INTERNATIONAL CONFERENCE
ON NUCLEAR POWER AND ITS FUEL CYCLE
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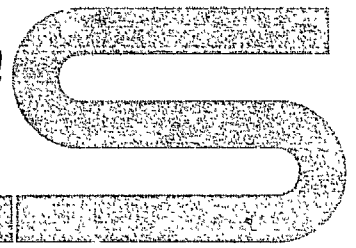

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ITALIAN EXPERIENCE WITH PILOT REPROCESSING PLANTS

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* From European Economic Community - Euratom

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1. THE ITALIAN PROGRAM ON FUEL REPROCESSING

Reprocessing activities at CNEN had a slow start in the early 60es as a consequence of the international situation and prospects existing at that time.
In fact Italy's participation to the Eurochemic joint venture could provide access to the essential know-how; furthermore, at that time, everyone was confident on the abundant availability of reprocessing services to be soon established on commercial basis in several countries.
This situation prompted us to think in terms of a limited national initiative, at R and D level, to be mainly oriented towards special reprocessing problems as an integration of those international activities aimed at power reactor fuel processing, such as Eurochemic.
Two programs were than started, the Eurex (Enriched URanium EXtraction) Program at Saluggia in the north and the PCUT (Programma Ciclo Uranio-Torio) Program at the Trisaia Center, in the south.
The Eurex Program aimed at the recovery of enriched uranium from material testing reactors, which seemed to be a promising market; the PCUT Program was centered on fuel reprocessing and refabrication techniques related to the Th-U^{233} cycle, as an alternative to the U-Pu cycle.
Each of the two programs called for the construction of a pilot plant of adequate size, so that the experience gained during its design, construction and operation could be of use in possible future industrial activities.
The process in the Eurex plant being based on a new chemical flowsheet, laboratory scale R & D took some time

4. CONCLUSIONS

and only in 1963 was the decision taken to build the plant. The EEC joined the program in 1964 with a financial contribution. A total of seven years was necessary to design, build and commission the plant since Eurex went into first hot operation on October 1970, with the first MTR campaign.
The PCUT plant had a more troubled story.
Its construction, too, ended within 1970, but the commissioning was considerably delayed because of operational difficulties experienced in the remote refabrication process of the mixed U-Th oxide.
On June '71 as a consequence of the decision taken at political level the PCUT program was phased-out, and CNEN was asked to look for alternative uses of the Trisaia Center facilities which included the reprocessing plant.
In the meantime from the early 70es onwards, technical and commercial problems started developing in the international reprocessing scene which suggested substantial revisions of the original domestic program.
For these reasons and in agreement with the interested industry a new assessment of the italian reprocessing program was made in 1974 according to which the two CNEN pilot plants, Eurex and ITREC (Impianto Trattamento Elementi Combustibili, the new name of the PCUT plant) were to start a wide range of experimental activities in the field of the power reactors oxide fuel reprocessing and, respectively, fast reactor fuel reprocessing.
To support these pilot plant activities an adequate R and D work at laboratory scale was also implemented.
The final objective was to gather with these two plants

and laboratory the necessary experience and knowledge which would allow domestic industry to design, build and operate a commercial size reprocessing plant when, by the late 80es, this plant will be justified by the extent of the italian nuclear program.

In fact the need of an extensive R and D work, to adapt the well known Purex technology to the case of high burnup power reactor fuels was demonstrated by the progressive outage of most operating plants.

In addition, under the synergetic combination of international policy, public opinion pressure, issues such as safeguarding of nuclear materials, physical protection of the plants, radioactive effluents control, environment protection, waste final disposal, refuse of reprocessing activities by local communities gained momentum and resulted in further difficulties and limitation.

The once favorable ratio of reprocessing service supply to demand was rapidly upset as a result of reprocessing plants shut-down and lack or delay of new plants coming on stream.

2. THE ITALIAN PILOT REPROCESSING PLANTS

2.1. The Eurex Plant

The Eurex plant located at Saluggia (Vercelli), in Northern Italy is essentially a two solvent extraction cycles plant.

It has been designed to be of "direct maintenance" type and it consists of a block of process cells, housing the process equipment. The "all welded" principle has been followed, in connecting equipment and piping, whereby equipment components requiring maintenance are located in cubicles accessible thru shielded doors.

Already during construction, several additions and improvements, in view of extending reprocessing to other than MTR fuel elements, were incorporated in the plant. For instance a Saint Gobain type chopping machine was installed in an "ad hoc" headend cell located on top of the dissolver cell, and connected, through a shielded tunnel with the storage pool.

The Eurex plant went into hot operation on October 1970. Since then until 1974, three MTR fuel elements reprocessing campaigns have been carried out with basically two chemical flowsheets totalling a cumulative of over 500 fuel elements.

At the end of these campaigns, the plant has been shut down to introduce major improvements and modifications in order to improve its flexibility, operability and maintenance, and to meet most recent safety requirements. These changes include both process (such as new automated make up, pneumatic transfer of analytical sample,

computer controlled analytical methods, off gas treatment, cladding material monitoring and disposal system) and protective measures.

It is also worth mentioning that next year the construction will start of a facility for LLW and MLW treatment and conditioning based on a bitumen embedding process.

With the above modifications and additions Eurex will be converted to a truly industrial pilot plant, where experimental reprocessing campaigns with significant amounts (50 to 100 kg of U per day) of power reactor fuels will be possible.

Later in 1977 a joint AECL-CNEN reprocessing experiment on CANDU type fuel coming from the Pickering reactor will begin. This experiment will test an amine based flowsheet, jointly developed by AECL and CNEN, to recover only Pu in two solvent extraction cycle.

Once the CANDU campaign will be over, the plant will undergo further changes, now in the design stage.

These consist of a new partition cycle, a final Pu purification unit, followed by a sol-gel unit to convert plutonium nitrate to mixed plutonium-uranium oxide.

All these activities will be housed in new process cells and laboratories already existing as a bare concrete structure.

The reprocessing campaigns which will follow, shall be devoted to experimental processing of batches of power reactor fuels: some of them, coming from the Latina and Trino ENEL power plants are already stored in the pool.

2.2. The ITREC Plant

The ITREC plant, located at the Trisaia Center (Matera, in Southern Italy), has been designed and constructed as an integrated reprocessing and refabrication unit. It consists of a chemical process section and of a remote fabrication section. After the dismantling of this last, only the chemical process section of the plant, and its related services, has been put into hot operations.The chemical process section consists, essentially, of a large hot cell, where the process equipment (dissolver, mixer settlers and vessels for one solvent extraction cycle, plus evaporators) is assembled on remotely removable racks and where the connection between the adjacent racks is obtained with swagelock type fast connectors housed in cubicles protected by shielded doors. This flexible arrangement allows, once connectors are disengaged after a light decontamination, to remotely maintain the process equipment by umplugging the different racks and taking them remotely to a decontamination cell. The head-end features an underwater fuel dismantling machine to disassemble fuel clusters and a single-pin cutting machine provided on the top of the dissolver.

The ITREC plant started hot operation on July 1975, when an experimental reprocessing campaign to recover Th and U from UO_2-ThO_2 fuel elements irradiated in the Elk River reactor was initiated.

This campaign is still in progress.

When the present phase of the first reprocessing campaign will be over (end of this year) the ITREC plant will

be used to test under active condition prototypes of advanced equipment such as the solvent extraction fast contactor now under development as a joint research program between CNEN and the Polish Atomic Energy Authority.

The long term task assigned to the ITREC plant is the reprocessing of the irradiated PuO_2-UO_2 mixed oxide fuel, which will be discharged from the experimental fast fuel testing italian reactor, named PEC (Prova Elementi Combustibili).

The modification is now in the stage of a preliminary design. It involves a new mechanical head-end section, a second extraction cycle and a Pu storage and conversion unit.

Coupled to the reprocessing plant, an activity on pilot scale will be started in the next years on HLW conversion to glass. It's worth mentioning that the Trisaia Center is the candidate site where the full range of industrial activities of the back end of the fuel cycle will be located.

3. EUREX AND ITREC OPERATIVE EXPERIENCE.

3.1. General

Before presenting the first results gained from the Eurex and Itrec plants operating experience, it is usefull to give an indication of the amount of financial resources poured in the two programs from 1960 so far (as to Dec.76) which is outlined in table 1 in non actualized money.

TABLE 1

Amount of financial resources (in US$ x 10^6) invested in EUREX and ITREC programs from 1960 to Dec. 76

	Personnel and operating costs	Investments	
		EUREX	ITREC
Research & Development (Lab scale)	2.3		
Design & construction of plants	1.7	22.0	9.0
Plant operation	16.5		
	20.5	22.0	9.0
Total	51.5		

3.2. Eurex plant operating experience

3.2.1. Operation.

In the first active campaign the plant reprocessed 112 MTR fuel elements, from the Ispra-1 reactor, containing appr. 21 kg of highly enriched uranium using a tertiary amine based (tricaprylamine, TCA) solvent extraction scheme to recover and purify uranium.

Active operations were stopped at the end of 1971.

An alternative flowsheet for recovering enriched U with TBP was then adopted and the second campaign was started using this flowsheet. 394 MTR fuel elements were processed and ~ 90 kg of enriched uranium were recovered during the period February 73 - June 1974.

In total, 506 elements, from Euratom and italian research reactors were processed, the cooling time of which varied from two to ten years, whilst the overall fission product content was about 250,000 Ci.

U recovery was higher than 98% with the TBP process whereas with the TCA process losses were slightly higher because of extensive testing of flowsheet conditions with the aim of improving especially purification from transuranic alpha (plutonium) activity (plutonium and neptunium).

Actually with the TCA process the alpha specs of the final uranium product (15,000 disintegration/min gram) were achieved only by a supplementary treatment with a new plutonium removing agent, named HX-70*, developed

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* Organic phase soluble hydroxamic acid.

in research labs, whereas, with the TBP process, two extraction cycleswere enough to obtain a final U product with the requested alpha decontamination (see table 3 which shows the level of the residual impurity in four different batches of final U product).

On the contrary, the TCA flowsheet has shown a higher separation of U from fission products than TBP, except for Ru (as shown in table 2).

More than 99.9% of F.P. was left in 1AW raffinate, which in presently stored in liquid form, awaiting for future conditioning.

All other process liquid effluents are concentrated and stored in liquid form, awaiting for further conditioning in the above mentioned bitumen embedding facility (table 4 shows the amount of the liquid waste produced and stored in the Eurex site).

The distillate, after a second evaporation, is collected together with cooling waters and condensates from the plant, in two open air ponds of 1,000 m^3 each to be monitored and discharged in nearby Dora Baltea river.

Liquid effluents discharged up to date in the Dora Baltea river add up to 180,000 m^3; on the basis of analytical control, the total radioactivity released with this volume is only a few percent of the maximum permissible environmental load for each radioisotope.

Even less is the environmental impact of the radioactivity release associated with the ventilation air discharge (440 millions of m^3 per year); by continuous monitoring techniques, a total release of $\sim$ 11,500 Ci of Kr^{85} over a three years period has been determined, which corresponds

to a few percent of maximum permissible discharge in air for this nuclide.

The reliability of the measurements at the release point has been confirmed by environmental radioactivity measurements on different materials and samples, taken within a range of few kilometers from the plant.

No radiological abnormal situations has occurred during routine operation nor during maintenance or special interventions on active parts or areas of the plants.

The most significant maintenance and modification operations in hot areas have been: decommissioning of chopping machine and its refurbishing with installation of hydraulic actuators outside the cells; the replacement of the impeller rotating mechanism in mixer settlers batteries; replacement of copper-silver iodine scrubbing columns by a new one, of silver zeolite type and installation of a vessel off-gas scrubbing system; improvement of absolute filters system on vessel vent line of 1AW storage tanks; modifications of final product cell, to install new final product storage vessels; replacement of α-contaminated analytical equipment in a direct intervention into the analytical hot cell.

Individual exposure level for plant personnel has been less than 10% of the average dose admitted by law for occupationally exposed workers.

3.2.2. Process control and safeguards.

In order to control chemical process conditions, to verify

the proper operation of the different units of the plants and to account the fertile and fissile material flow throughout the plant, for safety and safeguards purposes, almost 20,000 analytical measurements were made. Table 5 shows the various types of analytical measurements, from simple density or acidity determinations to isotopic analysis or special analysis on final product to detect trace impurities.

The accuracy of the analytical measurements is of particular importance where fissile material control for safeguards purposes is involved. Results of analytical determination, materials balance and physical inventories for the MTR campaigns et Eurex plant are given in paper IAEA-CN-36/315 and will not be described here.

3.3. ITREC plant operating experience

Following the decision to phase-out the remote fabrication activity, only the chemical process section of the plant, together with the mechanical head-end (fuel assembly, dismantling and individual pin cutting) has been put into active operation.

The reprocessing campaign which is still in progress concerns 20 mixed UO_2-ThO_2 fuel elements, irradiated in the Elk River (USA) reactor.

After the dismantling of the fuel element assembly, each pin is cut into 2 cm pieces, which fall into the dissolver. A Thorex "acid deficient" type flowsheet is being used for the codecontamination of U-Th in one extraction cycle.

The solution of thorium and uranyl nitrate concentrated up to 350 g/l is stored, in view of the subsequent U-Th partition campaign.

The fuel elements dismantling operations (today completed for all the elements), have pointed out the necessity of a few modification to the dismantling machine, in order to simplify its operation underwater in the storage pool.

Visual inspection of the leached "hulls" in the decontamination cell, and gamma spectrometric measurements, with Ge(Li), on the same "hulls", shows the very good operation of the pins cutting machine and the complete dissolution of the nuclear material contained. Further dissolution operations performed on the hulls showed no evidence of residuals.

In the extraction cycle a recovery of U and Th exceeding 99% was obtained.

Analytical determinations for process control and fissile material measurements, for safety and safeguards purposes, have shown the adequacy of the chemical flowsheet and, generally, the expected performances of the equipment.

To date, seven out of the 20 irradiated fuel elements have been reprocessed for a total of 15,000 Ci. Each element has a mixed Th-U oxide content of about 30 kg, of which 1.1 kg is U (6% U^{233} - 87% U^{235}); the average specific activity is about 90 Ci/kg of fuel.

For this first campaign the most cooled and less irradiated elements (cooling time: about 10 years; burn-up less than 8,000 MWd/ton) have been chosen.

The campaign has been stopped because the monitoring system on the condensate stream line detected a leak in the AISI 304 L heat exchange section of the high level

waste evaporator of the plant. This evaporator before the hot commissioning of the plant had been operated for about 18,000 hours.

The "rack removal system", which is the special feature of the ITREC plant has proven very effective, in that it has been possible to extract the rack with damaged equipment and decontaminate it to a level which allows the substitution of the damaged part by direct maintenance.

While operating on the rack it has been detected on the bottom of the settlers, the presence of Sb^{125}, adsorbed on MBP and DBP formed as a result of a stop of the batteries for more than 15 days.

The first extraction results show a DF of 10^4 for Cs and $\sim 10^2$ for Ru.

It is maybe worth mentioning here the use of a new level measurement technique on the input accountability tank of the plant, based on the TDR (Time Domain Reflectometry), which has been successfully tested and demonstrated. This technique, together with the gamma scanning on leached hulls have been developed in the frame of a research contract with IAEA and are reported in detail in paper CN 36/315 of this conference.

Liquid effluents discharged up to date in the Ionio sea add up to 10,000 m^3; on the basis of analytical control the total radioactivity released with this volume is only a few percent of the maximum permissible environmental load for each radioisotope.

4. CONCLUSIONS

The italian participation to the Eurochemic research activities and plant design, construction and operation, from one side, and the design construction and operation of the Eurex and Itrec plants with the related R and D activities at laboratory level, the continuous improvements and updating of the two plants, already carried out or foreseen in the next future, with the adoption of the more advanced technology, from the other side, contistutes an integrated "know-how" which can contribute to establishing in, in our country, a reprocessing service, at industrial scale.

According to the Italian Committee for the Economic Planning, this service will be carried out jointly by ENI (Agip Nucleare) and CNEN, in the frame of National Nuclear Energy Plan, as a necessary step for the penetration of the breeder reactors, the best utilisation of Uranium resources and finally for the safest conditioning and volume reduction of the fission product activity prior to final disposal.

TABLE 2

EUREX : INITIAL ACTIVITIES AND DECONTAMINATION FACTORS IN THE FIRST EXTRACTION CYCLE

FISSION PRODUCT	TCA Campaign		TBP Campaign	
	max act. Ci/g U Feed	D F	max act. Ci/g U Feed	D F
Cs - 137	0.7	10^6-10^7	1	10^5
Ce - 144	0.4	$> 10^7$	4	10^5-10^6
Ru - 106	0.2	$5 \cdot 10^3$	3	$3 \cdot 10^4$

TABLE 3

EUREX: URANIUM FINAL PRODUCTS

	1°(TCA)	2°(TBP)	3°(TBP)	4°(TBP)
n° of bottles	20	27	23	30
Volume, l	84.04	106.1	91.2	115.1
U tot., g	14,532.45	30,575	24,430.5	31,675.8
U g/l	173	287	258	276
U-235, %	84.44	83.03	82.95	18.22
Pu, dpm	2,500 *	1,950	15,300	10,600
Np, dpm	8,500 *	2,400	395	212
Ru-106, µCi/l	26.3	15	43	4.7
Cs-137, uCi/l	==	0.1	1.7	5.6
Cr, ppm	332	72	45	15.5
Fe, ppm	26 *	201	95	36.2
Na, ppm	1,700	45	200	18
Ni, ppm	783	155	94	29.2
P, ppm	traces	83	89	34
Br, Cl, I, ppm	15	9.5	55	14

* After treatment with EX-70

TABLE 4

EFFLUENTS STORED IN LIQUID FORM ON EUREX SITE

		m^3	Ci
1)	High level waste	85	~ 250,000
2)	Medium level waste	85	==
3)	Organic waste	19	==

TABLE 5

EUREX: NUMBER OF ANALYTICAL DETERMINATION

(As percent of total analyses)

Routine process control analysis	U	Spectro g/l	7%
		Spectro mg/l	7%
		Fluorim.	25%
		Potentiom.	1%
	Acidity		32%
	Al		8%
	TCA/TBP		1%
	Density		12%
	Gross gamma		2%
	Gross alpha		5%
Special and final product analysis	Coulombometry		20%
	Acidity		15%
	Fission Product		37%
	Transuranic		7%
	Metal impurities		9%
	Non metal impurities		5%
	Density		7%
Quality control and cold reagent analysis	Acids		61%
	Basis		8%
	Al		7%
	Iron		8%
	Mercury		8%
	Carbonates		8%