Aluminium – Iron – Uranium

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Introduction

Petzow *et al.* [1962Pet] as well as Lam *et al.* [1962Lam] investigated the quasibinary section UFe_2-UAl_2. [1963Pet] studied the partial ternary system U-UFe_2-UAl_2 in detail. 18 alloys for the quasibinary section and 84 ternary alloys were produced by arc melting from the pure elements (Al:99.99%, Armco-Fe, nuclear pure U) and studied by chemical, X-ray, microhardness, density, thermal and differential thermal (DTA) analyses and metallography. U and Fe were prealloyed. Samples were annealed at 1000°C (8 d), 800°C (16 d) and 600°C (24 d) and quenched in water.
[1963Kha, 1963Rus, 1964Dix] and [1966Sch] studied the solubilities of Al and Fe in the three solid modifications of pure U.
The homogeneity ranges and structures of the phases along the UAl_2-UFe_2 section were investigated by [1963Pet, 1964Ste, 1967Lam, 1971Kim].
In the U poor system structure and lattice parameters of a compound UFe_4Al_8 were determined by [1984Bar, 1984Ste]. In the following years this phase was investigated intensively for its exceptional magnetic properties using magnetic measurements, neutron diffraction at polycrystalline and single crystalline samples by [1986Bar, 1988Pta, 1989Sch, 1992Sus, 1994Gon, 1995God, 1995Gon, 1996God, 1996Wys, 1997Pai, 1997Rec, 1998Gon, 1999Kuz, 2000Car, 2000Rec, 2001Rec, 2001Wae, 2002Car, 2002Gon, 2002Rec, 2003Gon, 2003Li, 2003Tal]. It has an extended homogeneity range along Al-Fe exchange, UFe_xAl_{1-x} ($3 < x < 7$ at 850°C [1994Gon]). At high pressure, up to 26 GPa at composition UFe_5Al_7 the phase was found to remain stable [2002Hal]. Later several other phases were detected in the region with less than 33 at.% U, most of them in a systematic investigation of the 850°C isothermal section by [2005Gon]: $U_2Fe_{12}Al_5$ [1995Che], UFe_2Al_{10} [2002Mes, 2004Mes, 2004Noe], $U_2Fe_{3.6}Al_{12.4}$ [2005Gon], $U_3Fe_4Al_{12}$ [2005Gon] and $U_2Fe_xAl_{17-x}$ ($6.7 < x < 8.6$) [2005Gon]. Near the Al corner [2005Mes] found a further phase, U_2FeAl_{20}. No investigations were performed on temperature ranges of stability or on invariant reactions between these U poor phases.
The important experimental investigations are summarized in Table 1.

Binary Systems

[1963Pet] based the ternary investigation on the binaries U-UAl_2 and U-UFe_2 published by [H] with little corrections of the U transition temperatures ($l \rightarrow \gamma$: 776°C, $\beta \rightarrow \alpha$: 668°C) and the melting point of UAl_2 (1620°C). These are virtually equivalent to those of [Mas2], given in more detail by [1990Kas] and [1993Oka], respectively. In the binary systems UAl_2 and UFe_2 show no homogeneity ranges. UAl_4 was assumed to have some homogeneity range. However, in a very detailed comparison of samples with excess or deficient Al content [2004Tou] concluded from powder and single crystal X-ray diffraction measurements this phase to be strictly stoichiometric. As a third boundary system, the section UFe_2-UAl_2 after [1962Pet] (Fig. 1) was used by [1963Pet]. For the Al-Fe binary system the assessment adopted by [Mas2] was accepted, which is given in more detail by [1993Kat]. Complete thermodynamic datasets are assessed for the Al-Fe [1998Sei] and Fe-U [2003Cha] systems. For Al-U a partial dataset is given by [1990Kas].

Solid Phases

The solubilities of Fe and Al in the pure modifications of U were investigated by [1963Kha, 1963Rus, 1964Dix, 1964Rus, 1966Sch].
The maximum mutual solubility of the cubic Laves phases UFe_2 and UAl_2 is 19 mol% at 1000°C on both sides [1963Pet]. [1967Lam] gives solubilities of 27 mol% UFe_2 in UAl_2 and 15 mol% UAl_2 in UFe_2. The lattice parameters deviate from Vegards' law to smaller values. A ternary $MgZn_2$ type Laves phase

$U(Fe,Al)_2$ has been found between UFe_2 and UAl_2, ranging from 22 to 39.67 at.% Al [1964Ste]. It was later characterized to have an ordering of Fe and Al atoms on the different Wyckoff positions of this structure type (Mg_2Cu_3Si type [1971Kim]).
Another phase in the UFe_2-UAl_2 quasibinary system has a well-defined composition corresponding to the stoichiometric formula UAlFe; it is stable below 700°C [1967Lam]. Its structure is the Fe_2P-type, but with Al and Fe ordered on the two different Wyckoff positions of P-sites (ZrNiAl type) [1986And]. A phase called UFe_4Al_8 was first studied by X-ray diffraction [1984Ste, 1984Bar]. It has an extended homogeneity range along Al-Fe interchange, UFe_xAl_{12-x} ($3 < x < 7$ [2005Gon]). Up to the composition UFe_4Al_8, the Fe atoms are restricted to the 8f position of the $ThMn_{12}$ type structure. Until $x = 6$ further Fe atoms occupy preferably the 8j position, whereas for $x > 6$ also some Fe atoms go to the 8i position [1995Gon, 2005Gon]. Like other phases of the $ThMn_{12}$ type it is ferromagnetic and exhibits very large magnetic anisotropy with the hard magnetization along the c-axis. By spectroscopic investigation [2003Tal] found domination of the U 5f states at the Fermi level.
Five more ternary phases were identified to be stable at 850°C [2005Gon]. The crystal structure is known for all of them. An additional phase, U_2FeAl_{20} was reported without investigation of its stability range or details of its crystal structure [2005Mes].
The crystallographic data of all solid phases are summarized in Table 2.

Quasibinary Systems

The section UFe_2-UAl_2 was established to be quasibinary by [1962Pet, 1963Pet] and [1967Lam] (Fig. 1). [1967Lam] corrected their own results of 1962 [1962Lam]. [1967Lam] found additionally a ternary stoichiometric phase UFeAl. Above 800°C the phase relationships of [1962Pet] and [1967Lam] differ somewhat in the stability ranges of the phases. The major difference is at approximately 700°C, where UFeAl is formed peritectoidally and a concomitant narrowing of the $U(Fe,Al)_2$ phase field results. In Fig. 1 the section UFe_2-UAl_2 is mainly taken as published by [1962Pet], completed by the ternary compound UAlFe published by [1967Lam].

Invariant Equilibria

Five invariant four-phase equilibria are reported in the region U-UFe_2-UAl_2 [1963Pet] and listed with the phase compositions in Table 3. E_1 and E_2 are solid phase reactions resulting from the polymorphy of uranium. In Fig. 2 the reaction scheme is presented, mainly taken from [1963Pet]. The temperatures of E_1 and E_2 are based on small effects in the DTA curves and are not established exactly by [1963Pet] nor are the concentrations of the participating solid phases. At temperatures below the formation temperature of the stoichiometric phase UFeAl (700 ± 10°C) the phase equilibria given by [1963Pet] had to be corrected by adding equilibria with the phase UFeAl. As all three phases UAl_2, UFeAl and $U(Fe,Al)_2$ are restricted to the line with 33.3 at.% U, thus phase U_6Fe cannot participate in the four-phase reaction D_1: $UAl_2 + U(Fe,Al)_2 \rightleftharpoons UFeAl, U_6Fe$, which therefore is degenerate.

Liquidus Surface

Figure 3 shows the isotherms of the partial U-UFe_2-UAl_2 liquidus surface and the melting grooves separating five areas of primary crystallization: (γU), UAl_2, $U(Fe,Al)_2$, UFe_2 and U_6Fe after [1963Pet]. For the part with less than 33 at.% U no liquidus data are published.

Isothermal Sections

Figures 4 and 6 to 8 show isothermal sections of the U-UFe_2-UAl_2 partial system after [1963Pet] at 1000, 800, 700 and 600°C, respectively. Figure 5 presents the isothermal section at 850°C of the whole system. This is the only temperature, for which equilibria between phases with less than 33.3 at.% U are reported. The U rich part of Fig. 5 is based on [1963Pet], the U poor part on [2005Gon]. Some three-phase equilibria were given incompletely by [2005Gon], they are tentatively completed in Fig. 5. In Fig. 8 the stoichiometric ternary phase UFeAl is implemented into the diagram of [1963Pet]. In the binary systems the solubility

ranges of UFe_2 and UAl_2 are very small, therefore small deviations from exactly 33.3 at.% U, as drawn by [1963Pet] in the ternary system, are very improbable and omitted in Figs. 4 to 8.

Temperature – Composition Sections

Six isopleths (Figs. 9 to 14) at constant contents of 5, 10, 20 at.% Al, 25 at.% Fe, 40 and 60 at.% U are given after [1963Pet]. [1963Kha] published isopleths in the U corner, three at constant Fe:Al ratios (3:1, 1:1, 1:3) up to an amount of Al+Fe of 5 at.% (Figs. 15, 16, 17), one at constant 95 at.% U (Fig. 18) as well as a projection of the curves of double saturation of the (βU) and (γU) solid solutions (Fig. 19). The temperatures given by [1963Pet] and [1963Kha] differ slightly, but are kept in Figs. 9 to 19 as declared by the respective authors.

Notes on Materials Properties and Applications

The U rich phases were investigated as candidates for nuclear fuel materials. This may still be interesting for research reactors. In U with small amounts of Al and Fe ("adjusted uranium") small precipitates of UAl_2 or U_6Fe in (αU) diminish swelling [1964Dix].
Although its Curie temperature is near room temperature the phase UFe_xAl_{12-x} is of large theoretical interest to study the effect of f electrons (lanthanides and actinides) on magnetic structures and properties.

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Table 1: Key-Investigations of the Al-Fe-U Phase Equilibria and Crystal Structures

Reference	Method/Experimental Technique	Temperature/Composition/Phase Range Studied
[1962Pet] [1963Pet]	DTA, optical microscopy, X-ray diffraction	33-100 at.% U, 600-1150°C
[1962Lam] [1967Lam]	DTA, optical microscopy, X-ray diffraction	33.3 at.% U, 600-1100°C
[1963Kha]	Metallography of annealed and quenched samples	95-100 at.% U, 650-1050°C
[2005Gon]	scanning electron microprobe, X-ray powder diffraction	0-33 at.% U, 850°C

Table 2: Crystallographic Data of Solid Phases

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
γ, (γU)(h_2) 1132 - 772	*cI*2 *Im*$\bar{3}$*m* W	*a* = 352.4	[Mas2]
β, (βU)(h_1) 772 - 662	*tP*30 *P*4_2/*mnm* β$U(h_1)$	*a* = 1075.9 *c* = 565.6	[Mas2]
α, (αU)(r) < 662	*oC*4 *Cmcm* αU(r)	*a* = 285.37 *b* = 586.95 *c* = 495.48	[Mas2]
(γFe) 1390 - 910	*cF*4 *Fm*$\bar{3}$*m* Cu	*a* = 364.67	[Mas2]
(αFe) < 1535	*cI*2 *Im*$\bar{3}$*m* W	*a* = 286.65	[Mas2]
(Al)	*cF*4 *Fm*$\bar{3}$*m* Cu	*a* = 404.96	[Mas2]
$U(Fe_xAl_{1-x})_2$	*cF*24 *Fd*$\bar{3}$*m* $MgCu_2$		0 ≤ *x* ≤ 0.162 [V-C] maximal solubility of Fe (at 1000°C): 12.67 at.%
UAl_2		*a* = 776.6	[V-C2]
		a = 774.85	0 at.% Fe, quenched from 1000°C, (*x* = 0) [1964Ste]
		a = 769.6	5 at.% Fe, quenched from 1000°C, (*x* = 0.075) [1964Ste]
		a = 766.9	8.17 at.% Fe, quenched from 1000°C, (*x* = 0.122) [1964Ste]
		a = 759.5	10.87 at.% Fe, quenched from 1000°C, (*x* = 0.162) [1964Ste]
UAl_3	*cP*4 *Pm*$\bar{3}$*m* Cu_3Au	*a* = 426.5	[V-C2]
UAl_4	*oI*20 *Imma* UAl_4	*a* = 440.14 *b* = 625.52 *c* = 1372.79	[V-C2], lattice parameters [2004Tou]
U_6Fe < 805	*tI*28 *I*4/*mcm* U_6Mn	*a* = 1030.22 *c* = 523.86	[2005Gon]

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
$U(Fe_{1-x}Al_x)_2$	*cF*24 $Fd\bar{3}m$ $MgCu_2$		$0 \le x \le 0.19$ [V-C] maximal solubility of Al (at 1000°C): 12.7 at.%
UFe_2		a = 706.5	
		a = 704.7	0 at.% Al, 1000°C, (x = 0) [1964Ste]
		a = 706.96	5 at.% Al, 1000°C, (I = 0.05) [1964Ste]
		a = 711.8	12 at.% Al, 1000°C, (x = 0.18) [1964Ste]
Fe_4Al_5	*cI*16?	a = 598.0	at 61 at.% Al, [1993Kat]
$FeAl_2$ < 1156	*aP*18 *P*1 $FeAl_2$	a = 487.8 b = 646.1 c = 880.0 α = 91.75° β = 73.27° γ = 96.89°	at 66.9 at.% Al, solid solubility ranges from 65.5 to 67.0 at.% Al [1993Kat]
Fe_2Al_5 < 1169	*oC*24[a] *Cmcm* Fe_2Al_5	a = 765.59 b = 641.54 c = 421.84	at 71.5 at.% Al [1994Bur]
Fe_4Al_{13} < 1160	*mC*102 *C*2/*m* Fe_4Al_{13}	a = 1552.7 to 1548.7 b = 803.5 to 808.4 c = 1244.9 to 1248.8 β = 107.7 to 107.99°	solid solubility ranges from 74.5 to 75.5 at.% Al [1993Kat]
* $U(Fe_xAl_{1-x})_2$ < 1060	*hP*12 $P6_3/mmc$ $MgZn_2$	 a = 515 c = 798	$0.167 \le x \le 0.292$ (22 to 39.67 at.% Al) 41.67 at.% Fe, 25 at.% Al, 1000°C [1964Ste]
		a = 522 c = 816	30.67 at.% Fe, 36 at.% Al, 1000°C [1964Ste]
	(Mg_2Cu_3Si)		[1971Kim]
* UFeAl < 700	*hP*9 $P\bar{6}2m$ Fe_2P (ZrNiAl)	a = 667.2 c = 398.1	[1967Lam] water quenched from 675°C; [1986And]
* $U_3Fe_4Al_{12}$	*hP*38 $P6_3/mmc$ $Gd_3Ru_4Al_{12}$	a = 874.51 c = 925.88	[2005Gon]
* $U_2Fe_{12}Al_{15}$	*hP*38 $P6_3/mmc$ Th_2Ni_{17}	a = 856.31 c = 843.8	x = 12 [2005Gon]
* $U_2Fe_xAl_{17-x}$	*hR*57 $R\bar{3}m$ Th_2Zn_{17}	 a = 875.3 c = 1265.8	$6.7 < x < 8.6$ at 850°C [2005Gon] *a, c* at x = 8

Phase/ Temperature Range [°C]	Pearson Symbol/ Space Group/ Prototype	Lattice Parameters [pm]	Comments/References
* $U_2Fe_{3.6}Al_{13.4-x}$	*hP*38 $P6_3/mmc$ Th_2Ni_{17}	a = 885.89 c = 898.24	x = 3.6 [2005Gon]
* UFe_xAl_{12-x}	*tI*26 *I*4/*mmm* $ThMn_{12}$	 a = 874.9 c = 503.6	$3 < x < 7$ [2005Gon] a, c at x = 4, room temperature [1984Ste]
* UFe_2Al_{10}	*oC*52 *Cmcn* $YbFe_2Al_{10}$	a = 891.46 b = 1019.86 c = 901.14 a = 891.92 b = 1020.78 c = 901.83	[2004Noe] [2004Mes]
* U_2FeAl_{20}	*tI** *I*42*m*	a = 1240 c = 1030	[2005Mes]

[a] 12 Al-positions of the 24 total positions denoted in the Pearson symbol are occupied only partially. The mean total number of atoms in the unit cell is about 15.2 [1994Bur].

Table 3: Compositions of the Invariant Equilibria in the U-UAl_2-UFe_2 Partial System

Reaction	T [°C]	Type	Phase	Composition* (at.%)		
				Al	Fe	U
$L + UAl_2 \rightleftharpoons U(Fe,Al)_2$	1060	max_1	L	(33)	(34)	33.3
			UAl_2	(53)	(14)	33.3
			$U(Fe,Al)_2$	40	26.7	33.3
$L \rightleftharpoons U(Fe,Al)_2 + UF_2$	1015	max_2	L	20.5	46.2	33.3
			$U(Fe,Al)_2$	(22)	(45)	33.3
			UFe_2	(13)	(54)	33.3
$L + (\gamma U) \rightleftharpoons UAl_2 + U_6Fe$	805	U_1	L	6	19.5	74.5
			(γU)	3	2	95
			UAl_2	(64)	(2.7)	33.3
			U_6Fe	-	14.3	85.7
$L + UAl_2 \rightleftharpoons U(Fe,Al)_2 + U_6Fe$	780	U_2	L	6	23	71
			UAl_2	(57)	(9.7)	33.3
			$U(Fe,Al)_2$	(33.3)	(33.3)	33.3
			U_6Fe	-	14.3	85.7
$L + U(Fe,Al)_2 \rightleftharpoons UFe_2 + U_6Fe$	745	U_3	L	(2)	(33)	65
			$U(Fe,Al)_2$	(22 7)	(44)	33.3
			UFe_2	(4.7)	(62)	33.3
			U_6Fe	-	14.3	85.7

Reaction	T [°C]	Type	Phase	Composition* (at.%)		
				Al	Fe	U
$(\gamma U) \rightleftharpoons (\beta U) + UAl_2 + U_6Fe$	~ 680	E_1	(γU)	(< 1.5)	(1)	(> 97)
			(βU)	(< 0.5)	(< 1)	(> 98)
			UAl_2	(66)	(0.7)	33.3
			U_6Fe	-	14.3	85.7
$(\beta U) \rightleftharpoons (\alpha U) + UAl_2 + U_6Fe$	~ 610	E_2	(βU)	(< 0.2)	(< 0.2)	(> 99)
			(αU)	(< 0.01)	(< 0.1)	(> 99)
			UAl_2	(66.5)	(0.2)	33.3
			U_6Fe	(-)	14.3	85.7

* Values given in brackets are estimated

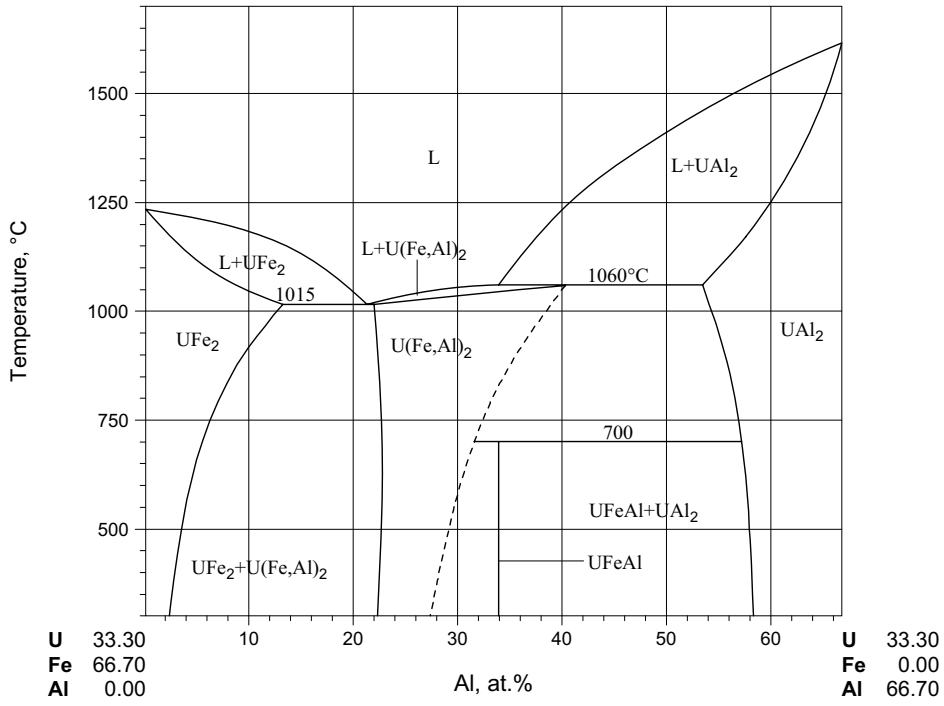

Fig. 1: Al-Fe-U. The quasibinary system UAl_2-UFe_2

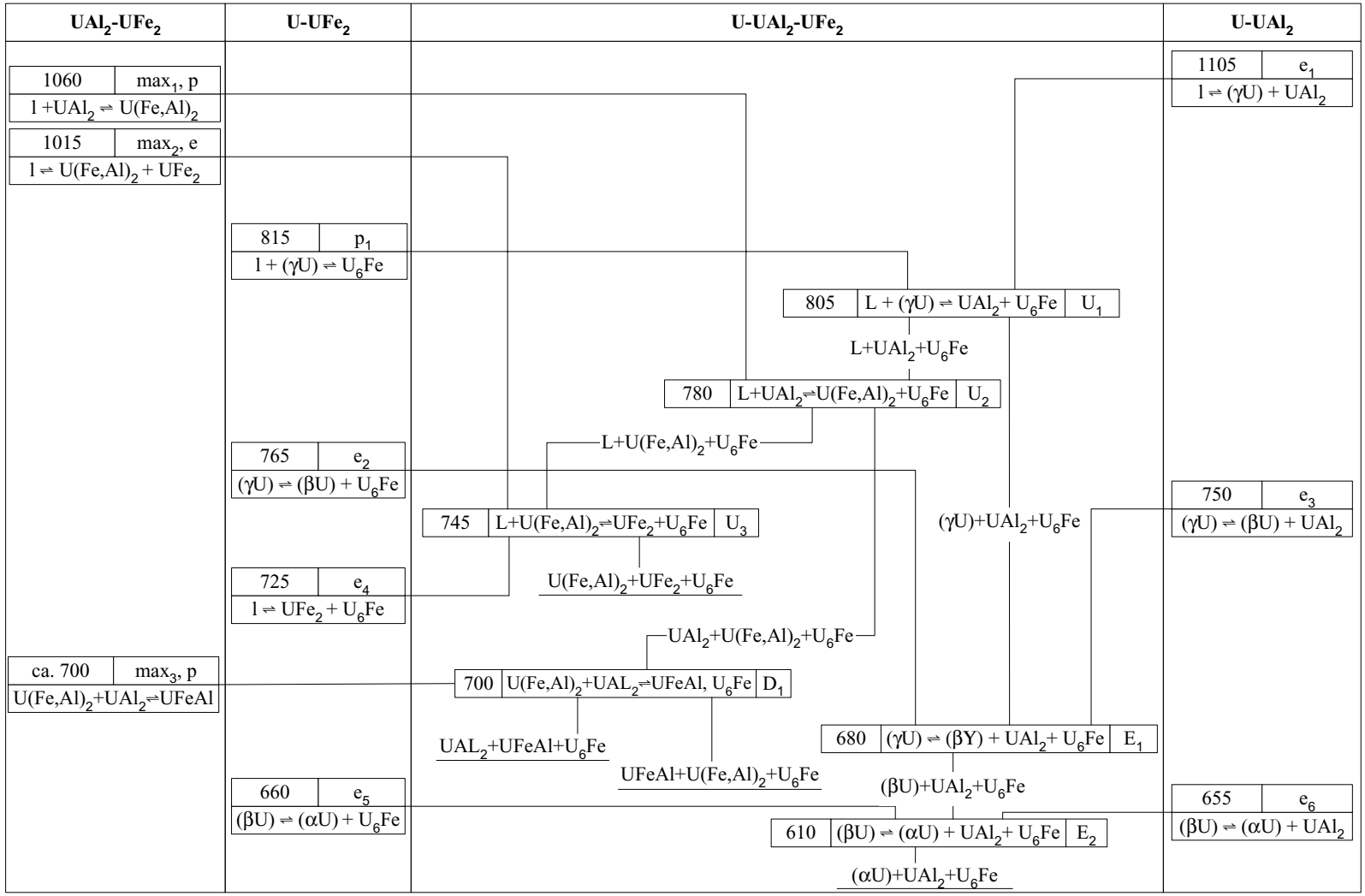

Fig. 2: Al-Fe-U. Partial reaction scheme

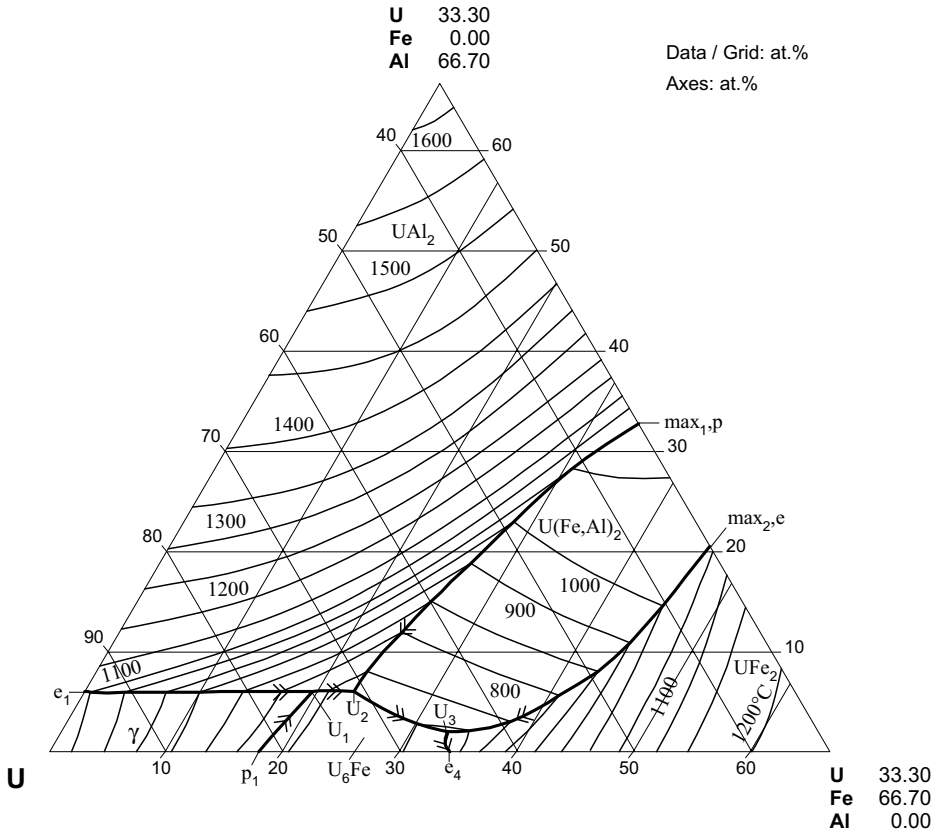

Fig. 3: Al-Fe-U. Partial liquidus surface projection

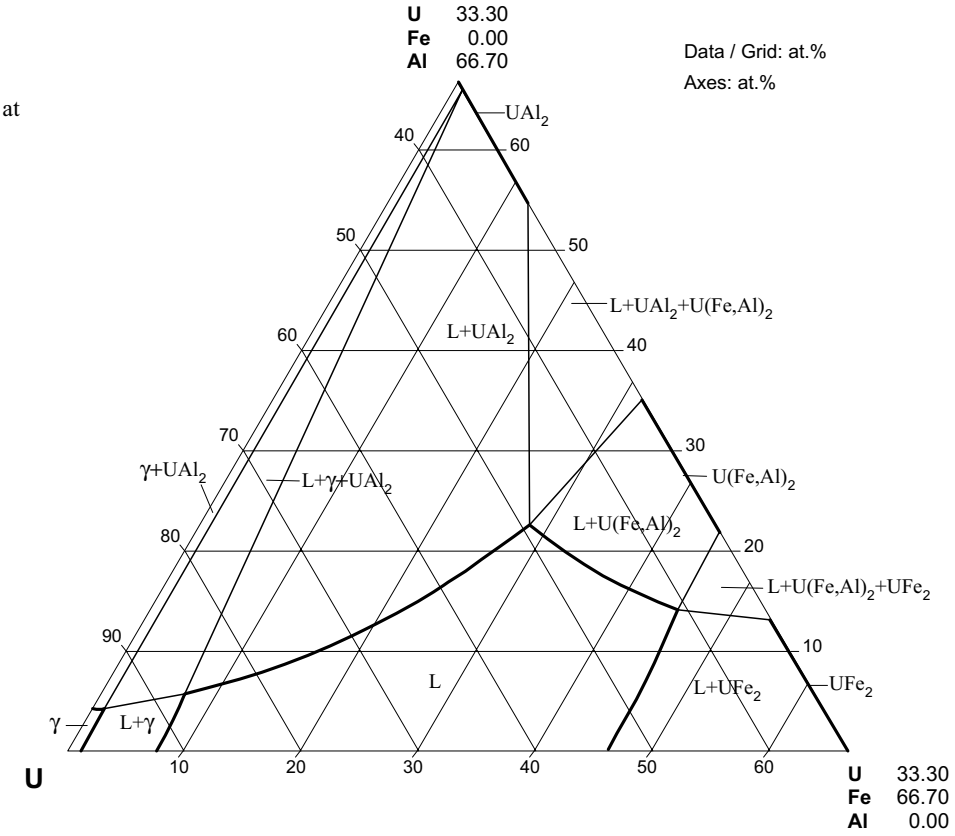

Fig. 4: Al-Fe-U. Isothermal section at 1000°C

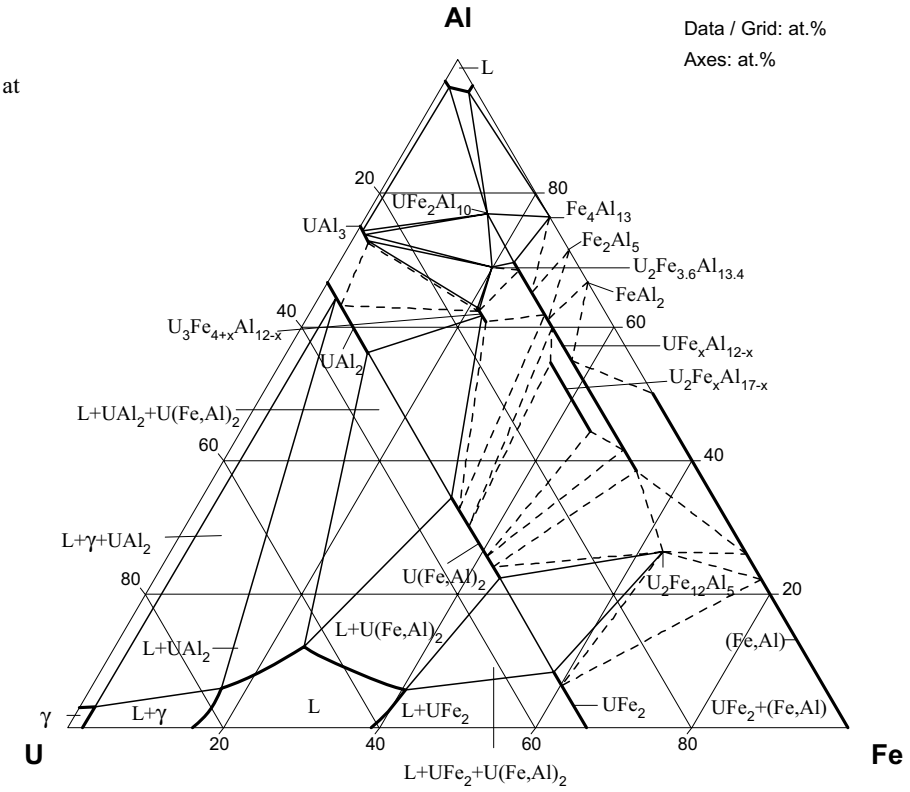

Fig. 5: Al-Fe-U. Isothermal section at 850°C

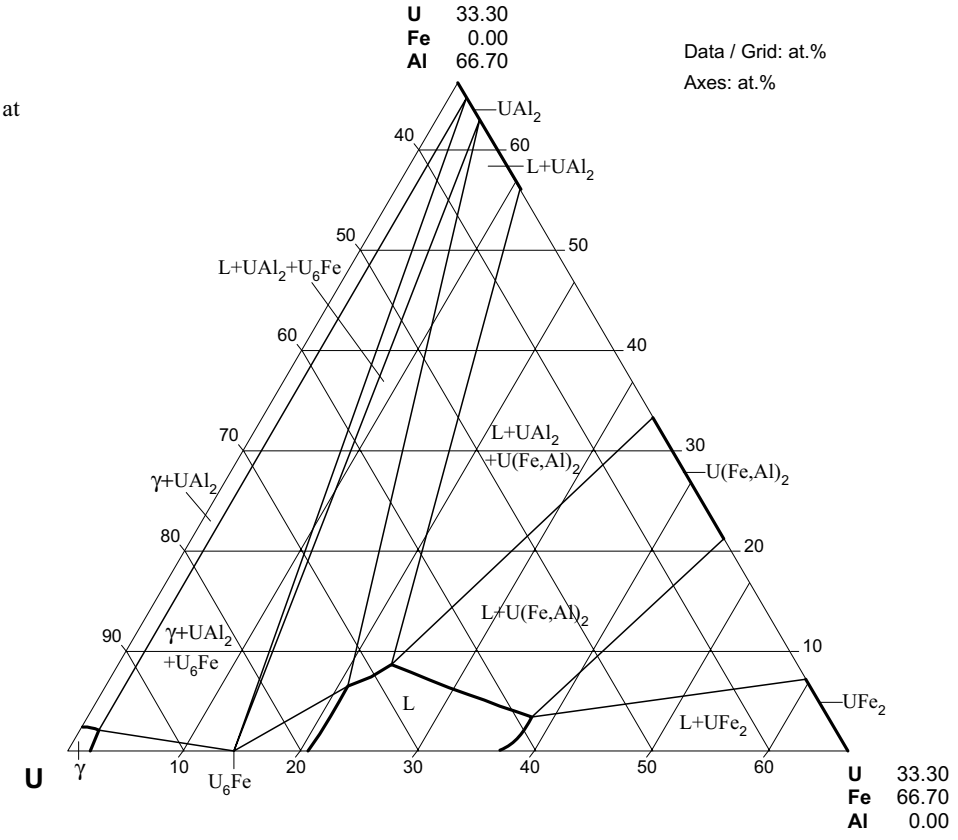

Fig. 6: Al-Fe-U. Isothermal section at 800°C

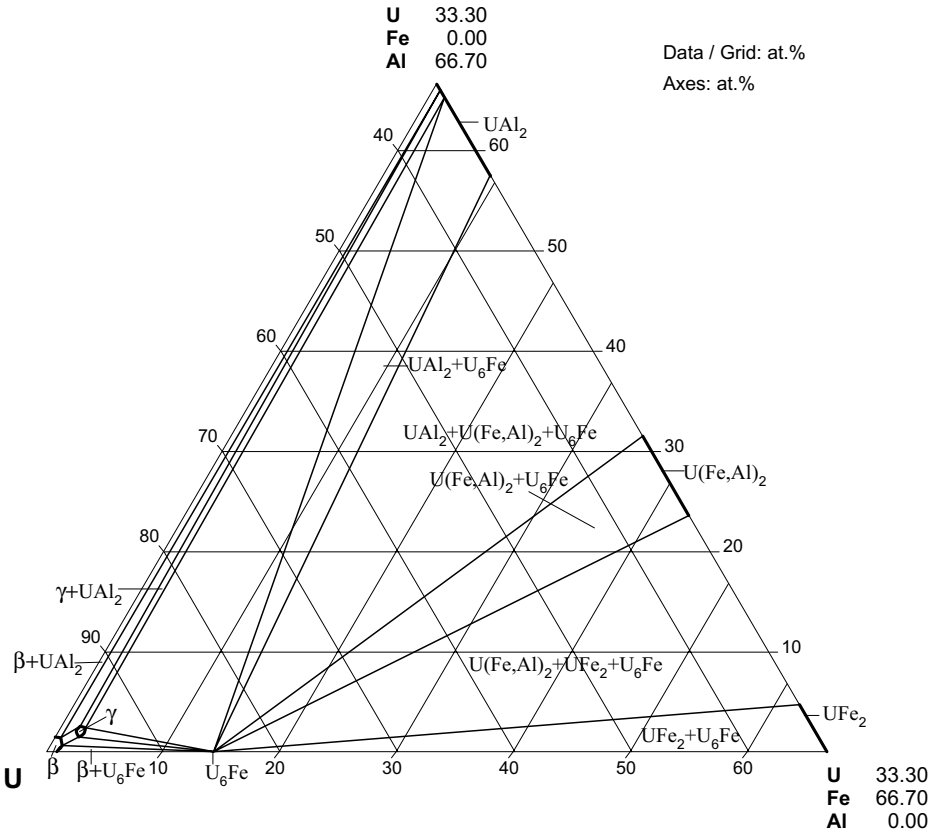

Fig. 7: Al-Fe-U. Isothermal section at 700°C

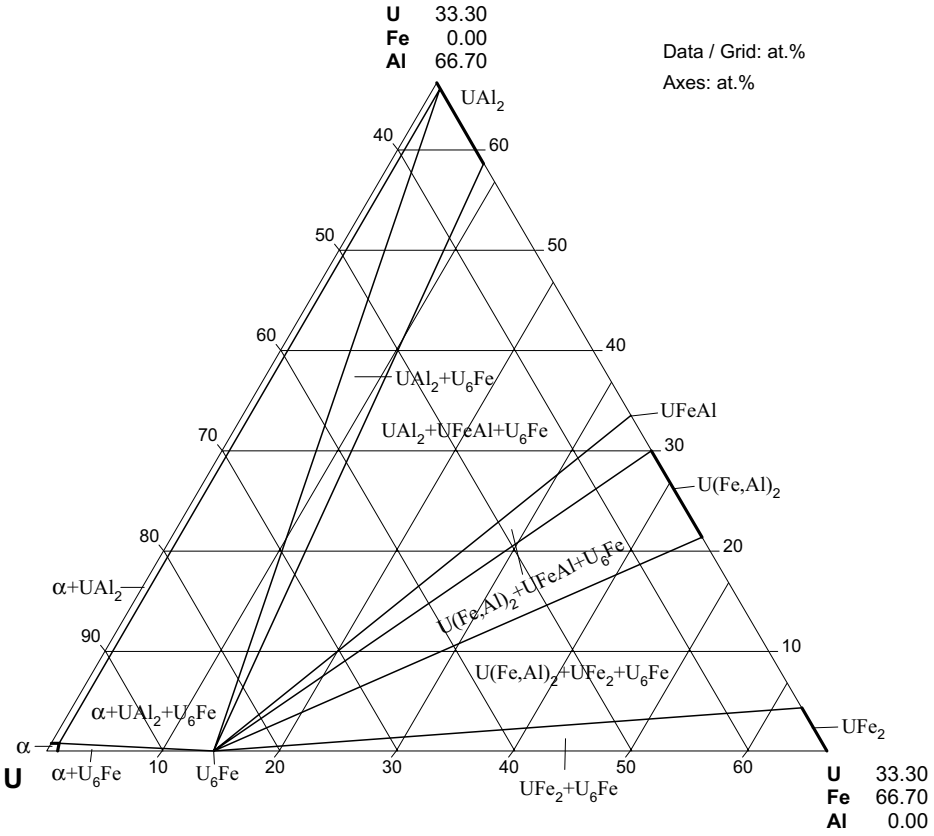

Fig. 8: Al-Fe-U. Isothermal section at 600°C

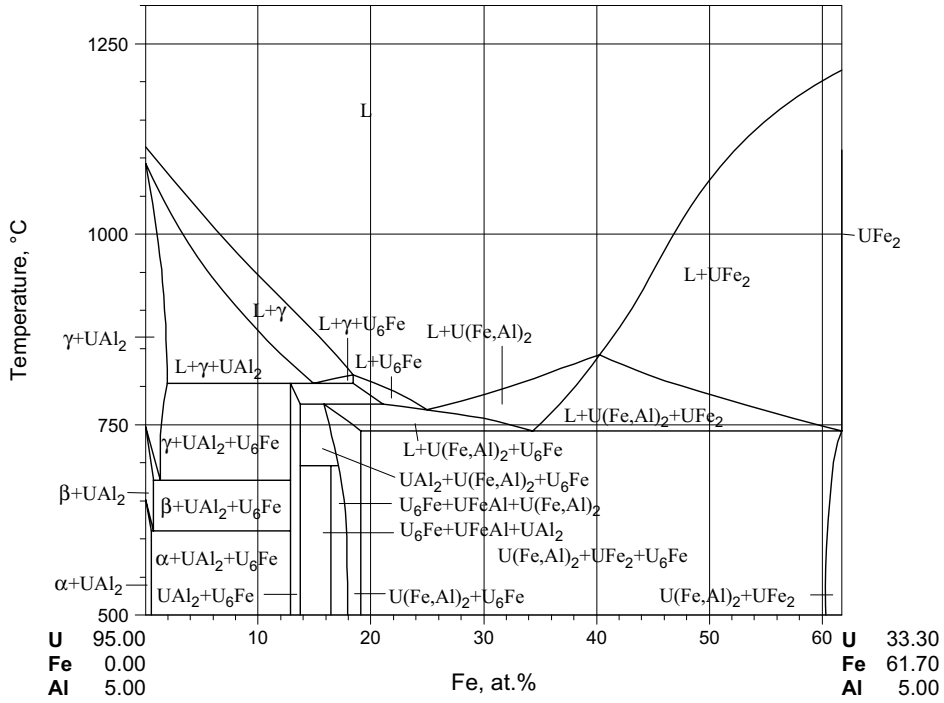

Fig. 9: Al-Fe-U. Temperature - composition cut for 5 at.% Al

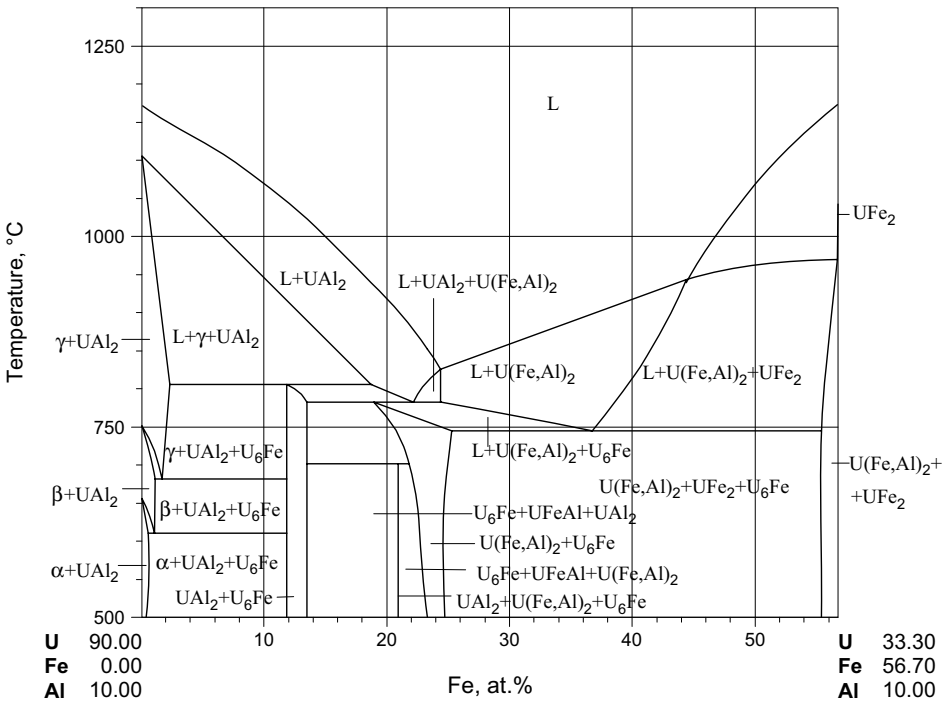

Fig. 10: Al-Fe-U. Temperature - composition cut for 10 at.% Al

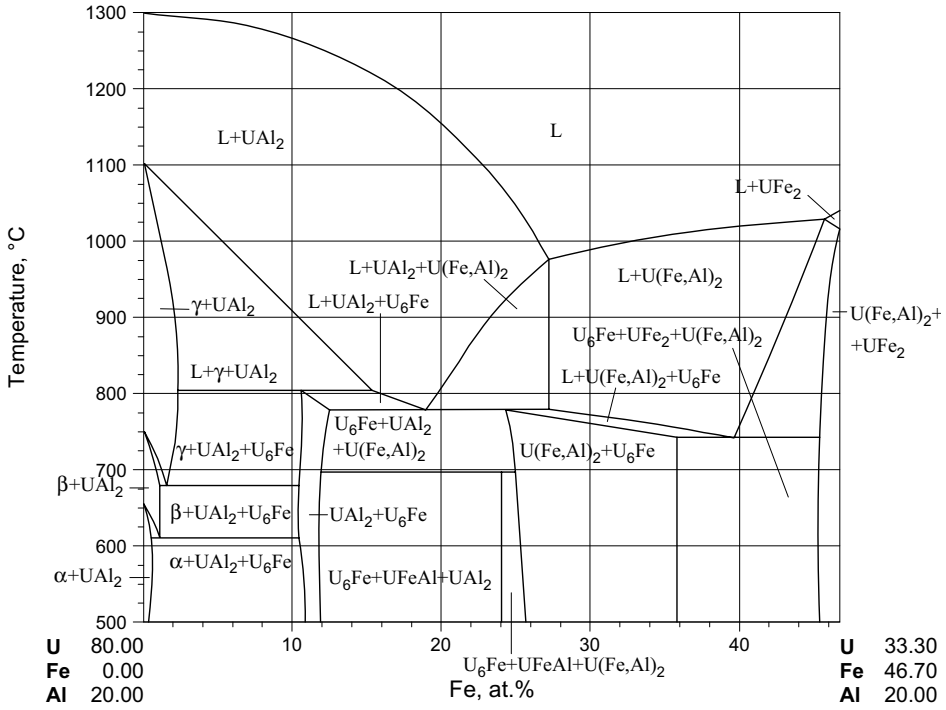

Fig. 11: Al-Fe-U. Temperature - composition cut for 20 at.% Al

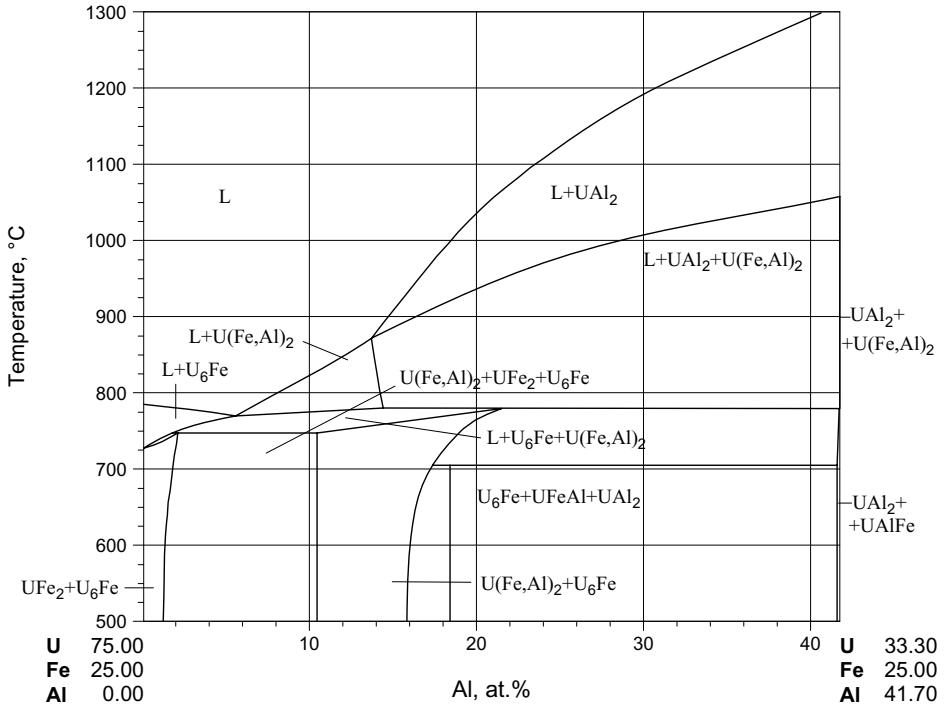

Fig. 12: Al-Fe-U. Temperature - composition cut for 25 at.% Fe

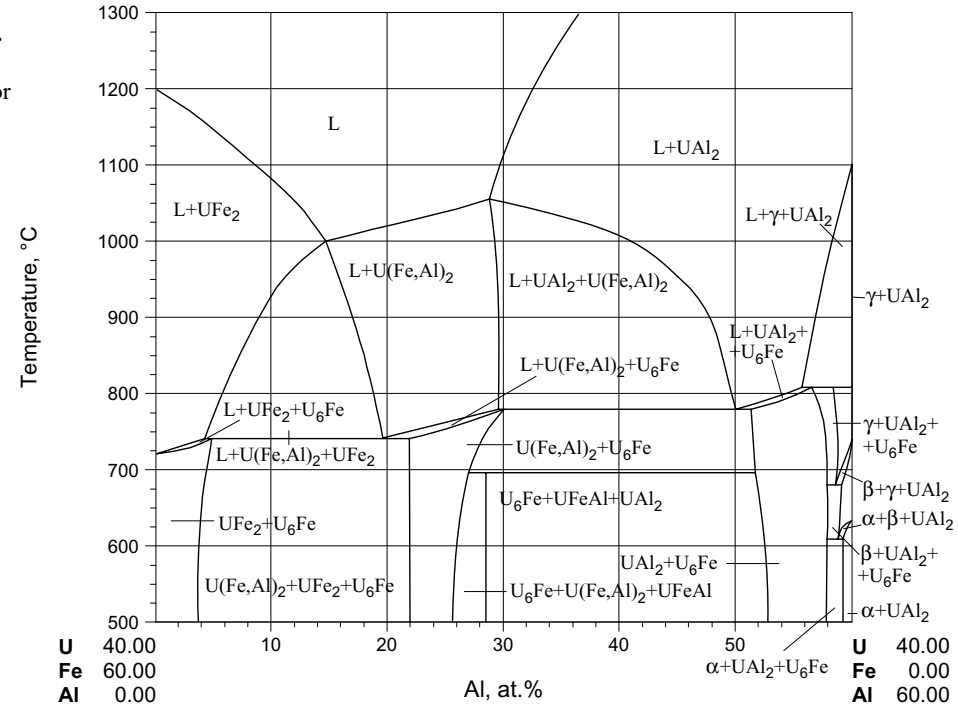

Fig. 13: Al-Fe-U. Temperature - composition cut for 40 at.% U

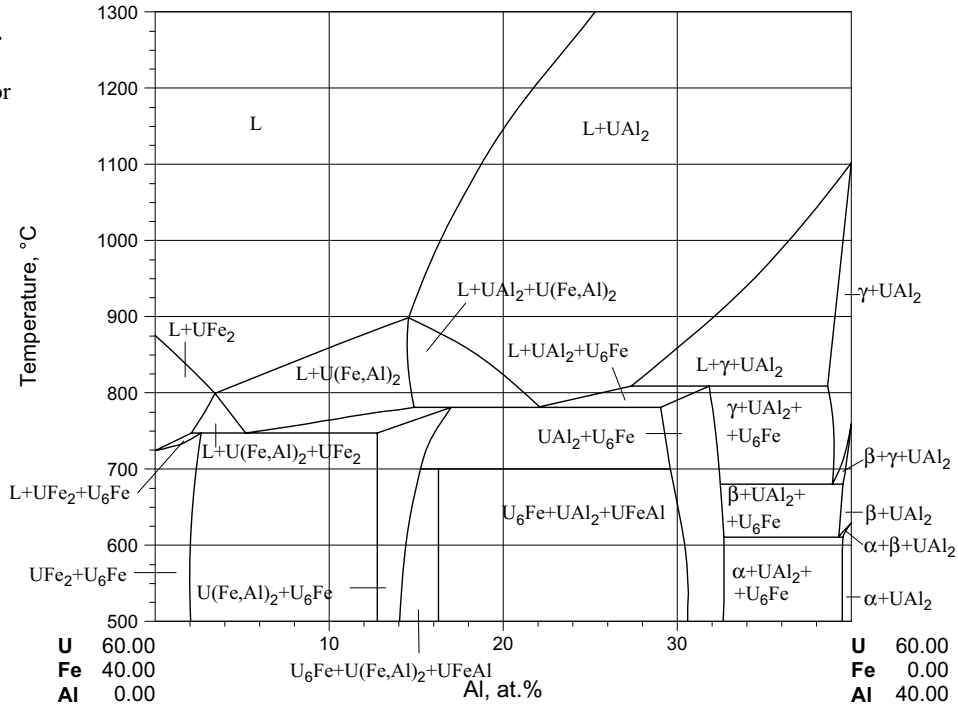

Fig. 14: Al-Fe-U. Temperature - composition cut for 40 at.% U

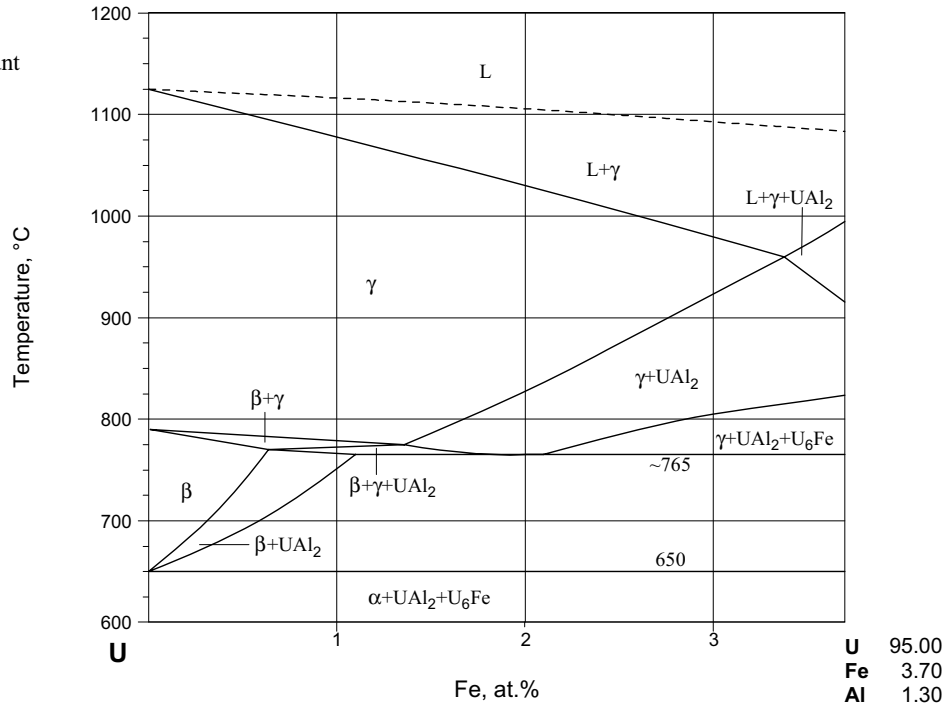

Fig. 15: Al-Fe-U. Isopleth with constant ratio Al:Fe = 3:1

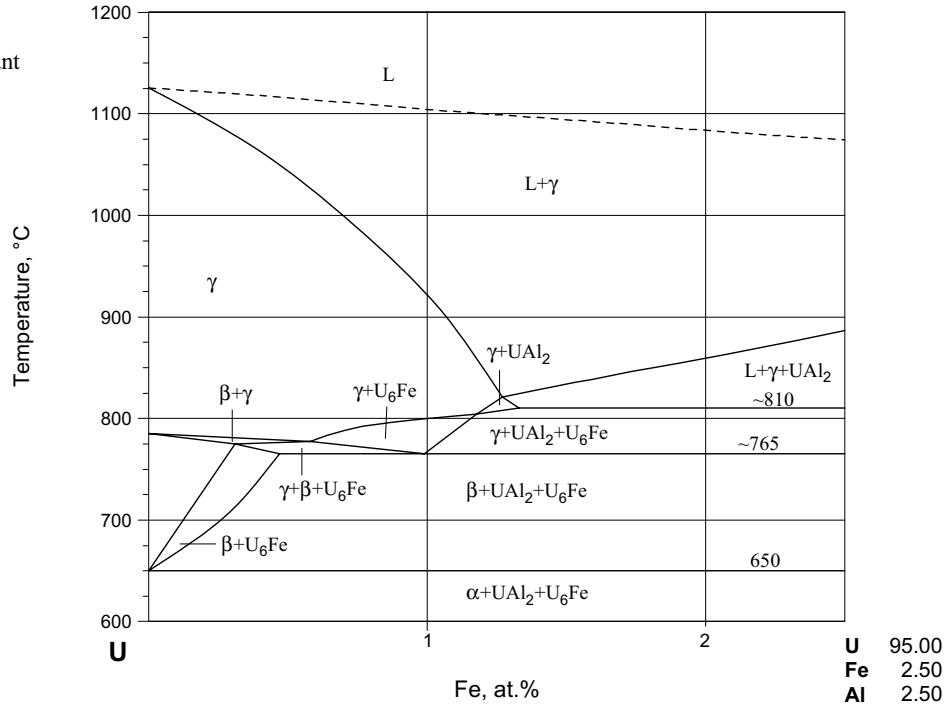

Fig. 16: Al-Fe-U. Isopleth with constant ratio Al:Fe = 1:1

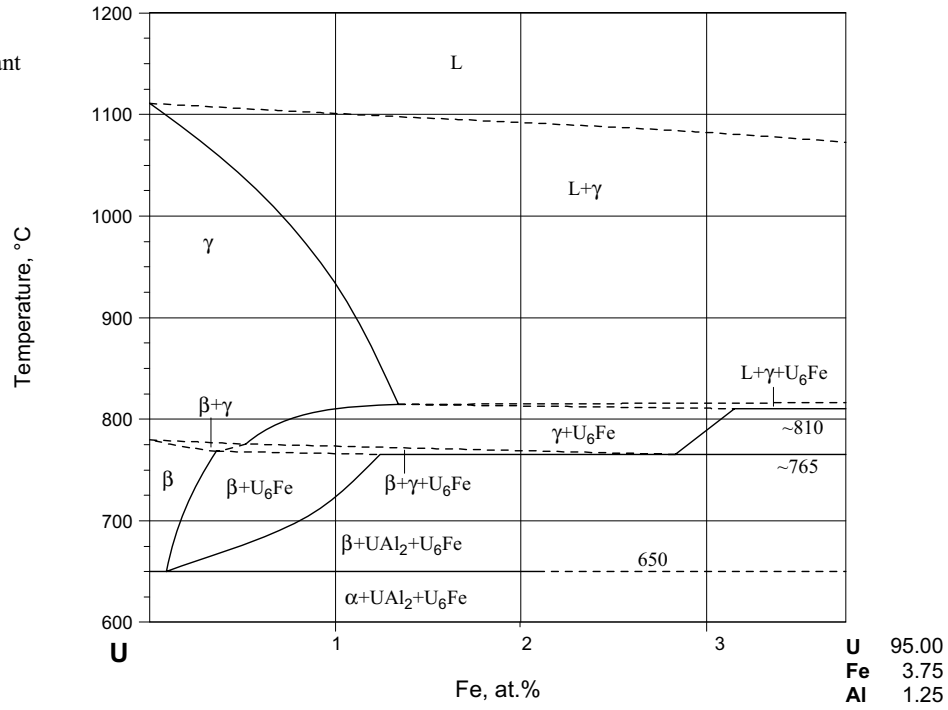

Fig. 17: Al-Fe-U. Isopleth with constant ratio Al:Fe = 1:3

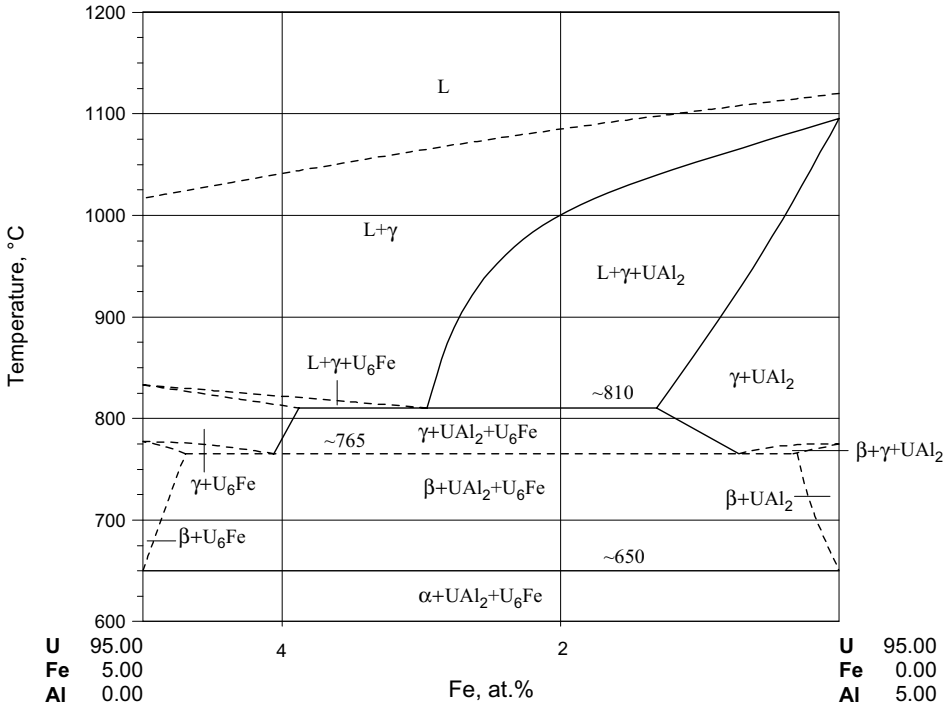

Fig. 18: Al-Fe-U. Isopleth at constant 95 at.% U

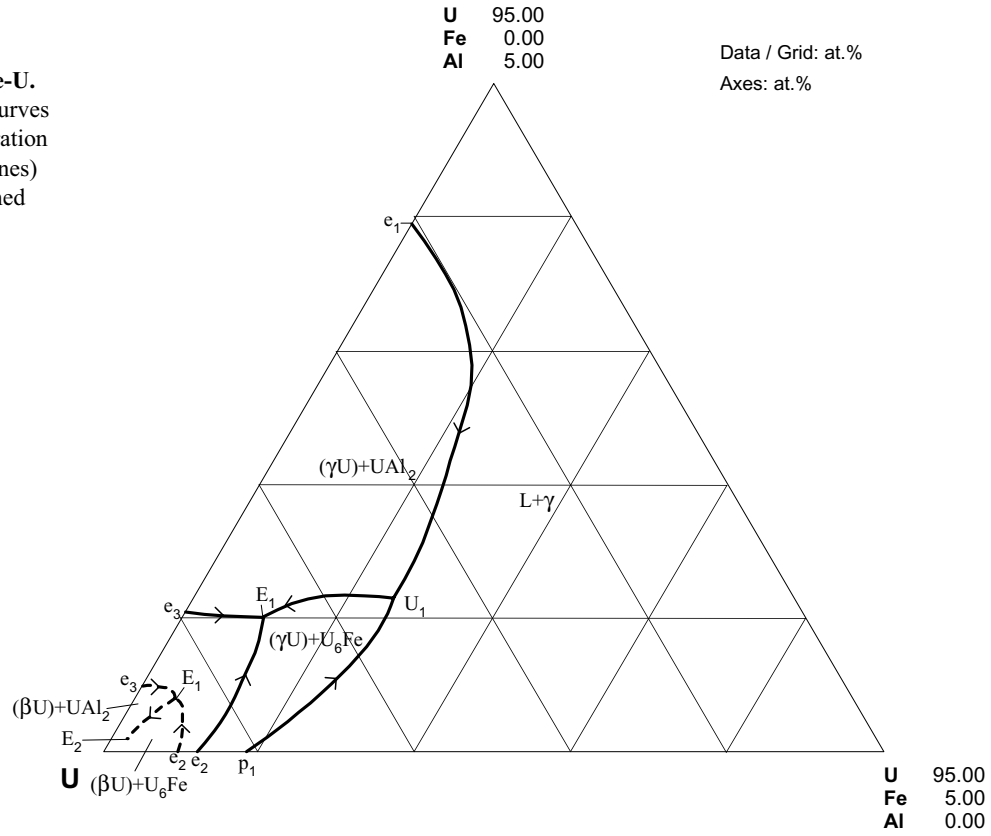

Fig. 19: Al-Fe-U. Projection of curves of double saturation of (γU) (full lines) and (βU) (dashed lines)