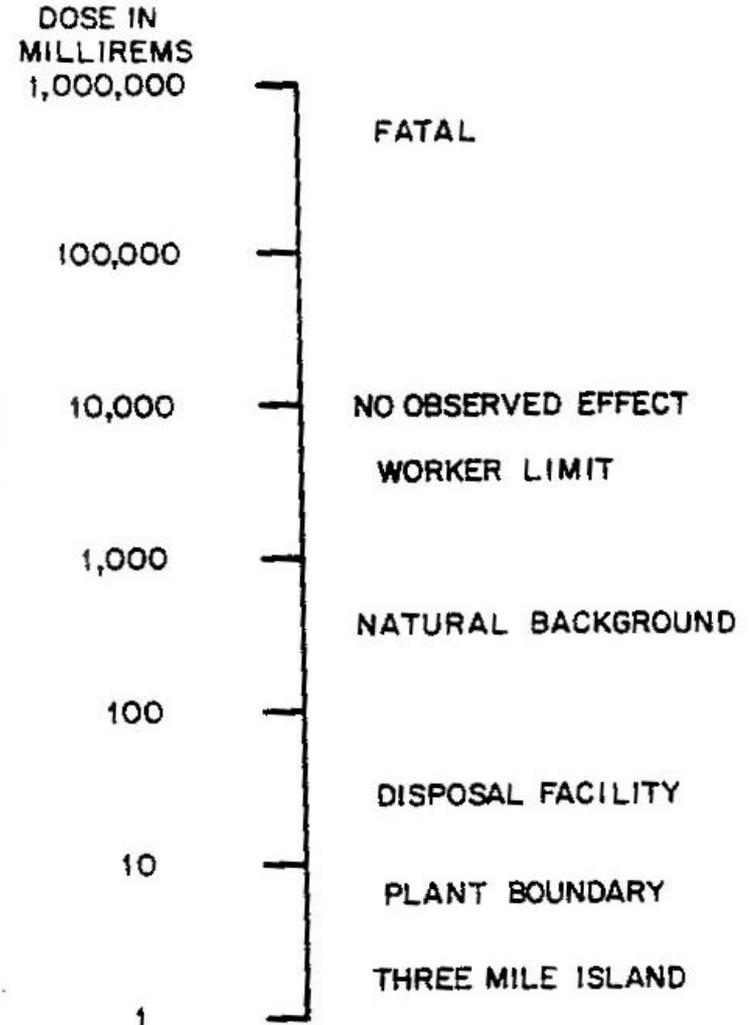
- Difference due to high versus low linear energy transfer (LET)
- Dose equivalent compensates for this difference
- $H$  (dose equivalent) =  $QD$
- $Q$  is dimensionless, has some different values
- $Q = \text{fn}(\text{particle, energy}); 1 \leq Q \leq 20$   
 →  $Q$  from NCRP Report 116
- uses LET (L) in keV/μm in water

## Q Dependence on LET

LET (L) (keV/μm in water)	Q
<10	1
10-100	$0.32L - 2.2$
>100	$300/L^{0.5}$

# Radiation Protection

- **3.6 mSv/y** Typical background radiation to US public ( $\approx$  80% from natural sources)
- **2.4 mSv/y** Average dose to US nuclear industry employees. (0.01mSv/y to public)
- **350 mSv in lifetime** Criterion for relocating people after Chernobyl
- **50 mSv annual worker dose limit** in US
- **15  $\mu$ Sv to public** from Three Mile Island in 50 mile radius
- **0.4 mSv** from dental x-rays
- **1000 mSv** as short term dose: causes (temporary) radiation sickness.
- **5000 mSv** as short term dose: would kill about half those receiving it within a month.
- **10,000 mSv** as short term dose: fatal within days



# Dose Equivalent

- **When dose in Gy, dose equivalent is Sv**
- **When dose in rad, dose equivalent is rem (roentgen-equivalent-man)**
- **1 Gy = 100 rad, 1 Sv = 100 rem**
- **Particle type and energy should be explicitly considered**
- **Biological distribution can depend on isotope**
  - **I goes to thyroid**
  - **Sr, Ra go to bone**
  - **Cs, H go all over**
  - **Metals go towards liver**
  - **Complexes can be released in kidneys**

# Biological Effects Concepts

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Time	Event
$10^{-18}$ seconds	Absorption of Ionizing Radiation
$10^{-16}$ seconds	Ionization, Excitation
$10^{-12}$ seconds	Radical formation, bond breakage
$10^{-12}$ to $10^{-6}$ seconds	Radical reaction
Min. to Hrs.	Cellular Processes
Hrs. to Months	Tissue Damage
Years	Clinical effects
Generations	Genetic Effects

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- **Linear Effect of Dose**

- **Any amount radiation above background is harmful**
- **Basis of radioisotope exposure limits**
- **<http://www.nrc.gov/reading-rm/doc-collections/cfr/part020/>**
- **Low level radiation effect not so clear**

# Intake limits

- **Air and water**
  - **nuclide specific (include daughter)**
  - **Class refers to lung retention (Days, Weeks, Years)**
  - **Annual limits on Intake (ALI) derived from 0.05 Sv total dose or 0.5 Sv dose to an organ or tissue**
  - **Derived air concentration (DAC) comes from ALI**  
 **$DAC = ALI / (2000 \text{ hr} \times 60 \text{ min/hr} \times 2E4 \text{ mL/min})$**

## Americium-241

Atomic No.	Radio-nuclide	Class	Table 1 Occupational Values			Table 2 Effluent Concentrations		Table 3 Releases to Sewers
			Col. 1	Col. 2	Col. 3	Col. 1	Col. 2	
			Oral Ingestion ALI (μCi)	Inhalation		Air (μCi/ml)	Water (μCi/ml)	Monthly Average Concentration (μCi/ml)
	ALI (μCi)	DAC (μCi/ml)						
95	Am-241	W, all compounds	8E-1 Bone Surf	6E-3 Bone Surf	3E-12	-	-	-
			(1E+0)	(1E-2)	-	2E-14	2E-8	2E-7

Isotope data found at: <http://www.nrc.gov/reading-rm/doc-collections/cfr/part020/appb/>

# Laboratory usage

- ALI and DAC basis of activities levels in the laboratory
  - [http://rms.unlv.edu/radiological/Form%202%20-%20Risk%20Assessment%20and%20Control%200Guideline%20for%20RAM%20\(2\).pdf](http://rms.unlv.edu/radiological/Form%202%20-%20Risk%20Assessment%20and%20Control%200Guideline%20for%20RAM%20(2).pdf)

ATTACHMENT 2  
UNLV RISK ASSESSMENT and CONTROL GUIDELINE for  
UNSEALED-RADIOACTIVE MATERIALS

Rad Safety Level	Risk Level	Activity per Experiment * (all apply)	Control Measures	Bioassay Requirement and Periodicity	Air Monitoring
1	<b>MINIMAL RISK:</b> Unlikely to produce a dose to a Worker greater than 100 mrem.  (1 ALI intake = 5000 mrem, 0.01 ALI intake = 50 mrem)	≤ .01 ALI-Ingestion  Max. = 50 µCi	<ul style="list-style-type: none"> <li>• General supervision by the Authorized User</li> <li>• Instruction to Workers on rad risks and proper handling procedures</li> <li>• In procedures and post use survey by Worker</li> <li>• Monthly inspection and quarterly survey by Radiation Safety Office</li> </ul>	None	None
2	<b>LOW RISK:</b> Possible to receive an annual dose in excess of 5 rem. Mitigated by the Worker: <ul style="list-style-type: none"> <li>• understanding, and applying good health physics work practices and procedures</li> <li>• use of engineering and contamination control measures</li> </ul> (1 ALI intake = 5000 mrem)	<u>Non Airborne</u> >.01 to ≤ 1.0 ALI-Ingestion  <u>Airborne</u> ≤ .01 ALI-Limiting  <u>All</u> Max. = 5 mCi	<ul style="list-style-type: none"> <li>• Instruction to Worker on rad risks and proper handling procedures</li> <li>• Review, understand and apply research protocol</li> <li>• Lab specific training by Authorized User followed by routine supervision</li> <li>• In-procedure monitoring and post use surveys by Worker</li> <li>• Monthly inspection and quarterly survey by Radiation Safety Office</li> </ul>	None	None
				None	None

ATTACHMENT 2  
UNLV RISK ASSESSMENT and CONTROL GUIDELINE for  
UNSEALED-RADIOACTIVE MATERIALS (cont.)

Rad Safety Level	Risk Level	Activity per Experiment * (all apply)	Control Measures	Bioassay Requirement and Periodicity	Air Monitoring
3	<p><b>MODERATE RISK:</b> Likely to receive an annual dose in excess of 5 rem. Mitigated by:</p> <ul style="list-style-type: none"> <li>the Worker has thorough knowledge of radiation safety principles and practices, plus task specific training</li> <li>use of engineering and contamination control measures</li> <li>consistent use of task specific control measures</li> <li>demonstrating ability to effectively control radiation hazards</li> </ul>	<p><u>Non-Airborne</u> &gt; 1.0 to ≤50 ALI-Ingestion</p> <p><u>Airborne</u> &gt; .01 to ≤50 ALI-Limiting</p> <p><u>All</u> Max. = 50 mCi</p>	<ul style="list-style-type: none"> <li>Protocol approval by Authorized User and RSO</li> <li>Lab specific training of Worker by Authorized User followed by routine supervision</li> <li>In-procedure monitoring and post use surveys by Worker</li> <li>Monthly inspection and survey by Radiation Safety Office</li> </ul>		
			<p><u>Non-airborne</u> &gt; 10 ALI (ingestion) requires fume hood</p> <p><u>Airborne</u></p> <ul style="list-style-type: none"> <li>≥ 0.01 ALI (limiting), requires fume hood</li> <li>≥ 10 ALI (limiting), requires negative pressure glove box</li> </ul>	<p><b>Baseline bioassay and quarterly bioassay required &gt;5 ALI (limiting) dispersible-airborne</b></p>	<p><b>Routine air monitoring required if &gt; 0.01 ALI (inhalation) of dry, dispersible material.</b></p> <p><b>Continuous air monitoring required if &gt;0.1 ALI (inhalation) of dry, dispersible material.</b></p> <p><b>Breathing Zone Air-sampling (BZA) is required when working with ≥ 1 ALI (inhalation) dry, dispersible materials (airborne).</b></p>

Rad Safety Level	Risk Level	Activity per Experiment * (all apply)	Control Measures	Bioassay Requirement and Periodicity	Air Monitoring
4	<p><b>HIGH RISK:</b> Very likely to receive an annual dose in excess of 5 rem. Mitigated by:</p> <ul style="list-style-type: none"> <li>the Worker has advanced knowledge in radiation safety principles and practices, plus task specific training and procedures</li> <li>consistently using task specific control measures</li> <li>demonstrating the ability to effectively control radiation hazards</li> </ul>	<p><u>Non-Airborne</u>  <b>&gt;50 to ≤ 1,000</b>  ALI-Ingestion</p> <p><u>Airborne</u>  <b>&gt; 50 to ≤1,000</b> ALI-Limiting</p> <p><u>All</u>  Max. = 1000 mCi</p>	<ul style="list-style-type: none"> <li>Protocol approval by Authorized User and RSO</li> <li>Authorized User MUST be present in lab</li> <li>Initial applied training of Worker by Authorized User followed by routine supervision</li> <li>In-procedure monitoring and post use surveys by Worker</li> <li>Weekly survey by Authorized User/Staff</li> <li>Monthly inspection and survey by Radiation Safety</li> </ul>		
			<p><u>Non-Airborne</u>  ≥100 ALI-(limiting), requires negative pressure glove box</p> <p><u>Airborne</u>  ≥ 10 ALI (limiting), requires negative pressure glove box - 1,000 ALI (limiting) maximum</p>	<p><b>Baseline bioassay and quarterly bioassay required.</b></p> <p>Work activity review by the Radiation Safety Office may increase bioassay frequency.</p>	<p><b>Continuous air monitoring required</b></p> <p><b>Breathing Zone Air-sampling (BZA) required</b></p>

**ATTACHMENT 1**  
**EVALUATION of AIRBORNE RADIOACTIVE MATERIALS (cont.)**

Nuclide	Limiting Values - Radiological Health*				Rad Level 1**	Rad Level 2	Rad Level 3		Rad Level 4	
	ALI Ingestion (μCi)	ALI Inhalation (μCi)	Ratio Ingestion /Inhalation	Limiting ALI (μCi)	Less Than (μCi)	Not Airborne & Less Than (μCi)	If NOT Airborne Less Than (μCi)	If Airborne Less Than (μCi)	If NOT Airborne Less Than (μCi)	If Airborne Less Than (μCi)
Am-241	0.8	0.006	133	0.01	0.000060	0.80	40	0.30	800	6.00
Am-242m	0.8	0.006	133	0.01	0.000060	0.80	40	0.30	800	6.00
Am-243	0.8	0.006	133	0.01	0.000060	0.80	40	0.30	800	6.00
Ba-133	2,000	700	2.86	700	7.00	2,000	50,000	35,000	1,000,000	700,000
C-14	2,000	2,000	1.00	2,000	20	2,000	50,000	50,000	1,000,000	1,000,000
Cd-109	300	40	7.50	40	0.400	300	50,000	2,000	300,000	40,000
Cl-36	2,000	2,000	1.00	2,000	20	2,000	50,000	50,000	1,000,000	1,000,000
Cm-244	1.0	0.010	100	0.01	0.00010	1.00	50	0.50	1,000	10
Cm-248	0.2	0.002	100	0.002	0.000020	0.20	10	0.10	200	2.00
Co-57	4,000	700	5.71	700	7.00	4,000	50,000	35,000	1,000,000	700,000
Co-60	200	30	6.67	30	0.300	200	50,000	1,500	200,000	30,000
Cs-137	100	200	0.50	100	1.00	100	50,000	5,000	100,000	100,000
Eu-152	800	20	40	20	0.200	800	50,000	1,000	800,000	20,000
Eu-154	500	20	25	20	0.200	500	50,000	1,000	500,000	20,000
Eu-155	4,000	90	44	90	0.900	4,000	50,000	4,500	1,000,000	90,000
Gd-148	10	0.008	1,250	0.01	0.000080	10	500	0.40	10,000	8.00
H-3	80,000	80,000	1.00	80,000	50	5,000	50,000	50,000	1,000,000	1,000,000
Hf-175	3,000	900	3.33	900	9.00	3,000	50,000	45,000	1,000,000	900,000
I-125	40	60	0.67	40	0.400	40	2,000	2,000	40,000	40,000
I-131	30	50	0.60	30	0.300	30	1,500	1,500	30,000	30,000
Mn-54	2,000	800	2.50	800	8.00	2,000	50,000	40,000	1,000,000	800,000

**ATTACHMENT 1**  
**EVALUATION of AIRBORNE RADIOACTIVE MATERIALS (cont.)**

Nuclide	Limiting Values - Radiological Health*				Rad Level 1**	Rad Level 2	Rad Level 3		Rad Level 4	
	ALI Ingestion (μCi)	ALI Inhalation (μCi)	Ratio Ingestion /Inhalation	Limiting ALI (μCi)	Less Than (μCi)	Not Airborne & Less Than (μCi)	If NOT Airborne Less Than (μCi)	If Airborne Less Than (μCi)	If NOT Airborne Less Than (μCi)	If Airborne Less Than (μCi)
Na-22	400	600	0.67	400	4.00	400	50,000	20,000	400,000	400,000
Np-237	0.5	0.004	125	0.004	0.000040	0.50	25	0.20	500	4.00
P-32	600	400	1.50	400	4.00	600	50,000	20,000	600,000	400,000
P-33	6,000	3,000	2	3,000	50	5,000	50,000	50,000	1,000,000	1,000,000
Pb-210	1.0	20	0.05	1.00	0.010	1.00	50	50	1,000	1,000
Po-210	3.0	0.60	5.00	0.60	0.0060	3.00	150	30	3,000	600
Pu-236	2.0	0.020	100	0.02	0.00020	2.00	100	1.00	2,000	20
Pu-238	0.9	0.007	129	0.01	0.000070	0.90	45	0.35	900	7.00
Pu-239	0.8	0.006	133	0.01	0.000060	0.80	40	0.30	800	6.00
Pu-240	0.8	0.006	133	0.01	0.000060	0.80	40	0.30	800	6.00
Pu-241	40	0.30	133	0.30	0.0030	40	2,000	15	40,000	300
Pu-242	0.8	0.007	114	0.01	0.000070	0.80	40	0.35	800	7.00
Ra-226	2.0	0.60	3.33	0.60	0.0060	2.00	100	30	2,000	600
Sb-125	2,000	500	4.00	500	5.00	2,000	50,000	25,000	1,000,000	500,000
Sm-147	20	0.070	286	0.07	0.00070	20	1,000	3.50	20,000	70
Sr-85	3,000	2,000	1.50	2,000	20	3,000	50,000	50,000	1,000,000	1,000,000
Sr-90	30	4.00	7.50	4.00	0.040	30	1,500	200	30,000	4,000
Tc-99	4,000	700	5.71	700	7.00	4,000	50,000	35,000	1,000,000	700,000
Tc-99m	80,000	200,000	0.40	80,000	50	5,000	50,000	50,000	1,000,000	1,000,000
Th-229	0.6	0.001	667	0.001	0.000009	0.60	30	0.05	600	0.90
Th-230	4.0	0.006	667	0.01	0.000060	4.00	200	0.30	4,000	6.00
Th-232	0.7	0.001	700	0.001	0.000010	0.70	35	0.05	700	1.00
Tl-204	2,000	2,000	1.00	2,000	20	2,000	50,000	50,000	1,000,000	1,000,000
U-232	2.0	0.008	250	0.01	0.000080	2.00	100	0.40	2,000	8.00
U-233	10	0.040	250	0.04	0.00040	10	500	2.00	10,000	40
U-235	10	0.040	250	0.04	0.00040	10	500	2.00	10,000	40
U-238	10	0.040	250	0.04	0.00040	10	500	2.00	10,000	40
Zn-65	400	300	1.33	300	3.00	400	50,000	15,000	400,000	300,000
Zr-95	1,000	100	10	100	1.00	1,000	50,000	5,000	1,000,000	100,000

# Hot Atom Chemistry

- Also called Szilard-Chalmers process
  - Activity of I extracted from water and ethyl iodide
    - Precipitated at AgI
- Chemical reactions produced by nuclear transformation
  - Neutron irradiation of ethyl iodide
    - Iodine extracted into aqueous phase
      - \*  $^{127}\text{I}(n,\gamma)^{128}\text{I}$
      - ✂ Possible to produce specific isotope
- Need to overcome bond energy
  - Neutron does not normally contain sufficient energy
  - Gamma decay can provide suitable energy from recoil
    - M is atom mass, E is gamma energy in MeV
    - \* Nucleus excited 6-8 MeV

**Table 11-3 Recoil Energies in Electron Volts Imparted to Nuclei by Gamma Rays of Various Energies**

M	$E_\gamma = 2 \text{ MeV}$	$E_\gamma = 4 \text{ MeV}$	$E_\gamma = 6 \text{ MeV}$
20	107	430	967
50	43	172	387
100	21	86	193
150	14	57	129
200	11	43	97

$$R(eV) = \frac{537 E_\gamma^2}{M}$$

# Hot Atom Chemistry

- **Bonds are broken due to reaction energy**
  - **Bond energies on the order of eV**
- **Conditions needed**
  - **Bond of produced atom must be broken**
  - **Should not recombine with fragments**
  - **Should not exchange with target molecule**
    - **Slow kinetics**
  - **Separation of new species**
- **Halogens produced in this method**
  - **CCl<sub>4</sub>**
  - **C<sub>2</sub>H<sub>2</sub>Cl<sub>2</sub>**
  - **C<sub>2</sub>H<sub>5</sub>Br**
  - **C<sub>2</sub>H<sub>2</sub>Br<sub>2</sub>**
  - **C<sub>6</sub>H<sub>5</sub>Br**
  - **CH<sub>3</sub>I**
    - **Used to produce <sup>38</sup>Cl, <sup>80</sup>Br, <sup>82</sup>Br, <sup>128</sup>I**

# Hot Atom Chemistry: Chemical Reactions

- Beta reactions can also be exploited
  - $\text{TeO}_3^{2-} \rightarrow \text{IO}_3^- + e^-$ 
    - Recoil is not quantized
      - \* Kinetic energy shared
      - \* E is maximum beta energy (MeV)

$$R_{\max} (eV) = \frac{537 E_{\beta} (E_{\beta} + 1.02)}{M}$$

→ 0.5 MeV in 100 amu is about 4 eV

- Energy is distributed
  - Translational, rotational, vibrational
- Bond usually not broken with only 4 eV
- Internal conversion set atom in excited state
  - Rearrangement of electrons and drive chemical reactions
  - Separation of isomers

**Table 11-4 Approximate Recoil Energies Expected with Various Nuclear Events (from reference C5)**

Nuclear Process	Range of Recoil Energy (eV) <sup>a</sup>
$\beta^-$ Decay	$10^{-1}$ – $10^2$
$\beta^+$ Decay	$10^{-1}$ – $10^2$
$\alpha$ Decay	$\sim 10^5$
IT	$10^{-1}$ –1
EC	$10^{-1}$ – $10^1$
$n_{th}, \gamma$	$\sim 10^2$
$n, p$	$\sim 10^5$
Fission	$\sim 10^8$

<sup>a</sup> Based on a hot-atom mass of  $\sim 100$ , the most probable kinetic energy for a given nuclear process, and a range of nuclear energies most frequently encountered.

# Radiochemistry in reactors: Chemistry coupled with nuclear reactions

- Fuel confined in reactor to fuel region
  - Potential for interaction with cladding material
    - Initiate stress corrosion cracking
  - Chemical knowledge useful in events where fuel is outside of cladding
- Some radionuclides generated in structural material
- Recoil length about 10 microns, diameter of 6 nm
  - About size of  $\text{UO}_2$  crystal
  - 95 % of energy into stopping power
    - Remainder into lattice defects
      - \* Radiation induced creep
  - High local temperature from fission
    - 3300 K in 10 nm diameter

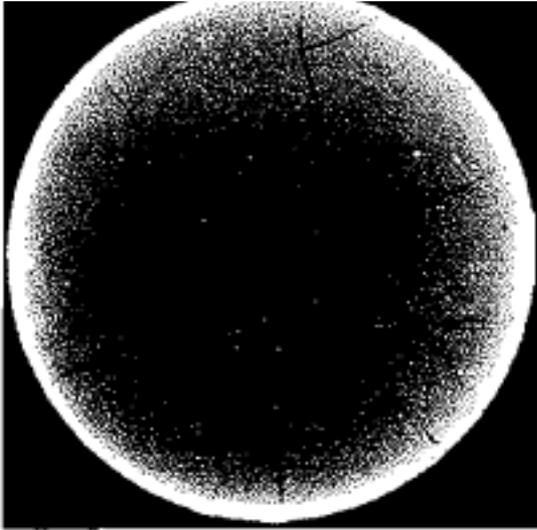
# Fuel variation during irradiation

- **Chemical composition**
- **Radionuclide inventory**
- **Pellet structure**
- **Higher concentrations of Ru, Rh, and Pd in Pu fuel**
- **Total activity of fuel effected by saturation**
  - **Tends to reach maximum**
- **Radionuclide fuel distribution studied**
  - **Fission gas release**
  - **Axial distribution by gamma scanning**
- **Radial distribution to evaluate flux**
- **Pu fuel has higher concentrations of:**
  - **Ru, Rh, Pd**

Table 3.3. Fission product element concentrations (g/kg HM) in irradiated LWR uranium fuel (initial enrichment 4.0% <sup>235</sup>U)  
(By courtesy of Siemens/KWU)

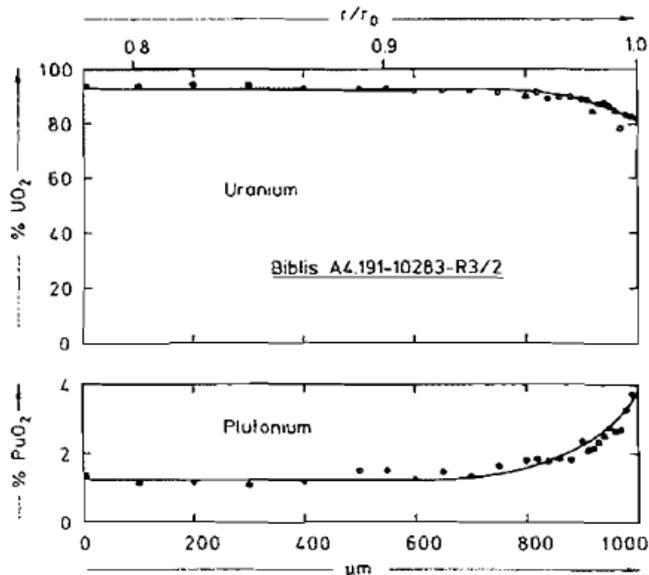
Element	Fuel burnup (MWd/kg HM)				
	13.0	26.0	39.0	52.0	65.0
Bromine	0.0093	0.018	0.026	0.034	0.041
Krypton	0.16	0.31	0.43	0.54	0.64
Rubidium	0.16	0.29	0.41	0.51	0.60
Strontium	0.47	0.82	1.11	1.36	1.57
Yttrium	0.24	0.42	0.58	0.71	0.82
Zirconium	1.56	2.97	4.27	5.48	6.62
Niobium	0.045	0.044	0.042	0.040	0.038
Molybdenum	1.23	2.57	3.89	5.18	6.46
Technetium	0.33	0.64	0.91	1.14	1.33
Ruthenium	0.84	1.76	2.76	3.85	5.00
Rhodium	0.17	0.35	0.50	0.60	0.66
Palladium	0.23	0.68	1.34	2.18	3.18
Silver	0.015	0.042	0.073	0.11	0.14
Cadmium	0.011	0.037	0.080	0.15	0.23
Indium	0.0007	0.0013	0.0016	0.0017	0.0018
Tin	0.014	0.032	0.054	0.079	0.11
Antimony	0.0058	0.013	0.020	0.027	0.034
Tellurium	0.16	0.34	0.53	0.74	0.96
Iodine	0.080	0.17	0.27	0.37	0.47
Xenon	2.02	4.07	6.16	8.28	10.4
Cesium	1.14	2.27	3.34	4.36	5.32
Barium	0.56	1.10	1.66	2.26	2.89
Lanthanum	0.51	0.99	1.45	1.90	2.32
Cerium	1.30	2.34	3.28	4.19	5.07
Praseodymium	0.43	0.87	1.30	1.71	2.11
Neodymium	1.38	2.89	4.42	5.93	7.41
Promethium	0.13	0.18	0.19	0.19	0.17
Samarium	0.23	0.51	0.81	1.10	1.36
Europium	0.036	0.10	0.19	0.27	0.34
Gadolinium	0.0094	0.037	0.10	0.22	0.40
Totals	13.5	26.9	40.3	53.6	66.8

# Distribution in Fuel

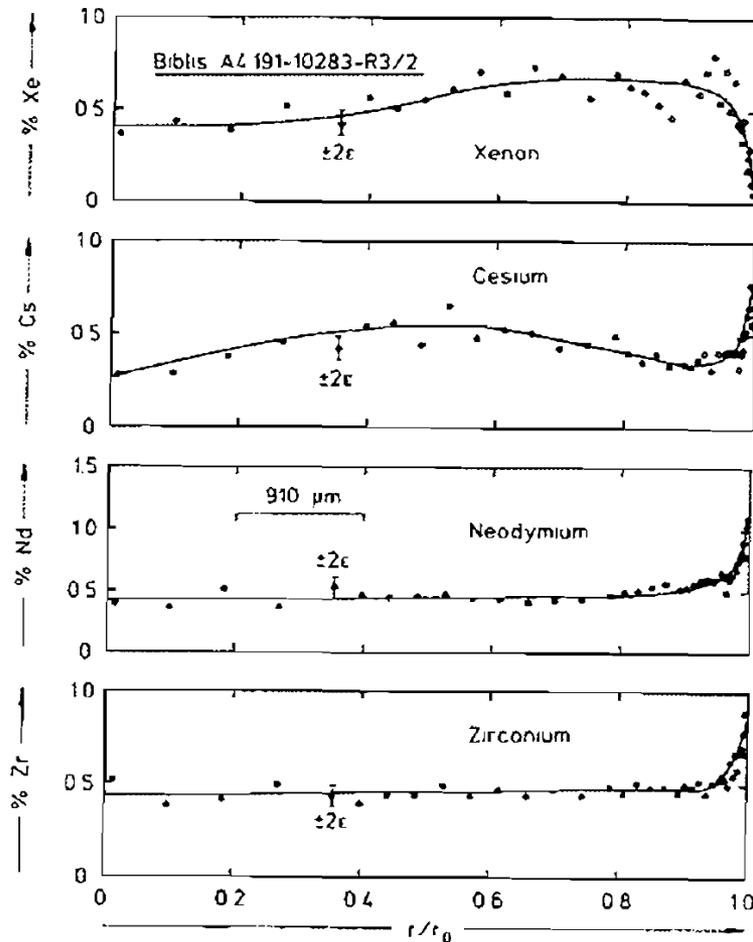


Transuranics on fuel rim

- Radial distribution of fission products mainly governed by thermal neutron flux profile .
- Higher Pu concentration in outer zone of fuel
  - Epithermal neutron capture on  $^{238}\text{U}$
  - Small influence of thermal migration on Cs
    - Gaseous and volatile fission products
    - Influence by fuel initial composition (O to M ratio)
  - Xe trapped in region with high gas bubble density



# Distribution in Fuel



- **Increased Pu concentration on rim leads to increased fission product density**
  - **Xe behavior influenced by bubble gas location**
- **Consumption of burnable poison**
  - **Gd isotopes 157 and 155 depleted in outer zone**

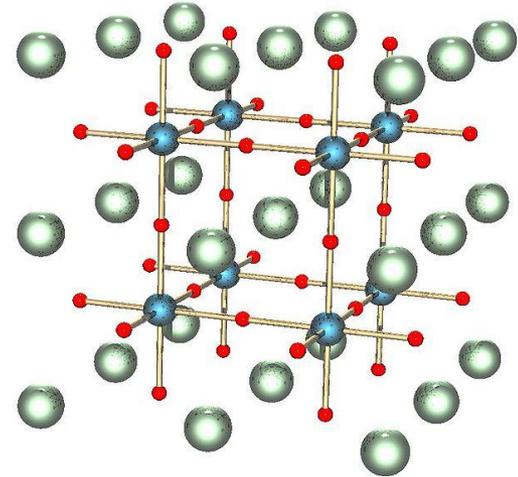
Figure 3.9. Fission product distribution as a function of the relative fuel pellet radius in a LWR high-burnup oxide fuel (Kleykamp, 1990 a)

# Distribution in fuel: Thermal behavior

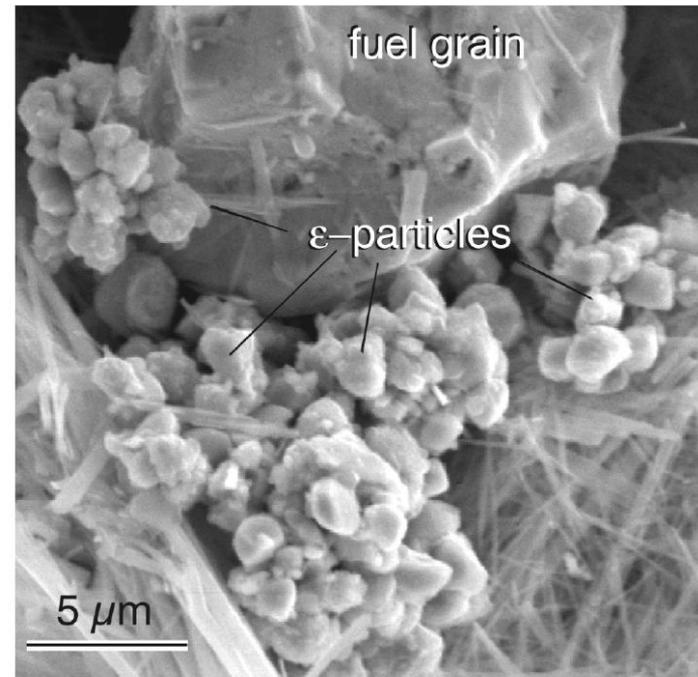
- **Mainly affects the gaseous and the volatile fission products**
  - **linear heat rating**
  - **pellet temperatures during reactor operation**
  - **stoichiometry of the fuel**
- **Halogens and alkali elements**
  - **Cs and I volatility**
    - **High fission yields**
    - **Enhanced mobility**
  - **Can be treated similarly, different chemical behavior**

# Separate phases in fuel

- **Perovskite ( $A^{2+}B^{4+}O_3$ ) identified oxide phase**
  - **U, Pu, Ba, Sr, Cs, Zr, Mo, and Lanthanides**
  - **Mono- and divalent elements at A**
- **Mechanism of formation**
  - **Sr and Zr form phases**
  - **Lanthanides added at high burnup**

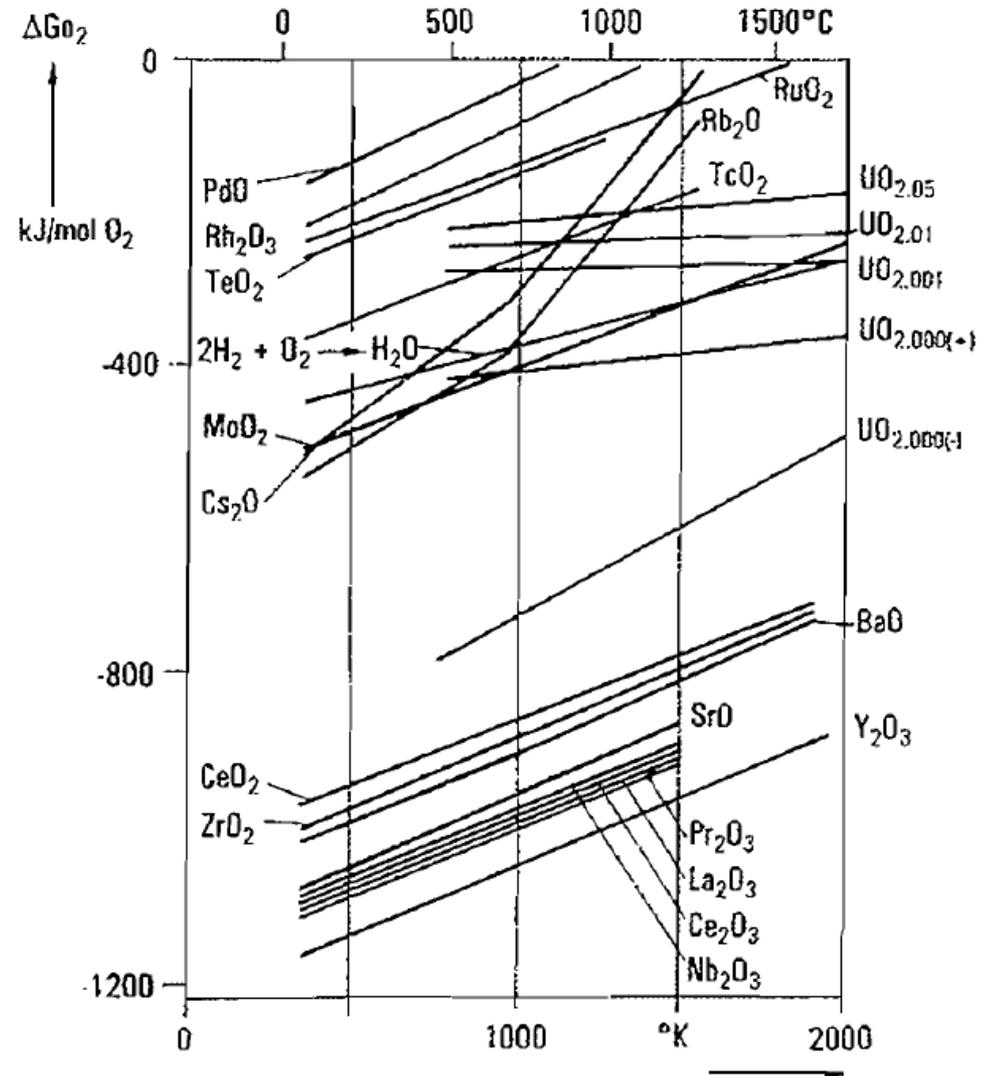


- **Metallic phase of fission products in fuel**
  - **Mo (24-43 wt %)**
  - **Tc (8-16 wt %)**
  - **Ru (27-52 wt %)**
  - **Rh (4-10 wt %)**
  - **Pd (4-10 wt %)**
- **Grain sizes around 1 micron**
- **Concentration nearly linear with fuel burnup**
  - **5 g/kg at 10MWd/kg U**
  - **15 g/kg at 40 MWd/kg U**
- **Catalytic property in used fuel**



# Epsilon Phase

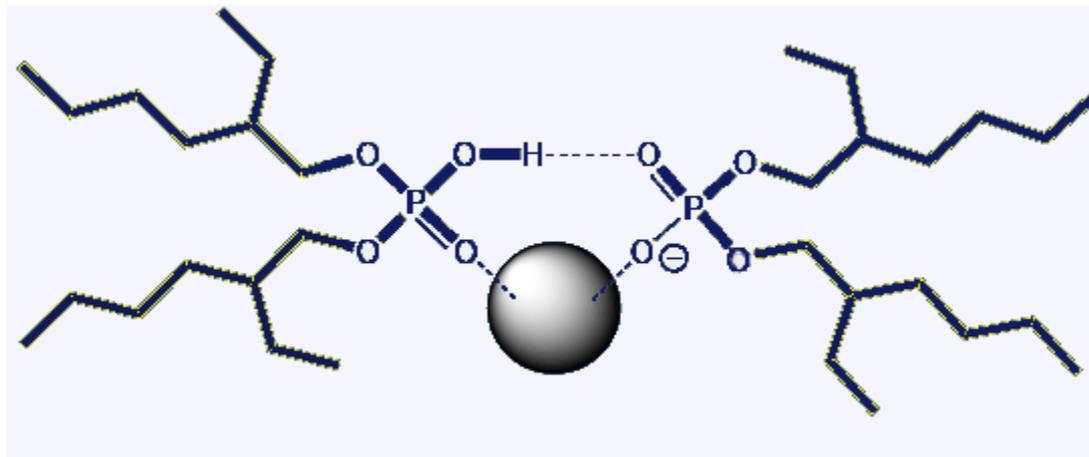
- **Formation of metallic phase promoted by higher linear heat**
  - **high Pd concentrations (20 wt %) indicate a relatively low fuel temperature**
  - **Mo behavior controlled by oxygen potential**
    - High metallic Mo indicates O:M of 2
    - O:M above 2, more Mo in  $\text{UO}_2$  lattice



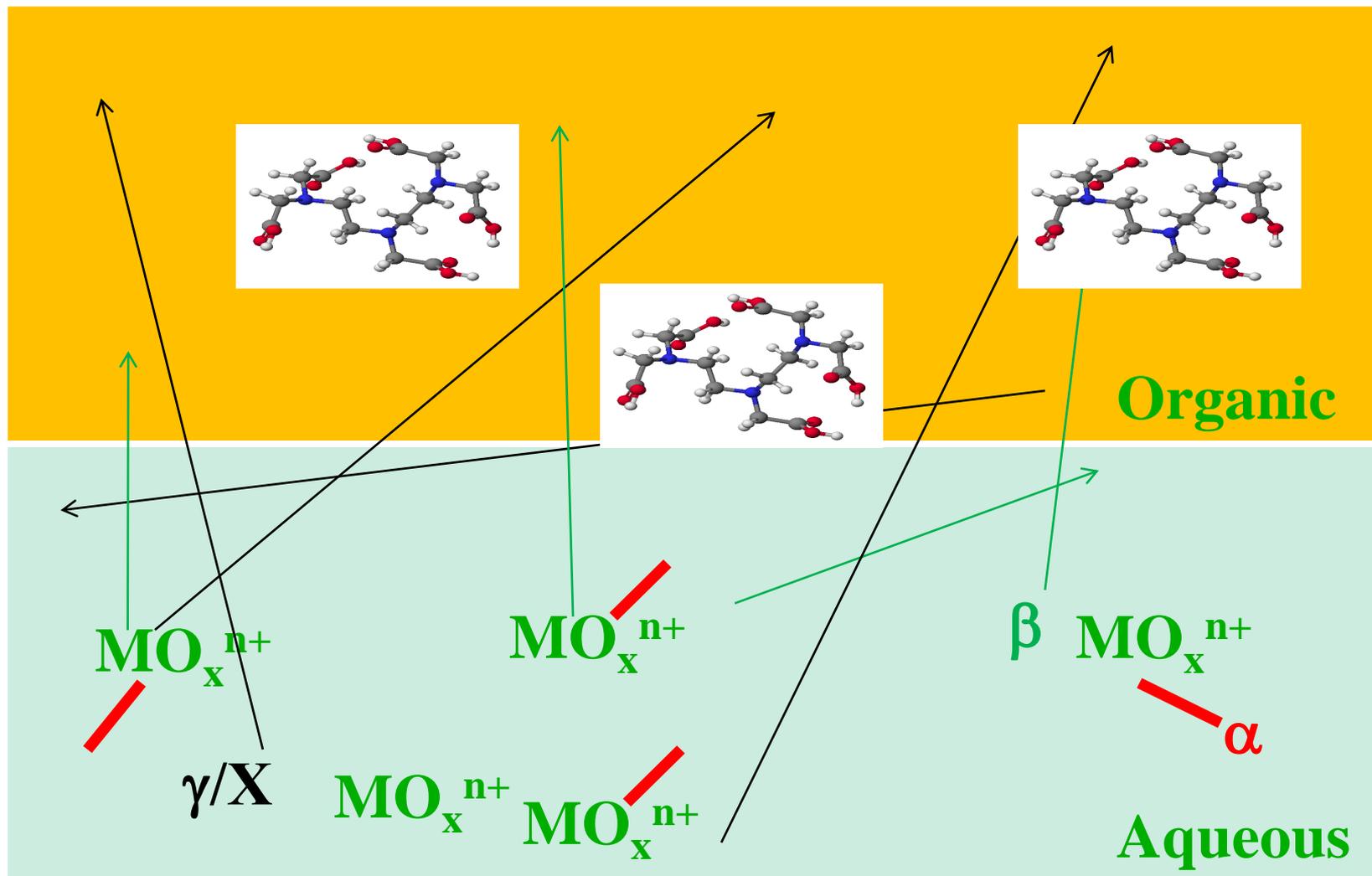
Relative partial molar Gibbs free energy of oxygen of the fission product oxides and  $\text{UO}_2$

# Radiolysis Effect on TALSPEAK

- **Trivalent Actinide Lanthanide Separations by Phosphorus-reagent Extraction from Aqueous Komplexes**
  - Weaver and Kappelman 1968
- Feed is buffered with lactic acid
  - pH must remain between 2-3
- DTPA in aqueous phase to complex with Actinides
- Lanthanides extracted with hard donor HDEHP (di-(2-ethylhexyl) phosphoric acid) in organic phase



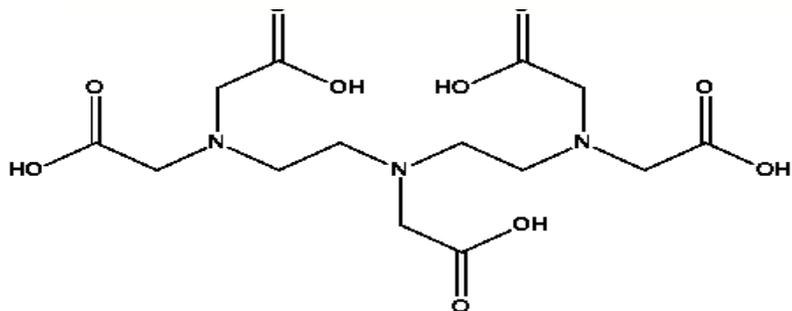
# Radiolysis of Solvents



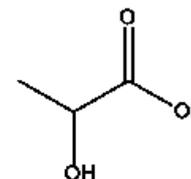
# Effects of Degradation

- **Accumulation of Degradation Products result in:**
  - **Lower Distribution coefficient**
    - locally modify redox conditions
  - **Formation of cruds and emulsions which disturb continuous extraction process**
    - product loss
- **Consider lactic acid and DTPA in TALSPEAK**

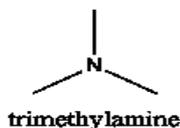
# Radiolysis byproducts



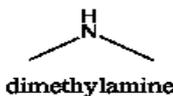
diethylenetriaminepentaacetic acid



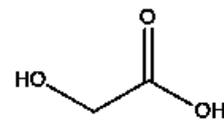
lactic acid



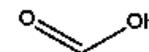
trimethylamine



dimethylamine



2-hydroxyacetic acid



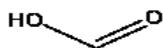
formic acid

Critical Temp: 457.96 [K]  
 Critical Pres: 43.97 [Bar]  
 Critical Vol: 221.5 [cm<sup>3</sup>/mol]  
 Gibbs Energy: 85.16 [kJ/mol]  
 Log P: 0.24

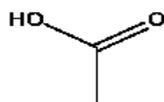
Critical Temp: 454.54 [K]  
 Critical Pres: 51.24 [Bar]  
 Critical Vol: 182.5 [cm<sup>3</sup>/mol]  
 Gibbs Energy: 55.35 [kJ/mol]  
 Log P: -0.13

Critical Temp: 660.57 [K]  
 Critical Pres: 66.21 [Bar]  
 Critical Vol: 190.5 [cm<sup>3</sup>/mol]  
 Gibbs Energy: -514.77 [kJ/mol]  
 Log P: -1.01

Critical Temp: 578.94 [K]  
 Critical Pres: 75.88 [Bar]  
 Critical Vol: 127.5 [cm<sup>3</sup>/mol]  
 Gibbs Energy: -278.8 [kJ/mol]  
 Log P: -0.42



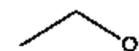
formic acid



acetic acid



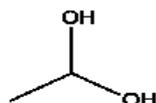
Methane



ethanol



carbon dioxide



ethane-1,1-diol

# $^{60}\text{Co}$ Source for irradiation

- Samples prepared and irradiated by Idaho National Laboratory
  - 0.001, 0.01 and 1M Sodium Lactate and DTPA
  - 0.3 kGy/hr dose rate
  - 0, 5, 10, 100, 200 kGy
- Irradiations also performed CNRS linear  $\text{He}^{2+}$  ion beam facility in Orleans, France



# HPLC Method Development

## System Components:

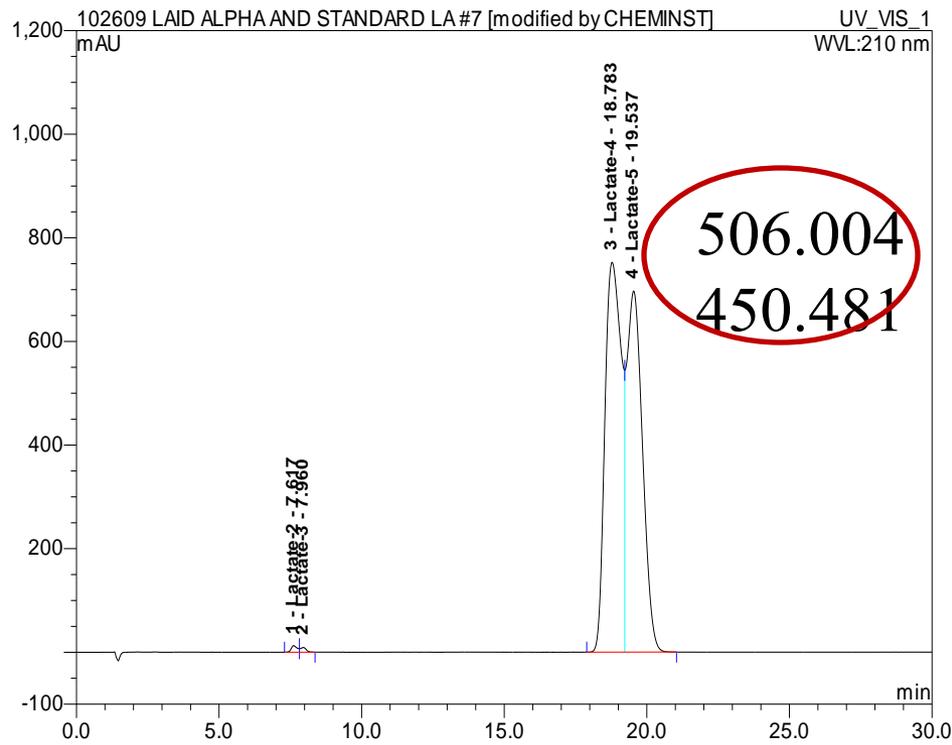
- **Dionex Ultimate 3000 HPLC system**
- **Column :**  
**Dionex Acclaim mixed-mode WAX-1**  
-Silica based packing material that has both hydrophobic and weak anion-exchange properties
- **Eluent :**  
**25 mM Potassium Phosphate buffer at pH 4.**

## Parameters:

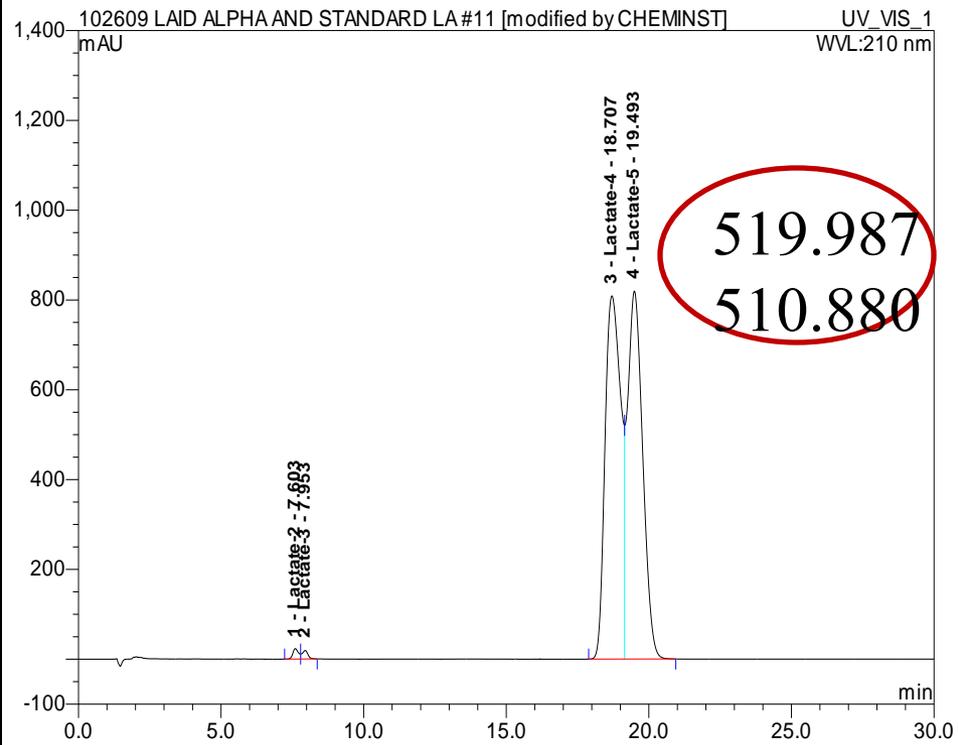
- **Eluent flow rate : 1 mL/min**
- **Injection Volume : 40 micro-liters**
- **Column Oven Temperature : 45 C**
- **UV wavelength : 210 nm**



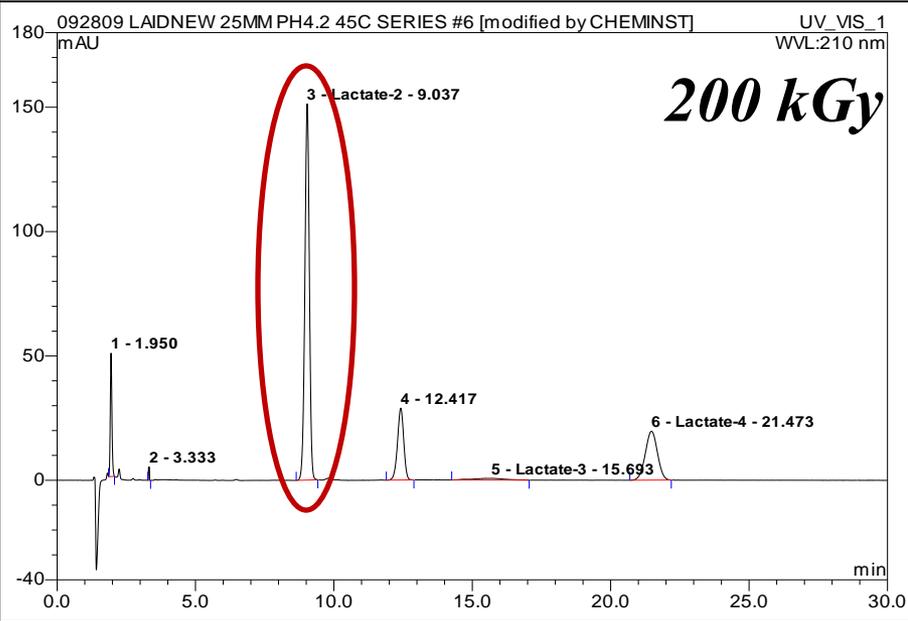
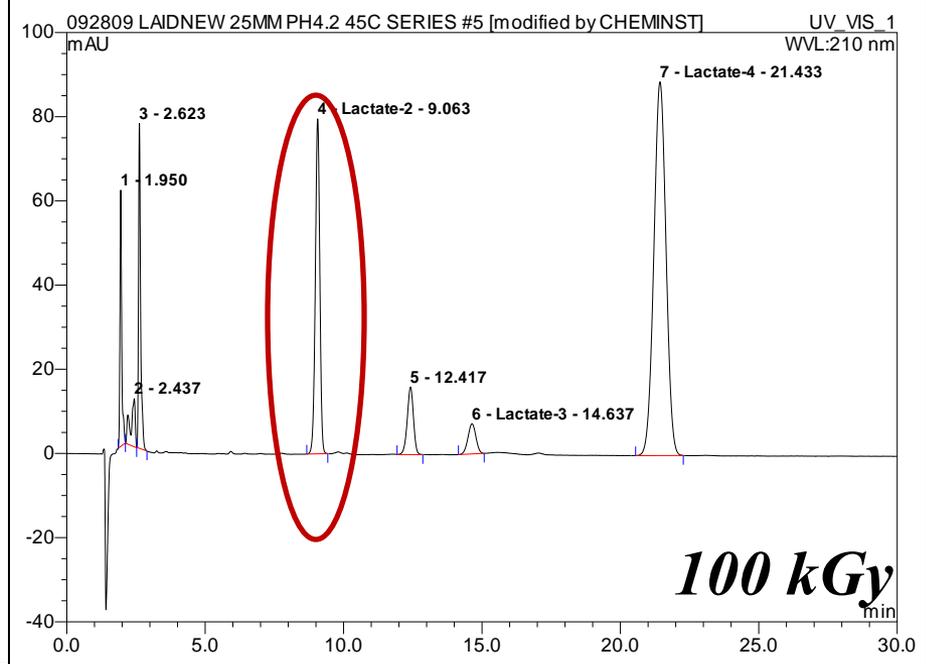
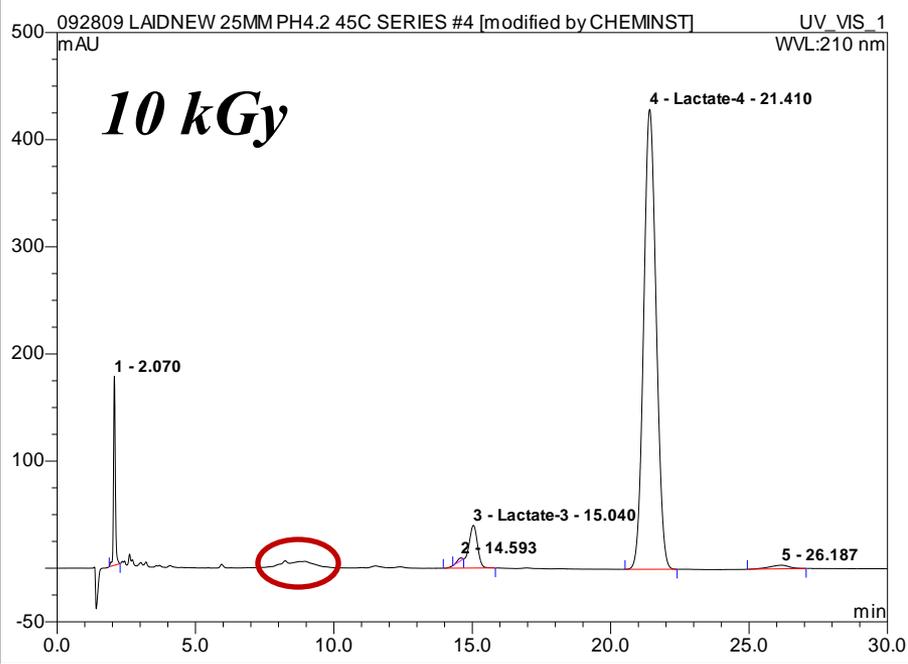
# Lactic Acid $\alpha$ Series



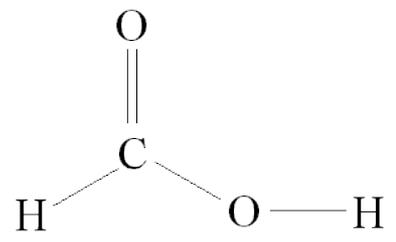
Time 0 minutes



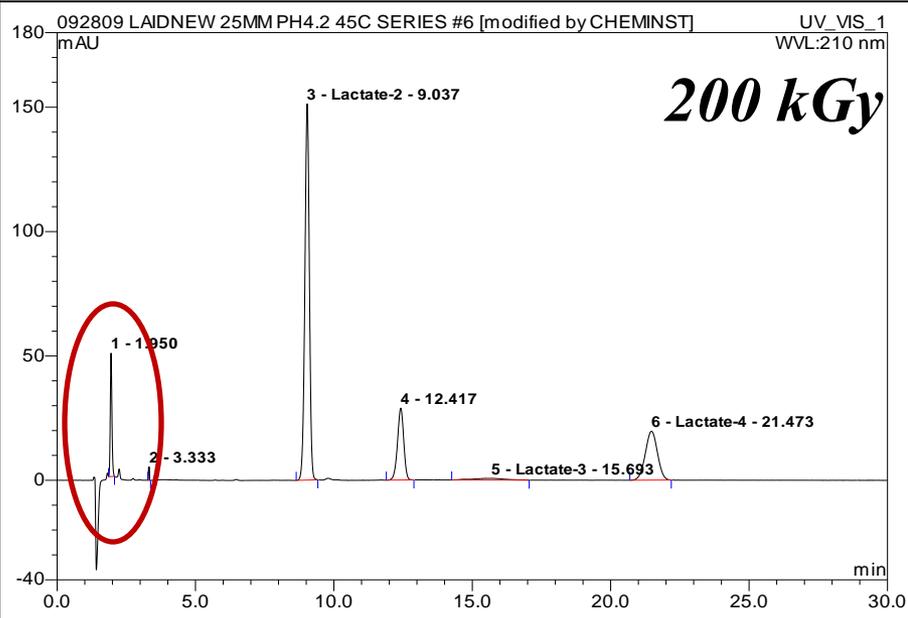
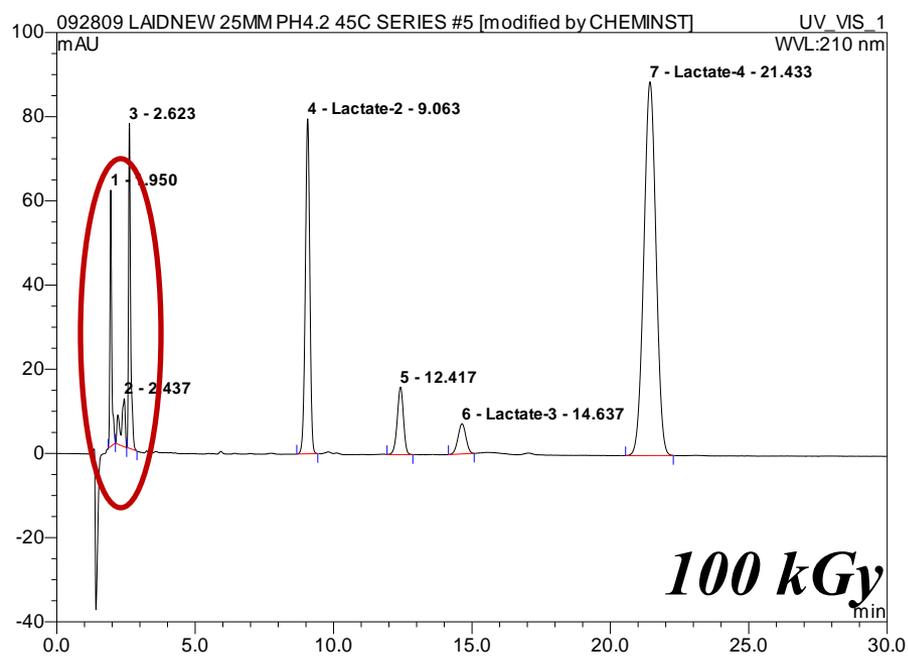
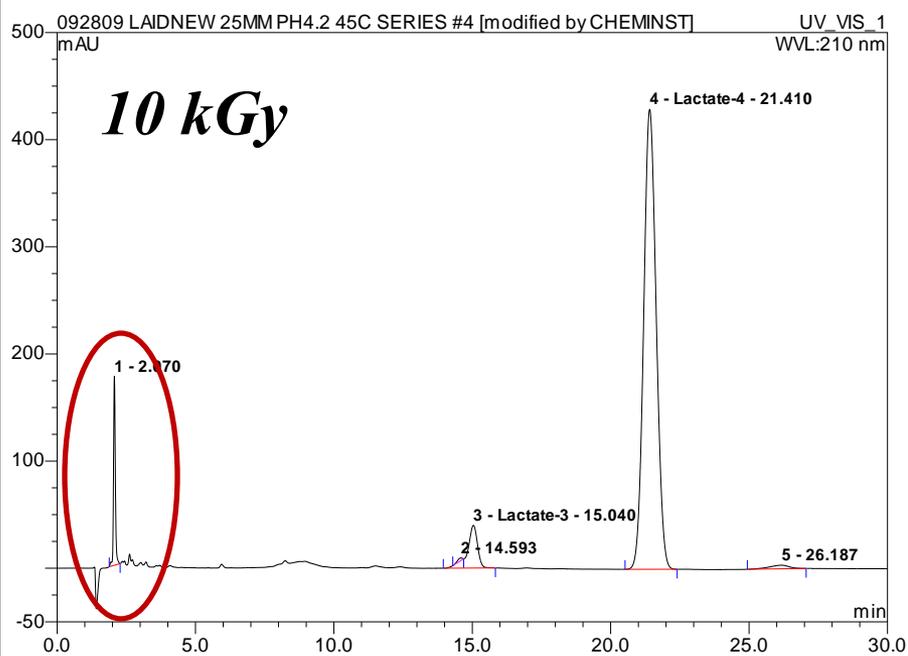
Time 120 minutes



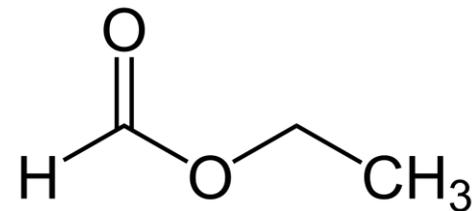
**Peak analysis of  
analyte eluting at 9  
minutes is Formic acid**

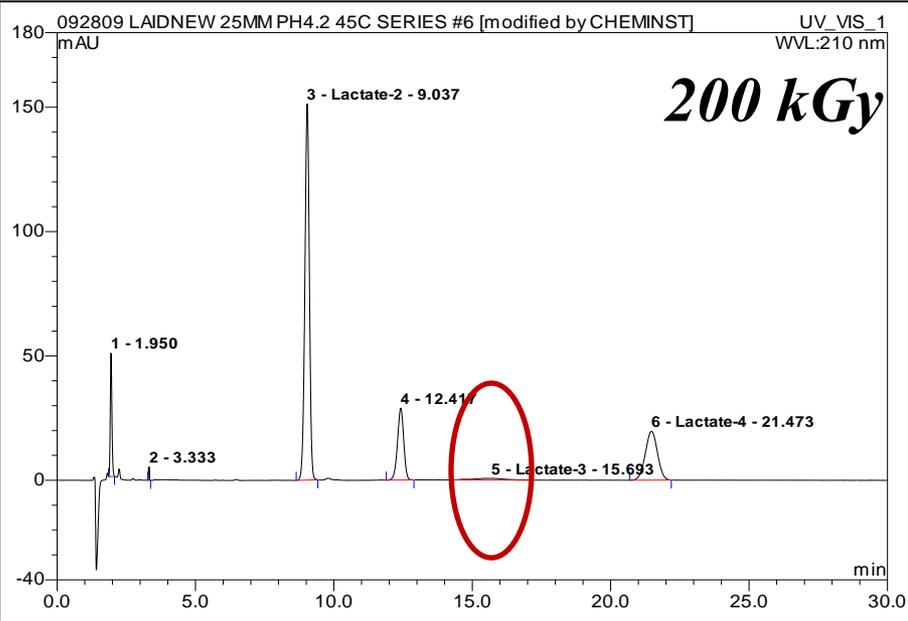
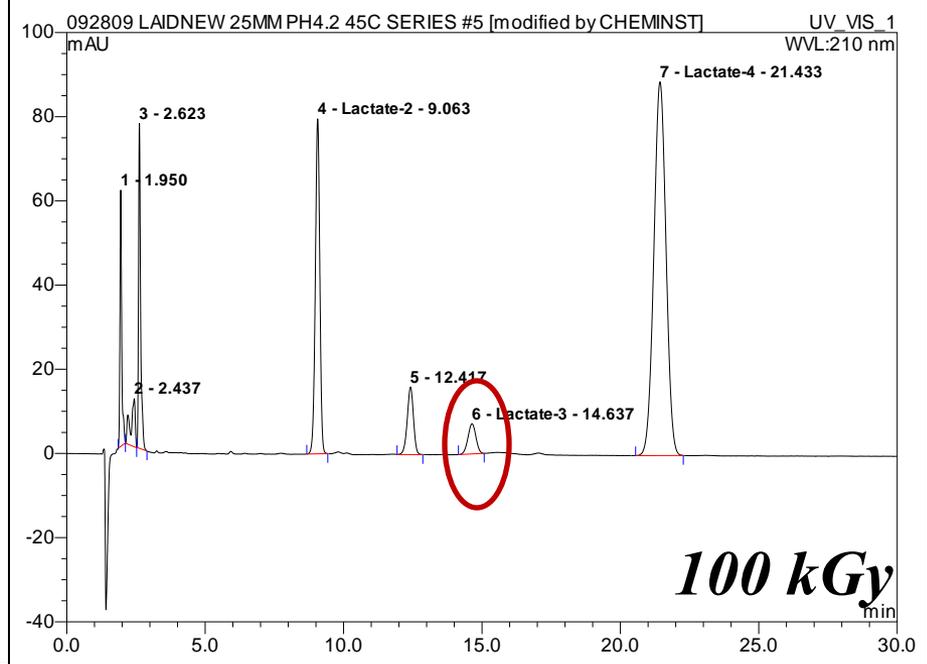
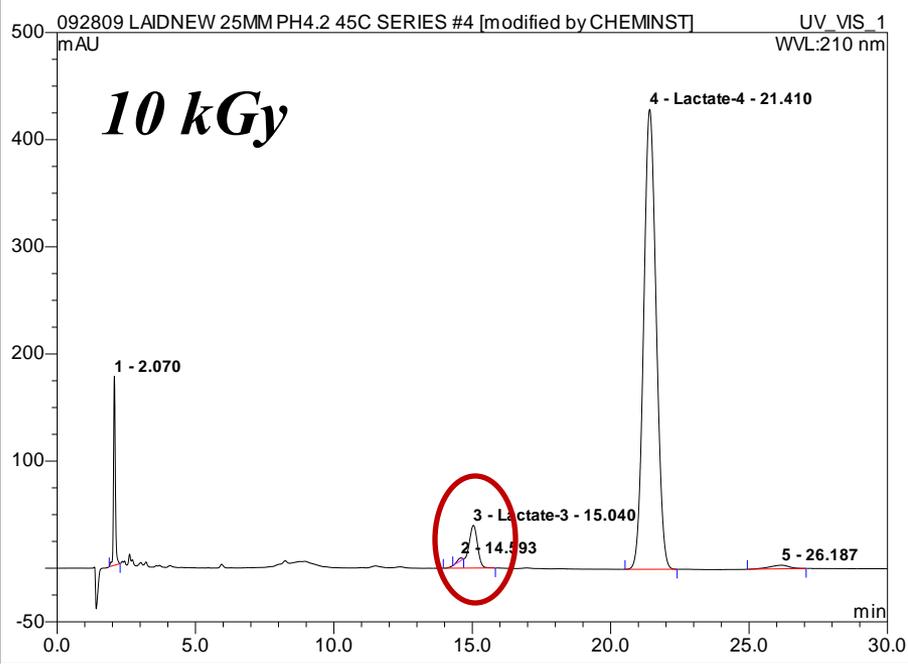


Formic Acid

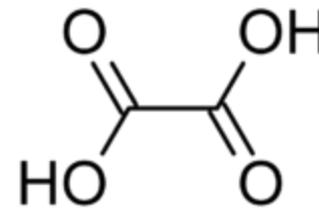


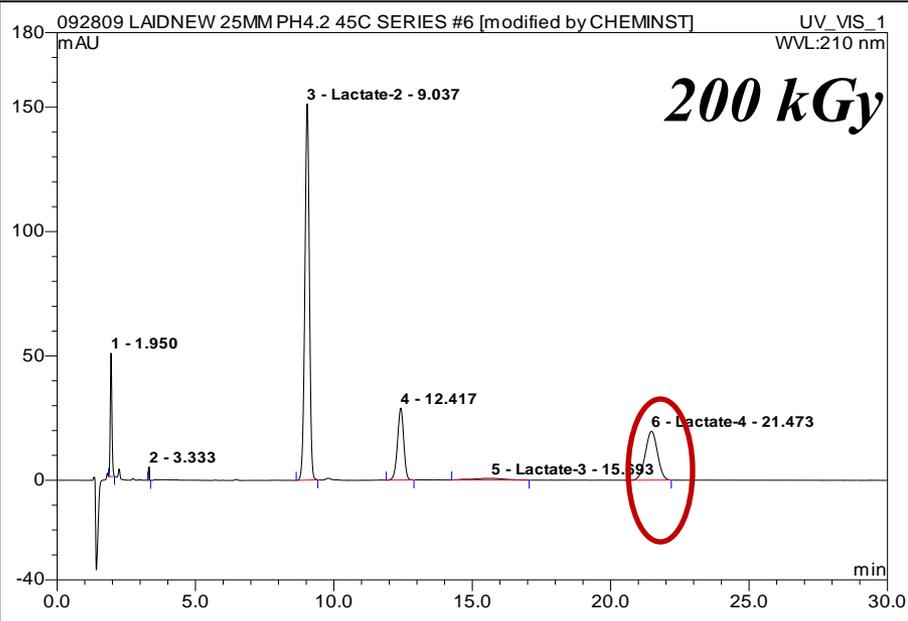
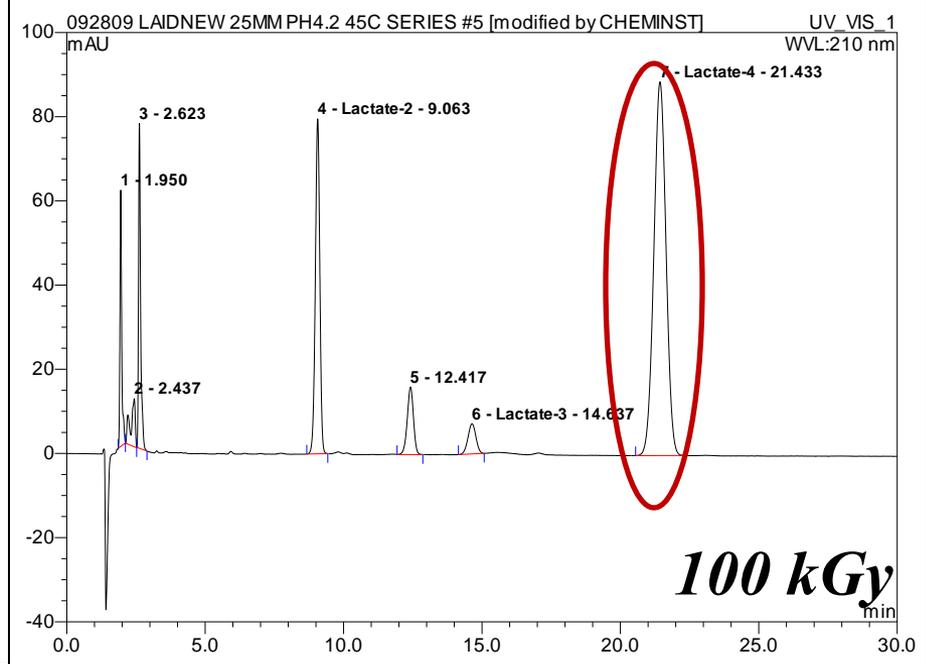
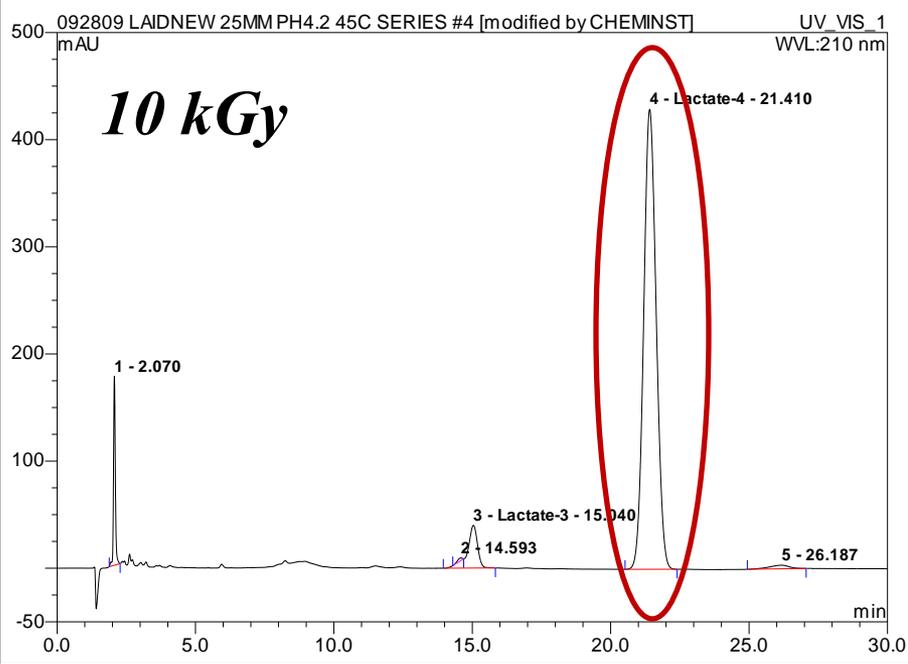
**Peak analysis of analyte eluting at 2 minutes is ethyl formate**





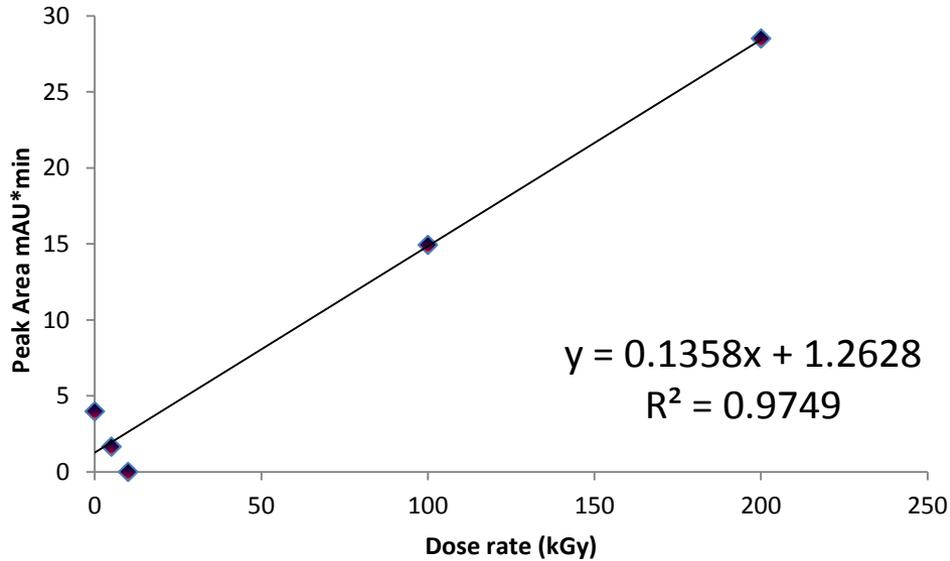
**Peak analysis of analyte  
eluting at 15 minutes is  
Oxalic acid**



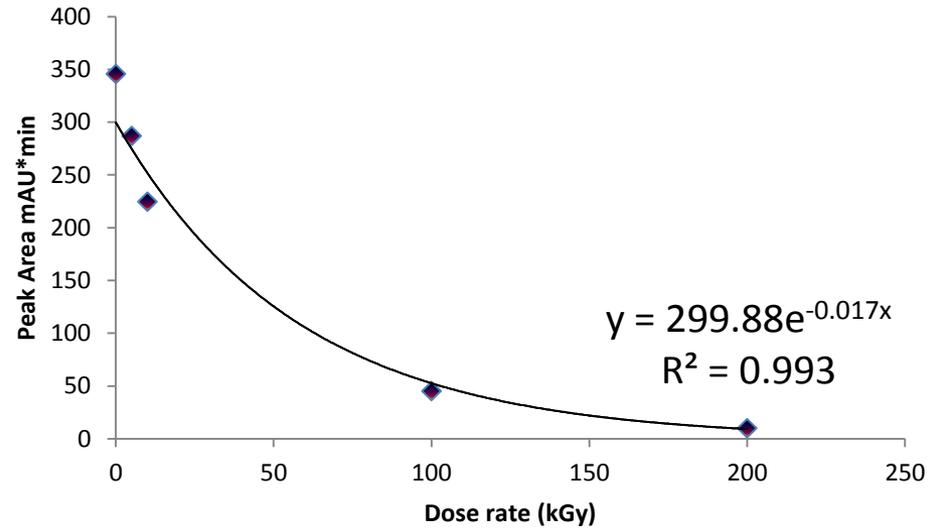


**Degradation of Lactic acid**

# Influence of dose on ligand

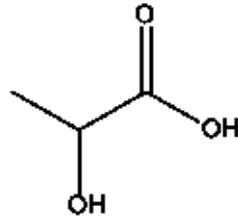


Formic Acid

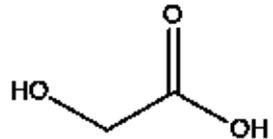


Sodium Lactate  
Decay

# Radiolysis: Potential By-products of lactic acid



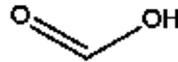
lactic acid



2-hydroxyacetic acid

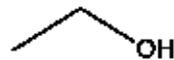
Critical Temp: 660.57 [K]  
Critical Pres: 66.21 [Bar]  
Critical Vol: 190.5 [cm<sup>3</sup>/mol]  
Gibbs Energy: -514.77 [kJ/mol]  
Log P: -1.01

CH<sub>4</sub>  
Methane



formic acid

Critical Temp: 578.94 [K]  
Critical Pres: 75.88 [Bar]  
Critical Vol: 127.5 [cm<sup>3</sup>/mol]  
Gibbs Energy: -278.8 [kJ/mol]  
Log P: -0.42

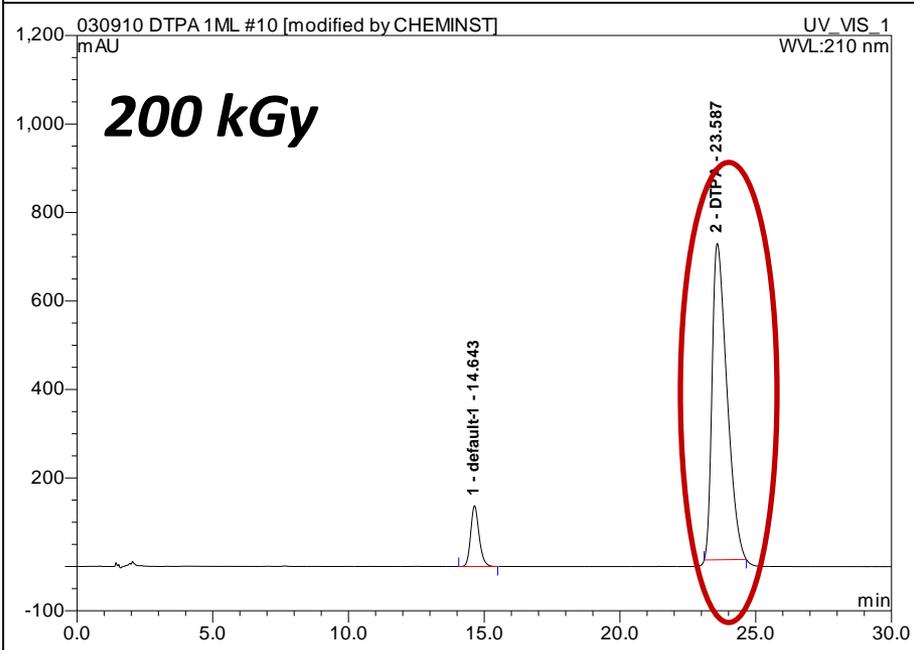
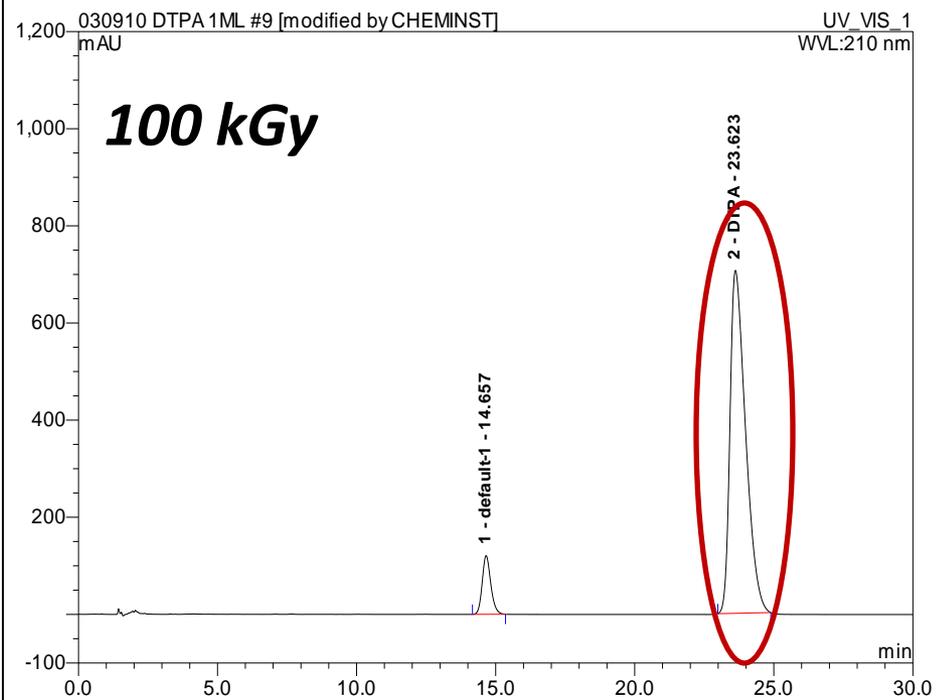
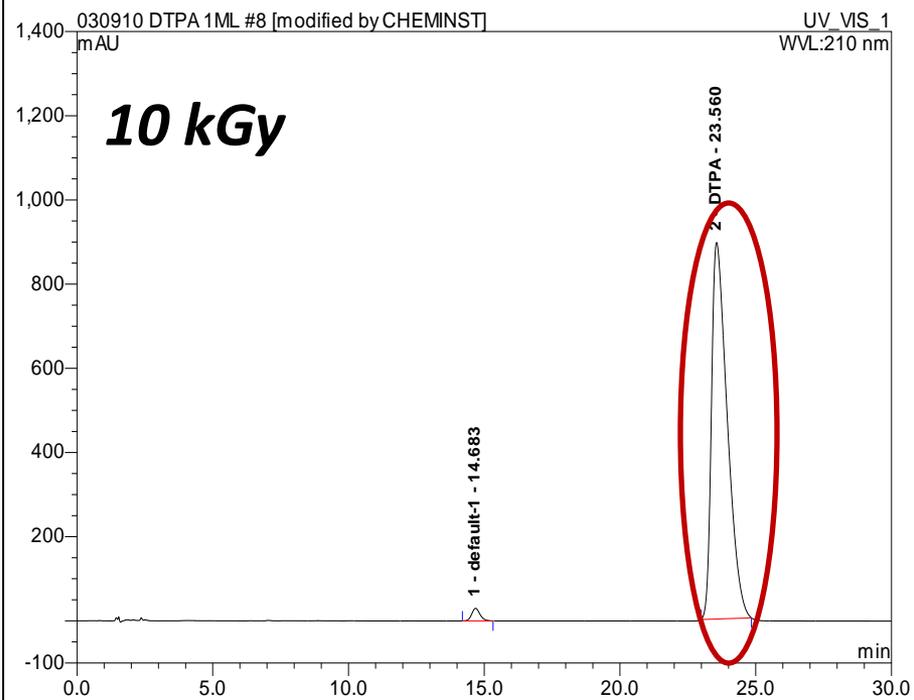


ethanol

## Gibbs Energy

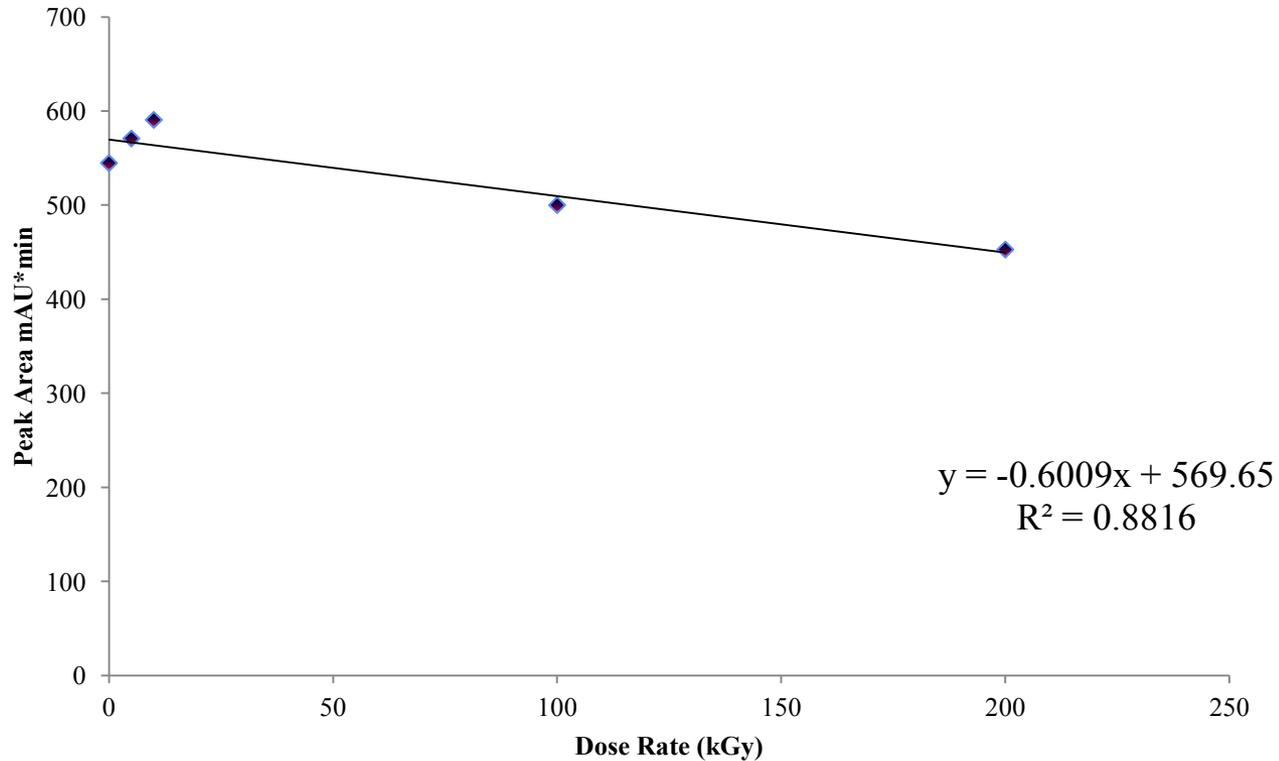
Left Pathway: -514.77 kJ/mol

Right Pathway: -278.8 kJ/mol

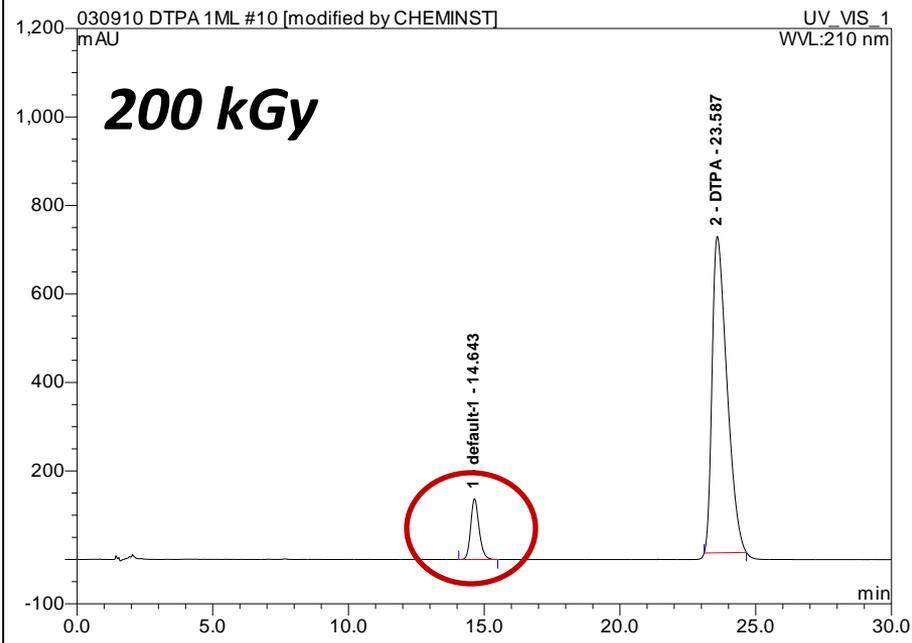
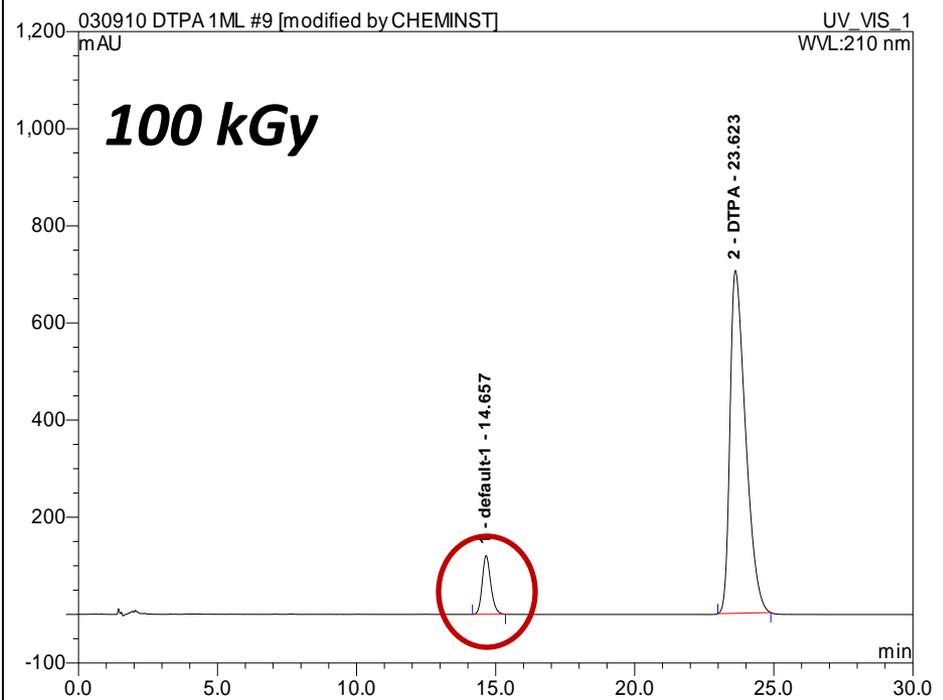
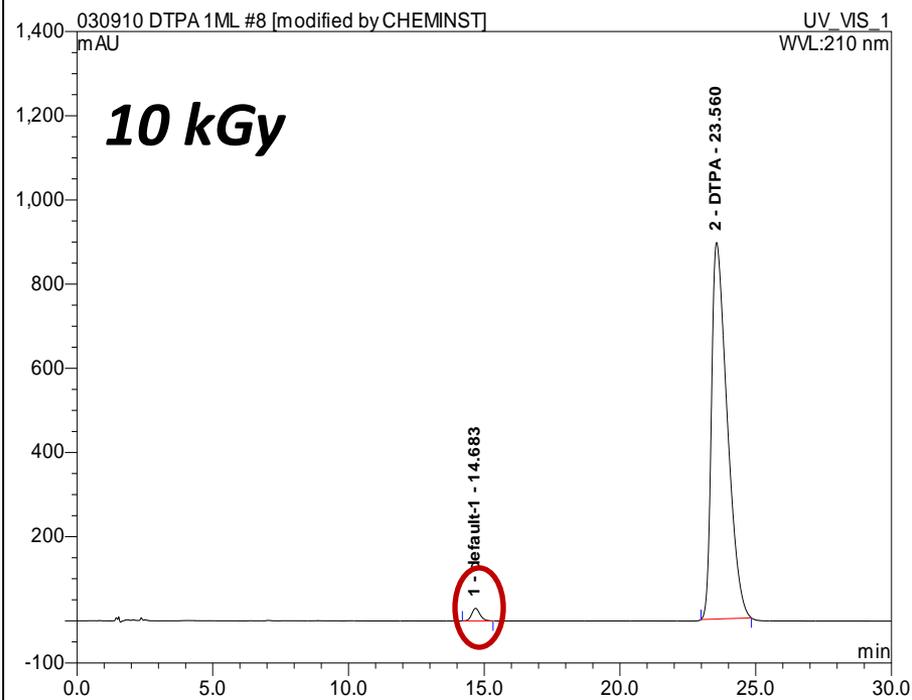


## Degradation of DTPA

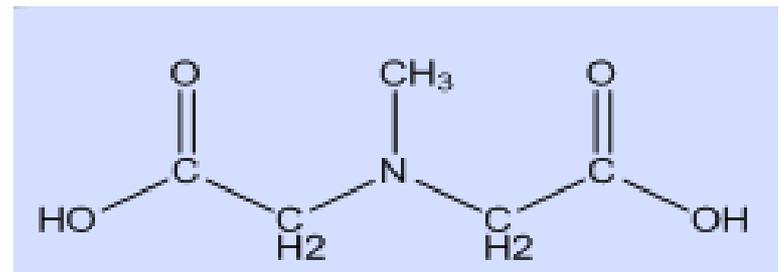
# Impact of Dose on DTPA Concentration



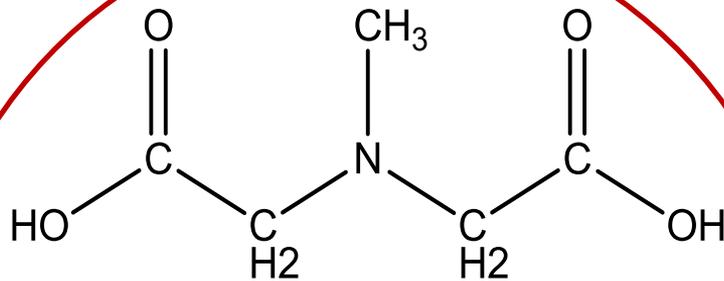
## DTPA Decay



**Peak analysis of analyte eluting at 15 minutes is Methyliminodiacetic acid**



# Radiolysis: Potential By-products of DTPA revisited



2,2'-(methylazanediyl)diacetic acid

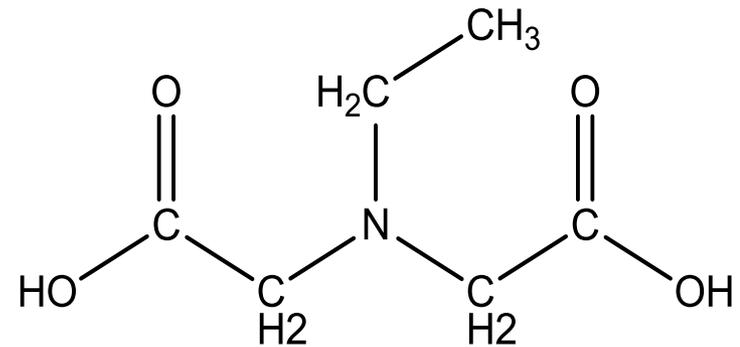
Critical Temp: 775.85 [K]

Critical Pres: 43.17 [Bar]

Critical Vol: 381.5 [cm<sup>3</sup>/mol]

Gibbs Energy: -585.82 [kJ/mol]

Log P: -1.22



2,2'-(ethylazanediyl)diacetic acid

Critical Temp: 784.25 [K]

Critical Pres: 38.2 [Bar]

Critical Vol: 437.5 [cm<sup>3</sup>/mol]

Gibbs Energy: -577.4 [kJ/mol]

Log P: -0.89

# Targetry coupled separations

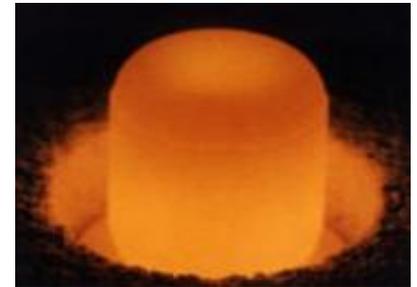
- **production of radioisotopes involves irradiation of targets followed by dissolution and separation**
  - **vast majority of the target is unreacted**
  - **minuscule amount of product must be separated from large amount of target**
  - **target must be reformed for further isotope production**
    - **losses and waste generation**
- **Targetry coupled separations exploit techniques to acquire the reaction product radionuclide without target destruction**
  - **advances in material synthesis**
  - **novel separations that do not require target dissolution**
- **Discussion provided for  $^{238}\text{Pu}$  production**
- **Can be applied to  $^{99}\text{Mo}$** 
  - **Porous uranium metal targets can be produced**
  - **$^{99}\text{Mo}$  is produced with a fission high yield.**
  - **$^{99}\text{Mo}$  can be separated from the uranium metal by the formation of volatile  $\text{Mo}(\text{CO})_6$** 
    - **Uranium does not react and remains in a form suitable for further irradiation**
    - **After treatment the uranium target can be reused to produce more  $^{99}\text{Mo}$**

# $^{238}\text{Pu}$ Production

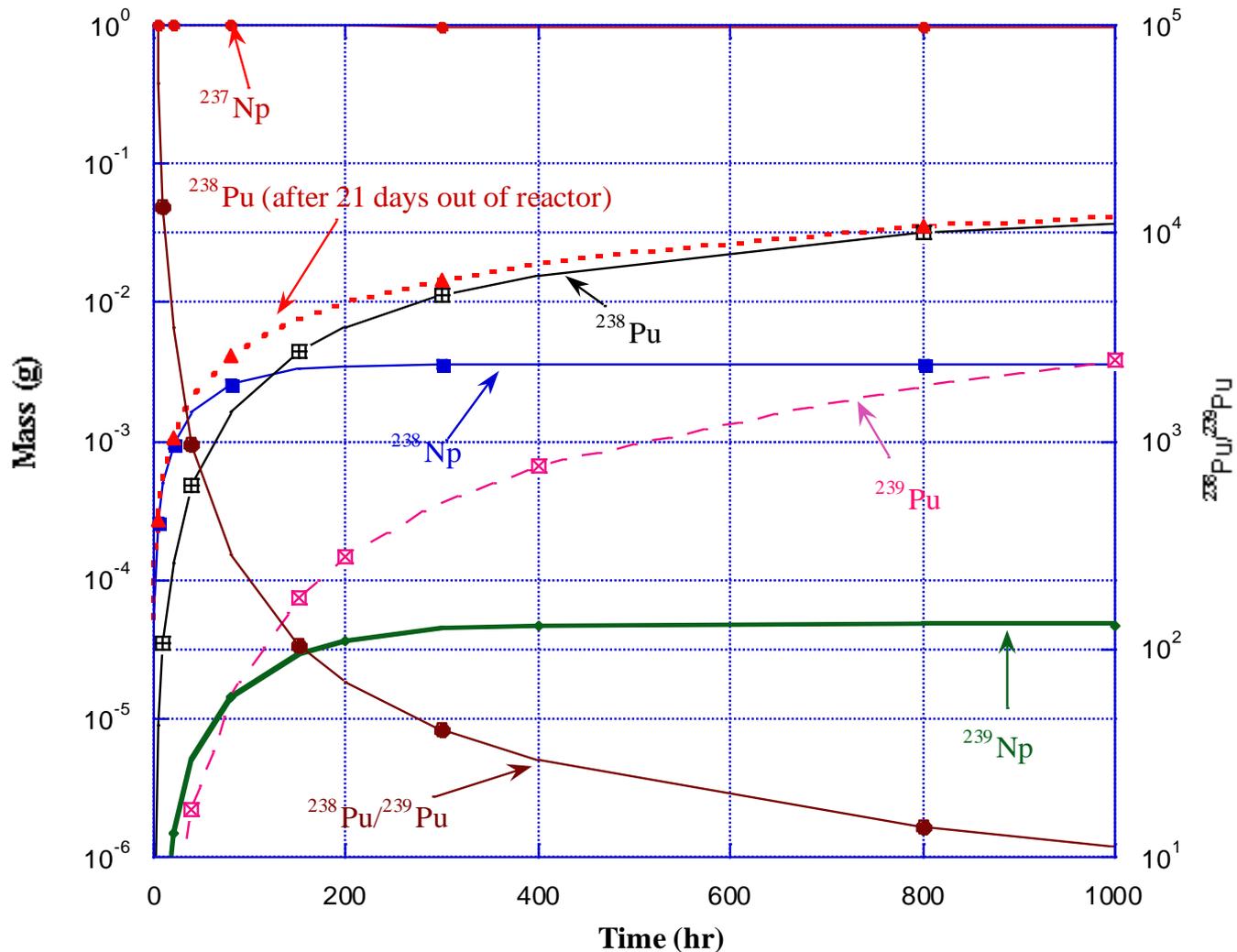
- **Target**
  - $^{237}\text{Np}$ 
    - (n, $\gamma$ ) reaction to form  $^{238}\text{Np}$  ( $t_{1/2} = 2.117$  d)
    - $^{238}\text{Np}$  beta decay to produce  $^{238}\text{Pu}$
  - $^{241}\text{Am}$ 
    - (n, $\gamma$ ) reaction to form  $^{242}\text{Am}$  ( $t_{1/2} = 16.02$  h) and  $^{242\text{m}}\text{Am}$  ( $t_{1/2} = 141$  a)
    - \* Large (n,f) cross section for 242 isotopes
      - ✂ 7000 b and 2100 b for m and g states
    - $^{242}\text{Am} \rightarrow \beta + ^{242}\text{Cm}$  ( $t_{1/2} = 162.8$  d) →  $\alpha + ^{238}\text{Pu}$
  - **Can exploit decay to tune isotopics and coupled with separation**
- **Separations**
  - **Coupled with target**

# $^{238}\text{Pu}$ properties

- $^{237}\text{Np}(n,\gamma)^{238}\text{Np}$ 
  - $^{238}\text{Pu}$  from beta decay of  $^{238}\text{Np}$
  - Separated from unreacted Np by ion exchange
- Decay of  $^{242}\text{Cm}$
- 0.57 W/g
- Power source for space exploration
  - 83.5 %  $^{238}\text{Pu}$ , chemical form as dioxide
  - Enriched  $^{16}\text{O}$  to limit neutron emission
    - 6000 n s<sup>-1</sup>g<sup>-1</sup>
    - 0.418 W/g PuO<sub>2</sub>
- 150 g PuO<sub>2</sub> in Ir-0.3 % W container



# Production of Np and Pu isotopes from 1 g of $^{237}\text{Np}$ in a neutron flux of $10^{14} \text{ n cm}^{-2}\text{s}^{-1}$



# $^{238}\text{Pu}$ production

- $^{238}\text{Pu}$  prepared by irradiating  $\text{NpO}_2$  particles embedded in an aluminum matrix
- Anion exchange in nitric acid solution was used to separate the Np and Pu from the fission products
  - Then separate Pu from Np
- Irradiated aluminum targets dissolved in 10 M nitric acid containing small amounts of KF and mercuric nitrate
  - nitric acid solution was adjusted to 8 M nitric acid concentration
  - ferrous sulfamate fixed Np and Pu to tetravalent state
- first stage of anion exchange on a Dowex I type resin separated Np(IV) and Pu(IV) nitrates from aluminum and fission products
- Pu selectively eluted as Pu(III) using 5.5 M nitric acid, 0.05 M ferrous sulfamate and 0.05 M hydrazine.
- Np(IV) was eluted 0.35 M nitric acid, precipitated as the oxalate complex, and then calcined to  $\text{NpO}_2$  for reuse in new targets
- $^{238}\text{Pu}$  was purified by anion exchange formed into  $\text{PuO}_2$

# Novel targets

- Previous method relied upon large target and highly shielded system
- Propose novel targets
  - **On-line separation**
    - Solution target
  - **Smaller sample sizes incorporated into automated separation**
    - Nanoparticle targets of  $^{237}\text{Np}$
- Can exploit hot atom chemistry of separation
  - **5.5 MeV gamma from capture**
    - 69 eV in recoil energy
    - May be sufficient for bond breaking

# Solution system

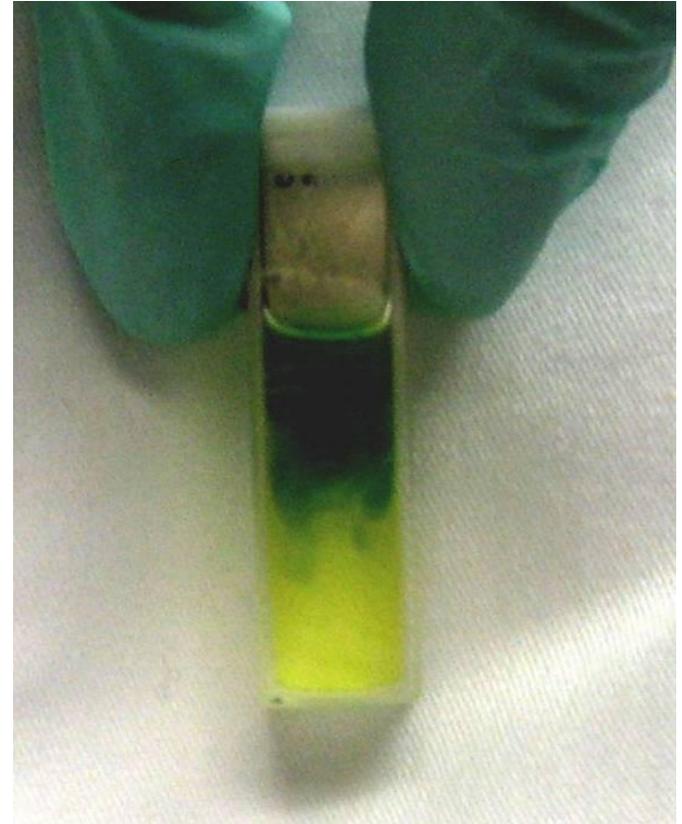
- **Non-aqueous systems**
  - **Molten salts**
  - **Ionic liquids**
- **Np in nitrate solution**
  - **Solution circulated into neutron flux**
  - **Produced  $^{238}\text{Pu}$  isolated on line**
- **Need to investigate**
  - **Selective separation of Pu**
    - Not practical to vary Np redox
    - Fission product interference
  - **Np redox chemistry under irradiation**
    - Np oxidation state can vary with accompanying impact on chemistry
    - Ability to separate Np from Pu controlled by redox

# Nanoparticle target

- **Nanoparticle target in holder, irradiated, then treated**
  - **Variation of irradiation time**
    - Static or dynamic placement in flux
  - **Fission recoil outside of particle**
  - **Possibility to elute Pu from NpO<sub>2</sub> matrix**
    - Selective separation with major change to matrix
    - Direct recycling of Np target
    - automation
- **Oxide nanoparticles**
  - **Based on abundant literature for ZrO<sub>2</sub> nanoparticles**
  - **Ability to vary size, porosity**

# Nanoparticle formation: Uranyl photoreduction

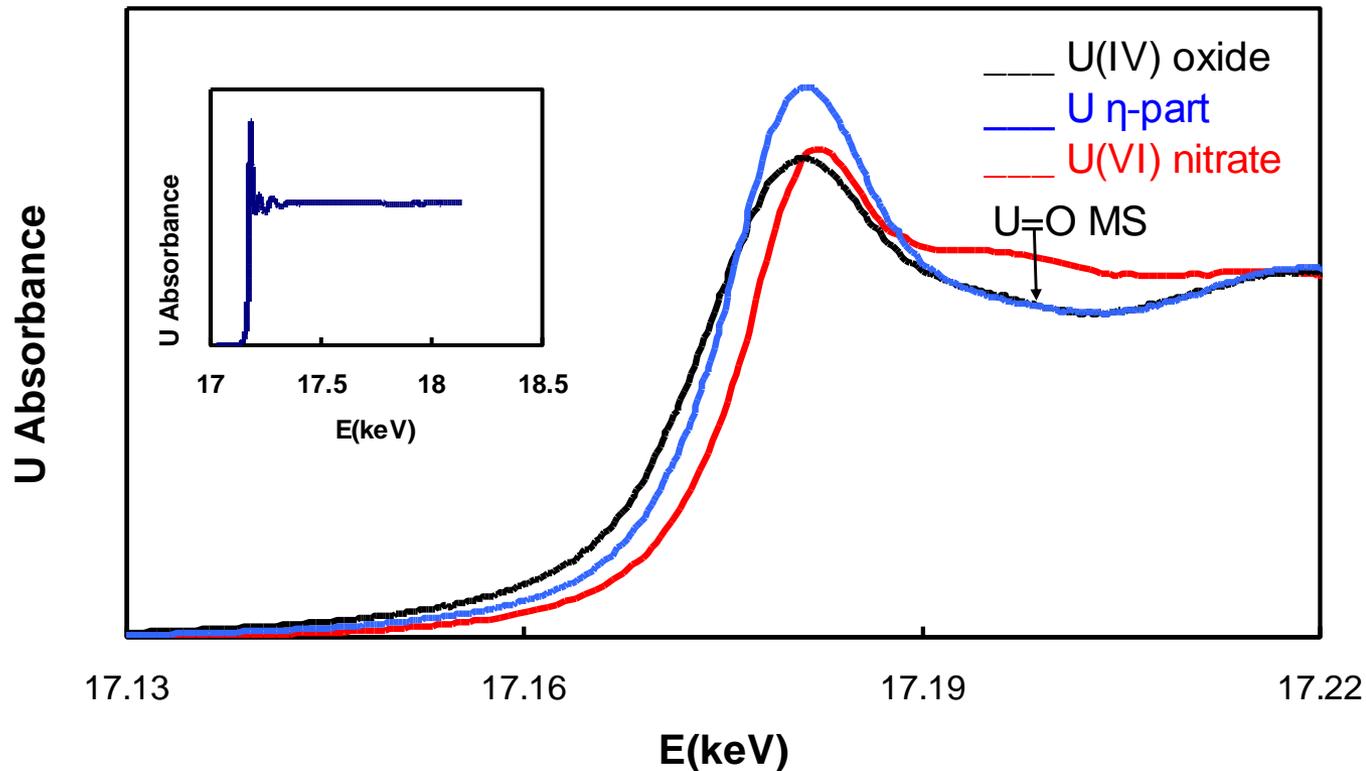
- **Rediscovered a system of photoreduction of uranyl in TBP-dodecane**
  - **Extraction of higher valent actinides into organic phase**  
→ U, Np, Pu
- **Uniform particle formation using laser**
  - **Can be used for rapid preparation of samples for alpha spectroscopy or laser ablation**
- **Properties appear tunable based on different parameters**
  - **Photon energy**
  - **acid/U concentration**
  - **concentration in alcohol for deposit**



**Nanoparticles form in the path of laser (414 nm)**

# EXAFS characterization of U nanoparticles

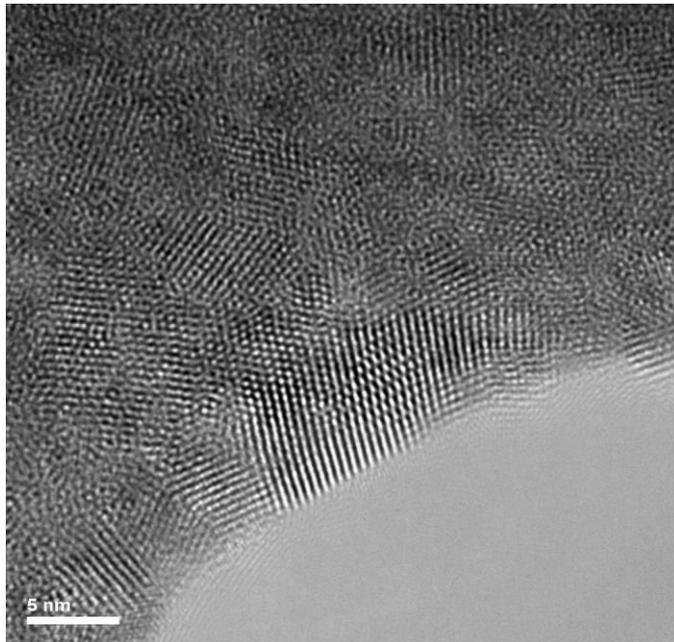
- First direct observation of U(IV) oxidation state
  - Confirmation of uranium reduction upon nanoparticle formation
- Initial analysis is consistent with U(IV)(DBP)<sub>4</sub> (8 symmetrical O ligands)



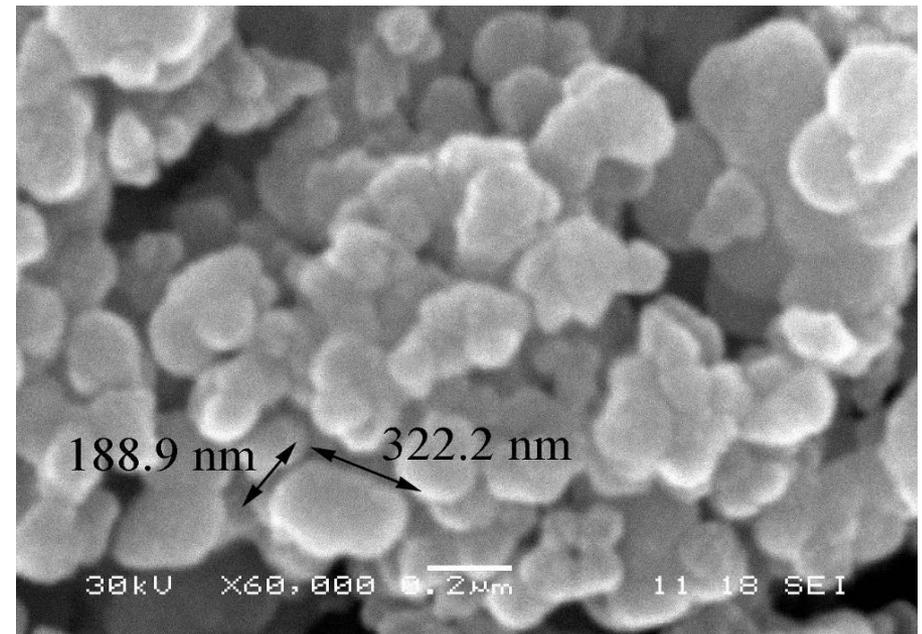
1 wt% U in BN; Advanced Photon Source Beamline BM-12  
(BESSRC-CAT), October 2007, Argonne National Laboratory

# Microscopy characterization

- **TEM** – shows that when the gel is in a dilute methanol suspension, the uranium compound forms self-assembling nanocrystalline monolayers

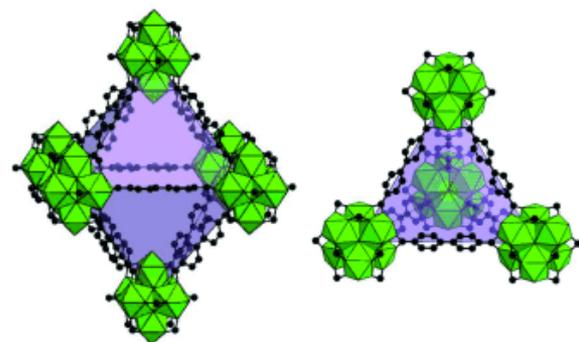


- **SEM** – shows that when the gel is in a concentrated methanol suspension, the uranium compound forms aggregated nanoscale amorphous granules

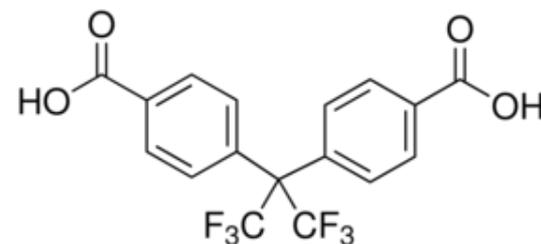


# Target Synthesis: MOF

- Literature review for Ln and Ac metal-organic framework materials under hydrothermal conditions
  - **Initial experiments on Y, lanthanides**
    - La, Y, Eu, Tb as carbonates or hydroxides
    - Reacted with 4,4'-(hexafluoroisopropylidene)bis(benzoic acid) due to its flexibility and ability to establish bridge between metal centers
    - Up to 200 °C for 72 hours, varied metal:ligand ratio
  - **10 different compounds formed**
    - Initial evaluation by X-ray diffraction
    - Primary product  $M_2L_3$
  - **Performing studies with uranium**
    - Aspartic and gluteric acid

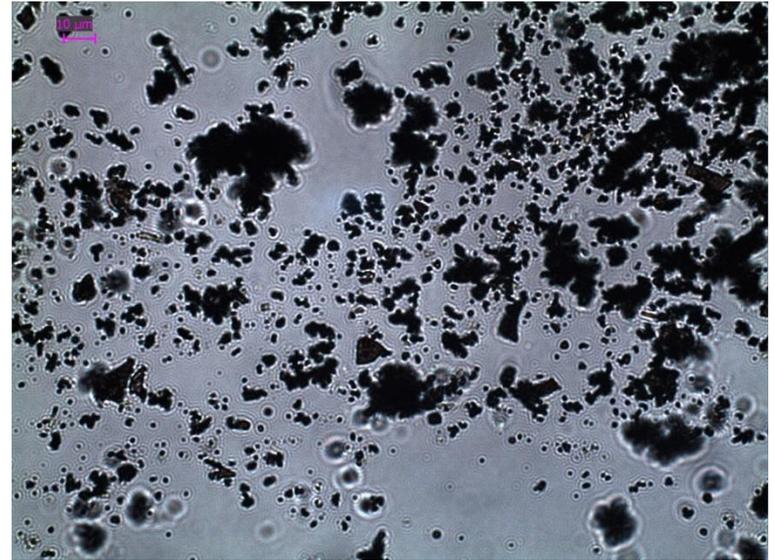


- Uranium MOF with N,N-dimethylformamide
- Cavities form 5 to 10 Å
- Chemistry 2013



# Recent Targetry

- **Formation of Uranium particles**
  - **Smaller than 10 micron**
- **Compact with KBr**
- **Irradiate with fast neutrons**
- **Dissolve KBr**



# Overview

- **Interaction of radiation with matter**
  - **Charged particles**
  - **Electrons**
  - **Photons**
  - **Neutrons**
    - **Role of energy, charge and mass**
- **Dosimetry**
  - **Influence of particles**
- **Hot Atom Chemistry**
  - **Energetic processes**
- **Experiments and utilization**
  - **Behavior in spent fuel**
  - **Organic phase degradation**
  - **Separations**