Oxygen – Plutonium – Zirconium

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Introduction

The O-Pu-Zr system is a key system in the nuclear waste and fuel applications where zirconium oxide is used to dilute the fissile ceramic material PuO_2 as ZrO_2 has a low thermal neutron absorption cross section and a high melting point. In addition, it is resistant to fission fragment, ion irradiation and neutron irradiation. The knowledge of the phase equilibria of zirconia forming solid solutions with lanthanides and actinides is of great importance in fuel systems and plutonium disposition. However, there exists very limited experimental information about systems containing plutonium oxides. Experimental investigations on the present system have been carried out by [1963Car], [1969Mar] and [2001Ser]. This is summarized in Table 1.

Binary Systems

The binary boundary system O-Zr is accepted from the thermodynamic assessment of [1998Che] since it was used to calculate ternary O-Pu-Zr diagrams by [2003Kin], which are accepted in the present evaluation. [1999Kur] have performed thermodynamic modeling on the Pu-Zr binary boundary system taking into account the phase behavior and thermodynamic data after performing a critical assessment of the available literature. The calculated results are in very good accordance with experimental data and the Pu-Zr system has been accepted from [1999Kur]. This is shown in Fig. 1. Despite a number of studies, the phase equilibrium in the O-Pu system is not well established. After [Mas2], a Calphad assessment of the O-Pu binary was carried out by [2003Kin] using thermodynamic modeling for the phase behavior and thermodynamic properties. The results are in very good agreement with experimental data in this assessment. This phase diagram is included in the O-Pu-U evaluation report in the present volume.

Solid Phases

The crystallographic data for the O-Pu-Zr phases and their ranges of stability are summarized in Table 2. With regards to the O-Zr system, both the allotropes of Zr show large solubilities of oxygen. ZrO_2 is stable up to a temperature of 2707°C with the cubic modification showing a homogeneity range towards oxygen deficiency formed by the vacancies of oxygen sublattice. The other two allotropes have no significant range of solubility. The amount of experimental data available on the Pu-Zr system is limited. PuO_2 and ZrO_2 are the major oxides formed in the respective systems.

Quasibinary Systems

[1963Car] proposed the first version of the PuO_2-ZrO_2 quasibinary phase diagram through experimental techniques but the solubility limit of PuO_2 in ZrO_2 was arbitrarily assigned. Moreover, they only considered two allotropes of ZrO_2. [1969Mar] experimentally determined the quasi binary system in the range ($1.61 < O/M < 2$). They observed that in the fully oxidized state, the solubility of PuO_2 in βZrO_2 passes through a maximum at about 1000°C and then decreases rapidly. In addition, at high temperatures, βZrO_2 is stabilized over a wide range of PuO_2 contents. But there still remains an uncertain region. The phase relationships in the low PuO_2 region were examined by [2001Ser]. They have concluded from their experiments the presence of a eutectoid point in the composition range of less than 3.1 mol% PuO_2. [2003Kin] have calculated the phase diagram for PuO_2-ZrO_2 quasibinary system and presented a consistent thermodynamic database for the O-Pu-Zr system. The resulting thermodynamic dataset reproduces experimental data in the best possible way. Therefore, the calculated quasibinary diagram is accepted here, Figs. 2a and 2b. This diagram differs from the experimental investigations of [1969Mar] but reflects the one suggested by [2001Ser]. The calculated diagram also shows similarity with the UO_2-ZrO_2 and CeO_2-ZrO_2 systems as should be the case because all of these phases have the same crystal structure.

Isothermal Sections

No experimental data for the ternary system are known besides the quasibinary section PuO_2-ZrO_2. [2003Kin] have performed thermodynamic modeling to assess the phase relation in the O-Pu-Zr system. In the first stage, they attempted a preliminary thermodynamic modeling for the O-Pu system with thermodynamic data and phase diagram information available in the literature. The assessed data reproduced the general feature of the system with respect to phase diagram, oxygen potential and heat capacity. In the second stage, a possible set of phase diagrams for the O-Pu-Zr system was calculated using obtained data for the O-Pu system together with those for the O-Zr [1998Che] and Pu-Zr [1999Kur] subsystems available in the literature. In general, the ternary system starts forming oxides when the oxygen composition in the system increases. Moreover, plutonium tends to oxidize more readily than zirconium as is concluded by the presence of Pu_2O_3 rather than ZrO_2 at the calculated ternary sections. All the accepted diagrams are internally consistent as they are based upon the same database for calculation. These are shown in Figs. 3-5. However it should be noted that the (αZr)′+(αZr)′′+Pu_2O_3 three-phase equilibrium in Fig. 3 seems to be doubtful considering the shape of the (αZr) field, which is shifted very close to the binary sides. Therefore tie-lines (αZr)′ - (αZr)′′ are shown by dashed lines in Fig. 3.

Notes on Materials Properties and Applications

[2004Gib] have synthesized small ternary oxide clusters of zirconium and plutonium oxide in order to study plutonium chemistry. In other cases, zirconia based ceramics are used in high temperature applications such as engine components due to their low thermal conductivity and high fracture toughness [2002Arr]. The O-Zr system is also important in solid oxide fuel cells. [1965Far1, 1965Far2, 1965Far3, 1965Far4, 1966Bar1, 1966Bar2, 1966Far1, 1966Far2, 1967Far1, 1967Far2, 1968Far1, 1968Far2, 1968Far3, 1968Far4, 1968Far5, 1969Fac1, 1969Fac2, 1969Far] undertook a review of mechanical properties, method of fabrication and irradiation influence on plutonium oxides.

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Table 1: Investigations of the O-Pu-Zr Phase Relations, Structures and Thermodynamics

Reference	Method/Experimental Technique	Temperature/Composition/Phase Range Studied
[1963Car]	X-ray diffraction Metallography	1550°C PuO_2-ZrO_2
[1967Ack]	X-ray diffraction, metallography examination, emf	up to 1000°C from 64 to 73 at.% O and 40 to 33 at.% Pu
[1969Mar]	X-ray powder diffraction dilatometry	1500°C to 1900°C
[2001Ser]	High temperature X-ray diffraction	1000°C to 1200°C 3.1 to 11.2 mol% PuO_2

Table 2: Crystallographic Data of Solid Phases

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
(αPu) < 125	*mP*16 $P2_1/m$ αPu	*a* = 618.30 *b* = 482.20 *c* = 1096.3	[V-C2]
(βPu) 215 - 125	*mC*34 *C*2/*m* βPu	*a* = 1183.0 *b* = 1044.9 *c* = 922.70	[V-C2]
(γPu) 320 - 215	*oF*8 *Fddd* γPu	*a* = 315.87 *b* = 576.82 *c* = 1016.2	[V-C2]
(δPu) 463 - 320	*cF*4 $Fm\bar{3}m$ Cu	*a* = 463.47	[V-C2]
(δ'Pu) 483 - 463	*tI*2 *I*4/*mmm* In	*a* = 333.90 *c* = 444.67	[V-C2]
(εPu,βZr)	*cI*2 $Im\bar{3}m$		
(εPu) 640 - 483	W	*a* = 365.70	[V-C2]
(βZr) 1855 - 863		*a* = 356.80	[Mas2]
(αZr) < 863	*hP*2 $P6_3/mmc$ Mg	*a* = 323.17 *c* = 514.83	[V-C2] [Mas2]
Pu_2O_3 < 2020	*hP*5 $P\bar{3}m1$ La_2O_3	*a* = 383.88 *c* = 595.94	lattice parameters from [V-C2] existence of an ordered superstructure [1967Ack]
αZrO_2 < 1205	*mP*12 $P2_1/c$ $CoSb_2$	*a* = 515.05 *b* = 521.16 *c* = 531.73	[V-C2] [Mas2]
βZrO_2 2377 - 1205	*tP*6 $P4_2/mnm$ TiO_2	*a* = 529.28 *c* = 365.26	[V-C2] [Mas2]
γ, $(Pu,Zr)O_2$	*cF*12 $Fm\bar{3}m$		continuous solid solution
PuO_2 < 2467	CaF_2	*a* = 539.7	62.8 to 66.7 at.% O [2003Kin] lattice parameters [V-C2]
γZrO_2 2710 - 2377		*a* = 509.00	[V-C2] [Mas2]

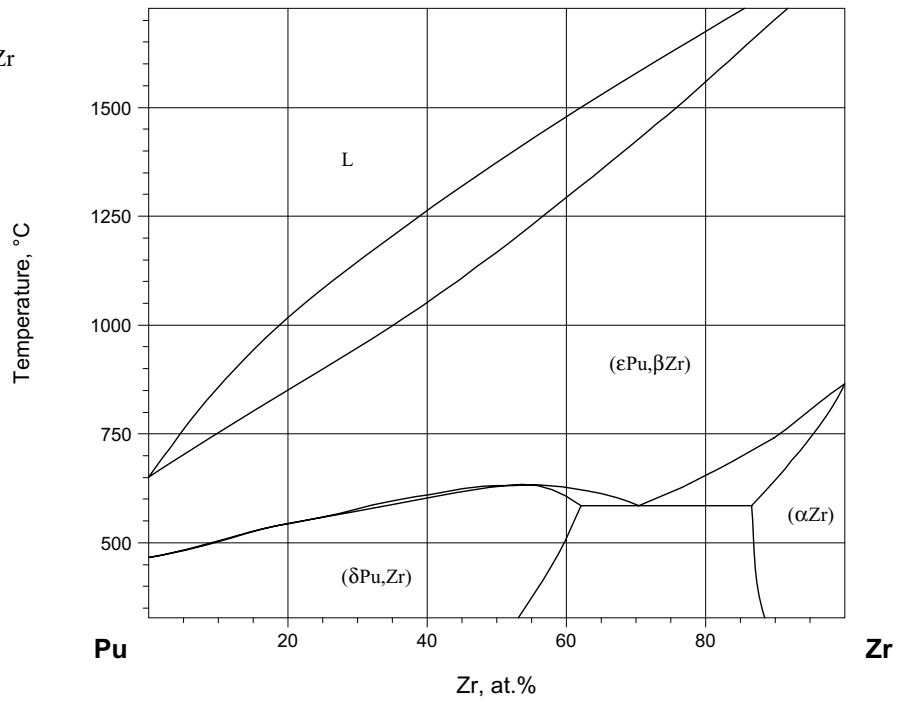

Fig. 1: O-Pu-Zr. The calculated Pu-Zr equilibrium phase diagram

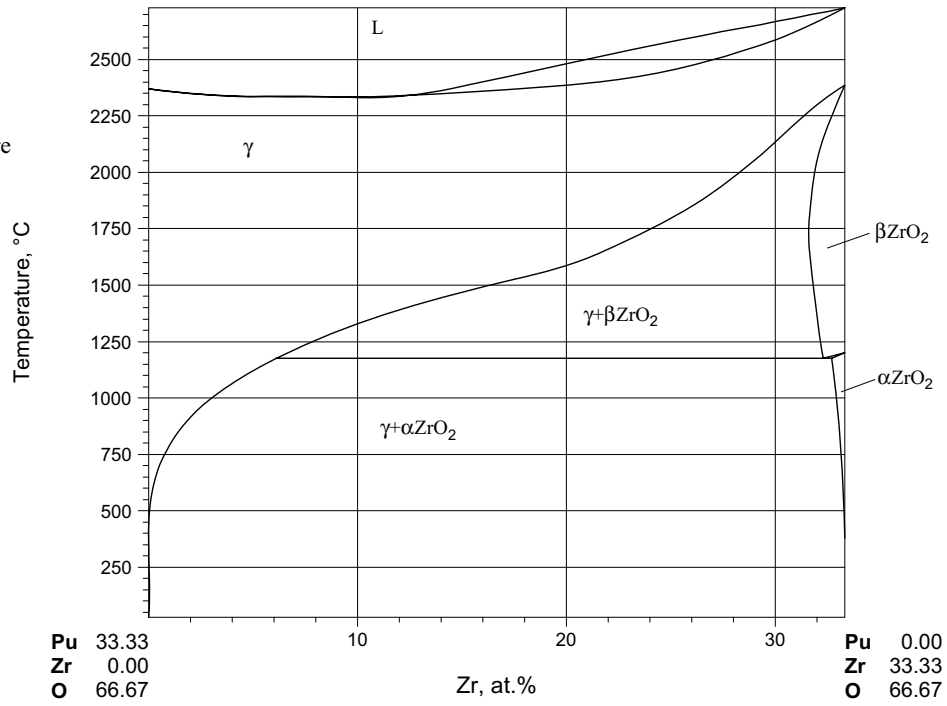

Fig. 2a: O-Pu-Zr. The calculated PuO_2-ZrO_2 quasibinary phase diagram in the entire composition range

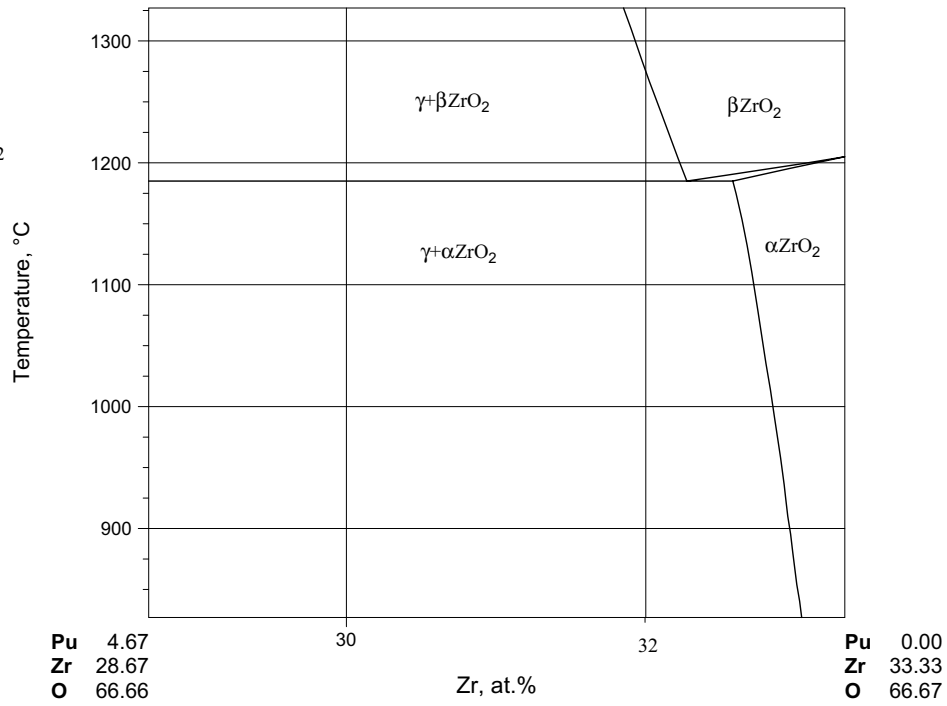

Fig. 2b: O-Pu-Zr. The calculated PuO_2-ZrO_2 quasibinary phase diagram at the ZrO_2 rich end; enlarged part of Fig. 2a

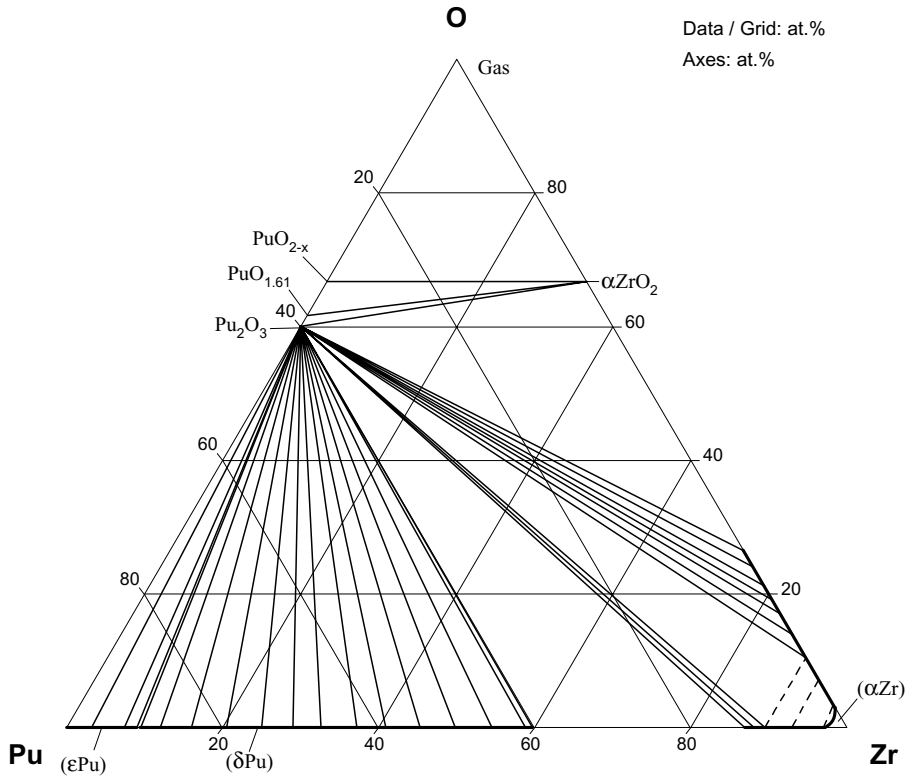

Fig. 3: O-Pu-Zr. The calculated isothermal section 500°C

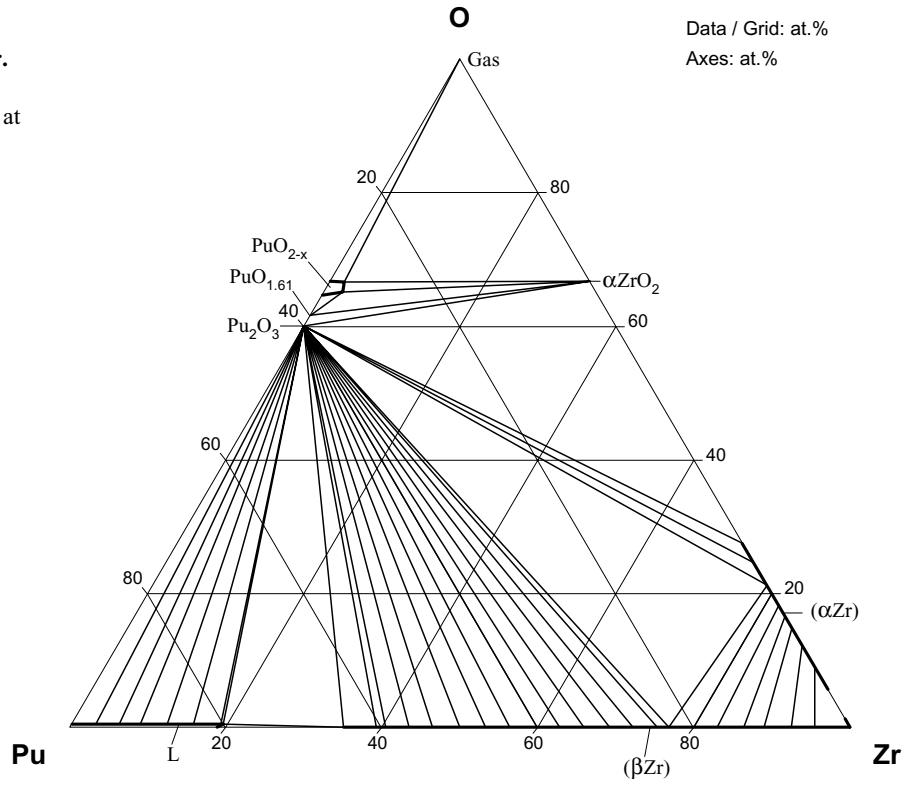

Fig. 4: O-Pu-Zr. The calculated isothermal section at 1000°C

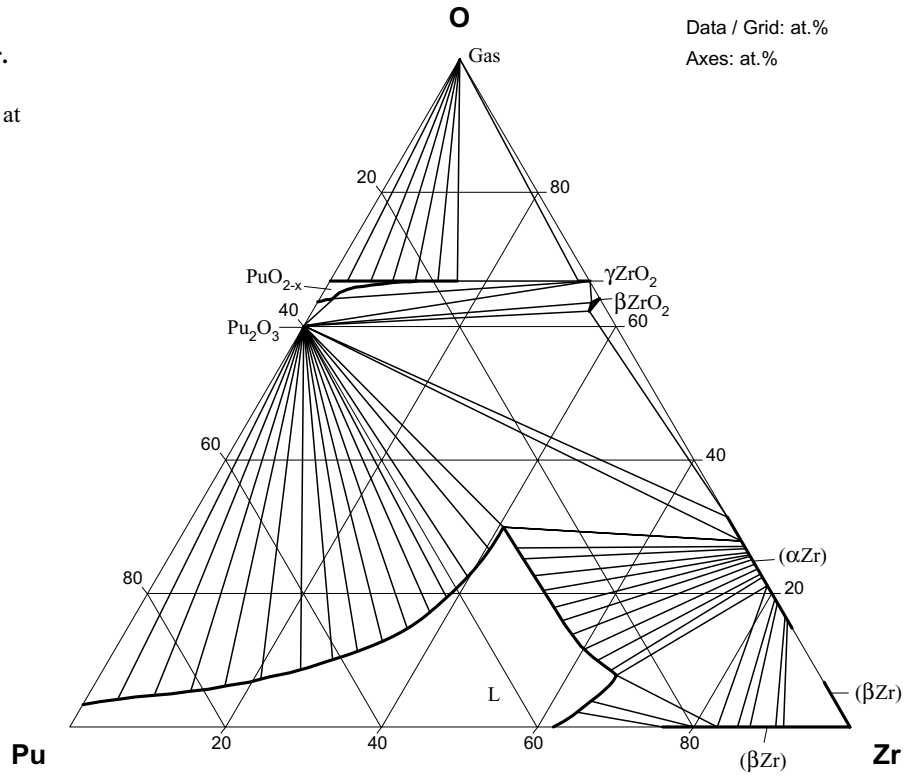

Fig. 5: O-Pu-Zr. The calculated isothermal section at 1500°C