RECENT ACTIVITIES OF THE NUCLEAR FUEL TECHNOLOGY DEPARTMENT OF ÇEKMECE NUCLEAR RESEARCH AND TRAINING CENTER

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ABSTRACT

The Nuclear Fuel Technology Department (NFTD) in ÇNRTC is a unique unit in Turkey in charge of performing all activities in nuclear fuel field. It has a pilot plant on uranium refining and conversion to UO_2 since 1986. Presently, its R&D activities are focused on pellet manufacturing and characterization: UO_2, ThO_2 and $(Th,U)O_2$. The studies on thorium dioxide fuel include to obtain ThO_2 pellets from thorium nitrate and mixed $(Th,U)O_2$ pellets.A study on evaluation of different fuel cycle options in accordance with nuclear energy planning in Turkey is also going on.

BACKGROUND

The works on the nuclear fuel cycle at Cekmece Nuclear Research and Training Center (ÇNRTC) were started at the beginning of 1970. At the beginning works were at laboratory scale and related to uranium ores and compounds. As the volume of work increased and expanded it became necessary to establish a new department. Thus Nuclear Fuel Technology Department (NFTD) was established on 12.02.1977 at ÇNRTC.

In 1985 Turkish Atomic Energy Authority (TAEA) made a decision to have the whole technology of the front-end of nuclear fuel cycles either by technology transfer and/or technology development by its own staff. In March 1985, design and construction of a pilot plant on refining of uranium concentrate, conversion to UO_2, and manufacturing of sintered pellets, were started. The objectives of this pilot plant are to design and develop the necessary processes and equipment, to install and operate them, to learn and demonstrate the production processes, to solve the problems encountered, to make the possible improvements, to provide the technological and economical data which will serve as the basis for the nuclear fuel plant to be constructed in future, and to provide experienced manpower.

During the laboratory scale experiments many valuable results were obtained. Under the light of the experimental results obtained during the previous works at the NFTD, by their own design and using the utilities of ÇNRTC, the pilot plant was constructed in the period of 1985-1986.The implementation of the design and construction of the pilot plant was realized by domestic resources; i.e. most of the equipment and material and all the manpower including engineering. The pilot plant started operation on 30-Oct-1986.

At the beginning, the pilot plant produced sinterable nuclear grade uranium dioxide powder through refining by solvent extraction, ADU (ammonium diuranate) precipitation, and reduction in a static furnace. It was expanded to include pellet manufacture through powder preparation, pressing and sintering at 1700°C in 1990s. Required quality control tests in all the steps of the process mentioned are being successfully performed by the department staff. The pilot plant is subjected to IAEA Safeguard since 1994.

PRESENT SITUATION

Presently, Turkey has no nuclear power reactors, but the nuclear power option is under serious consideration. The Energy Ministry of Turkey has issued an invitation to bid for the Akkuyu power reactor.

Turkish Atomic Energy Authority (TAEA) is in charge of regulating the nuclear activities, and performing research and development (R&D) on nuclear field. TAEA has two research centers: one in Istanbul, and the other in Ankara. The first research center in Istanbul, ÇNRTC, has 10 departments (in the R&D area) and two research reactors: TR-1 and TR-2.

The NFTD in ÇNRTC is a unique unit in Turkey in charge of performing all activities in nuclear fuel field. It has a pilot plant on uranium refining and conversion to UO_2. Presently, its R&D activities are focused on pellet manufacturing and characterization: UO_2, ThO_2 and $(Th,U)O_2$. The UO_2 powder produced in the pilot plant is pressed into pellets and sintered under hydrogen at 1700°C. Densities of sintered pellets reach around 97% of TD (theoretical density).

Up to date the samples taken from several parts of the installation have been analyzed. Every lot of yellow cake, intermediates and end product have been analyzed successfully for U content and for the following impurities: Cd, Ni, Mo, Cr, Fe, Co, Cu, Zn, Al, Mn, Na, Mg, Pb, K, SO_4^{2-}, PO_4^{3-}. Metallurgical quality control tests and characterization are performed on UO_2 and ThO_2 powders and pellets.

STUDIES ON URANIUM FUEL

Production Processes of Uranium Refining, Conversion to UO_2 and Pellet Manufacturing

The flowsheet of general production process is shown in Figure-1. The main production steps of the pilot plant are:

- dissolution of yellow cake by nitric acid
- uranium purification by solvent extraction using TBP in sieve-plate pulsed columns
- precipitation of nuclear-grade uranyl nitrate as ADU by ammonia solution

- filtration using rotary-drum vacuum filter
- reduction of ADU to UO_2 powder in static furnaces
- calcination at 350°C for 1½-hour in air
- reduction at 600°C for 4-hour in H_2
- stabilization at 100°C for 1-hour in 2%O_2 + 98% N_2
- UO_2 powder preparation for pressing
- milling of powder in ball-mill, and screening to <75μm
- Lubricant blending (0.2% zinc stearate) and homogenization
- Pressing with the hydraulic press at a diameter of 15 mm with green density ranges of 50 to 55 percent of TD.
- Sintering

In the sintering process the pellets are sintered in a continuous sintering furnace under hydrogen atmosphere at 1700°C. A typical sintering cycle is at 600°C for 3-hour, at 1500°C for 2-hour and at 1700°C for 4-hour, and total residence time of the pellets in the furnace is approximately 25 hours, allowing for the times the tray travels in the intermediate zones of the furnace. Required H_2 is produced by a hydrogen generator. The densities of the sintered pellets are in the ranges of 95 to 98 percent of TD.

Quality Control of the Products

Chemical Analysis

Chemical analysis of uranium and the impurities have been carried out in every lot of yellow cake, product solutions and end products. Samples have been analyzed successfully for U content and for the following impurities: Cd, Ni, Mo, Cr, Fe, Co, Cu, Zn, Ca, Al, Mn, Na, Mg, Pb, K and rare earth elements. An automatic titrator is used for uranium analysis; AAS and ICP-AES for impurities.

Metallurgical Inspections

The following inspections are applied:

<u>UO_2 Powder characterization</u>
Particle size distribution
Surface area measurement
Flowability
Bulk density
O/U ratio determination

<u>UO_2 pellet characterization</u> for green pellets
Visual inspection
Weighing
Geometrical dimension measurement
Determination of pellet density

Inspection of Sintered Pellets

Diameter	- micrometer
Weighing	- digital balance
Density	- immersion in Xylene
Surface imperfections	- visual
Cracks	- visual and 100% ultrasonic
Impurities	- chemical analysis (AAS and ICP)
O/U ratio	- thermogravimetry
Microstructure	- ceramography, optical, image analysis
Hourglass	- blade micrometer

The sintered pellet densities are given in Figure 2.

The diameter distribution of pellets are given in Figures 3. The statistical values of the pellets produced in the same production campaign are as follows:

Green Pellets (Count 331)

	Length (mm)	Weigth (g)	Diameter (mm)	Density (g/cm^3)
AVG	16.529	17.177	15.125	0.528
STD	0.2055	0.2046	0.002	0.002

Sintered Pellets (Count 330)

				GEOMETRIC		IMMERSION	
	Diam. (mm)	L (mm)	Weigth (g)	Density (g/cm^3)	% TD	Density (g/cm^3)	% TD
AVG	12.293	13.395	16.808	10.571	0.965	10.679	0.974
STD	0.0212	0.1651	0.252	0.024	0.002	0.015	0.001

STUDIES ON THORIUM FUEL

Utilization of thoria-based fuels is possible for some reactor types. A number of possible fuel cycles exists. There are several processes to manufacture thoria fuel, such as conventional blended powder, cold-pressed and sintered pellet route; sphere-pac fuel which consists of almost fully dense mixed-oxide microspheres from sol-gel process vibration-packed into fuel cladding; extruded sol-gel-derived clays; etc.

The majority of experiments have been performed on fuels with less than 10% UO_2. Data on fuels with UO_2 content greater than 20% are almost nonexistent. More work is required to demonstrate conclusively the performance capability of thoria-based fuels, particularly under off-normal operating conditions, and to provide quantitative data required for modelling fuel pin behaviour for design and licensing purposes.

Turkey has thorium resources amounting to 380,000 tonnes (ThO_2). The average Th content of this complex ore is about 0.2%. Other main constituents are rare earth elements. Recovery of thorium from the ore, and production of thoria powder were studied in the past.

TAEA carried out a project, namely "Thorium Studies", to study the feasibility of the utilization of thorium in power reactors, options of thorium fuel cycle and manufacturing of thorium fuel. The department staff were involved in this project. In the conclusion section of this project's final report, after discussion of availability of fissile material for thorium fuel doping under NPT regulations, it was stated that "under these conditions, if Turkey wants to assess its own thorium potential, once-through thorium cycle seems to be the only applicable option for Turkey". The it was suggested that "It would be realistic to evaluate and implement the thorium studies such as cycle calculations, reactor design and safety studies, manufacturing of and studies on fuel and material from this viewpoint.

Studies on thorium in the department are carried out upon the above mentioned recommendations.The studies on thorium dioxide fuel in the department include precipitation of thorium nitrate as oxalate and calcination of thorium oxalate to obtain ThO_2. The main focus of these studies is on obtaining homogenous mixture of UO_2-ThO_2 powders and on sintering of ThO_2-based pellets under different atmospheres.

Mixed $(Th,U)O_2$ pellets are prepared by mechanical blending in various proportions. The preparation procedure is shown in Figure 5, and X-ray diffraction patterns in Figure 6. The pellets have reached densities higher than 90% of TD.

The first-stage sintering kinetics of these pellets and high temperature sintering are being carried out. The apparent activation energy has found 338.9±40.6 kJ/mole for $(U_{0.8},Th_{0.2})O_2$ in 75% H_2 + 25% N_2 at 900-1000°C.

FUTURE OBJECTIVES

The further steps such as design, fabrication, irradiation and PIE of fuel elements and assemblies containing UO_2 and $(U,Th)O_2$, are under planning.

- Automatic process control using computers and PLC
- Designing of an experimental fuel element
- Fabrication of the fuel element
- Inspection of welds
- Irradiation and performance tests of the fuel elements
- Pre- and post-irradiation examinations

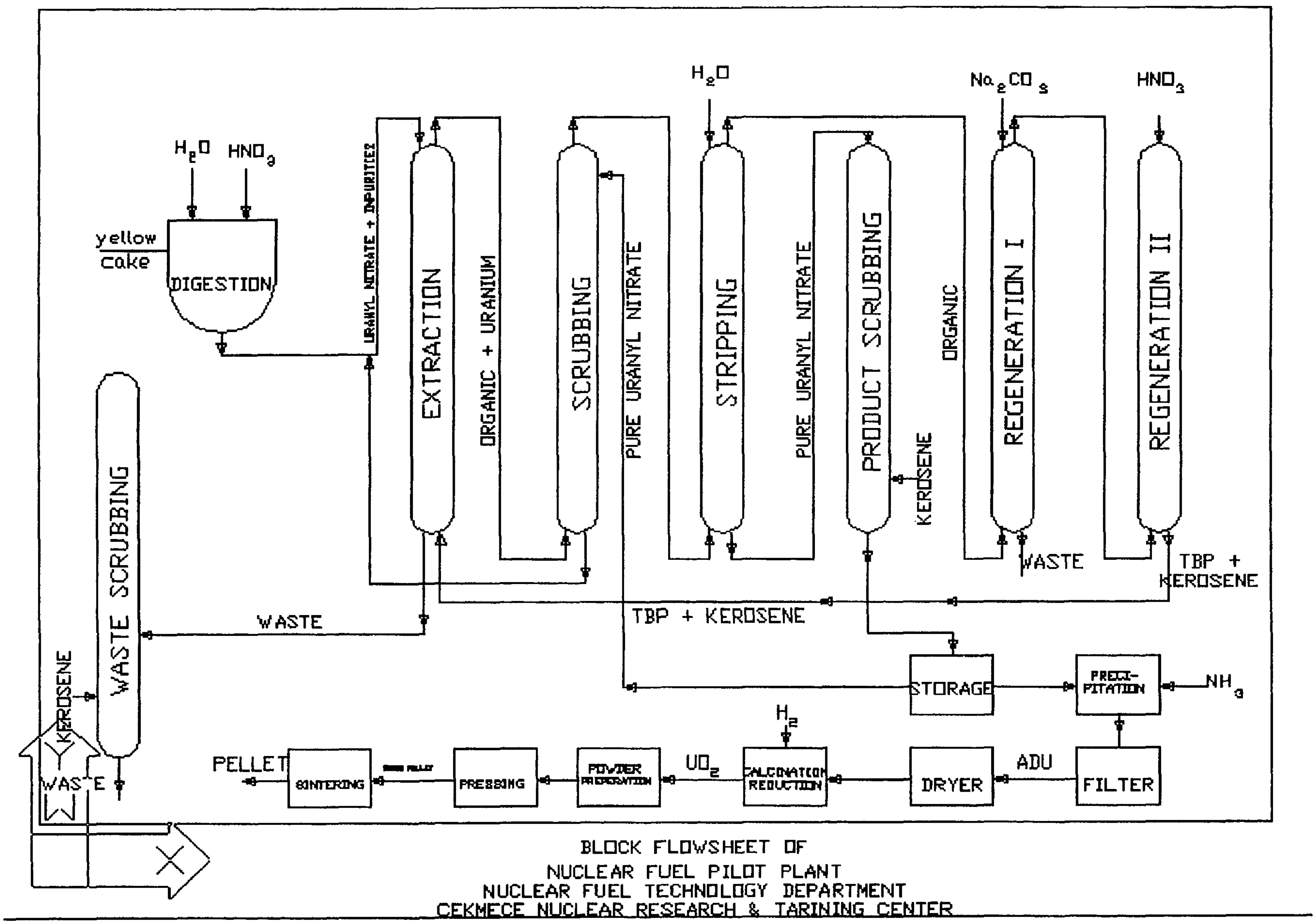

Figure 1 Flowshhet of General Production Process

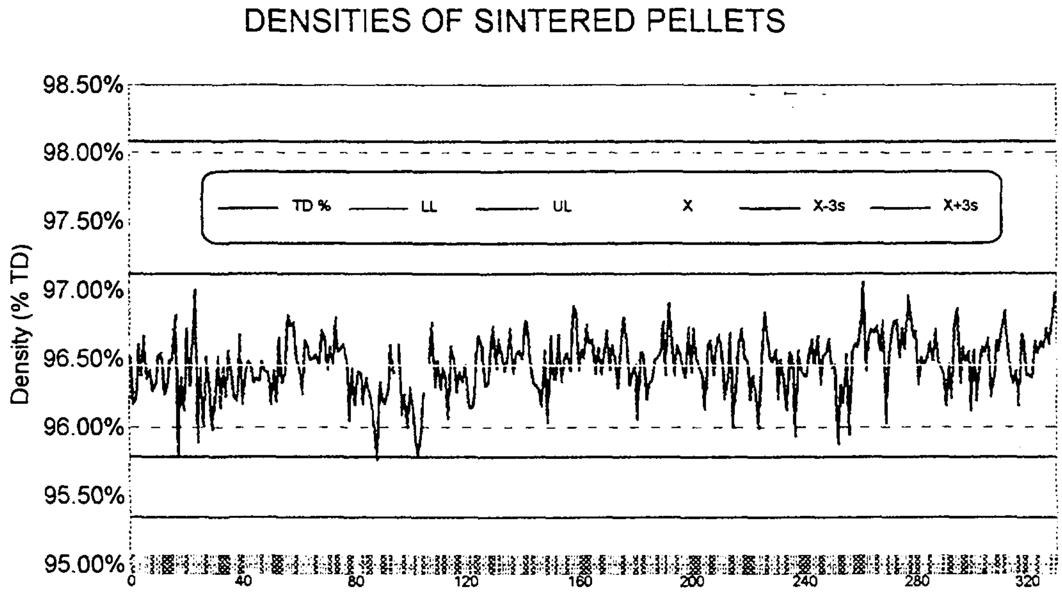

Figure 2 Densities of Sintered Pellets

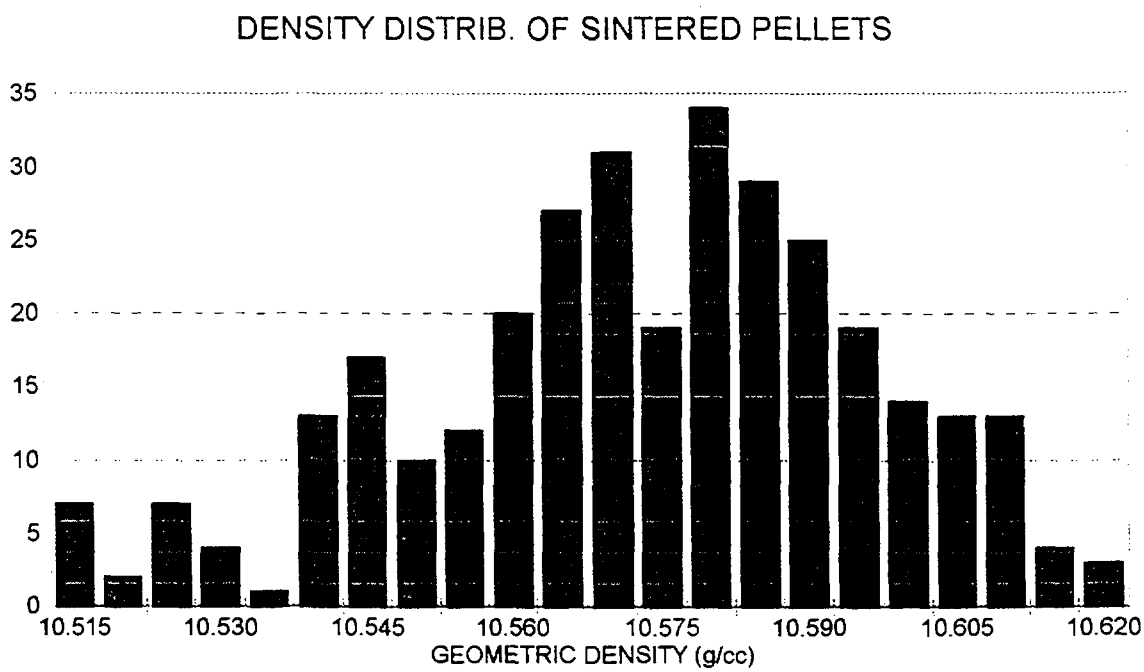

Figure 3 Density Distribution of Sintered Pellets

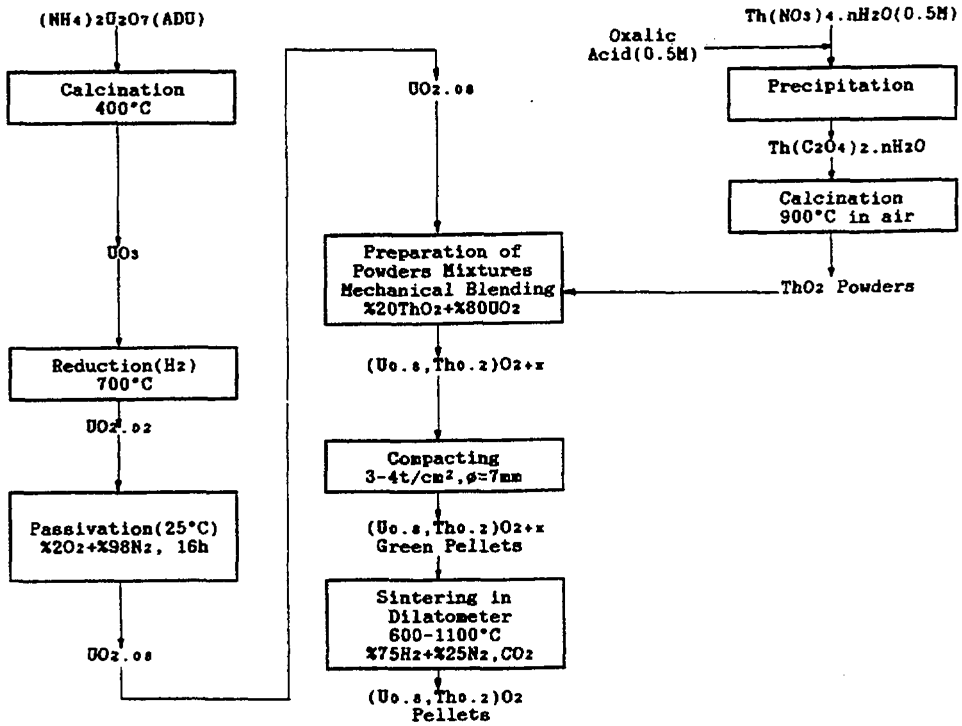

Fig. 4. Preparation of the mixed oxide pellets ($(U_{0.8}, Th_{0.2})O_2$).

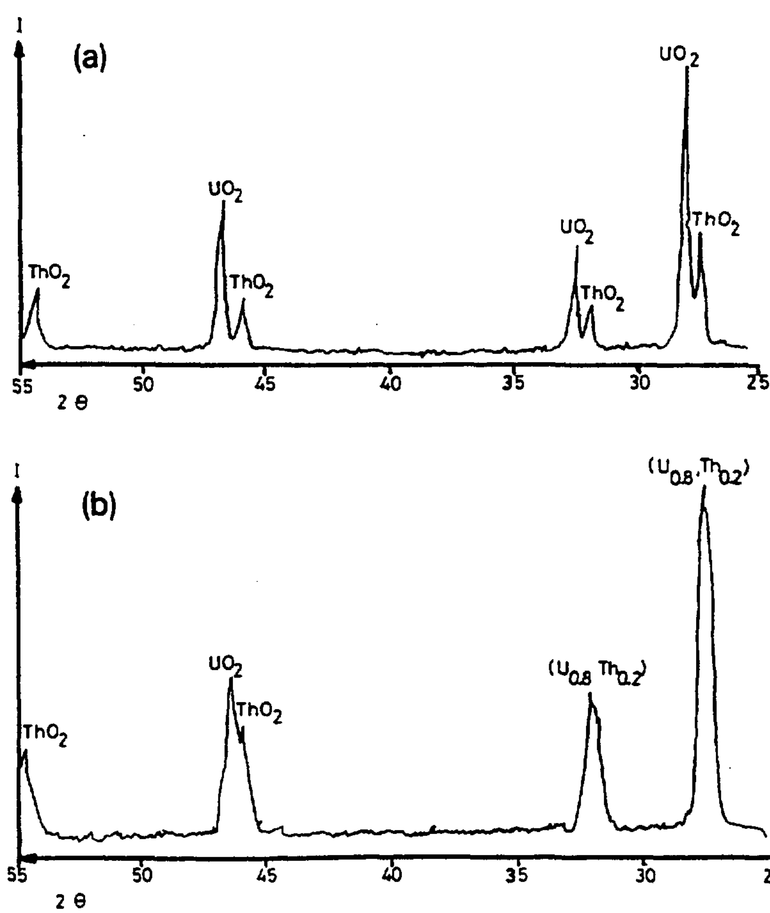

Fig. 5. X-Ray diffraction pics of UO_2–ThO_2 powders mixtures and solid solution. (a) $T = 1150°C$, 75% H_2 + 25% N_2 ($P_{O_2} = 10^{-15}$ atm: solid electrolyte sensor. (b) $T = 1150°C$, CO_2 ($P_{O_2} = 10^{-6}$ atm: solid electrolyte sensor).