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DESIGN OF A RESEARCH LABORATORY FOR THE REPROCESSING OF IRRADIATED FUEL

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ABSTRACT

A general design of a research laboratory for the reprocessing of irradiated fuel is undertaken. The fuel under consideration is natural UO_2 pellets clad in zircaloy-4. Reprocessing of irradiated fuel is a very complicated and advanced technology in which many aspects of mechanical, chemical, nuclear, control and metallurgical engineering are involved. A complete design work of such a laboratory, therefore, is a team work in related fields. In this study, the chemical process for reprocessing is selected. Shielding calculations are carried out. The equipment to be used and facilities to be provided are described in detail and a general layout is prepared. The layout is so arranged that the basement of the Nuclear Technology Laboratory of Mechanical Engineering Department of METU can be used for this purpose /1/

ÖZET

Kullanılmış nükleer yakıtların ayrıştırılmasını sağlayacak olan bir laboratuvar tasarımı ana hatları ile gerçekleştirildi. Söz konusu nükleer yakıt zirkaloy-4 zarfı içindeki doğal UO_2 peletleridir. Ayrıştırma işlemi son derece karışık ve gelişmiş bir teknoloji ürünüdür. Bu teknoloji makina, nükleer, kontrol, metallurji vb., mühendislik dallarının pekçok kısmını içermektedir. Bundan ötürü de söz konusu laboratuvarın eksiksiz tasarımı bu sahalarda çalışan kişilerin ortak çabaları ile mümkündür. Bu çalışma kapsamı içinde, söz konusu laboratuvar için bir kimyasal proses seçildi. Radyasyon ışınlarından korunmak için gerekli zırh hesapları yapıldı. Sistemde kullanımı öngörülen ekipmanlarla, kullanılacak çeşitli tesisatın detaylı açıklamaları verildi ve ayrıca genel bir yerleştirme planı hazırlandı. Tasarımı yapılan laboratuvar, ODTÜ Makina Mühendisliği Bölümü, Nükleer Teknoloji Laboratuvarının alt katına yerleştirilebilecek biçimdedir /1/.

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INTRODUCTION

The recovery and purification of fissionable materials in irradiated fuel elements is the principal reason for the reprocessing of fuel. Pu^{239} which is the fissionable material produced in the reactor is the most desirable fuel for the fast breeder reactors. Aside from its fissionable character, Pu^{239} is also the most strategic material for nuclear weapons which makes its recovery vital for military reasons. The amount of fissionable material in nuclear core decreases gradually as the reactor operated. Reactivity decrease can be compensated to certain extend by burnable poisons or control rods. Ultimately, the decrease of the fissionable material will make the reactor subcritical and fuel should be removed from the reactor. The remaining fissionable material in the depleted fuel is too valuable to discard. A less important reason for the reprocessing is the recovery of some fission products, which are potentially useful as radiation sources.

FUEL CYCLE

Fuel cycle consists of (a) Production of fuel material, (b) Fabrication of fuel elements, (c) Use of fuel elements in the reactor, (d) Reprocessing of the irradiated fuel elements and (e) Discharge or storage of radioactive waste. In this cycle, MTA has built a pilot scale plant for the production of yellow-cake. No other experience is available in Turkey.

The most important factors in reprocessing are separation of fission products. These fission products include all the elements with mass numbers from 72 through 161. Most of these products decay by beta emission to more stable isotopes. In reprocessing plants thermal energy and radiation effects of these fission products play an important role. These factors shall depend on fission rate, irradiation duration and cooling period after irradiation. For any isotope, for an irradiation period of "T" and a cooling period of "t" number of atoms is given by

$$N_i = \frac{\Sigma_f \phi y_i}{\lambda_i} \cdot (1-e^{-\lambda_i T})\, e^{-\lambda_i t}$$

where Σ_f : fission cross section, y_i : yield, λ_i : decay constant. This equation is valid for almost all of the fission products. At the end of six month's

cooling period only long lived fission products survive.

A common specification of the permissible activity remaining in separated and decontaminated uranium is that activity not exceed that of natural uranium. The cooling period usually depend on either the decay of U-237 to Np-237 or decay of I-131 to negligible amounts. Cooling period for the selected reference fuel which is considered as being irradiated 585 days at 10^{14} n/cm^2-sec neutron flux is 160 days. To be on safe side cooling period set at minimum of one year.

REPROCESSING

The fuel bundle to be processed is made of 19 of about 50 cm long, 1,5 cm diameter fuel elements. Fuel bundle has about 8 cm diameter and weigh 17 kg. Fuel elements are joined by end-plates at the bottom and top. For all of the reprocessing methods separation of end-plates by mechanical means is required. Cladding on the fuel elements may either be removed mechanically or chemically. In any case chopping of the fuel element is required. For this laboratory fuel elements shall be chopped together with cladding and dissolved.

The following methods may be used after mechanical chopping:

(a). HF-HNO_3 dissolution,
(b). Hydrochlorination-HNO_3 dissolution,
(c). Oxidation-HNO_3 leaching,
(d). Hydrating-HNO_3 dissolution

etc.

The selection of any one of these methods should be decided by chemical engineers. However, Oxidation-HNO_3 leaching method is more suitable for this laboratory. This method is more favorable for Zirconium cladded, natural or low enriched fuel. After the oxidation of the cladding, fuel is dissolved in HNO_3. Zirconium Oxide settles-down and uranium stays in HNO_3.

Solvent extraction method is usually used for aqueous separation. In this method either TBP (Purex Method) or Hexone (Redox Method) is used. The advantages of both methods are not significant. Europian Countries usually prefer to use Purex method therefore Purex method is also proposed for this laboratory study. TBP is used in kerosene type diluent to decon-

taminate, separate and purify uranium and plutonium from fission products and from each other. Generally, all the Purex variations have been demonstrated to be consistently operable without substantial product losses. Table-1 provides the approximate over-all logarithmic gamma decontamination factors and product recoveries, which are expected to be obtained. In the table dF is logarithmic impurity reduction ratio.

TABLE-1. Over-all decontamination factors and recovery of fissile elements /2/

Product Stream	Three-Cycle		Two-Cycle Plus Head-End		Two-Cycle Plus Tail-End	
	dF	Recovery (%)	dF	Recovery (%)	dF	Recovery (%)
Uranium	7.2	99.95	7.2	99.95	6.6	99.95
Plutonium	7.6	99.7	7.6	99.7	7.5	99.7

REQUIRED FACILITIES

Shielding is designed for different zones of the laboratory not to exceed radiation levels given in Table-2. For the calculations of shield thicknesses the calculated values of dose rates given in Table-3 have been used.

TABLE-2. Permissible levels of radiation in different zones of a reprocessing laboratory /2/

Type of Zone	Type of Activity	Permitted level of radiation (mrem/hr)
Hot Cells	Decanning, chopping, dissolution, first-cycle solvent-extraction	2.5
Warm Cells	Second-cycle solvent-extraction	1
Regulated Work Zone	Decontamination, hot maintenance, recovered solvent storage	1
Service	Regulated change rooms, toilets, cold chemical storage areas	0.1

A ventilation system including ducting is proposed not to exceed dose rates of radio-active gases given in Table-4.

TABLE-3. Non-volatile fission products

Isotope	Half Life	Yield (%)	Level at the end of irradiation		Level at the end of 1 yr. cooling	
			of atoms per kg of U	Activity (Ci/kgU)	of atoms per kg of U	Activity (Ci/kgU)
Sr^{89}	53 days	4.8	2.10×10^{20}	859	1.78×10^{18}	8
Sr^{90}	28 yr.	5.8	1.91×10^{21}	40	1.86×10^{21}	39
Y^{91}	58 days	5.8	2.78×10^{20}	1040	3.55×10^{18}	13
Zr^{95}	63 days	6.3	3.27×10^{20}	1125	5.90×10^{18}	20
Nb^{95}	35 days	——	1.87×10^{20}	1158	7.52×10^{18}	47
Ru^{103}	41 days	2.9	9.83×10^{19}	520	2.06×10^{17}	1
Ru^{106}	1 yr.	0.38	7.69×10^{19}	46	3.85×10^{19}	23
Te^{127}	105 days	0.06	5.09×10^{18}	11	4.58×10^{17}	1
Cs^{137}	30 yr.	5.9	1.94×10^{21}	39	1.90×10^{21}	38
Ce^{141}	32 days	5.7	1.51×10^{20}	1023	5.57×10^{16}	1
Ce^{144}	280 days	5.8	1.03×10^{21}	798	4.17×10^{20}	323
Pm^{147}	2.6 yr.	2.6	7.09×10^{20}	162	5.43×10^{20}	124

Study gives details of lighting, fire prevention, materials to be used for construction of interiors and prevention of critical mass formation.

A circular saw operating inside the laboratory pool is proposed for separation of the end plates on the fuel bundle. A chopping machine (pneumatically operated) is proposed to be operated inside the hot cell to chop fuel elements into 12 mm pieces. These small pieces shall be dissolved after oxidation, in HNO_3 and fed into a five stage pump-mix type mixer-settler. Two liter per minute feed rate can easily be obtained in

such a mixer-settler.

Off-gas treatment equipment and equipment for solvent recovery is also proposed and some details are given.

TABLE-4. Permissible limits of some gases in air

Isotope	Half-Life	μc/ml of air
Xe^{133}	5.27 days	$4x10^{-6}$
Xe^{135}	9.2 hr.	$5x10^{-7}$
Kr^{85}	10.6 yr.	$2x10^{-6}$
I^{131}	8.05 days	$3x10^{-9}$
Ru^{106}	1 yr.	$3x10^{-8}$

THE PROPOSED LABORATORY

The laboratory is proposed to be located in the basement of Nuclear Technology Laboratory. Changes inside the laboratory is given without changing civil structure and design. Diesel Generator room, water treatment cell and waste disposal system is supposed to be outside of the building since no space is available to be used for these purposes.

For safety purposes, one element at a time shall be processed. Entrance to the chemical reactive room is provided from the first floor. Chemicals prepared in Nuclear Chemistry laboratory can be transferred to this room.

Circular sawing machine, chopping machine, dissolution tank can be manufactured locally. Manipulators, mixer-settlers and electronic equipment should be imported.

CONCLUSIONS

A general design have been completed. Details of the design require a good team work. Instrumentation required for the laboratory remain to be investigated. It is our opinion that with a good team work this laboratory can be designed, built and operated without much difficulty.

REFERENCES

/1/ Yasav, F., Design of a Research Laboratory for the Reprocessing of Irradiated Fuel, M.Sc. Thesis METU. (1979).

/2/ Stoller, S.M. and Richards, R.B., Handbook V.2, Interscience Publishers, (1961).