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MOX - TDB : Nuclear Thermodynamic DataBase

Bertrand Cheynet

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NUCLEAR THERMODYNAMIC DATABASE

« MOX-TDB »

Ba-Fe-La-O-Pu-Ru-Sr-U-Zr

+

Ar-H

Version 2006-01

MOX-TDB

Nuclear Thermodynamic Database
Version 2006-01

developed by

THERMODATA - INPG - CNRS

&

AEA-Technology

Editor

B. Cheynet

with the participation of

P-Y. Chevalier, E. Fischer, P. Mason, M. Mignanelli.

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I. INTRODUCTION

A thermodynamic database collecting critical assessments made for nuclear applications has been built since 2003 for the MOX fuel.

MOX-TDB

is a thermodynamic database for in-vessel applications containing 9 + 2 elements :

Ba-Fe-La-O-Pu-Ru-Sr-U-Zr + Ar-H

This database covers the entire field from metal to oxide domains. It allows the user to calculate the thermodynamic equilibrium states and to use the results of the thermodynamic approach for improving the predictions of thermo-hydraulic or other codes.

The CALPHAD method, has been used here. It consists in describing the Gibbs energy of each phase in a given system, as a function of temperature, pressure and its composition taking into account various properties like crystal structure, solubility of elements on various types of sites etc. Thermodynamic databases provide consistent and accurate information for a better understanding of very complex phenomena.

2. DATA in the SYSTEM Ba-Fe-La-O-Pu-Ru-Sr-U-Zr + Ar-H

2.1. LIST of the SYSTEMS.

The lists of the assessed binary and quasi-binary systems are respectively presented in Table 1 and Table 2.

Table 1 Binary systems (36) in MOX-TDB.

| Element | Ba | Fe | La | O | Pu | Ru | Sr | U | Zr |
|---------|----|----|----|----|----|----|----|----|----|
| Ba | - | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 |
| Fe | - | - | 9 | 10 | 11 | 12 | 13 | 14 | 15 |
| La | - | - | - | 16 | 17 | 18 | 19 | 20 | 21 |
| O | - | - | - | - | 22 | 23 | 24 | 25 | 26 |
| Pu | - | - | - | - | - | 27 | 28 | 29 | 30 |
| Ru | - | - | - | - | - | - | 31 | 32 | 33 |
| Sr | - | - | - | - | - | - | - | 34 | 35 |
| U | - | - | - | - | - | - | - | - | 36 |
| Zr | - | - | - | - | - | - | - | - | - |

Table 2 Quasi-binary systems (28) in MOX-TDB.

| Oxide | BaO | FeO | FeO _{1.5} | LaO _{1.5} | OSr | O ₂ Pu | O ₂ U | O ₂ Zr |
|--------------------|-----|-----|--------------------|--------------------|-----|-------------------|------------------|-------------------|
| BaO | - | 1 | 2 | 3 | 4 | 5 | 6 | 7 |
| FeO | - | - | 8 | 9 | 10 | 11 | 12 | 13 |
| FeO _{1.5} | - | - | - | 14 | 15 | 16 | 17 | 18 |
| LaO _{1.5} | - | - | - | - | 19 | 20 | 21 | 22 |
| OSr | - | - | - | - | - | 23 | 24 | 25 |
| O ₂ Pu | - | - | - | - | - | - | 26 | 27 |
| O ₂ U | - | - | - | - | - | - | - | 28 |
| O ₂ Zr | - | - | - | - | - | - | - | - |

Only the most important ternary systems were critically assessed. The list is presented in Table 3.

Table 3 Ternary assessed systems (10) in MOX-TDB.

| | | | | | |
|---------|----------|---------|---------|--------|---------|
| Fe-O-Pu | Fe-Pu-U | Fe-U-Zr | O-Pu-U | O-U-Zr | Pu-U-Zr |
| Fe-O-U | Fe-Pu-Zr | | O-Pu-Zr | | |
| Fe-O-Zr | | | | | |

For all the other ternary systems including one or more of the elements Ba, La, Ru and Sr, the ternary parameters have been estimated by analogy with other systems of the same type or considered as null.

2.2. LIST of PHASES.

2.2.1. Condensed solutions.

The analysis of the binary and ternary phase diagrams allowed the listing of all the possible phases in the complex system.

The solution phases are the following :

Liquid (L) All the elements and pure oxides, except O_2Ru , are present in the liquid state. The associated non-ideal model is based on the 18 liquid following species : [Ba, BaO, Fe, FeO, $FeO_{1.5}$, La, $LaO_{1.5}$, O, OPu, OSr, O_2Pu , O_2U , O_2Zr , Pu, Ru, Sr, U, Zr]<L>.

The existence of miscibility gaps in the metal-oxygen systems Fe-O, La-O, O-Pu and O-U, the liquid has to be separated in 2 domains :

- ⇒ Liquid <Ba, Fe, La, O, Pu, Ru, Sr, U, Zr> (1) metallic liquid phase, with a limited solubility of oxygen.
- ⇒ Liquid <Ba, Fe, La, O, Pu, Ru, Sr, U, Zr> (2) oxide liquid phase, with a large solubility of oxygen.

The shape of the miscibility gap highly depends of the binary and ternary interaction parameters in the metal-oxygen systems, which have been estimated from the available experimental information.

More, few oxides systems may present a miscibility gap, such as $Fe_2O_3-ZrO_2$ (to confirm) and $Fe_2O_3-PuO_2$. There is a possibility for a second liquid phase with an oxide type :

- ⇒ Liquid <Ba, Fe, La, O, Pu, Ru, Sr, U, Zr> (3) liquid phase, with a large solubility for oxygen.

Some metallic systems (Ba-Fe, Ba-La, Ba-Pu, Ba-Ru, Ba-Sr, Ba-U, Ba-Zr, Fe-Sr, La-Pu, La-Sr, La-U, La-Zr, Pu-Sr, Ru-Sr, Sr-U, Sr-Zr) also presents a miscibility gap, but the elements Ba, La, Ru and Sr are in small quantity (« reactor » case) and the demixion in the metallic systems cannot appear.

Thus, it should be possible to imagine theoretically the coexistence of three liquid phases at equilibrium, two with an oxide type and one metallic. Evidently, it is not possible to distinguish « a priori » which liquid phase will be more or less rich in oxygen, or which metallic phase will be more or less rich in one element. Consequently, the liquid phase should be written three times with the same name, LIQUID. The thermodynamic data of these liquids are the same.

The melting temperatures (K) of the elements and pure oxides are : Ba [1000], Fe [1811], La [1193], Pu [913], Ru [2607], Sr [1050], U [1408], Zr [2127.85] : BaO [2286], $Fe_{0.947}O$ [1650], Fe_2O_3 [1925], La_2O_3 [2586], PuO_2 [2715], SrO [2805], UO_2 [3120], ZrO_2 [2987].

Bcc_A2 The metal elements such as Ba, Fe, La, Pu, Sr, U and Zr, have the cubic standard structure centered (bcc_A2) as allotropic form, in the intervals of following temperature (K) : Ba [298.15 - 1000], Fe [298.15 - 1184.8], [1667.5 - 1811], La [1134 - 1193], Pu [755.67 - 913], Sr [820 - 1050], U [1049 - 1408], Zr [1139.45 - 2127.85].

The solubility of oxygen in these metals [Ba, Fe, La, Pu, Sr, U] is limited or negligible

and consequently, this phase is primarily of metallic type, except for the system Zr-O, where maximum solubility in oxygen reaches approximately 13 at % O.

The equilibrium diagrams of phases of the binary systems shows various homogeneity fields for the solid solutions of bcc_A2 type :

- ⇒ Bcc_A2(1) <Fe, Pu, Ru, Zr> Final solid solution rich in iron, with a low solubility of Pu and Zr and limited Ru (approximately 7 at % Ru).
- ⇒ Bcc_A2(2) <Fe, O, Pu, Ru, Sr, U, Zr> Final solid solution rich in plutonium, uranium and zirconium, with a limited solubility of oxygen (approximately 13 at % O in O-Zr) and of Ru (7 at % Ru in Ru-U and 11.4 at % Ru in Ru-Zr) and a solubility of the other elements weak (Fe, Sr) or negligible (Ba, La). The solid solution uranium-zirconium presents a miscibility gap at low temperature (K) [970 - 1013].
- ⇒ Bcc_A2(3) <Ba, Sr> Final solid solution rich in barium and strontium.
- ⇒ Bcc_A2(4) <La, Pu> Final solid solution rich in lanthanum (< 25 at % Pu).

In the field of composition and temperature corresponding to the « reactor » cases, the elements like Ba, Sr, La and Ru are in limited content, and consequently, the phases bcc_A2(3) and bcc_A2 (4) are not likely to appear.

There are thus primarily two possible phases bcc_A2, the one rich in iron with a limited solubility of Ru, bcc_A2(1) and the other rich in Pu, U, Zr, with a limited solubility of Ru, bcc_A2(2). This phase can demix at low temperature ($T < 1013$ K).

Consequently, we decided to duplicate the solid solution bcc_A2 in the database, simultaneously to represent the solid solution rich in iron and the solid solution rich in Pu, U, Zr. The only notable differences between the two phases relate to the maximum temperature of stability, respectively 1820 K for the phase rich in iron (Fe-Ru) and 2127.85 K for the phase rich in Pu, U, Zr (Zr).

Fcc_A1

The metallic elements such as Fe, La, Pu, Sr, have the cubic standard structure with faces centered (fcc_A1) like allotropic form, in the following intervals of temperature (K) : Fe [1184.8 - 1667.5], La [550 - 1134], Pu [593.06 - 736.40], Sr [298.15 - 820].

The solubility of oxygen in these metals is negligible and consequently, this phase is primarily of metallic type.

The diagrams of phases of the binary systems shows various homogeneity fields for the solid solutions of fcc_A1 type :

- ⇒ Fcc_A1(1) <Fe, Pu, Ru, Zr> Final solid solution rich in iron, with negligible solubility of Ba, La, O, Sr, U, a low solubility of Pu and Zr (lower than 2 at %), and a wide solubility of Ru (25 at % maximum).
- ⇒ Fcc_A1(2) <Fe, Pu, Ru, U, Zr> Final solid solution rich in plutonium and zirconium, with limited solubility of Fe, Ru, U (1 at %), extended from Zr (60 at %), negligible of Ba, La and Sr, O.
- ⇒ Fcc_A1(3) <Ba, Sr> Final solid solution rich in Sr (up to 25% Ba).

⇒ Fcc_A1(4) <Ba, La, Pu> Final solid solution rich in La (< 25 at % Pu, 1.5 at % Ba).

In the field of composition and temperature corresponding to the « reactor » cases, the elements like Ba, Sr, La and Ru are in limited content and consequently, the phases fcc_A1(3) and fcc_A1(4) are not likely to appear.

There are thus primarily two possible phases fcc_A1, the one rich in iron with a wide solubility of Ru, fcc_A1(1) and the other rich in Pu and Zr, with a limited solubility of Fe, Ru, U, fcc_A1(2).

Consequently, we decided to duplicate the solid solution fcc_A1 in the database, simultaneously to represent the solid solution rich in iron and the solid solution rich in Pu, Zr. The only notable differences between the two phases relate to the maximum temperature of stability, respectively 1850 K for the phase rich in iron (Fe-Ru) and 900 K for the phase rich in Pu, Zr (Zr).

Hcp_A3

The metallic elements such as Ru and Zr have the hexagonal standard structure compact hcp_A3 like allotropic form, in the intervals of following temperature (K) : Ru [298.15 - 2607], Zr [298.15 - 1139].

Solubility of oxygen in Ru is negligible (unknown), while that in Zr is limited (30 at %). This phase is thus in metallic matter or metal-oxygen.

The diagrams of phases of the binary systems shows various homogeneity fields for the solid solutions of hcp_A3 type :

⇒ Hcp_A3(1) <Fe, Ru, Zr> Final solid solution rich in ruthenium, with negligible solubility of Ba, La, O, Pu, Sr, U, a low solubility of Zr (lower than 2 at %) and a wide solubility of Fe (60 at % maximum).

⇒ Hcp_A3(2) <O, Pu, Zr> Final solid solution rich in zirconium, with negligible solubility of Ba, La, Fe, Sr, U, Zr, and a wide solubility of Pu (15 at % maximum) and O (30 at %).

The phase hcp_A3 was thus duplicated in the database, the only difference between the two final solid solutions rich in Ru or rich in Zr being the maximum temperature of stability, respectively 2607 K and 2413 K.

Tet

Metallic uranium U has the standard structure tetragonal, tet, like allotropic form, in the temperature interval (K) [942 - 1049]. It dissolves small quantities of Fe, Ru, Zr, and large quantities of Pu (25 at %).

⇒ Tet <Fe, Pu, Ru, U, Zr> Final solid solution rich in uranium.

Ort_A20

Metallic uranium U has the orthogonal standard structure ort_A20, like allotropic form, in the temperature interval [298.15 - 942 K]. It dissolves small quantities of Fe, Zr, and large quantities of Pu (15 at %).

⇒ Ort_A20 <Fe, Pu, U, Zr> Final solid solution rich in uranium.

Laves

The binary systems Fe-Pu, Fe-U and Fe-Zr shows the existence of Laves compounds, Fe₂Pu(S) (T^m = 1514 K), Fe₂U(S) (T^m = 1507 K) and Fe₂Zr(S) (T^m = 1947 K). Consequently, there is a quaternary phase of Laves :

⇒ Laves <Fe, Pu, U, Zr> Inter metallic quaternary solid solution.

FeM₆

The binary systems Fe-Pu and Fe-U shows the existence of compounds of type FePu₆ (T^m = 703 K) and FeU₆ (T^m = 1077 K) of the same structure. Consequently, it exists a ternary phase with the type FeM₆.

⇒ FeM₆ <Fe, Pu, U> Ternary solid solution.

β

The binary systems Pu-U and Pu-Zr shows the existence of final solid ternary β<Pu, U> and β<Pu, Zr> with a very weak solubility of U and Zr. It exists a final solid ternary solution presenting the same characteristics :

⇒ β<Pu, U, Zr> Final solid ternary rich in β-Pu with a very weak solubility of U and Zr.

γ

The binary systems Pu-U and Pu-Zr shows the existence of final solid solutions γ<Pu, U> and γ<Pu, Zr> with a very weak solubility of U and Zr. It thus exists a final ternary solid solution presenting the same characteristics.

⇒ γ<Pu, U, Zr> Final solid ternary solution rich in γ-Pu with a very weak solubility of U and Zr.

Tet_A6

The binary systems Pu-U and Pu-Zr shows the existence of final solid ternary solutions Tet_A6<Pu, U> and Tet_A6<Pu, Zr> with a very weak solubility of U and Zr. It thus exists a final solid ternary solution presenting the same characteristics :

⇒ et_A6<Pu, U, Zr> Final solid ternary solution rich in Tet_A6-Pu with a very weak solubility of U and Zr.

ζ

The binary system Pu-U shows the existence of an intermediary solid solution rich in Pu-U, ζ<Pu, U>. Experimental information shows an extension of that phase in the ternary system Pu-U-Zr. It thus exists a solid ternary solution rich in Pu-U :

⇒ ζ<Pu, U, Zr> Solid ternary solution rich in Pu-U with a very weak solubility of Zr.

η

The binary system Pu-U shows the existence of an intermediary solid solution rich in Pu-U, η<Pu, U>. Experimental information shows an extension of that phase in the ternary system Pu-U-Zr. It thus exists an solid ternary solution rich in Pu-U :

⇒ η<Pu, U, Zr> Solid ternary solution rich in Pu-U with a very weak solubility of Zr.

α

The binary system Pu-Zr shows the existence of a final solid solution α<Pu, Zr> with a very weak solubility of Zr :

⇒ α<Pu, Zr> Final solid binary solution rich in α-Pu with a very weak solubility of Zr.

- θ** The binary system Pu-Zr shows the existence of an intermediary solid solution rich in Pu-Zr, $\theta\langle\text{Pu}, \text{Zr}\rangle$:
- ⇒ $\theta\langle\text{Pu}, \text{Zr}\rangle$ Intermediary solid binary solution rich in Pu.
- δ** The binary system U-Zr shows the existence of an intermediary solid solution rich in Zr, $\delta\langle\text{U}, \text{Zr}\rangle$:
- ⇒ $\delta\langle\text{U}, \text{Zr}\rangle$ Intermediary binary solid solution rich in Zr.
- Fcc_C1** The pure oxides O_2Pu , O_2U and O_2Zr have the structure centered face cubic, fcc_C1, with fluorite type (CaF_2), in the temperature range [298.15 - 2715 K], [298.15 - 3120 K] and [2650 - 2987 K] respectively. They are completely miscible at high temperature and can partially dissolve BaO, La_2O_3 and SrO. The solubility of oxides of Ba, La and Sr in O_2Pu is unknown and was assimilated to the one in O_2U . O_2U presents a field of sub and over stoichiometry, O_{2+x}U , while O_2Pu and O_2Zr are only sub-stoichiometric, O_{2-x}Pu and O_{2-x}Zr . Due to a tendency to demixion in the quasi-binary system $\text{O}_2\text{U}-\text{O}_2\text{Zr}$, the phase fcc_C1 may present a miscibility gap in the fields of non-stoichiometry.
- ⇒ Fcc_C1<Ba, La, O, Pu, Sr, U, Zr> Oxides solid solution, type fcc_C1 (O_2Pu , O_2U and O_2Zr).
- Fcc_B1** The pure oxides BaO, $\text{Fe}_{0.947}\text{O}$ and SrO have the structure centered face cubic, fcc_B1, in the temperature range [298.15 - 2286 K], [298.15 - 1650 K] and [298.15 - 2805 K] respectively. The wustite $\text{Fe}_{0.947}\text{O}$ is non-stoichiometric and his melting point can go to 1693 K with the content in oxygen. BaO and SrO are completely miscible at high temperature. It exists a very weak mutual solubility of $\text{Fe}_{0.947}\text{O}$ and SrO. In consequence, it exists two solid solutions with the fcc_B1 type, the one rich in BaO and SrO with a very weak solubility of $\text{Fe}_{0.947}\text{O}$, the second with the wustite type FeO- $\text{FeO}_{1.5}$ with a very weak solubility of SrO. We have represented these two phases with the same dataset :
- ⇒ Fcc_B1<Ba, Fe, O, Sr> Oxides solid solution fcc_B1 type (BaO, FeO, $\text{FeO}_{1.5}$, OSr). The one is a final one (rich in BaO and OSr), the other is an intermediary one with the wustite type (rich in FeO).
- Cc** The pure oxide La_2O_3 has three structures, hex, fcc and bcc versus temperature, which have been combined in the same denomination, cc, in the temperature range [298.15 - 2586 K] because the lack of experimental information concerning the heats of transition between these structures. This oxide can dissolve partially BaO, OSr and O_2Zr .
- ⇒ Cc<Ba, La, O, Sr, Zr> Final oxides solid solution, type cc (rich in La_2O_3).
- Tetragonal** The pure oxide O_2Zr has the tetragonal structure, in the temperature range [1478 - 2650 K]. It can partially dissolve O_2Pu and O_2U .
- ⇒ Tetragonal<O, Pu, U, Zr> Oxides solid solution, type tetragonal (rich in O_2Zr).
- Monoclinic** The pure oxide O_2Zr has a monoclinic structure, in the temperature range [298.15 - 1478 K]. It can dissolve partially O_2Pu .

⇒ Monoclinic<O, Pu, Zr> Oxides solid solution, type monoclinic (rich in O₂Zr).

Bcc The binary metal-oxygen system O-Pu shows an intermediary oxide solid solution with the bcc structure at low temperature :

⇒ Bcc<O, Pu> Intermediary oxide solid solution, type bcc.

Perovskite(1) The quasi-binary systems BaO-O₂Pu, BaO-O₂U, BaO-O₂Zr, OSr-O₂Pu, OSr-O₂U and OSr-O₂Zr all present perovskite type compounds, BaO₃Pu (T^m = 2399 K), BaO₃U (T^m = 2569 K), BaO₃Zr (T^m = 2815 K), O₃PuSr (T^m = 2408 K), O₃SrU (T^m = 2506 K) and O₃SrZr (T^m = 3023 K). In consequence, we have built a perovskite phase, having for reference species all these six compounds :

⇒ Perovskite(1) <Ba, O, Pu, Sr, U, Zr> Oxides solid solution, type perovskite (BaO₃Pu, BaO₃U, BaO₃Zr, O₃PuSr, O₃SrU and O₃SrZr).

Perovskite(2) Six complex oxides (ordered perovskites) with alkaline rare earths and hexavalent actinides have been reported : Ba₂SrUO₆, Ba₂SrPuO₆, Ba₃PuO₆, Sr₃PuO₆, Ba₃UO₆ and Sr₃UO₆. In consequence we should build a perovskite phase having for references the last four compounds :

⇒ Perovskite(2) <Ba, O, Pu, Sr, U> Oxides solid solution, type perovskite (Ba₃PuO₆, Sr₃PuO₆, Ba₃UO₆ and Sr₃UO₆).

But due to the lack of sufficient thermodynamic data, that phase has not been modelised.

The list of the solution phases taken into account in MOX-TDB is presented in Table 4.

Table 4 List of the solution phases in MOX-TDB.

| Phase | Elements | Formula | T | MG | Tmax |
|------------------|--------------------------|---|----|----|------|
| Liquid (L) | Ba-Fe-La-O-Pu-Ru-Sr-U-Zr | [Ba, BaO, Fe, FeO, FeO _{1.5} , La, LaO _{1.5} , O, OPu, OSr, O ₂ Pu, O ₂ U, O ₂ Zr, Pu, Ru, Sr, U, Zr]<L> | MO | 1 | 6000 |
| fcc_C1 | Ba-La-O-Pu-Sr-U-Zr | [BaO, LaO _{1.5} , O, OSr, O _{1.5} Pu, O ₂ Pu, O ₂ U, O ₂ Zr, U, Zr]<fcc_C1> | O | 1 | 3120 |
| fcc_B1 | Ba-Fe-O-Sr | [BaO, FeO, FeO _{1.5} , OSr]<fcc_B1> | O | 1 | 2805 |
| cc | Ba-La-O-Sr-Zr | [BaO, LaO _{1.5} , OSr, O ₂ Zr]<cc> | O | 0 | 1693 |
| perovskite(1) | Ba-O-Pu-Sr-U-Zr | [Ba, Sr][O] ₃ [Pu, U, Zr] | O | 0 | 2586 |
| tetragonal | O-Pu-U-Zr | [O ₂ Pu, O ₂ U, O ₂ Zr]<tetra> | O | 0 | 3023 |
| monoclinic | O-Pu-Zr | [O ₂ Pu, O ₂ Zr]<mono> | O | 0 | 2650 |
| bcc | O-Pu | [O, O _{1.5} Pu, O ₂ Pu, Pu]<bcc> | O | 0 | 1478 |
| bcc_A2 | Ba-Fe-La-O-Pu-Ru-Sr-U-Zr | [O, Va] ₃ [Ba, Fe, La, Pu, Ru, Sr, U, Zr]<bcc_A2> | MO | 1 | 1600 |
| hcp_A3 | Fe-O-Pu-Ru-U-Zr | [O, Va] _{0.5} [Fe, Pu, Ru, U, Zr]<hcp_A3> | MO | 1 | 1820 |
| fcc_A1 | Ba-Fe-La-Pu-Ru-Sr-U-Zr | [Ba, Fe, La, Pu, Ru, Sr, U, Zr]<fcc_A1> | M | 1 | 2128 |
| tet | Fe-Pu-Ru-U-Zr | [Fe, Pu, Ru, U, Zr]<tet> | M | 1 | 2607 |
| ort_A20 | Fe-Pu-U-Zr | [Fe, Pu, U, Zr]<ort_A20> | M | 1 | 2450 |
| Laves | Fe-Pu-U-Zr | [Fe] ₂ [Pu, U, Zr]<Laves> | M | 1 | 1850 |
| FeM ₆ | Fe-Pu-U | [Fe][Pu, U] ₆ <FeM _{6 <td>M</td> <td>1</td> <td>900</td>} | M | 1 | 900 |
| β | Pu-U-Zr | [Pu, U, Zr]<β> | M | 0 | 1049 |
| γ | Pu-U-Zr | [Pu, U, Zr]<γ> | M | 0 | 942 |
| tet_A6 | Pu-U-Zr | [Pu, U, Zr]<tet_A6> | M | 0 | 1947 |
| ζ | Pu-U-Zr | [Pu, U, Zr]<ζ> | M | 0 | 1077 |
| η | Pu-U-Zr | [Pu, U, Zr]<η> | M | 0 | 488 |
| α | Pu-Zr | [Pu, Zr]<α> | M | 0 | 593 |
| θ | Pu-Zr | [Pu, Zr] ₆ [Pu, Zr]<θ> | M | 0 | 756 |
| δ | U-Zr | [Zr][U, Zr] ₂ <δ> | M | 0 | 900 |
| | | | | | 1000 |

NOTE :

T = Type (O = Oxide, M = Metal, MO = Metal-Oxygen).

MG = Miscibility Gap (1 = Yes, 0 = No).

Tmax = Temperature max of stability.

The lists of the condensed and gaseous substances are presented in Table 5 and Table 6.

Table 5 List of the condensed substances in MOX-TDB.

| N° | SUBSTANCES | J / mole | J / g-atom |
|----|-------------------|--------------|------------|
| 1 | BA1(SER) | 0 | 0 |
| 2 | BA1FE12O19(S) | -5768158.27 | -180254.95 |
| 3 | BA1FE2O4(S) | -1502694.8 | -214670.69 |
| 4 | BA1H2(C) | -208883.49 | -69627.83 |
| 5 | BA1H2O2(C) | -972184.89 | -194436.98 |
| 6 | BA1LA2O4(S) | -2624374.86 | -374910.69 |
| 7 | BA1O2(S) | -666582.79 | -222194.26 |
| 8 | BA1O4U1(S) | -2043006.15 | -340501.03 |
| 9 | BA2FE2O5(S) | -2128609.36 | -236512.15 |
| 10 | BA2FE6O11(S) | -3873018.41 | -203843.07 |
| 11 | BA7FE4O13(S) | -6074799.94 | -253116.66 |
| 12 | FE1(SER) | 0 | 0 |
| 13 | FE10O22SR7(S) | -8939581.75 | -229220.04 |
| 14 | FE12LA1O19.5(S) | -6054445.14 | -186290.62 |
| 15 | FE12O19SR1(S) | -5927749.78 | -185242.18 |
| 16 | FE1H1O2(S) | -576696.07 | -144174.02 |
| 17 | FE1H2O2(S) | -600241.45 | -120048.29 |
| 18 | FE1H3O3(S) | -863802.49 | -123400.36 |
| 19 | FE1LA1O3(S) | -1404196.83 | -280839.37 |
| 20 | FE1O4U1(S) | -1623035.11 | -270505.85 |
| 21 | FE1ZR2(S) | -69427.85 | -23142.62 |
| 22 | FE1ZR3(S) | -99821.26 | -24955.32 |
| 23 | FE2H2O4(S) | -1153392.65 | -144174.08 |
| 24 | FE2O3(S) | -850469.83 | -170093.97 |
| 25 | FE2O5SR2(S) | -2176992.73 | -241888.08 |
| 26 | FE2O6SR3(S) | -2822811.84 | -256619.26 |
| 27 | FE333U250ZR417(e) | -27027.11 | -27027.11 |
| 28 | FE3O4(S) | -1160676.23 | -165810.89 |
| 29 | FE50U18ZR32(k) | -30356.85 | -30356.85 |
| 30 | FE6U71ZR23(l) | -17793.96 | -17793.96 |
| 31 | FE735ZR265(S) | -26835917.79 | -26835.92 |
| 32 | H2LA1(S) | -204522.91 | -68174.3 |
| 33 | H2O1(L) | -306684.99 | -102228.33 |
| 34 | H2O2SR1(C) | -992218.88 | -198443.78 |
| 35 | H2O4U1(S) | -1574966.17 | -224995.17 |
| 36 | H2PU1(S) | -157165.8 | -52388.6 |
| 37 | H2SR1(C) | -195503.66 | -65167.89 |
| 38 | H2ZR1(S) | -181153.17 | -60384.39 |
| 39 | H3LA1O3(S) | -1447228.46 | -206746.92 |
| 40 | H3PU1(S) | -157407.62 | -39351.91 |
| 41 | H3U1(S) | -146154.99 | -36538.75 |
| 42 | H4O5U1(S) | -1876492.03 | -187649.2 |
| 43 | LA1(DHCP) | -16965.45 | -16965.45 |
| 44 | LA1(SER) | 0 | 0 |
| 45 | LA1RU2(S) | -174054.54 | -58018.18 |
| 46 | LA2O5ZR1(S) | -2966288.16 | -370786.02 |
| 47 | LA2O7ZR2(S) | -4150169.84 | -377288.17 |
| 48 | LA3RU1(S) | -158894.75 | -39723.69 |

| | | | |
|----|------------------|--------------|------------|
| 49 | LA4O7SR1(S) | -4130654.4 | -344221.2 |
| 50 | LA4O9SR3(S) | -5311300.38 | -331956.27 |
| 51 | LA5RU2(S) | -290549.62 | -41507.09 |
| 52 | LA5RU3(S) | -364435.37 | -45554.42 |
| 53 | LA7RU3(S) | -421715.33 | -42171.53 |
| 54 | O1(SER) | 0 | 0 |
| 55 | O2RU1(S) | -327959.62 | -109319.87 |
| 56 | O2SR1(S) | -651046.78 | -217015.59 |
| 57 | O3.04PU2(S) | -1723956.34 | -342054.83 |
| 58 | O3PU2(HEXAGONAL) | -1704604.37 | -340920.87 |
| 59 | O3U1(S) | -1252455.16 | -313113.79 |
| 60 | O4SR2ZR1(S) | -2449213.55 | -349887.65 |
| 61 | O7SR3ZR2(S) | -4233460.37 | -352788.36 |
| 62 | O8U3(S) | -3657757.71 | -332523.43 |
| 63 | O9U4(S) | -4611611.91 | -354739.38 |
| 64 | PU1(SER) | 0 | 0 |
| 65 | PU19RU1(S) | -396792.18 | -19839.61 |
| 66 | PU1RU1(S) | -88908.52 | -44454.26 |
| 67 | PU1RU2(S) | -149185.59 | -49728.53 |
| 68 | PU3RU1(S) | -136608.75 | -34152.19 |
| 69 | PU5RU3(S) | -322803.91 | -40350.49 |
| 70 | RU1(SER) | 0 | 0 |
| 71 | RU1U2(S) | -102970.62 | -34323.54 |
| 72 | RU1ZR1(S) | -153269.53 | -76634.77 |
| 73 | RU2ZR1(S) | -140564.4 | -46854.8 |
| 74 | RU3U1(S) | -167385.3 | -41846.33 |
| 75 | RU474U526(S) | -35067025.96 | -35067.03 |
| 76 | RU4U3(S) | -239055.24 | -34150.75 |
| 77 | RU5U3(S) | -284872.9 | -35609.11 |
| 78 | SR1(SER) | 0 | 0 |
| 79 | U1(SER) | 0 | 0 |
| 80 | ZR1(SER) | 0 | 0 |

Hydrogen was only taken into account in the stoichiometric substances. It was not introduced in the solution phases.

Table 6 List of the gaseous substances in MOX-TDB.

| N° | SUBSTANCES | J / mole | J / g-atom |
|----|------------|------------|------------|
| 1 | AR1(G) | -46156.01 | -46156.01 |
| 2 | BA1(G) | 147398.23 | 147398.23 |
| 3 | BA1H1(G) | 138270.27 | 69135.14 |
| 4 | BA1H1O1(G) | -342305.72 | -114101.91 |
| 5 | BA1H2O2(G) | -719984.02 | -143996.8 |
| 6 | BA1O1(G) | -197909.36 | -98954.68 |
| 7 | BA2O1(G) | -327217.82 | -109072.61 |
| 8 | FE1(G) | 357670.39 | 357670.39 |
| 9 | FE1H2O2(G) | -414938.69 | -82987.74 |
| 10 | FE1O1(G) | 178944.32 | 89472.16 |
| 11 | FE2(G) | 469463.25 | 234731.63 |
| 12 | H1(G) | 183796.42 | 183796.42 |
| 13 | H1O1(G) | -15399.56 | -7699.78 |
| 14 | H1O1SR1(G) | -264116.81 | -88038.94 |
| 15 | H1O2(G) | -65748.6 | -21916.2 |
| 16 | H1SR1(G) | 155860.43 | 77930.22 |
| 17 | H1ZR1(G) | 451889.28 | 225944.64 |
| 18 | H2(G) | -38929.31 | -19464.66 |
| 19 | H2O1(G) | -298082.19 | -99360.73 |
| 20 | H2O2(G) | -205539.12 | -51384.78 |
| 21 | H2O2SR1(G) | -673915.64 | -134783.13 |
| 22 | LA1(G) | 361792.21 | 361792.21 |
| 23 | LA1O1(G) | -192451.31 | -96225.65 |
| 24 | LA2O1(G) | -141915.24 | -47305.08 |
| 25 | LA2O2(G) | -715506.87 | -178876.72 |
| 26 | O1(G) | 201160.56 | 201160.56 |
| 27 | O1PU1(G) | -135848.95 | -67924.48 |
| 28 | O1SR1(G) | -70257.88 | -35128.94 |
| 29 | O1U1(G) | -45138.27 | -22569.14 |
| 30 | O1ZR1(G) | 14500.96 | 7250.48 |
| 31 | O2(G) | -61164.58 | -30582.29 |
| 32 | O2PU1(G) | -495164.16 | -165054.72 |
| 33 | O2U1(G) | -553546.13 | -184515.38 |
| 34 | O2ZR1(G) | -477564.43 | -159188.14 |
| 35 | O3(G) | 70540.6 | 23513.53 |
| 36 | O3RU1(G) | -160573.12 | -40143.28 |
| 37 | O3U1(G) | -897302.47 | -224325.62 |
| 38 | O4RU1(G) | -269585.7 | -53917.14 |
| 39 | PU1(G) | 292391.6 | 292391.6 |
| 40 | RU1(G) | 600987.7 | 600987.7 |
| 41 | SR1(G) | 111785.53 | 111785.53 |
| 42 | SR2(G) | 228192.96 | 114096.48 |
| 43 | U1(G) | 476549.41 | 476549.41 |
| 44 | ZR1(G) | 535132.84 | 535132.84 |
| 45 | ZR2(G) | 851510.77 | 425755.38 |

3. STATE of VALIDATION

The state of validation of a thermodynamic database is characterised by the good agreement between calculated and available experimental results (phase diagrams and thermodynamic properties) concerning basic sub-systems (binary, ternary, ...) or practical global experiments, made in similar conditions and at thermodynamic equilibrium.

For user information, a quality criterion, based on comparison between calculation and available experimental data, has been established for each assessed sub-system.

- * **Estimated**
No experimental data available.

- ** **Perfectible**
Some domains need more experimental information (phase diagram or thermodynamic properties).

- *** **Acceptable**
The system is well known and satisfactorily modelled.

- **** **High quality**
The system is quite known and modelled.

The complete list of the metal-metal or metal-oxygen binary systems based on pure elements and oxide pseudo-binary systems based on pure oxides are presented in tables 7 and 8 respectively.

Due to the very high number of possible ternary and pseudo-ternary systems, it is completely unimaginable to assess all of them in a human delay. For that reason it was decided to assess only the most important ternary systems for practical applications. The list of the selected ternary systems is presented in table 9 .

The quality criterion takes into account both phase diagram and thermodynamic properties, and thus cannot be indicated only on a phase diagram. This point is fundamental for the modelling of multi-component systems.

Moreover, it must kept in mind that the set of a quality criterion remains somewhere subjective, and the improvement of existing sub-systems with newly available experimental results is a continuous task, which is part of the database management and updating.

3.1. BINARY and PSEUDO-BINARY SYSTEMS.

Table 7 Binary systems.

| System | Quality | Date of issue |
|--------|---------|---------------|
| Ba-Fe | ** | 03/2003 |
| Ba-La | ** | 03/2003 |
| Ba-O | ** | 03/2003 |
| Ba-Pu | * | 03/2003 |
| Ba-Ru | * | 03/2003 |
| Ba-Sr | ** | 03/2003 |
| Ba-U | * | 03/2003 |
| Ba-Zr | * | 03/2003 |
| Fe-La | *** | 03/2003 |
| Fe-O | *** | 03/2003 |
| Fe-Pu | ** | 03/2003 |
| Fe-Ru | *** | 03/2003 |

| System | Quality | Date of issue |
|--------|---------|----------------|
| Fe-Sr | * | 03/2003 |
| Fe-U | ** | 01/2005 |
| Fe-Zr | ** | 01/2005 |
| La-O | * | 03/2003 |
| La-Pu | ** | 03/2003 |
| La-Ru | ** | 03/2003 |
| La-Sr | * | 03/2003 |
| La-U | *** | 03/2003 |
| La-Zr | * | 03/2003 |
| O-Pu | ** | 03/2003 |
| O-Ru | * | 03/2003 |
| O-Sr | * | 03/2003 |

| System | Quality | Date of issue |
|--------|---------|----------------|
| O-U | *** | 01/2006 |
| O-Zr | *** | 01/2005 |
| Pu-Ru | ** | 03/2003 |
| Pu-Sr | * | 03/2003 |
| Pu-U | ** | 03/2003 |
| Pu-Zr | ** | 03/2003 |
| Ru-Sr | * | 03/2003 |
| Ru-U | *** | 03/2003 |
| Ru-Zr | *** | 03/2003 |
| Sr-U | * | 03/2003 |
| Sr-Zr | * | 03/2003 |
| U-Zr | *** | 01/2005 |

Table 8 Pseudo-binary systems.

| System | Quality | Date of issue |
|------------------------------------|---------|---------------|
| BaO-Fe ₂ O ₃ | *** | 03/2003 |
| BaO-La ₂ O ₃ | ** | 03/2003 |
| BaO-O ₂ Pu | * | 03/2003 |
| BaO-OSr | ** | 03/2003 |
| BaO-O ₂ U | ** | 03/2003 |
| BaO-O ₂ Zr | *** | 03/2003 |
| FeO-OSr | ** | 03/2003 |
| FeO-O ₂ U | ** | 03/2003 |

| System | Quality | Date of issue |
|--|---------|---------------|
| FeO-O ₂ Zr | ** | 03/2003 |
| Fe ₂ O ₃ -La ₂ O ₃ | ** | 03/2003 |
| Fe ₂ O ₃ -OSr | ** | 03/2003 |
| Fe ₂ O ₃ -O ₂ U | * | 03/2003 |
| Fe ₂ O ₃ -O ₂ Zr | * | 03/2003 |
| La ₂ O ₃ -O ₂ Pu | * | 03/2003 |
| La ₂ O ₃ -OSr | ** | 03/2003 |
| La ₂ O ₃ -O ₂ U | ** | 03/2003 |

| System | Quality | Date of issue |
|---|---------|----------------|
| La ₂ O ₃ -O ₂ Zr | *** | 03/2003 |
| OSr-O ₂ Pu | * | 03/2003 |
| OSr-O ₂ U | ** | 03/2003 |
| OSr-O ₂ Zr | *** | 03/2003 |
| O ₂ Pu-O ₂ U | ** | 03/2003 |
| O ₂ Pu-O ₂ Zr | ** | 03/2003 |
| O ₂ U-O ₂ Zr | *** | 01/2005 |

3.2. SELECTED TERNARY SYSTEMS.

Table 9 Ternary systems.

| System | Quality | Date of issue |
|----------|---------|----------------|
| Fe-O-Pu | | 03/2003 |
| Fe-O-U | ** | 01/2005 |
| Fe-O-Zr | ** | 01/2005 |
| Fe-Pu-U | ** | 03/2003 |
| Fe-Pu-Zr | ** | 03/2003 |

| System | Quality | Date of issue |
|---------|---------|----------------|
| Fe-U-Zr | ** | 01/2005 |
| O-Pu-U | * | 03/2003 |
| O-Pu-Zr | * | 03/2003 |
| O-U-Zr | *** | 01/2006 |
| Pu-U-Zr | ** | 03/2003 |

3.3. DOMAIN of APPLICATION and LIMITATIONS.

In this version of MOX-TDB most of the binary and pseudo-binary systems have been analysed and critically assessed.

However, some of them are still insufficiently known, and further experimental work is needed for the ones interesting the nuclear field.

In a very high-number component system such as the one covered by MOX-TDB, the number of possible ternaries and pseudo-ternaries is very important, while the time cost to assess only one is about one man year. Few experimental data are available in open literature for ternaries of high interest for nuclear field. Consequently, the complete assessment of all ternaries is not achievable in a human delay and only some selected ternary systems of main importance were assessed at this time.

4. ASSESSMENT of O-Pu-U-Zr

An important experimental and modelling work was done in the last years by many people in the world to obtain now a rather good knowledge and representation of that main quaternary system.

In the present version of the database all the available information (equilibrium points of phase diagrams, activities measurements, vapour pressures etc...), up to the year 2005, was taken into account in the optimisation procedure.

To allow the user to have an idea of the validity of the obtained results the calculated phase diagrams are compared, in the following pages, with some experimental points. But if anybody is interested more in detail by the experiments or by critical assessment and optimisation work he has to look at the numerous scientific papers published in the recent years.

4.1. BINARY SYSTEMS.

O-Pu,
O-U,
O-Zr,
Pu-U,
Pu-Zr,
U-Zr.

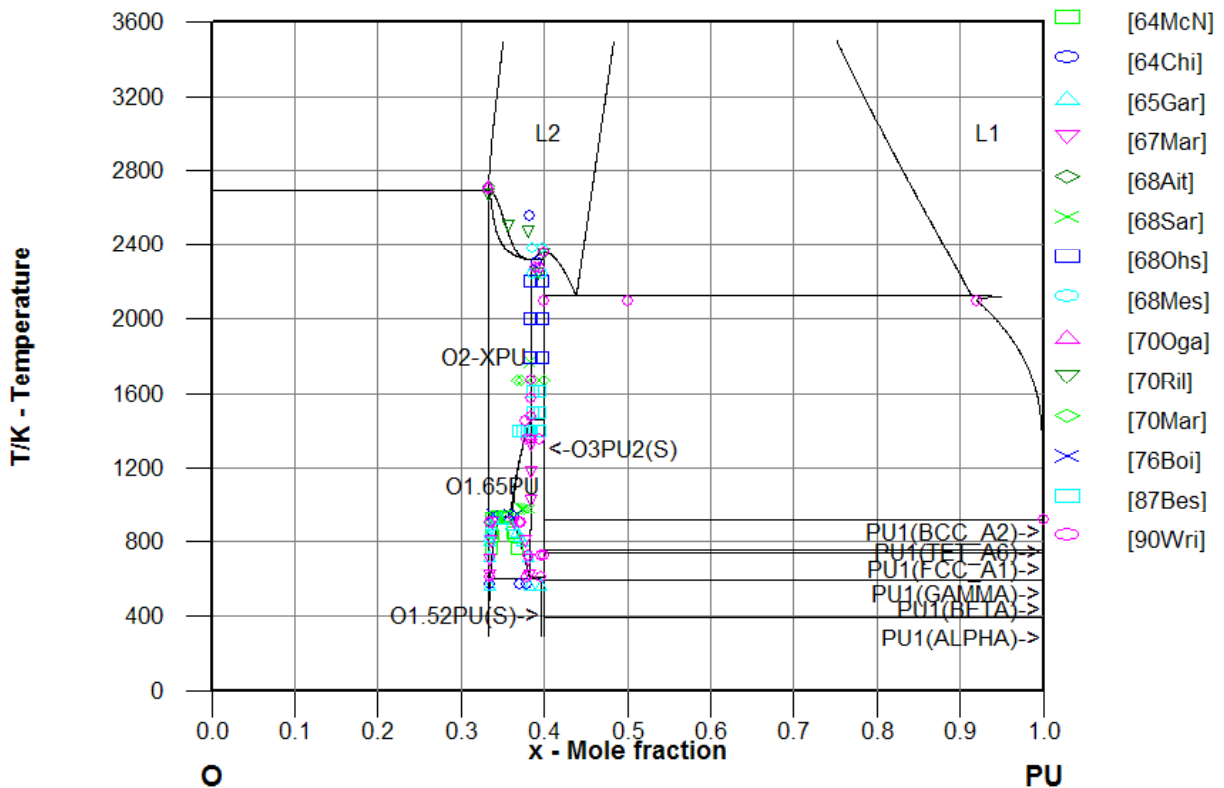
4.2. PSEUDO-BINARY SYSTEMS.

O₂Pu-O₂U,
O₂Pu-O₂Zr,
O₂U-O₂Zr.

4.3. TERNARY SYSTEMS.

O-Pu-U,
O-Pu-Zr,
O-U-Zr.
Pu-U-Zr

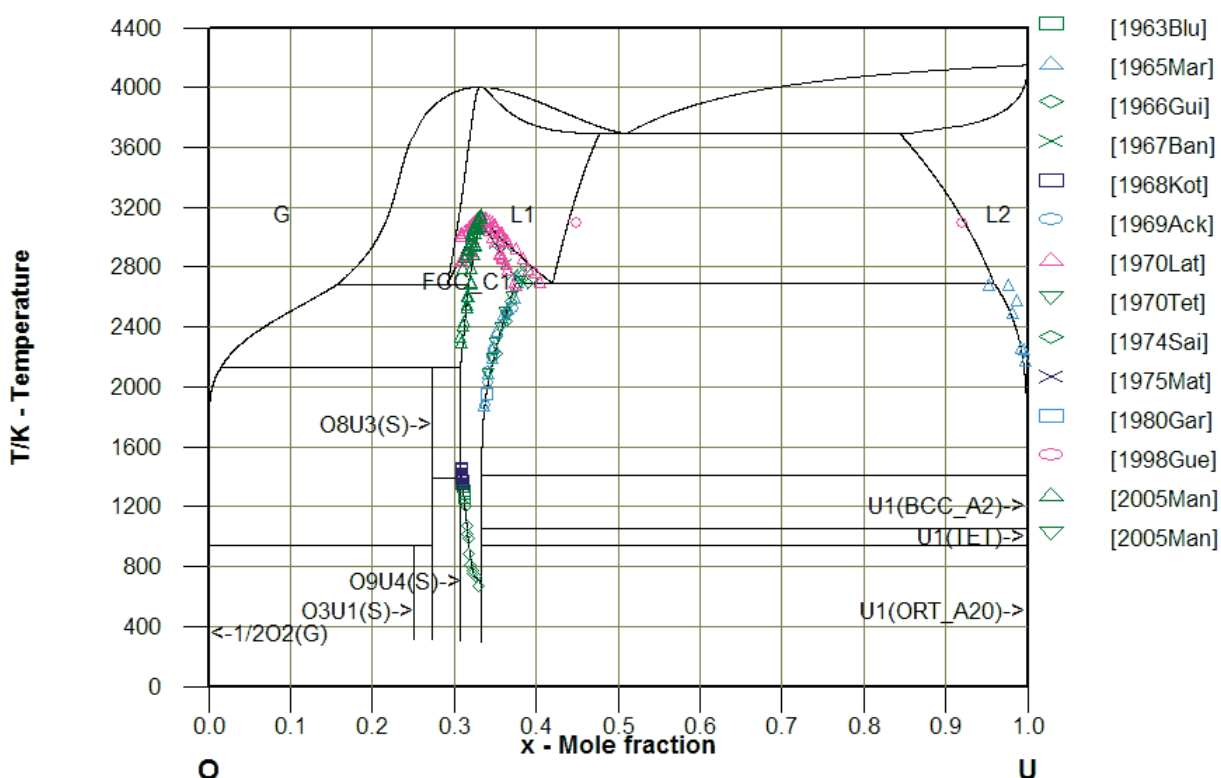
O-Pu



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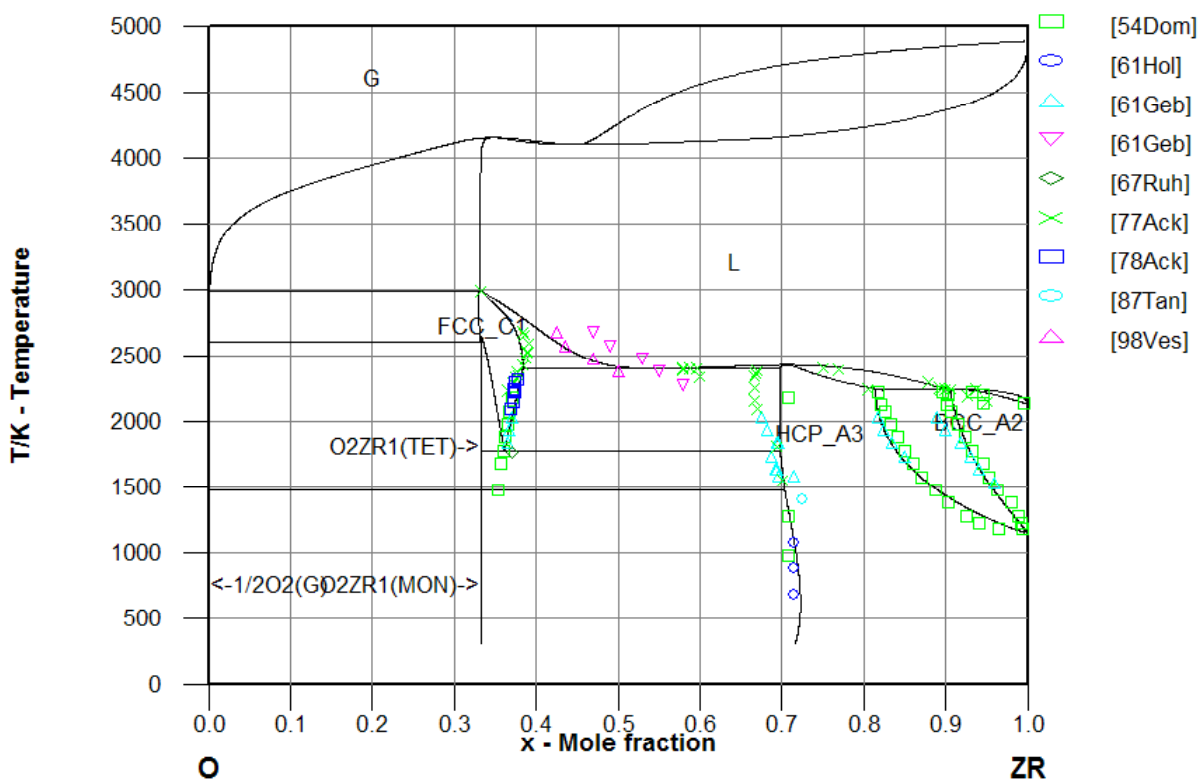
O-U



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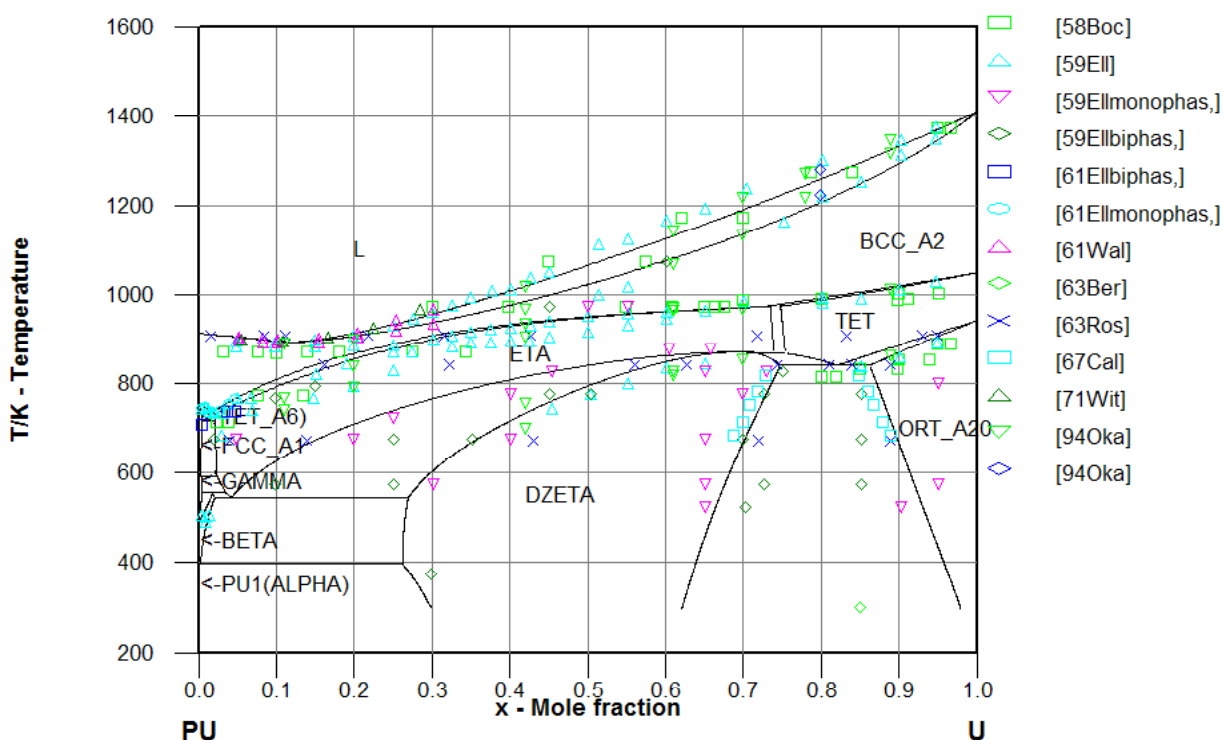
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O-Zr



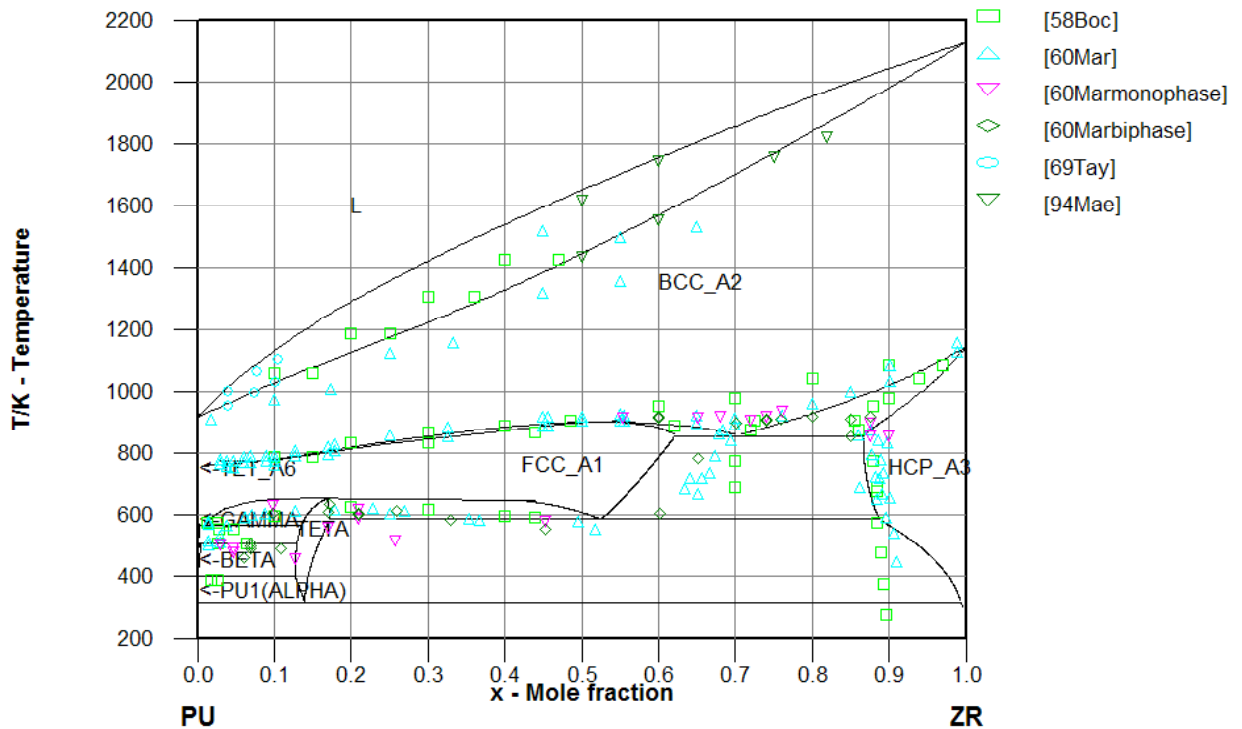
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Pu-U



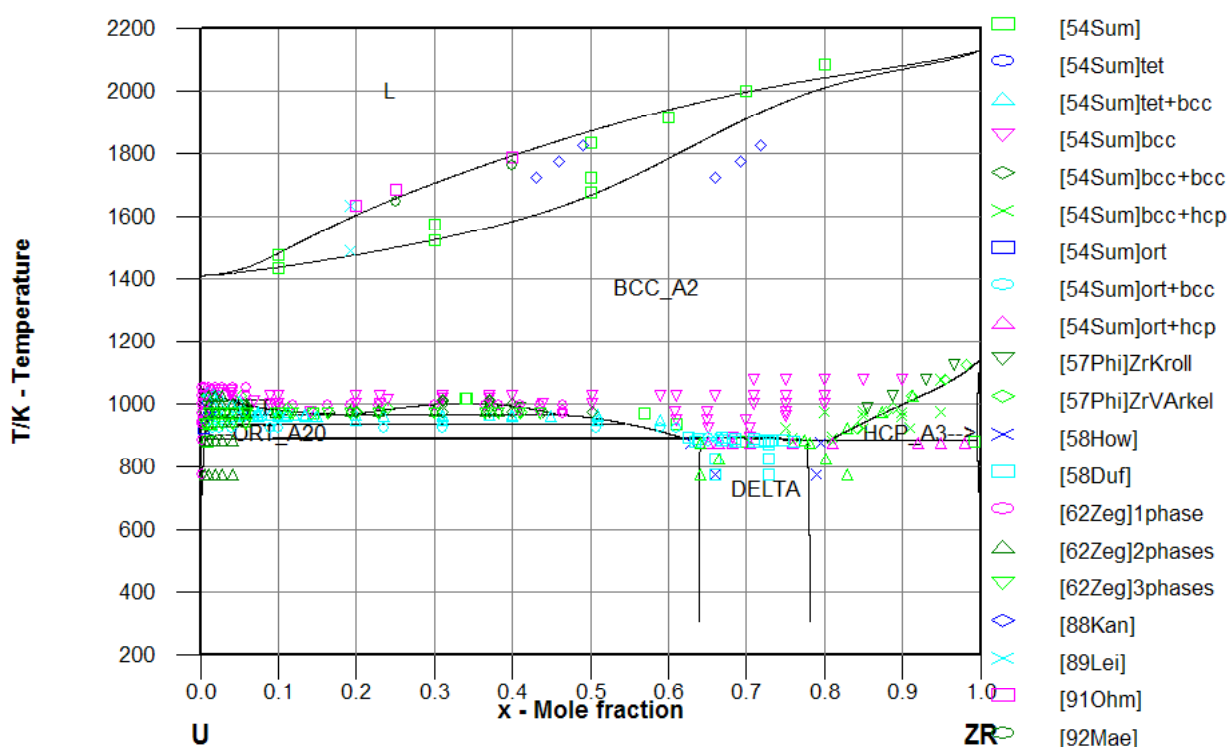
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Pu-Zr



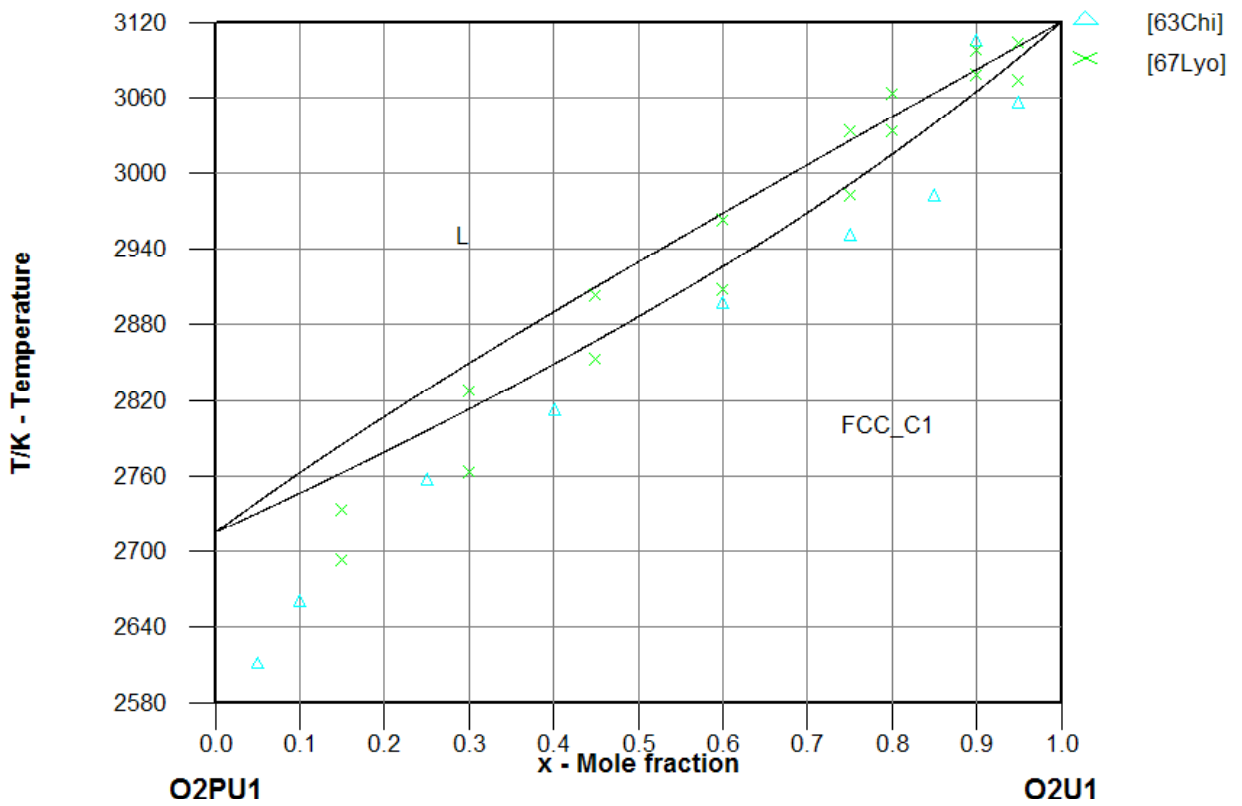
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U-Zr

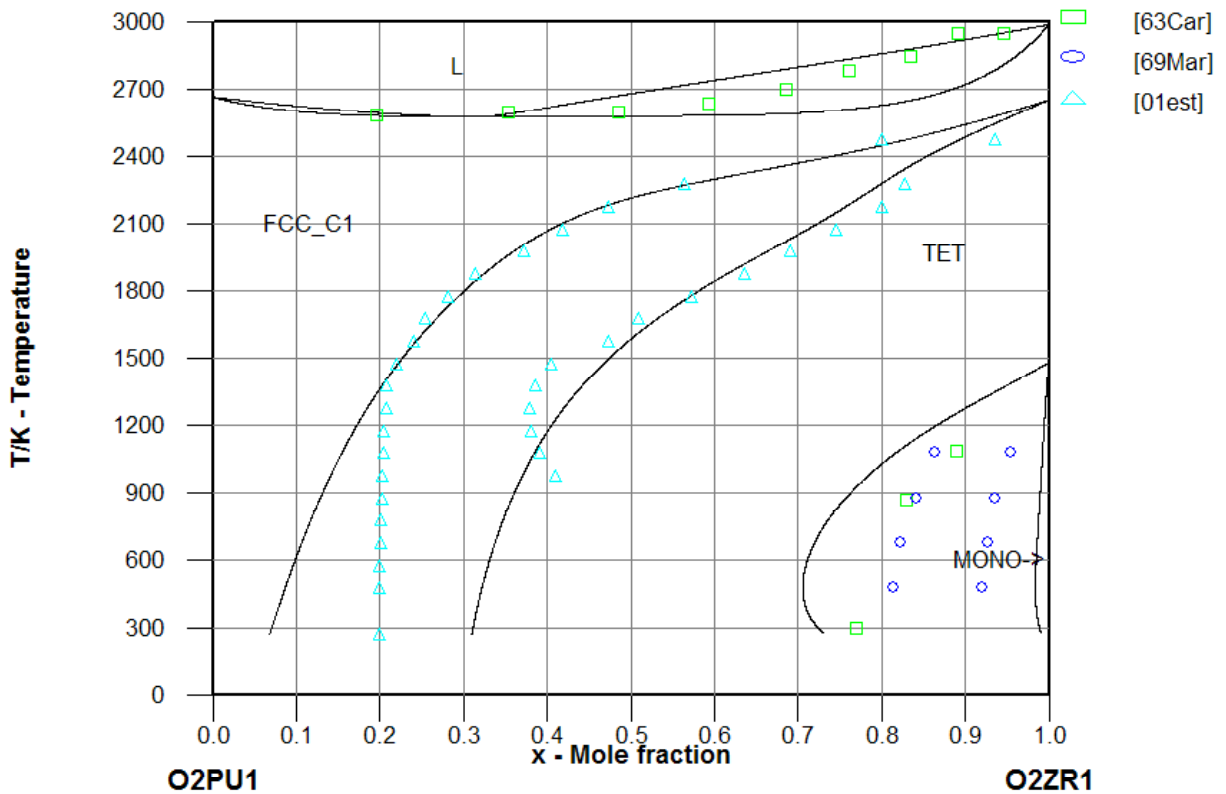


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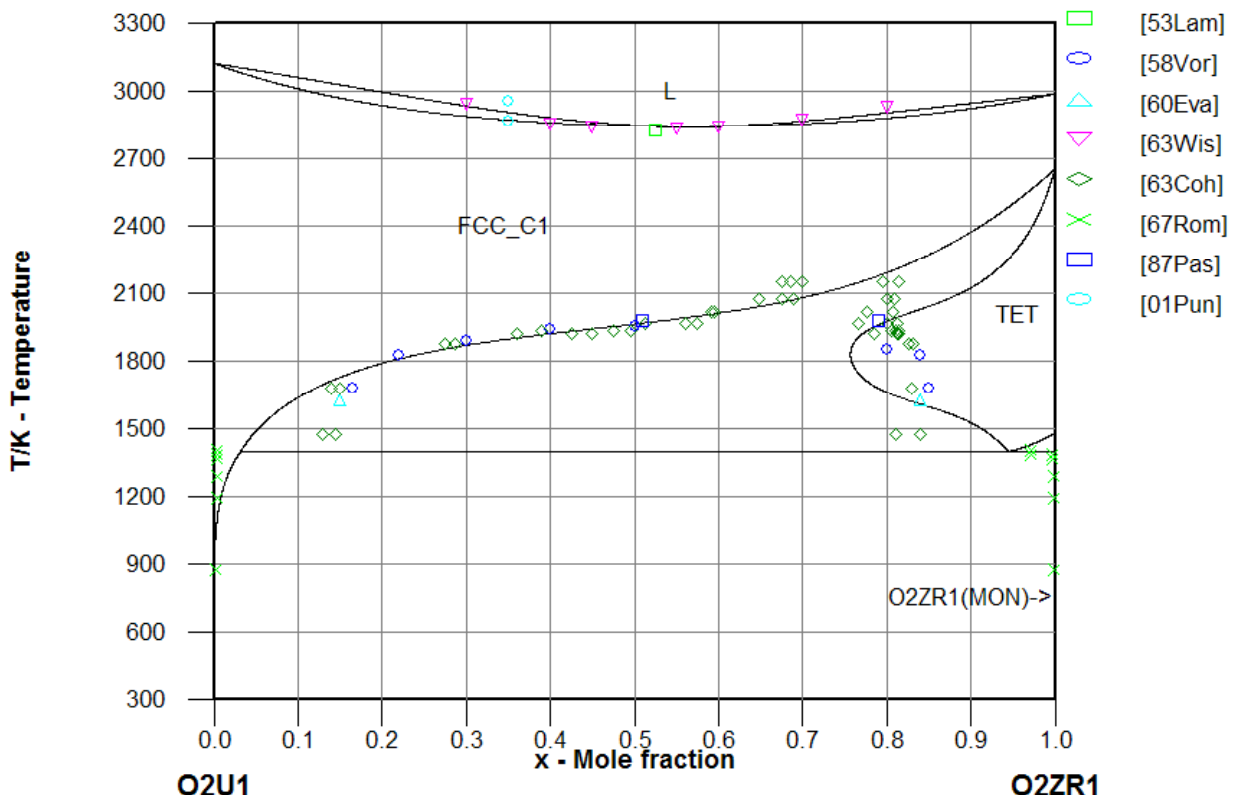
O₂Pu-O₂U



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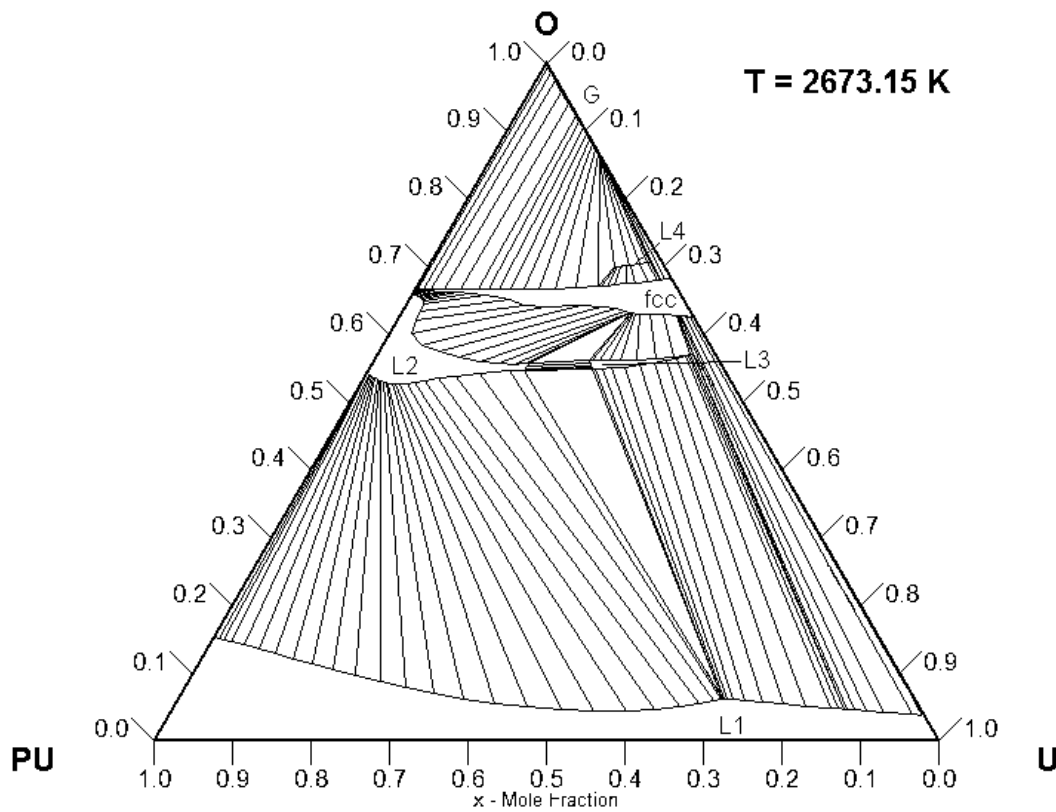
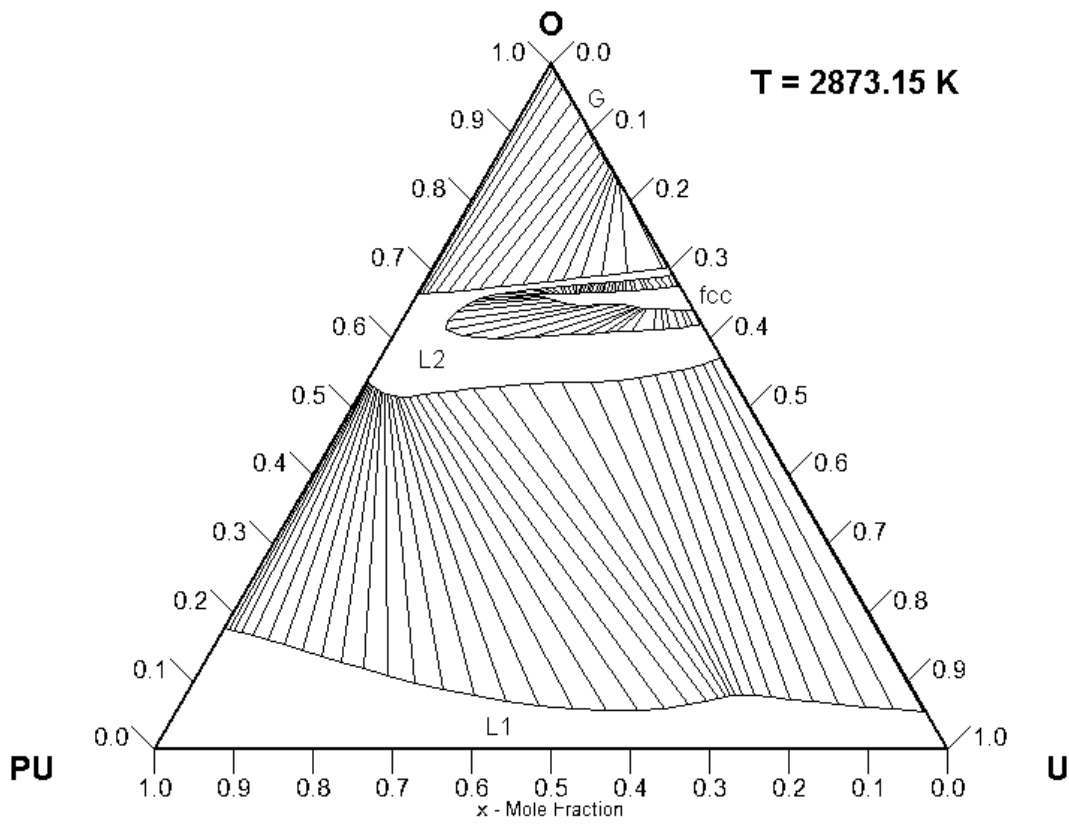
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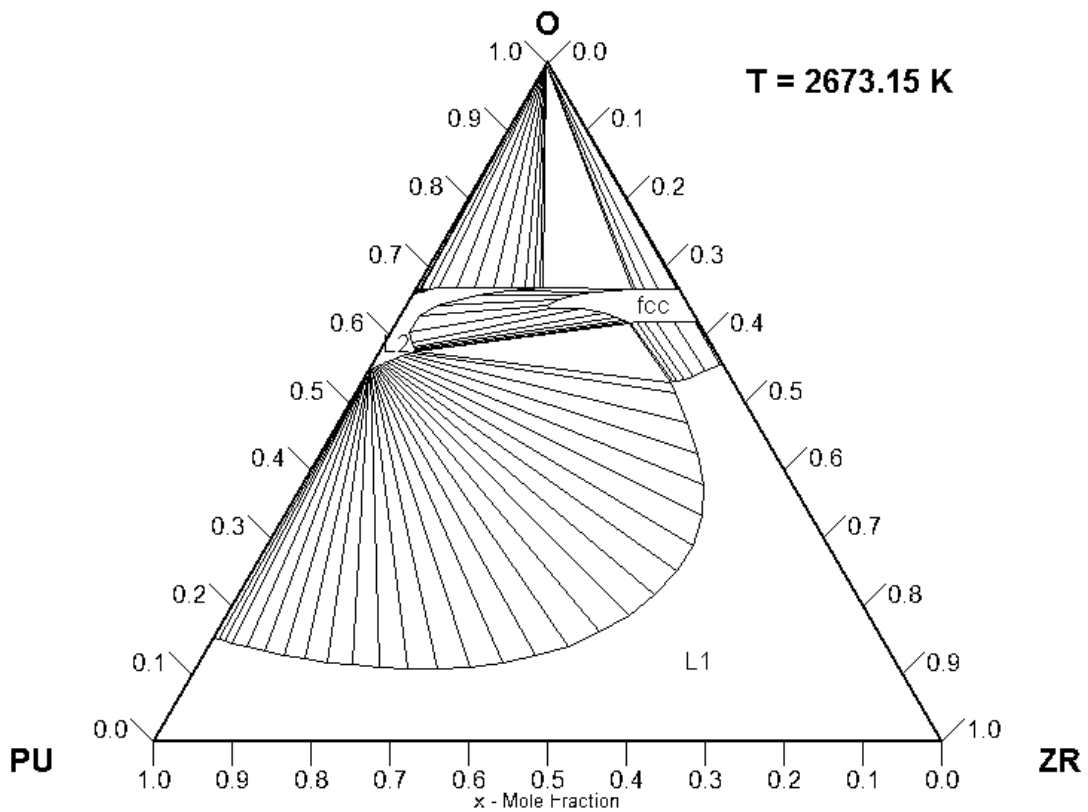
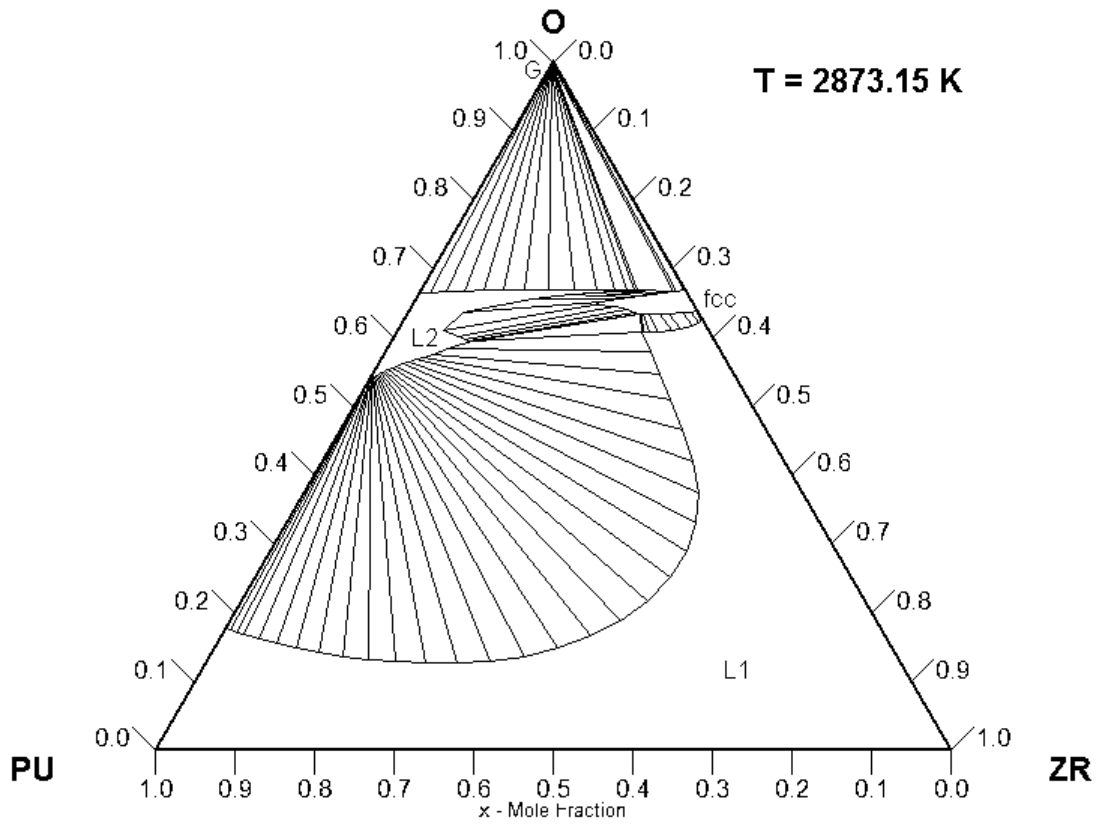
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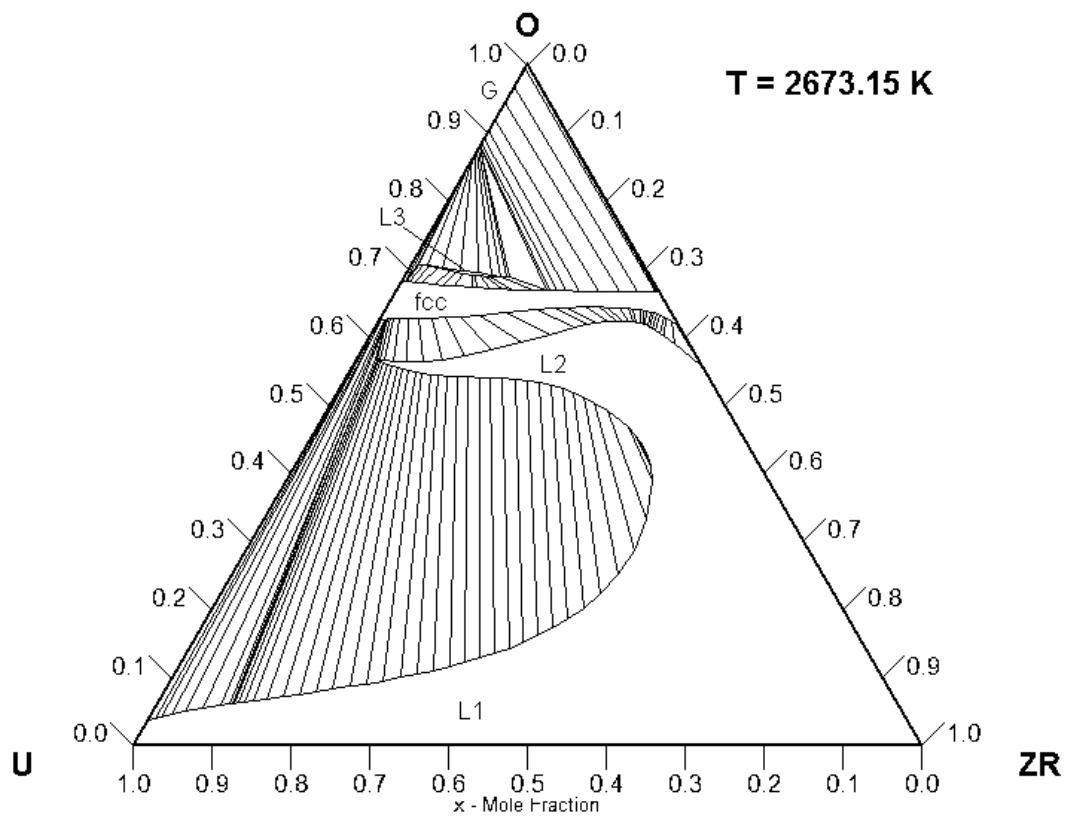
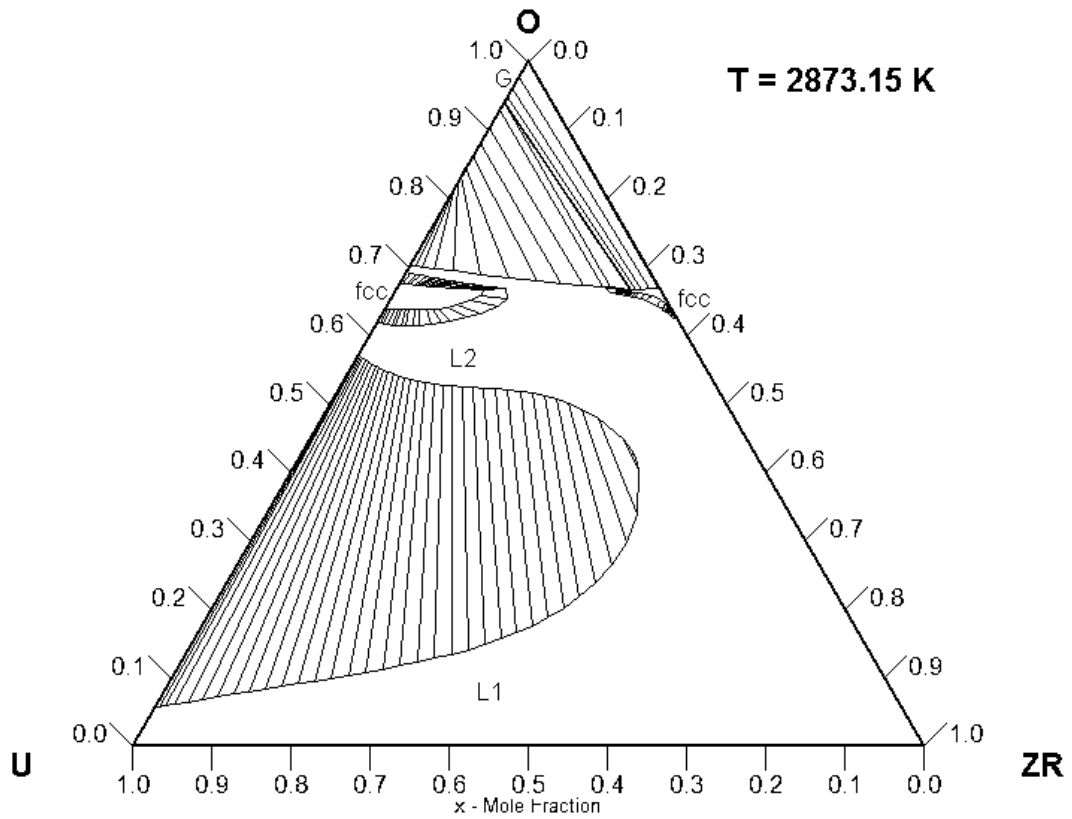
O-Pu-U



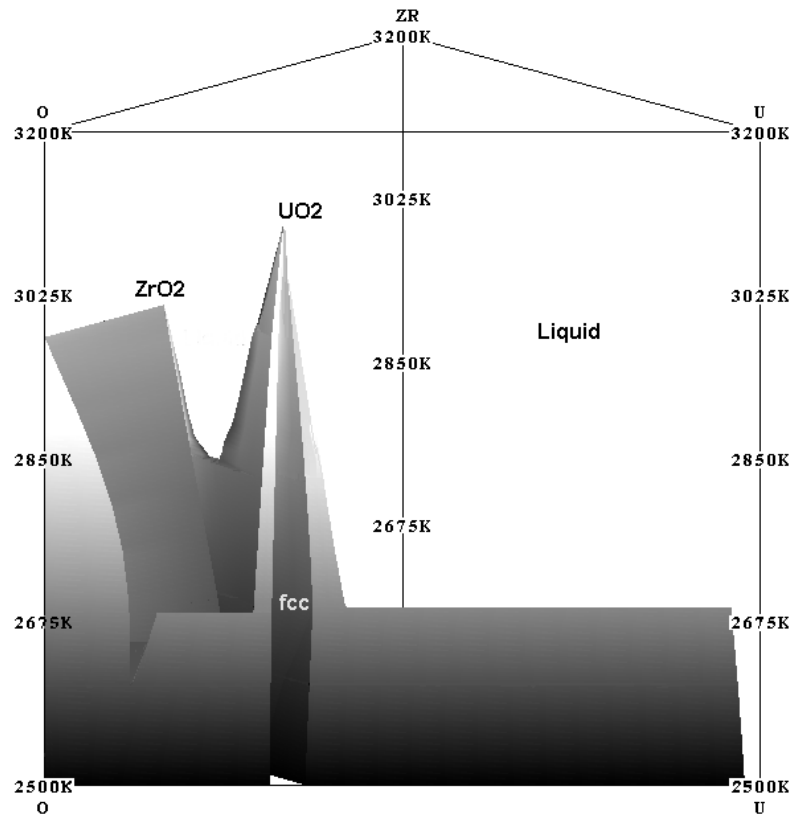
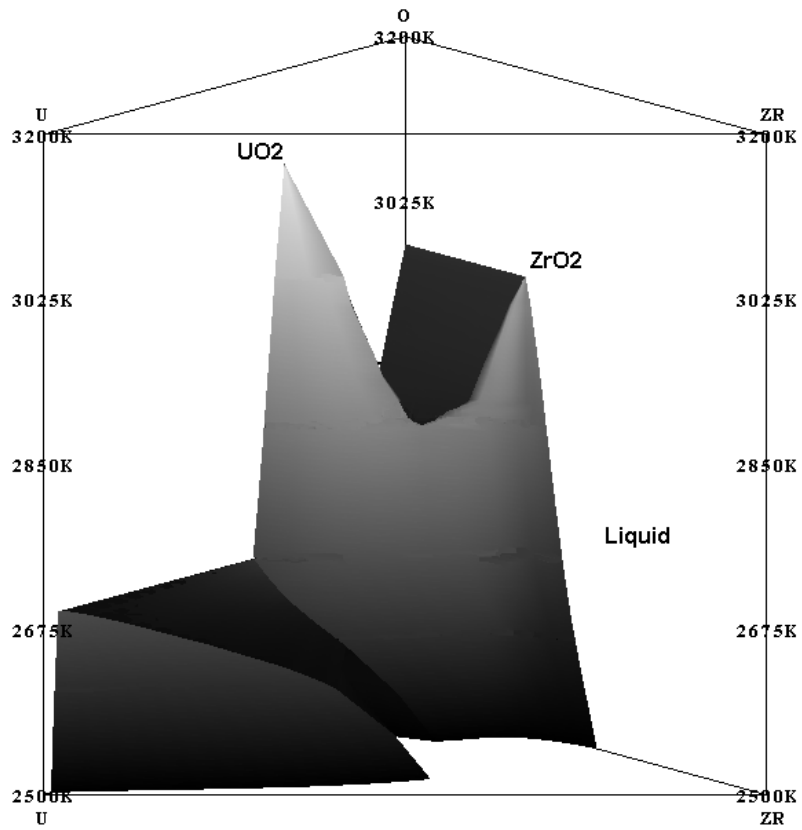
O-Pu-Zr



O-U-Zr



O-U-Zr - Liquidus



Pu-U-Zr

