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On the characterization of $(U,Np)_xT_yX_z$ intermetallic solid solutions

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Abstract

Recent studies of pseudoquaternary, doped systems or solid solutions of heavy fermion and superconducting materials have been started in an attempt to understand their unusual behavior. To resolve the physics, the characterization of the studied samples, and particularly their homogeneity, is of crucial importance. We present here a detailed characterization of $(U_{1-x}Np_x)Ru_2Si_2$ and $(U_{1-x}Np_x)Pd_2Al_3$ alloys by X-ray diffraction, metallography and microprobe analysis. The U and Np atoms were shown to be homogeneously distributed and the samples were found to be single phase of uniform composition.

Keywords: Metallurgy; Actinide compounds; $(U,Np)_xT_yX_z$; EPMA

1. Introduction

In the last decade, heavy fermion and superconducting materials with f electrons have presented a challenge in solid state physics. Among these materials, uranium intermetallic compounds are of great interest. More recently, in attempts to understand their unusual behavior, studies were made of systems doped with rare-earths or Th. This ‘doping’ approach allows use of the Doniach’s Kondo-necklace model [1] and its extension (see e.g. Ref. [2]) for understanding the physics. A natural extension of this approach is doping with 5f elements which was started in recent years [3,4]. We present here a detailed characterization of $(U_{1-x}Np_x)Ru_2Si_2$ and $(U_{1-x}Np_x)Pd_2Al_3$ alloys by X-ray diffraction, metallography and microprobe analysis.

2. Experimental details

Large batches of $(U,Np)_xT_yX_z$ were prepared by arc melting together stoichiometric amounts of metals of high purity. Solid solutions, with x as listed in Table 1 were then prepared by arc melting stoichiometric amounts of pure U and Np compounds. The ingots were melted at least four or six times to optimize their homogeneity. Weight losses were checked after fusion and found to be negligible. Samples obtained were characterized by X-ray diffraction, metallography and microprobe analysis (EPMA) to verify their homogeneity.

X-ray diffraction patterns on powder were obtained with a Debye–Scherrer camera (114.6 mm diameter) using a Phillips type PW1120/90 diffractometer and Cu Kα radiation.

Representative pieces of each sample were extracted by fragmentation from the polycrystalline ingots and prepared for metallography and EPMA. Microprobe analysis was performed on a machine which combines the crystal spectrometer of the CAMECA MS46 analyzer and a three electron lens optical system of the CAMEBAX analyzer [5]. The analyses were performed with an acceleration potential of 20 keV and a beam current of 100 nA. The conventional EPMA matrix corrections were carried out using the QUAD2 program of Farthing et al. [6].

Table 1
Lattice parameters of $(U_{1-x}Np_x)Ru_2Si_2$ of $ThCr_2Si_2$ structure type and $(U_{1-x}Np_x)Pd_2Al_3$ of $PrNi_2Al_3$ structure type

Compound	Lattice parameters	
	a (pm)	*c* (pm)
URu_2Si_2	413.1 (1)	957.4 (1)
$U_{0.99}Np_{0.01}Ru_2Si_2$	413.2 (1)	957.6 (1)
$U_{0.9}Np_{0.1}Ru_2Si_2$	413.1 (1)	957.5 (1)
$U_{0.7}Np_{0.3}Ru_2Si_2$	413.2 (1)	957.8 (1)
$U_{0.5}Np_{0.5}Ru_2Si_2$	413.4 (1)	958.3 (1)
$U_{0.3}Np_{0.7}Ru_2Si_2$	413.6 (1)	958.4 (1)
$U_{0.1}Np_{0.9}Ru_2Si_2$	413.5 (1)	958.8 (1)
$NpRu_2Si_2$	413.7 (1)	959.3 (1)
$U_{1-x}Np_xPd_2Al_3$	See Ref. [9]	

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3. Results and discussion

Results of the crystallographic characterization indicate single phase samples with the lattice parameters listed in Table 1.

In Figs. 1–4 some typical results obtained for various compositions are presented. These are considered to be representative of the whole range investigated.

Fig. 1(a), (b) and Fig. 2(a), (b) show electron absorption micrographs for $(U_{1-x}Np_x)Ru_2Si_2$. Samples are very homogeneous and only traces of grain boundaries are seen. Particular care was taken to verify that U and Np (An) were uniformly distributed in the sample and that no clusters were present. For this purpose, X-ray scans, in which a profile of the distribution of a selected element is obtained by scanning the electron beam along a single line, were made. A good homogeneity and the absence of clusters is seen in Fig. 1(c), (d) and Fig. 2(c), (d). Quantitative analyses revealed that the sample composition was in agreement with the nominal composition to within roughly 1–3% (absolute).

Fig. 3(a), (b) and Fig. 4(a), (b) show electron absorption micrographs for $(U_{1-x}Np_x)Pd_2Al_3$. It is interesting to see how these pictures reveal the different texture of these 1:2:3 intermetallic compounds compared to the 1:2:2 ones. Grain boundaries are clearly seen between the acicular crystallites. This is intrinsic to the hexagonal crystal structure of the 1:2:3 compounds. In the samples with a higher content of Np, EPMA revealed some enrichment of Np at the grain boundaries (Fig. 4(c), (d)). However, this observation should not be interpreted as evidence of inhomogeneity or of the formation of An clusters in the samples. Earlier observations have shown that the secondary phases formed at the grain boundaries in pure 1:2:3 compounds are An rich [7]. Quantitative analysis revealed again that the relative compositions were in agreement with the nominal ones to within roughly 1–3% (absolute), but that there was a slight deficit in Al (2.8–2.9 instead of 3). A similar systematic deficit in Al is often seen in samples of this type of compounds and is explained by preferential Al evaporation during the melting process (see e.g. [8]).

4. Conclusions

The EPMA results of the $(U,Np)_xT_yX_z$ compounds presented here show that the samples obtained may be considered as homogeneous and free of An clusters. It further shows that homogeneous solid solutions are formed

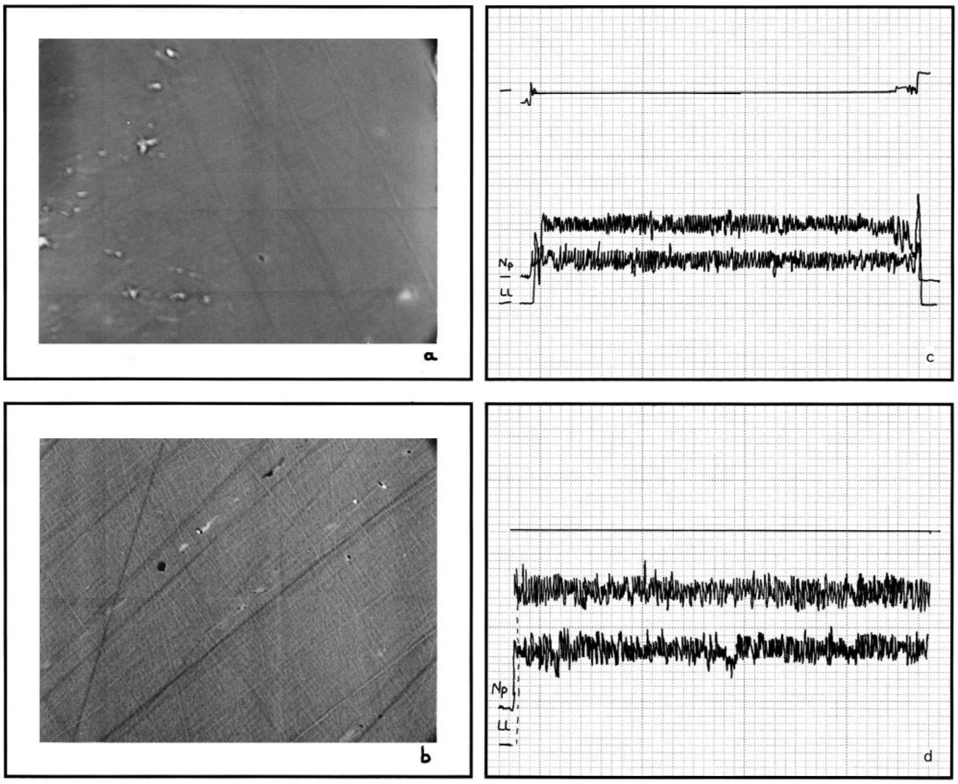

Fig. 1. (a) Electron absorption micrograph of $U_{0.99}Np_{0.01}Ru_2Si_2$; (b) electron absorption micrograph of $U_{0.9}Np_{0.1}Ru_2Si_2$; (c) and (d) respective distribution of U and Np analyzed by EPMA on an arbitrary scale (top line is the absorbed electron current).

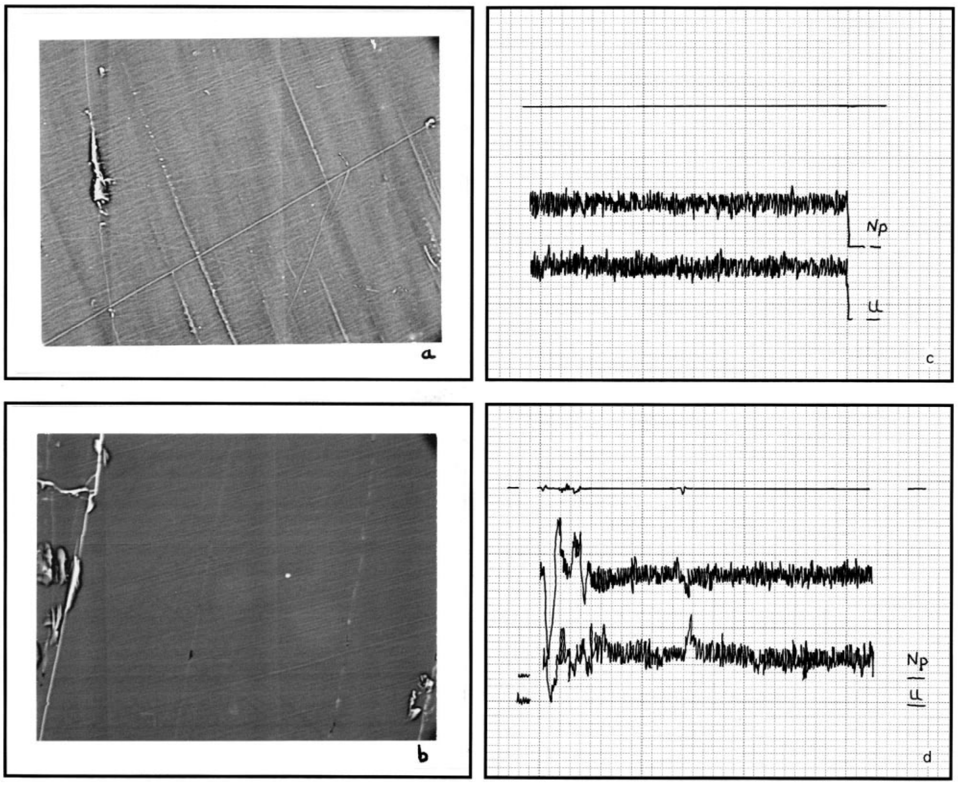

Fig. 2. (a) Electron absorption micrograph of $U_{0.5}Np_{0.5}Ru_2Si_2$; (b) electron absorption micrograph of $U_{0.1}Np_{0.9}Ru_2Si_2$; (c) and (d) respective distribution of U and Np analyzed by EPMA on an arbitrary scale (top line is the absorbed electron current).

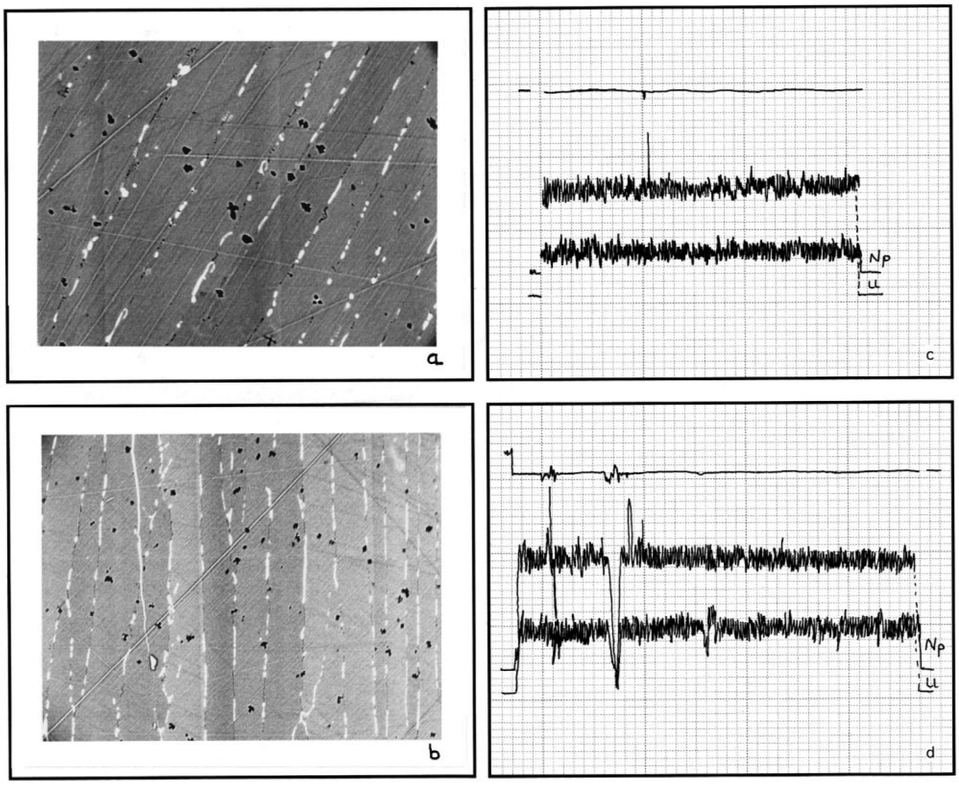

Fig. 3. (a) Electron absorption micrograph of $U_{0.97}Np_{0.03}Pd_2Al_3$; (b) electron absorption micrograph of $U_{0.93}Np_{0.08}Pd_2Al_3$; (c) and (d) respective distribution of U and Np analyzed by EPMA on an arbitrary scale (top line is the absorbed electron current).

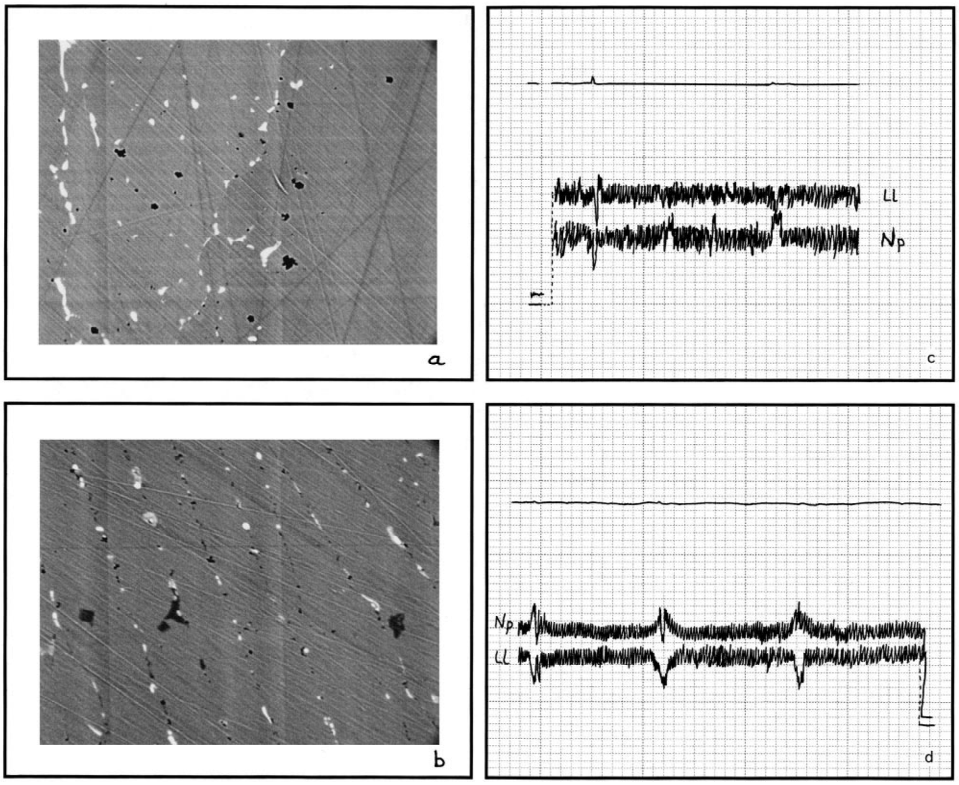

Fig. 4. (a) Electron absorption micrograph of $U_{0.9}Np_{0.1}Pd_2Al_3$; (b) electron absorption micrograph of $U_{0.5}Np_{0.5}Pd_2Al_3$; (c) and (d) respective distribution of U and Np analyzed by EPMA on an arbitrary scale (top line is the absorbed electron current).

for the whole range of Np doping. The $(U_{1-x}Np_x)Ru_2Si_2$ samples are showing less grain boundaries and therefore are of better quality than the $(U_{1-x}Np_x)Pd_2Al_3$ samples, and this is directly related to the ease with which the parent compounds can be synthesized. Thus, it is confirmed that the intermetallic solid solutions of 1:2:2 and 1:2:3 systems presented here are sufficiently pure for the purpose of comparative studies of the influence of the dopants on the electronic properties.

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