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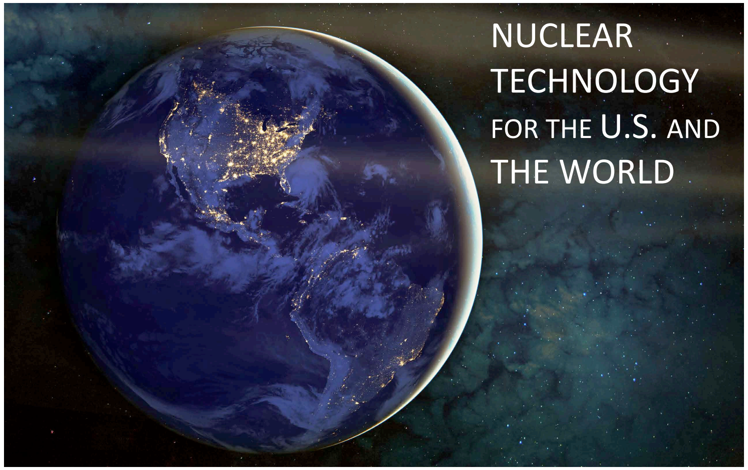
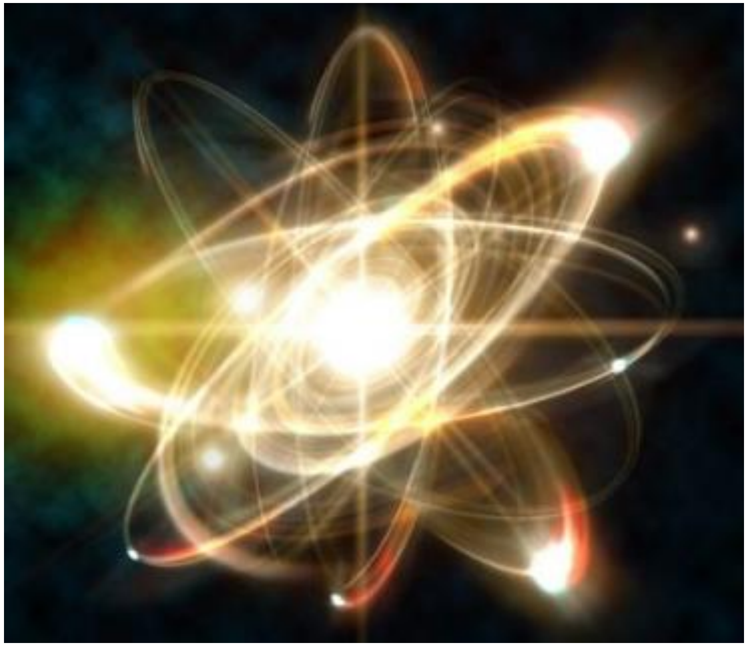
K-Infinite Comparison of Uranium Compounds at 5 wt. % U235

NCSD Technical Session
Data, Analysis and Operations in Nuclear Criticality Safety - II

November 20, 2019

ANS Winter Meeting & Expo 2019
Washington D.C. USA
Marriott Wardman Park





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Agenda

- Intro
- Uranium
- Uranium Speciation
- SCALE6.1 Model
- Results
- Conclusions

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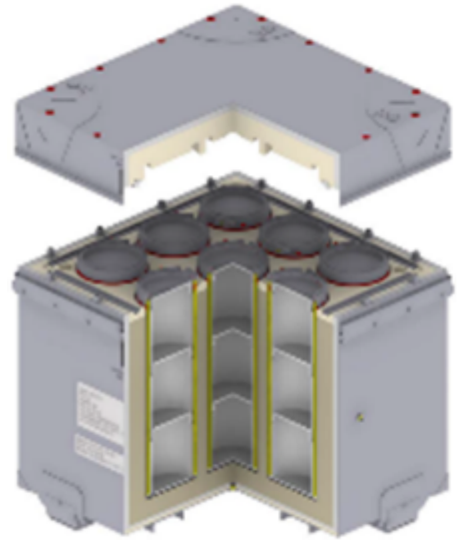
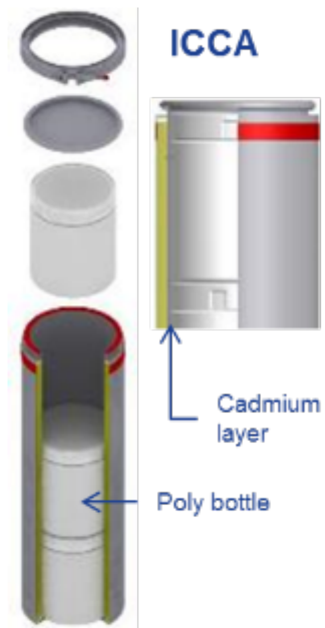
Introduction

Recent efforts to expand authorized contents of existing NPC Type A, Fissile nuclear package resulted in further study of select uranium compounds

- GEH/GNF owned design
- Licensed in the U.S. by NRC to 10CFR71 & IAEA requirements [USNRC CofC 9294]
- Type A container used to transport unirradiated fissile uranic material
- A cubic stainless steel and foam outer packaging with nine (9) ~ 8 inch ID inner containment canister assemblies (ICCAs)
- Currently used for transport of Type A quantities of low-enriched uranium oxide powder, pellets, and compounds of uranium
- Unique encased cadmium sheet used for reactivity control
- Planned future expansion of authorized content to include Type B material quantities (>5%, HALEU material forms)





Uranium

- The **element uranium** was discovered in 1789 in the pitchblende ores of Saxony by M. H. Klaproth (1743-1817), a notable analytic chemist and professor and the university of Berlin. Klaproth name the new element "Uranit", after planet Uranus, discovered earlier in 1781. A year later he changed the name to uranium.

- **Uranium is a soft**, silvery white metal. Its atomic number is 92 and its atomic weight is 238.03; thus it is the heaviest element found in nature.

- Certain **chemical characteristics** of uranium govern its behavior in chemical / geochemical processes. The element uranium has six valence electrons in the configuration $[Rn]5f^3 6d^1 7s^2$. The most common oxidation state of +6 involve the loss of all outer electrons; but the element may also exist in lower oxidation states including +3 ($5f^3$), +4($5f^2$), +5($5f^1$) and +6($5f^0$).

- Uranium is recognized as a **ubiquitous element**. Its concentration is sea water (3.34 µg/l) appears remarkably constant. The earth's crust contains 0.0004% uranium., which is more than gold, silver, mercury contents.

- Uranium has an **ion size** of about 1.05 A and is never found in its elemental state, but always in chemical combinations with other elements.

Uranium Speciation - I

- Aqueous solutions of uranium salts have an acid reaction as a result of hydrolysis; for instance $U^{4+} + H2O$ «» $U(OH)^{3+} + H^{+}$. The order of increasing hydrolysis depend on the charge and the size of the ion and is indicated by $U^{4+} > UO2^{2+} > U^{3+} > UO^{2+}$. Thus the U^{4+} ion has a strong tendency for hydrolysis, and when present in solution, form much stronger complexes with a given ligand than does the uranyl ion.

- Fuel fabrication facility take advantage of these chemical characteristics; precipitation recipes are vast and the resulting speciated uranium compounds involve complicated chemical reactions that continue to be studied to the present day.

- Depending on the byproduct waste streams, liquid lime slurry ($Ca(OH)_2$), nitric acid (HNO_3^-), ammonia (NH_4), ammonium bicarbonate ($(NH_4)HCO_3$), and sodium hydroxide (NaOH) may be used to precipitate uranium and ultimately dried into solid form.

- U3O8 is considered to be the more attractive form for disposal purposes because, under normal environmental conditions, U3O8 is one of the most kinetically and thermodynamically stable forms of uranium.

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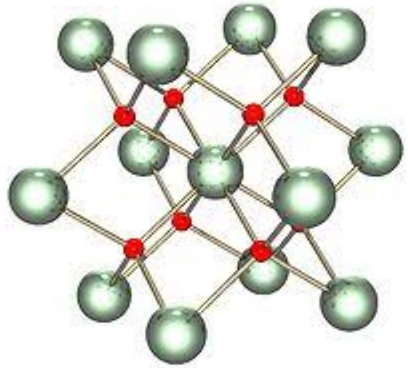
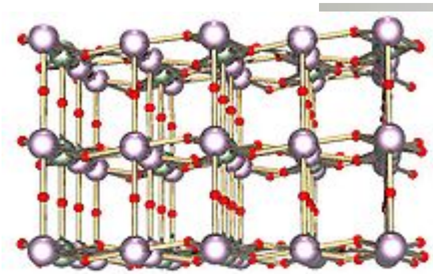
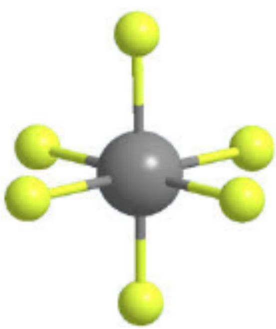
Uranium Speciation - II

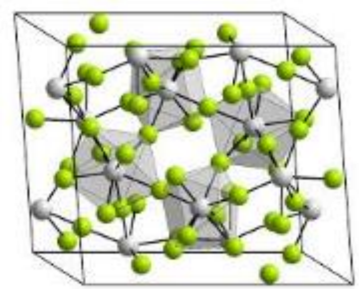
U_3O_8





UO_2





UF_6

UF_4

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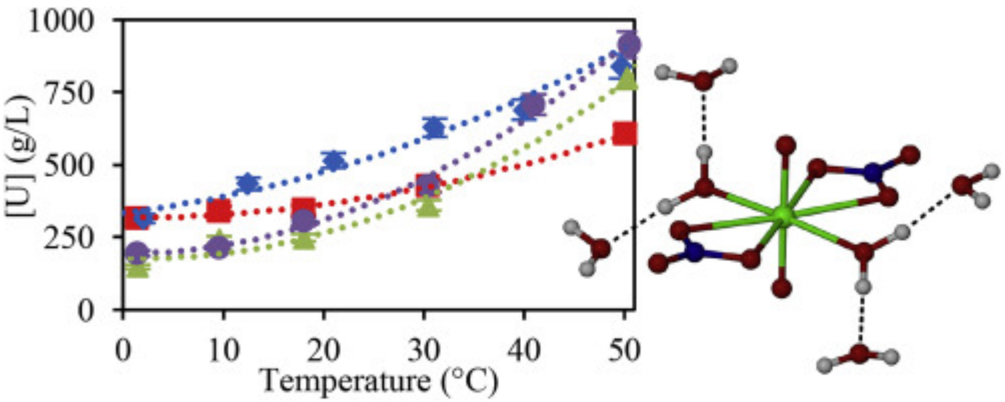
Uranium Speciation - III

$UO_2 (NO_3)_2$



$UO_2 (NO_3)_2*6H_2O$

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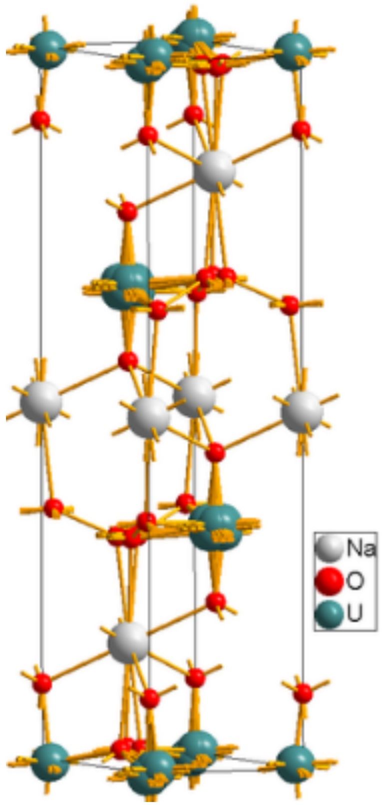
Uranium Speciation - IV



$Na_2U_2O_7*6H_2O$

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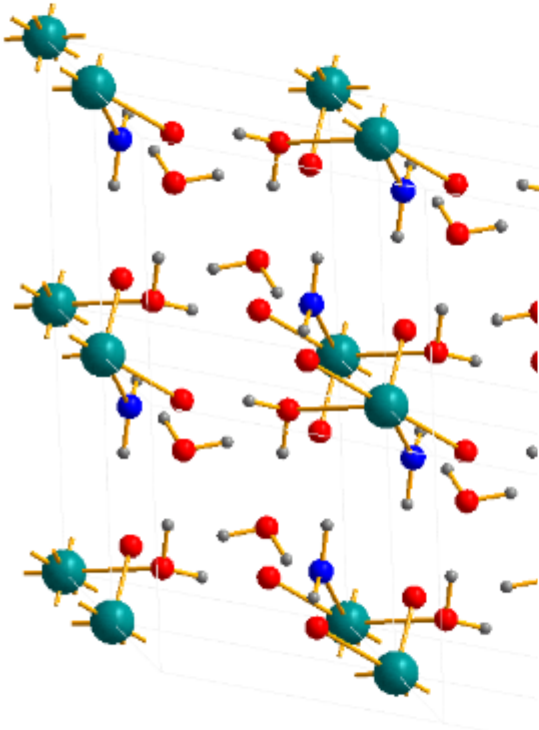
Uranium Speciation - V



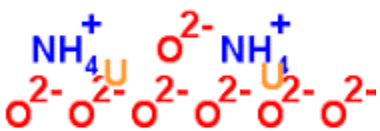


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Table A.1 Composition of ADU(I), (II), (III) and (IV)

Type	Chemical formula	Theoretical density*1), g/cm³	N / U atomic ratio	H / U atomic ratio
(I)	$UO_3 \cdot 2H_2O$	—	—	4.0
(II)	$3UO_3 \cdot NH_3 \cdot 5H_2O$	4.831	0.333	4.333
(III)	$2UO_3 \cdot NH_3 \cdot 3H_2O$	5.144	0.5	4.5
(IV)	$3UO_3 \cdot 2NH_3 \cdot 4H_2O$	5.201	0.667	4.667

*1) Natural uranium (^{235}U enrichment 0.711 wt %)

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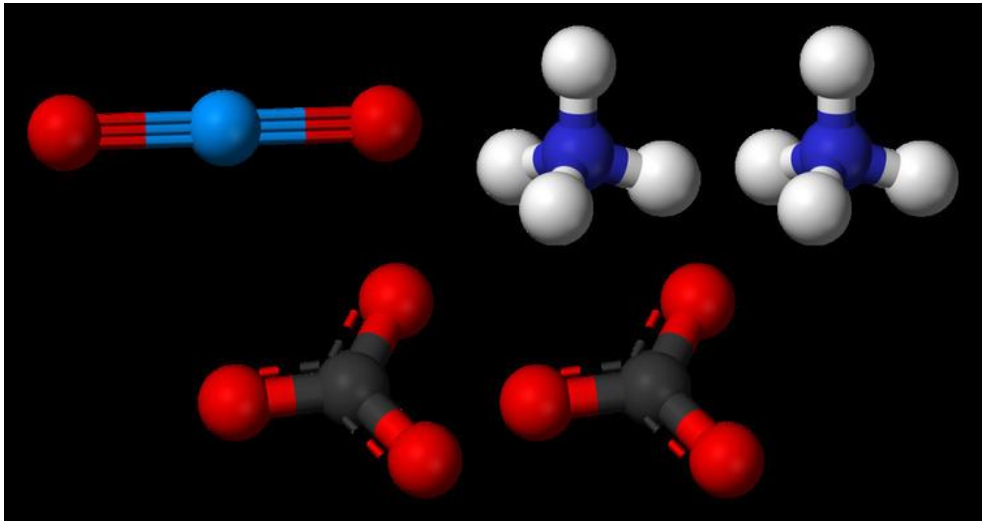
Uranium Speciation - VI



$(NH_4)_4*UO_2*(CO_3)_3$

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Uranium Compounds

Compound Name	Uranium Compound	Density (g/cc)	UFACT	Molecular Weight (g/mole)	Reference
Uranium oxides/ oxide bearing ash / sludges	UO_2	10.96	0.88144	269.8974	[1]
Uranium oxides/ oxide bearing ash / sludges	UO_2	4.5	0.88144	269.8974	[2]
Uranium oxides/ oxide bearing ash / sludges	U_3O_8	8.3	0.84793	841.691	[1]
Uranyl nitrate [UN]	$UO_2\ (NO_3)_2$	2.203	0.60395	393.9072	[1]
Uranyl nitrate hexahydrate [UNH]	$UO_2(NO_3)_2*6H_2O$	2.807	0.47390	501.9989	[3]
Calcium uranium oxides	$CaUO_3$	6.97	0.72981	325.9752	[4]
Calcium uranium oxides	$CaUO_4$	7.45	0.69566	341.9746	[4]
Calcium uranium oxides	Ca_2UO_5	5.67	0.59766	398.0524	[4]
Calcium uranium oxides	Ca_2UO_4	7.806	0.62268	382.053	[4]
Calcium uranium oxides	Ca_3UO6	5.337	0.52386	454.1302	[4]
Calcium uranium oxides	$CaU_3O_{10}*4H_2O$	5.25	0.72395	985.8294	[4]
Calcium uranium oxides	$CaU_6O_{19}*11H_2O$	5.16	0.72470	1969.6268	[4]
Calcium uranium oxides	$CaU_6O_{19}*10H_2O$	5.1	0.73152	1952.5245	[4]
Sodium diuranate	$Na_2U_2O_7$	6.57	0.75074	633.7726	[5]
Sodium uranate	Na_2UO_4	5.74	0.68386	347.8758	[6]
Uranium tetrafluoride (insoluble)	UF_4	6.7	0.75790	313.8922	[1]
Sodium diuranate – uranium tetrafluoride	$4\ Na_2U_2O_7*UF_4$	6.5862	0.75153	2848.9825	[5,7]
Ammonium diuranate [ADU]	$3UO_3*2NH_3*4H_2O$	5.201	0.74049	963.8126	[8]
Sodium diuranate hexahydrate	$Na_2U_2O_7*6H_2O$	6.57	0.64135	741.8642	[5]
Ammonium uranyl carbonate [AUC]	$(NH_4)_4*UO_2*(CO_3)_3$	2.77	0.45568	522.0780	[9]

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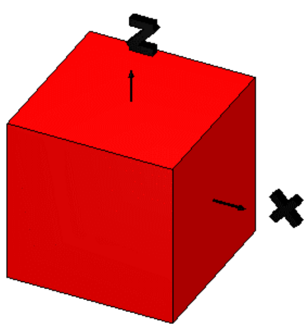
Ex. SCALE6.1 Model Constructs

```
=csas6
infinite medium of theoretical uo2 + h2o, 5% enr, var. wtfr_h2o
ce_v7_endf
read composition
 uo2        1 den=9.9655 0.99 300
                       92235 5
                       92238 95   end
 h2o        1 den=9.9655 0.01 300   end
end composition
read parameter
 gen=600
 npg=5000
 nsk=100
 htm=yes
end parameter
read geometry
global unit 1
com="global unit"
 cuboid 10      10     -10      10     -10      10     -10
 media 1 1 10
 boundary 10
end geometry
read bnds
  body=10
    all=mirror
  end bnds
end data
end
```




```
theoretical uo2(no3)2 + h2o, 5% enr, var. wtfr_h2o
read composition
 uo2(no3)2   1 den=2.1767 0.99 300
                      92235 5
                      92238 95   end
 h2o        1 den=2.1767 0.01 300   end
end composition
```

```
theoretical uo2(no3)2*6h2o + h2o, 5% enr, var. wtfr_h2o
read composition
 atomunh        1  2.7570  4
                      92000 1
                      8016 14
                      7014 2
                      1001 12
               0.99 300
                      92235 5
                      92238 95   end
 h2o        1 den=2.7570 0.01 300   end
```

```
theoretical cau6o19*10h2o + h2o, 5% enr, var. wtfr_h2o
read composition
 atomcauxoy_f8      1  4.8987  4
                      92000 6
                      8016 30
                      1001 22
                      20000 1
               0.99 300
                      92235 5
                      92238 95   end
 h2o        1 den=4.8987 0.01 300   end
```

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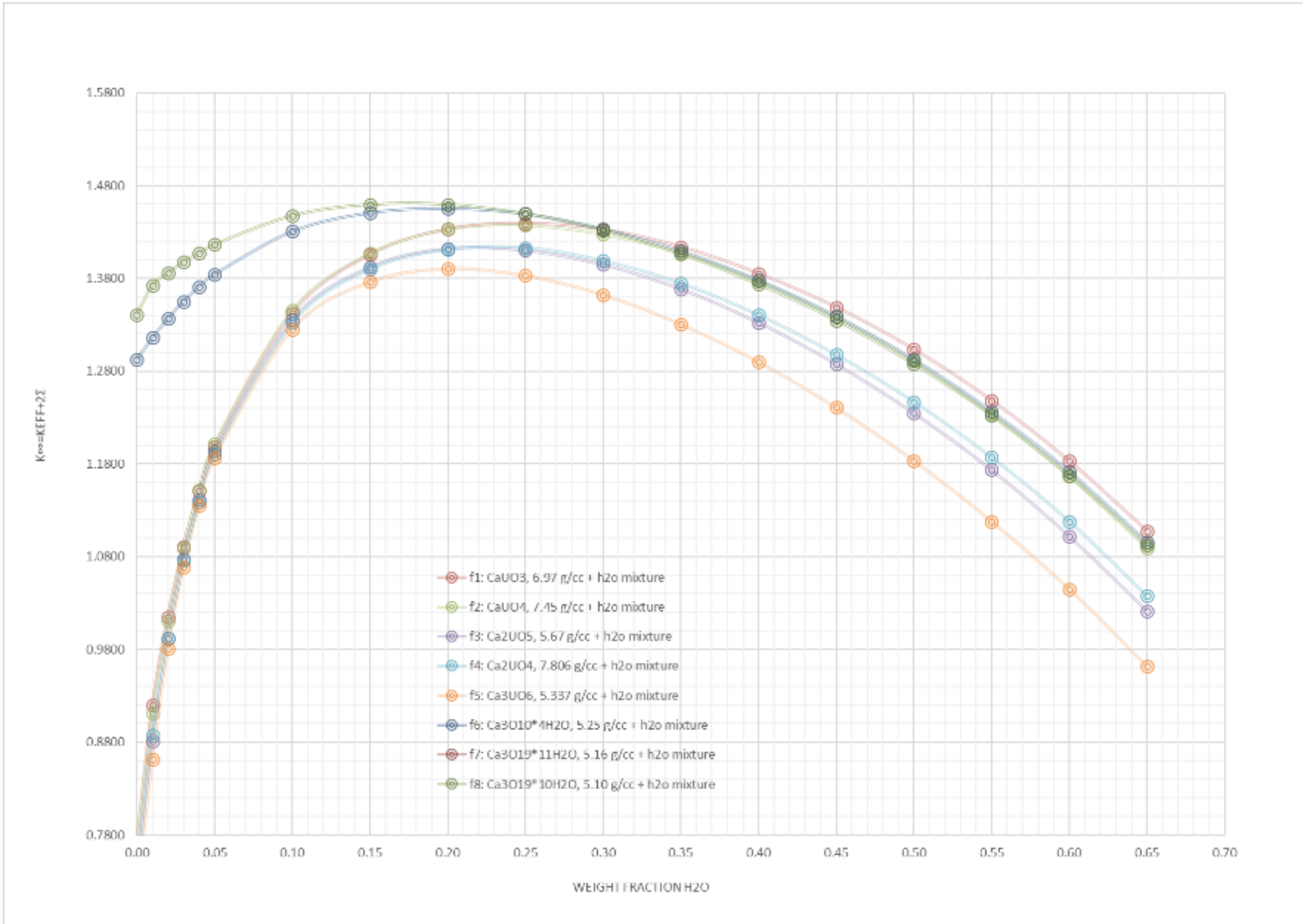
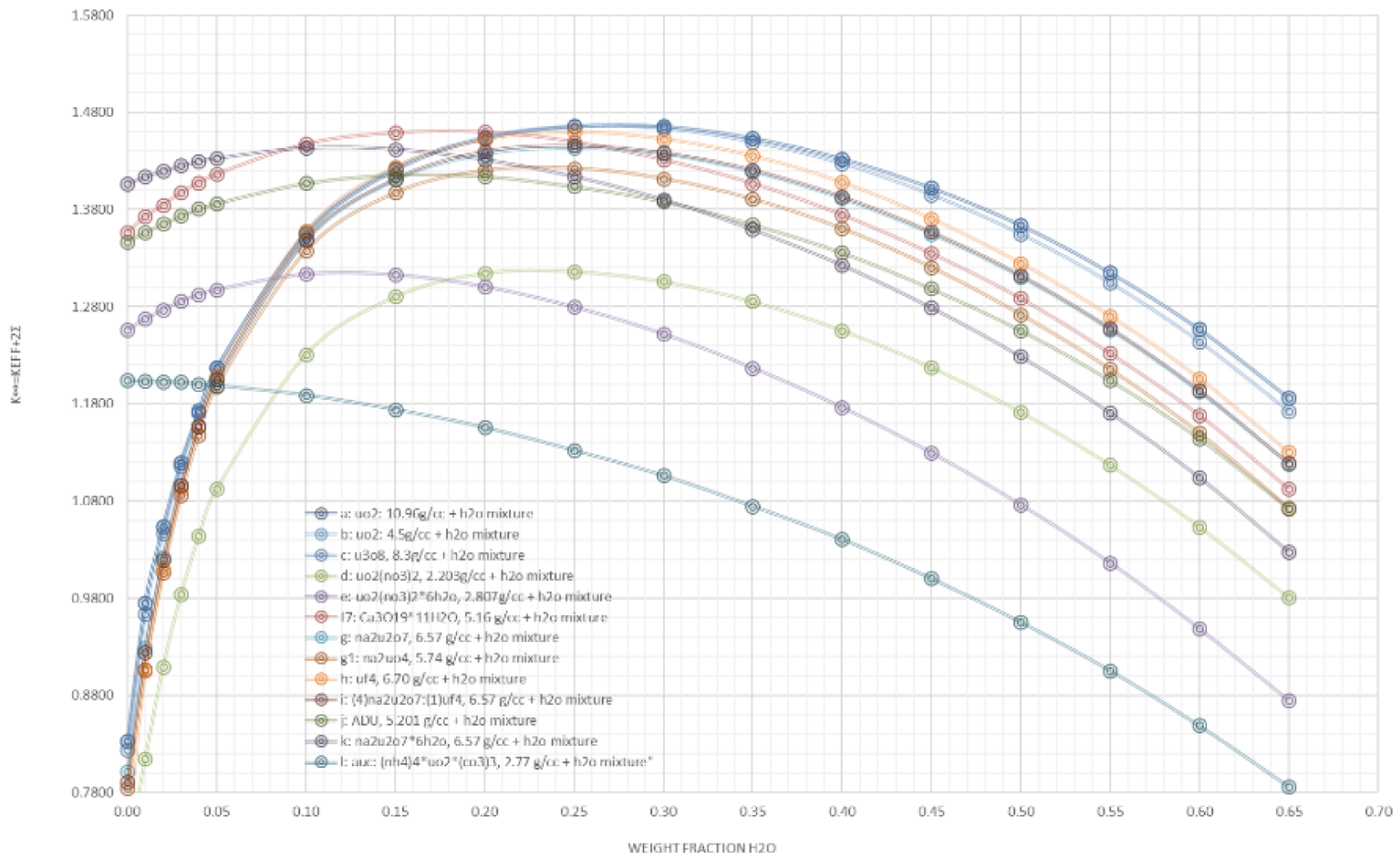
Results – I (Calcium uranium oxides)



Results – II (Uranium compounds)



Results – III (Tabulated Parato)

Compound	Keff	σ	K_{∞} [Keff + 2σ]
UO_2	1.46513	0.00040	1.46593
U_3O_8	1.46499	0.00048	1.46595
$CaU_6O_{19}*11H_2O$	1.45901	0.00040	1.45981
UF_4	1.45875	0.00045	1.45965
$4\ Na_2U_2O_7*UF_4$	1.44496	0.00040	1.44576
$Na_2U_2O_7$	1.44306	0.00042	1.44390
$Na_2U_2O_7*6H_2O$	1.44295	0.00041	1.44377
Na_2UO_4	1.42173	0.00041	1.42255
$3UO_3*2NH_3*4H_2O$	1.41478	0.00041	1.41560
$UO_2\ (NO_3)_2$	1.31546	0.00037	1.31620
$UO_2(NO_3)_2*6H_2O$	1.31270	0.00047	1.31364
$(NH_4)_4*UO_2*(CO_3)_3$	1.20346	0.00040	1.20426

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Conclusions - I

- Theoretical UO_2 is bounding when compared to other solid uranium compounds containing a uranium content less than a weight fraction of 0.88144.

- There is no statistically significant difference between UO_2 and U_3O_8 (as they are within 1σ)

- Uranium tetrafluoride compound is more reactive than the sodium diuranate compound. The representative 4:1 ratio evaluated for the complex compound of both substances (sodium diuranate – uranium tetrafluoride) is less reactive than UF_4. Thus, no molar ratio would be any more reactive than the bounding theoretical compound.

- The hexahydrate of sodium diuranate is less dense than the anhydrous form. It is therefore conservative to use the higher anhydrous density for this hexahydrate.

- Other modeled uranium compounds are demonstrated to be less reactive than UO_2 due to the presence of other neutron absorbing compounds (e.g., Ca, Na, N, F, etc.) or other diluents.

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Conclusions - II

- These results support the overall conclusion that theoretical uranium dioxide compound may be considered bounding insofar as authorized solid uranium scrap, sludge, ash, and residue byproduct compounds resulting from uranium recovery processes.

- Additional dry solid uranium byproducts not specifically evaluated herein that contain a uranium weight fraction less the theoretical UO2 would also be conservatively bounded.

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