THE STRATEGY OF CLOSED NUCLEAR FUEL CYCLE BASED ON FAST REACTOR AND ITS BACK-END R&D ACTIVITIES

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Abstract

To develop nuclear energy is inevitable choice for China to meet the requirement of decreasing greenhouse gas emission, at the same time of economic and society development. To ensure sustainable development of nuclear energy, closed nuclear fuel cycle strategy based on fast reactor has to be adopted. Both of recent and next R&D activities of nuclear fuel cycle back-end were introduced in the paper, such as:

— Nuclear energy development and spent fuel accumulation, including fast reactor and ADS development aiming at transmutation long-lived nuclides;
— Commissioning of Reprocessing Pilot Plant for PWR spent fuel, development of advanced PUREX process and hot test of separation both U and Pu in CRARL (China reprocessing and radiochemistry laboratory);
— Minor Actinides separation on laboratory scale;
— Investigation on vitrification of high-level liquid waste, high level waste disposal and its programme.

1. BACKGROUND

Nuclear units operated in China are 45 till 2018, electric power capacity is 45 GWe, and about 8 units may be approved this year. The great demand for energy, with the development of society and economy, makes it become significant strategic option to develop nuclear power actively and efficiently for China's energy security. According to the estimation of experts of the Chinese Academy of Engineering, nuclear capacity of China will achieve 150 GWe and 300 GWe in 2035 and 2050, respectively [1].

It is clearly clarifying that technical route of closed nuclear fuel cycle will be adopted in China. Building advanced nuclear fuel recycling system based on thermal and fast reactors combining with accelerator driven system (ADS) is the main task of next stage.

2. DEVELOPMENT OF NUCLEAR ENERGY AND NUCLEAR FUEL CYCLE

Nuclear energy development and accumulation of spent fuel is shown in Table 1, accumulated amounts of minor actinides (MA) is shown in Fig. 1. Till 2030, about 30000t spent fuel will be accumulated in China, including ~20t MA; Till 2050, 60000t spent fuel will be accumulated and amount of MA will grow up to ~55t.

TABLE 1. NUCLEAR ENERGY DEVELOPMENT AND ACCUMULATION OF SPENT FUEL

	2020	2035	2050
Capacity/GW	58+30	150-180	300
SF/t	7000	30000	60000

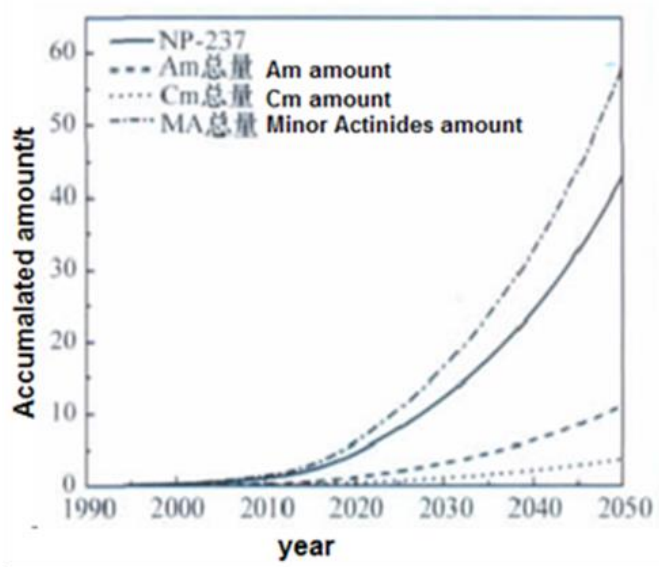

FIG. 1. Forecast for accumulated amounts of MA.

The technical development road-map of the closed fuel cycle was shown in Fig. 2 [2]. The next step is to focus on the second Generation II+ and Generation III of nuclear power to improve the safety of nuclear power systems. Besides the AP1000 in Sanmen county, disposition of independent development Generation III nuclear power Hualong One, No.3 and No.4 generating units in Fangchenggang City and No.5 and No.6 generating units in Fuqing City, are also ongoing, to further improve the Economy of Hualong one.

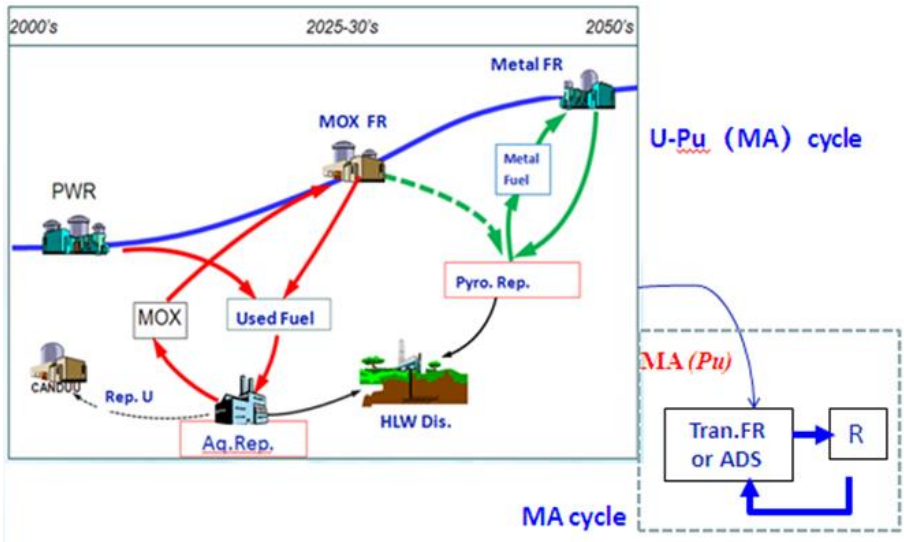

FIG. 2. Middle-to-long-term plan to develop the closed fuel cycle in China.

To improve the utilization ratio of uranium and reduce potential long-term environmental radiotoxicity hazard in radioactive wastes, FRs will be built while developing PWRs to build nuclear fuel cycle based on FR on the industrial scale during 2025-2030. From then, keeping development of FR with higher breeding ratio, meanwhile, and transmutation facility will be developed with a certain proportion. Nuclear fuel cycle based on metal fuel FR will be established around 2050, as shown in Fig. 2 [3]. Currently, the first 600 MWe CDFR has been approved to start construction in Xiapu, Fujian province.

At the same time, Accelerator-Driven Advanced Nuclear Energy System (ADANES) developed by CAS has been approved. ADANES consists of accelerator driven burner (ADB) and accelerator driven regeneration of used fuel (ADRUF). ADB is equal to a long refueling cycle (16-36 years) FR with accelerator driven system, which could burn the recycled fuel containing about 50% of FPs, ADRUF could separate 50% of FPs by means

of pyroprocessing separation method, the rest were converted to bred fuel, utilization ratio of fuel can reach more than 95% [4].

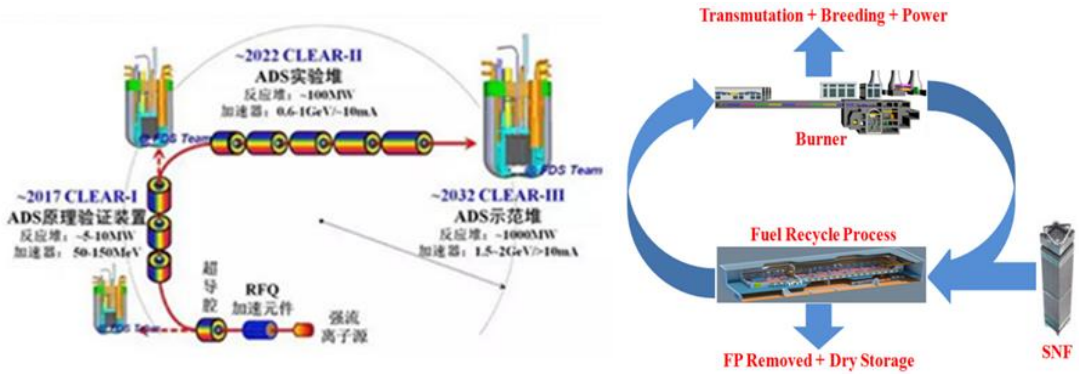

FIG. 3. Accelerator-Driven Advanced Nuclear Energy System (ADANES).

3. TRANSPORTATION AND TEMPORARY STORAGE OF SPENT FUEL

China's spent fuel implements centralized management. A 500t spent fuel storage pools was built in reprocessing pilot plant, and a 1200t spent fuel pools is being constructed in reprocessing demonstration plant in Jinta, Gansu province. The spent fuel transportation system from Daya Bay to Northwest of China has been established now. Spent fuel transport containers were independently developed successfully (Fig. 4).

In the following years, more spent fuel pools will be established in commercial reprocessing plant, and small scale dry storage facilities are planned to be built in Jiangsu and Guangdong province. A transportation system combining spent fuel highway-sea-rail will be further built.

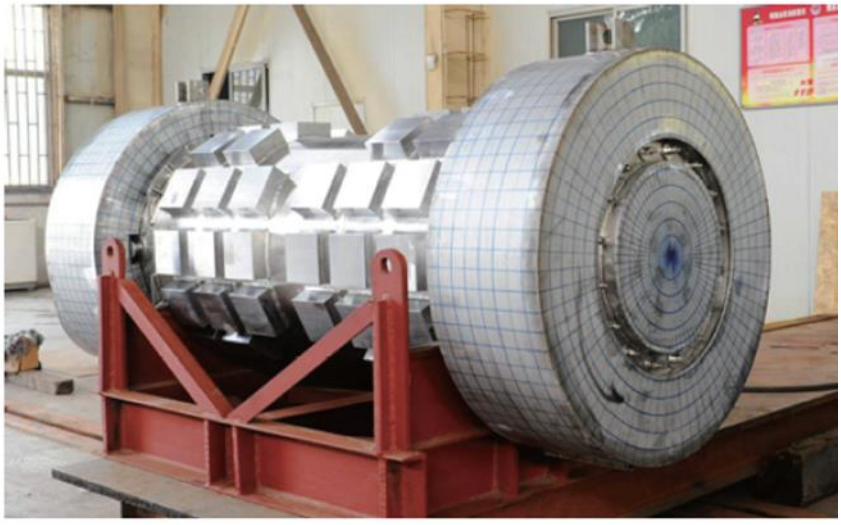

FIG. 4. 1:3 ratio of spent fuel transport container.

4. REPROCESSING ACTIVITIES AND TECHNOLOGY DEVELOPMENT

4.1. Reprocessing pilot plant

Reprocessing pilot plant (Fig. 5), which was currently in stable operation, after its hot-test carried out in 2010. Traditional PUREX process was employed in reprocessing pilot plant, 400 kg/d for head end and product finishing end, 300 kg/d for the chemical separation part, with PuO_2 and UO_3 as products. The purpose of operating reprocessing pilot plant was to demonstrate capability and stability of the used process, equipment and

instrumentation under radioactive condition, and to supply plutonium for MOX fuel used in the experimental FR (CEFR).

FIG. 5. Reprocessing pilot plant.

4.2. Technology R&D

Focus on the roadmap for building nuclear fuel cycle system (Fig. 2), technical research on advanced reprocessing process, equipment, materials and analysis for spent fuel reprocessing of high burnup PWRs were mainly carried out (Fig. 6). For this reason, the government had set up National Reprocessing Technology Research Project to provide technical support for the construction of commercial reprocessing plant around 2035. At the same time, research on pyroprocessing methods are also carried out, laying the foundation for fuel cycle systems based on FR (including ADS) around 2050.

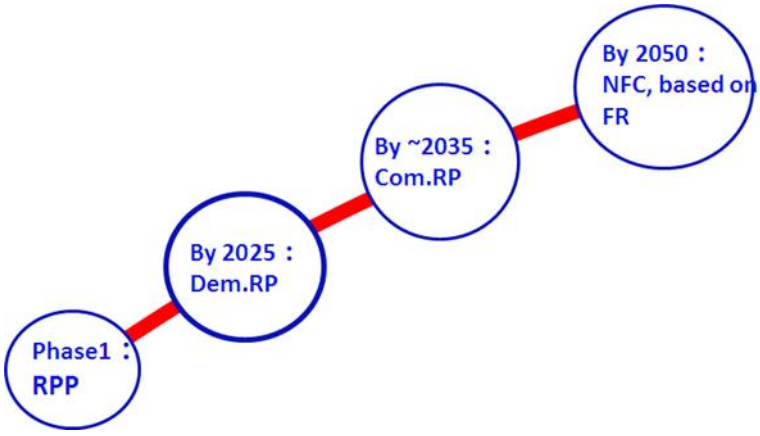

FIG. 6. Roadmap for building nuclear fuel cycle system.

4.2.1. CRARL

In order to perform hot test of reprocessing process, China Reprocessing And Radiochemistry Laboratory (CRARL) was built in Beijing in 2014 (Fig. 7) [5]. Its floor area is 10,000m^2, including five single building as shown in Fig. 7, D block was area of hot cell complex, containing 14 hot cells. The maximum operating radioactivity was 100,000Ci. There are more than 50 glove boxes in C block. The laboratory was put into service in 2015. Dissolution of single high burn-up spent fuel rods and hot test of separation process could be operated in CRARL.

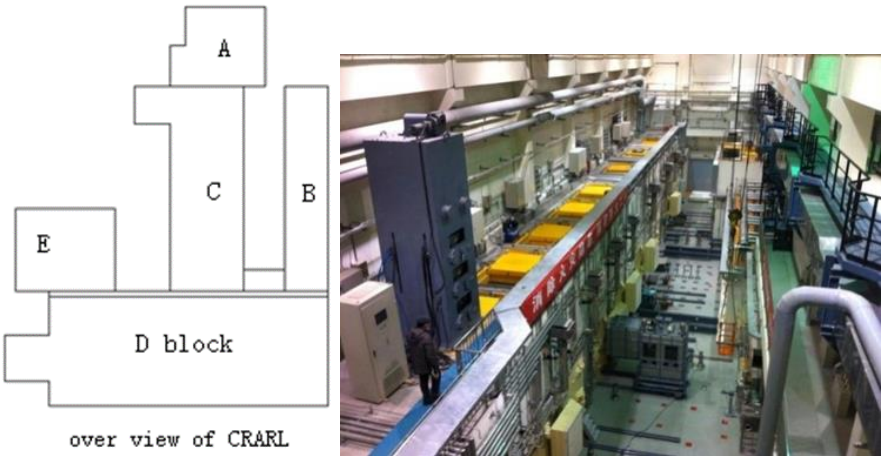

FIG. 7. Building plan of CRARL and hot cell complexes in D block.

4.2.2. Research on advanced process

In traditional Purex process, U(IV)-N2H4 has been successfully demonstrated as reductants in U/Pu separation section [6]. Catalytically oxidization of U(IV)-N2H4 leads to the excessive use of reductants, the existence of excess N_2H_4 leads to a potential explosion risk arising from hydrazoic acid produced by N_2H_4 and HNO2. Futhermore, U(IV) could reduce Np (VI, V) to Np(IV), leads to the dispersive distribution of Np in Purex process.

Experiment results showed organic reductants reduce Pu(IV) at a rapid rate, reduce Np(VI,V), however at a relatively low rate [7]. Thermodynamic and kinetic studies between a series of organic reductants and Pu and Np were studied in China Institute of Atomic Energy (CIAE). Preliminary experimental results showed that N, N-Dimethylhydroxylamine (DMHAN)/monomethylhydrazine (MMH) or hydroxyl-semicarbazide (HSC) were promising reductant reagents for advanced Purex process [8,9].

APOR reprocessing flowsheet:

Based on the primary results, Advanced Reprocessing Process based on Organic Reductants (APOR) had been established as shown in Fig. 8, organic reductants were employed in both U/Pu separation and Pu purification cycles [10].

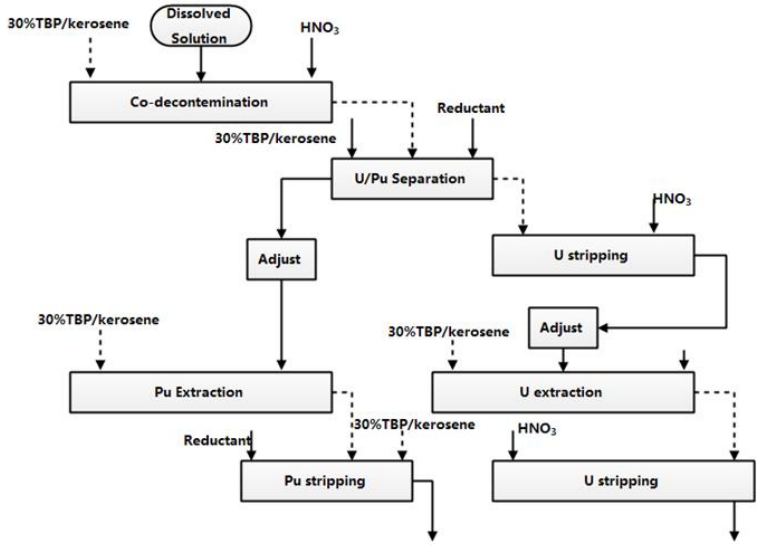

FIG. 8. The flow diagram of APOR.

Hot test of APOR process and results:

Spent fuel from research reactor (3.2% enrichment UO_2 fuel, Zr -2 alloy cladding, burn-up 10000MWd/tU), was used. Fuel rod shearing apparatus and extraction bench are showed in Fig. 9, the APOR process showed in Fig. 8 were employed using DMHAN/MMH as reductants.

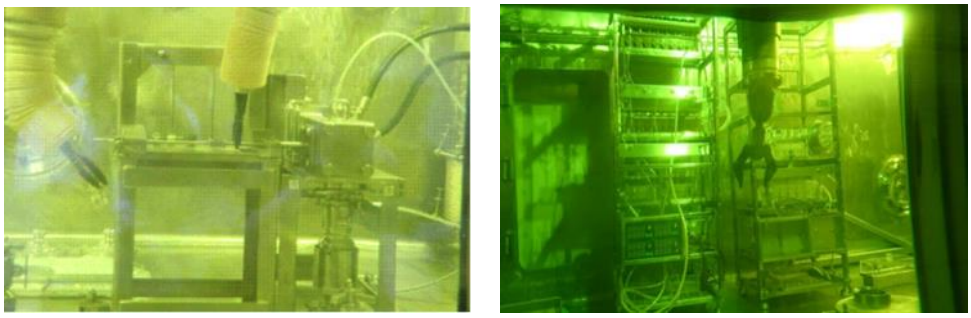

FIG. 9. Spent fuel shearing apparatus and extraction bench.

Hot tests were performed twice, total run time reached to 150 h and amount to 1.7 kgU, key technologies such as spent fuel rods transfer through sealed transfer cart, fuel dissolution and dissolution off-gas treatment, radioactive feed transfer, high efficiency separation and purification of U/Pu, preparation of over 50 analytic methods, storage of radioactive waste, control of liquid level and compressed air, environment emission monitoring and individual dose monitoring were solved during the experiment period.

Two hot tests run safely and smoothly, the experiment results achieved design targets of APOR process. Total uranium recovery rate was 99.94%, total plutonium recovery rate was 99.99%; and γ decontamination factor was more than 3.6×10^5, 1.1×10^6 for U and Pu respectively. Partial Np went to high-level liquid wastes (HLLW) and most of it was controlled into Pu purification cycle and into ILLW [6].

Operating results of CRARL:

Monitoring and analyzing of γ/n radioactive level, radioactive aerosol concentration, radioactive surface contamination, gas and liquid effluent, individual dose were performed, all data were lower than limits for safety analysis reports.

Monitoring of exhaust air from red area: α radioactive aerosol maintained at about 2.10×10^{-3}Bq/m^3 level, β radioactive aerosol maintained at about 2.33×10^{-2}Bq/m^3, below the management limit 6.00×10^{-2}Bq/m^3.

Monitoring of individual dose: No individual dose exceeded 0.03 mSv, all doses were lower than the management limit 2.5 mSv.

Design technology, key equipment performance indicator, safety and reliability of CRARL were comprehensively validated through the operation of hot test.

4.2.3. Minor actinides separation

TRPO process employing trialkylphosphine oxides as extractant to achieve group separation of HLLW had been established by Tsinghua University (Fig. 10) through hot test, tri-valent actinides and lanthanides in stripping solutions were separated by Cyanex301 after denitrification.

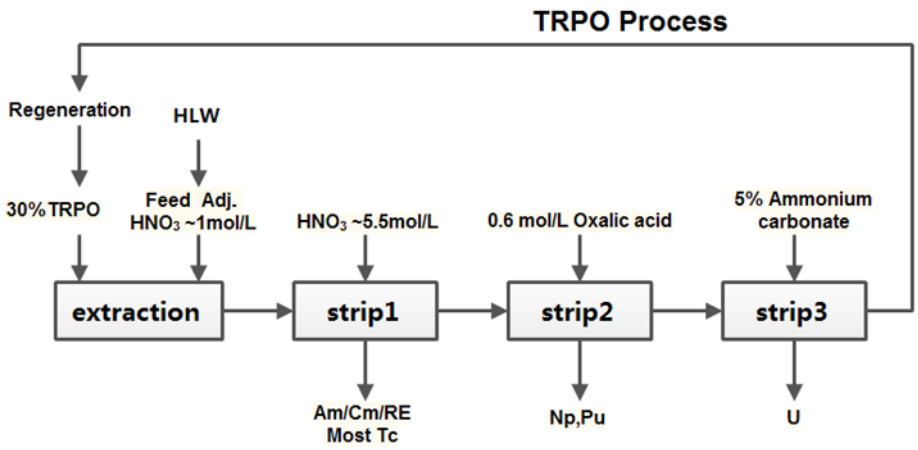

FIG. 10. The flow diagram of TRPO process [3].

HLLW group separated process using TODGA/DHOA extractants had been developed by CIAE (Fig. 11). Warm test results were showed below in Table 2 and 3, showing actinides recovery high and separation factor reasonable with ~4% remain of Am/RE in U/Np/Pu stream:

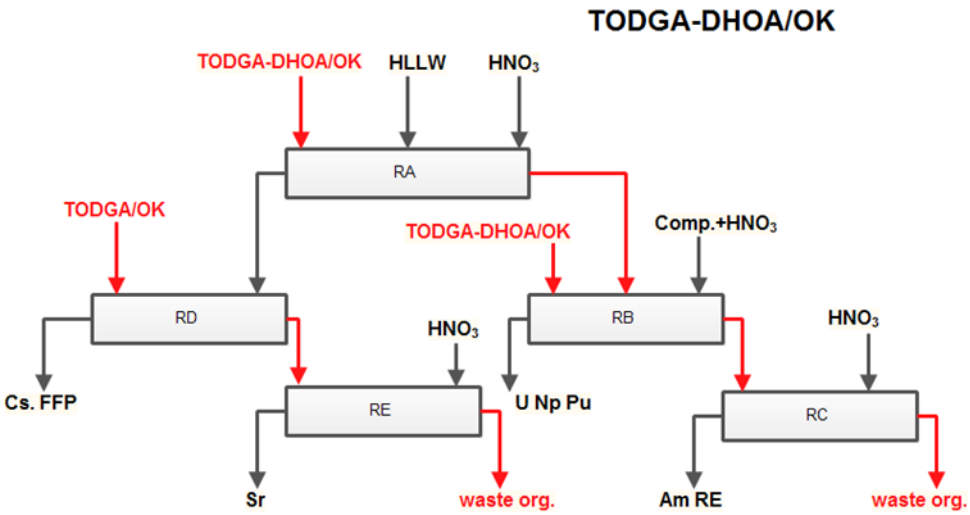

FIG. 11. The flow diagram of TODGA process.

TABLE 2. RECOVERY RATE AND MASS BALANCE OF EACH NUCLIDES IN RA EXTRACTION STAGE

Element	U	Np	Pu	Am	Eu
Recovery/%	99.98	99.99	＞99.99	＞99.99	99.91
Mass balance/%	99.60	96.28	101.23	97.60	99.24

TABLE 3. RECOVERY RATE OF EACH NUCLIDES IN RB AND RC STRIPPING STAGE

Stripping efficiencies in RB contactor %				
U	Pu	Np	Am	Eu
95.87	96.48	99.37	4.01	3.59
Stripping efficiencies in RC contactor %				
U	Pu	Np	Am	Eu
-	-	-	99.68	99.72

4.2.4. Pyroprocessing

Actinide separation over lanthanides via aluminium or gallium cathode-based electrolysis in LiCl-KCl eutectic had been carried out. High efficient separation of Ans over Lns could be achieved through forming An-Al or An-Ga alloys. Pyroprocessing separation method of classified fuels was proposed as shown in Fig. 12.

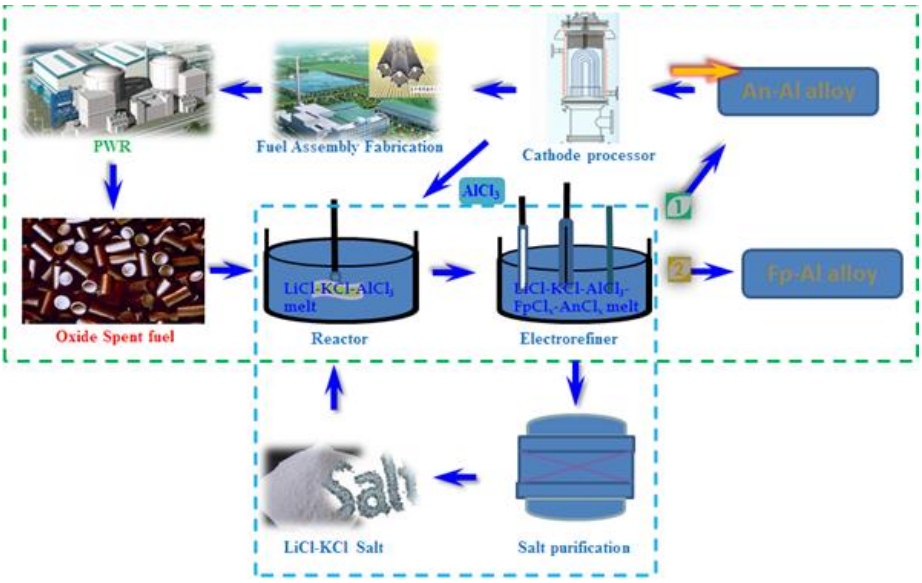

FIG. 12. The flow diagram of Pyroprocessing separation method.

5. RESEARCH OF MOX FUEL FABRICATION

An experimental MOX fuel fabrication facility [11], nearby the reprocessing pilot plant using mechanical blend (MIMAS process, experiment scale, capacity is 500 kgHM/y) has been constructed (Fig. 13). First batch of MOX fuel is under irradiation test in CDFR. Construction of a small MOX fuel fabrication plant at Northwest of China has been approved.

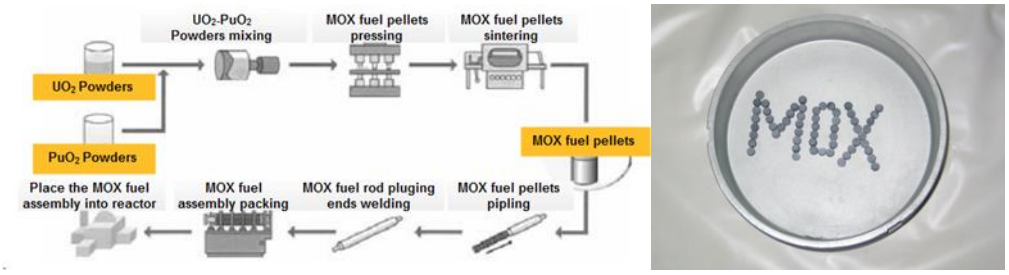

FIG. 13. Fabrication procedure of MOX Fuel assembly and 3.5%Pu-MOX pellet.

6. VITRIFICATION

First vitrification plant adopted electric furnace technology of Germany, cold test will be carried out this year in Guangyuan, old reprocessing plant location. During 2010-2014, the first principle CCM vitrification prototype was set up in China Institute of Atomic Energy. The CCM was Φ300, with its capacity 6 kg/h, max temperature around 1200°C. The bench testing was carried on continuous 24 hours, and accumulated running time was longer than 100 h. During the bench testing, several key technologies were obtained, such as start phase parameters, matching high frequency power with the melter structure, and design of drain device. From 2014～2017, the Φ500 CCM research prototype and HLLW transform system with 30L/h were developed and set up in China institute of Atomic Energy.

7. GEOLOGICAL DISPOSAL OF HIGH LEVEL WASTE

Geological repository will be completed by 2050 [12]. Underground laboratory was planned to be built by 2026, site selection of laboratory completed. Research on security analysis of related research, engineering research, nuclide migration and disposal chemistry are being carried out (Fig. 14).

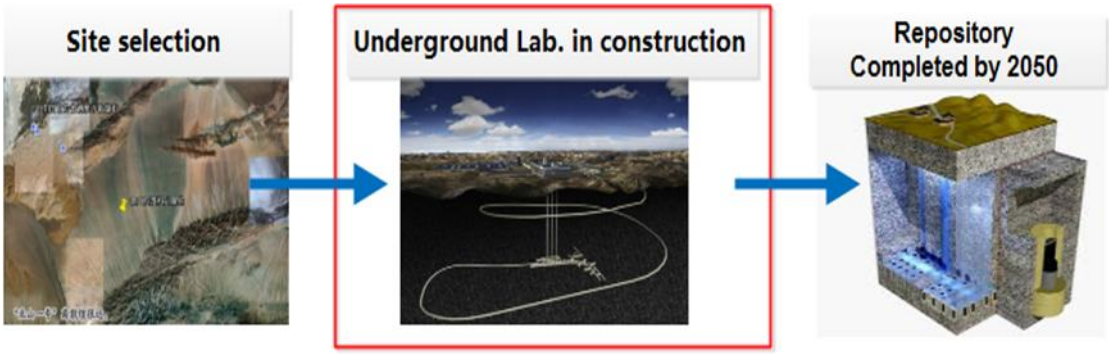

FIG. 14. 3-step strategy of repository development in China.

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