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Review article

Dissolution of mixed oxide(MOX) fuel in nitric acid:A review

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ABSTRACT

The fast neutron reactor is an internationally promising fourth-generation reactor. The main fuel for this reactor is a mixed oxide fuel, and its reprocessing is currently one of the technical challenges being tackled by various countries. One of the difficulties in the reprocessing of mixed oxide (MOX) fuel lies in the nitric acid dissolution process. The high Pu content in MOX fuel can lead to issues such as solvent radiolysis, nuclear criticality, increased insoluble residues, and slow dissolution rates during the nitric acid dissolution process. These challenges have yet to be effectively addressed. This article discusses the chemical aspects of nitric acid dissolution of MOX fuel and investigates the impact of fuel manufacturing processes, the addition of metal catalyst ions, hydrofluoric acid addition, fuel plutonium content, dissolution temperature, and ultrasonic assistance on the nitric acid dissolution of MOX fuel. A review of various countries' engineering practices related to MOX fuel dissolution is presented. Based on the research findings and experiences, a potentially feasible future industrial processing route for MOX fuel is proposed, and future research priorities are outlined.

1. Introduction

Fast reactors exhibit the distinctive capability to initiate a chain fission reaction utilizing fast neutrons [1,2]. They provide notable advantages, including enhanced utilization of uranium resources, transmutation of long-lived waste, and inherent safety features. Consequently, the development of the fast reactor fuel cycle stands as a pivotal element in ensuring sustainable nuclear energy [3–6]. Within the realm of fast reactors, uranium ceramic and metal fuels represent the primary categories employed internationally. Remarkably, mixed oxide (MOX) fuel, comprising uranium dioxide (UO_2) and plutonium dioxide (PuO_2) [7,8], emerges as the most prevalent and well-established fuel type [9–12]. In the context of the fast reactor fuel cycle, efficient reprocessing of spent fuel becomes imperative for the expeditious recovery of plutonium from the aforementioned spent fuel.

Plutonium, being a synthetic element with no natural existence, underscores the pivotal role of reprocessing technology in the fast reactor fuel cycle. Swift extraction of plutonium from spent fast reactor fuel and subsequent reprocessing into new fuel constitute essential steps in this cycle. The new fuel is subsequently reintroduced into the fast reactor, culminating in the realization of a closed-loop nuclear fuel cycle tailored for fast reactors [13]. Typically, hot MOX fuel encompasses 3%–5% PuO_2, occasionally reaching levels of up to 10%. In contrast, fast reactor MOX fuel contains higher proportions, ranging between 20% and 30% PuO_2 [14,15]. In contrast to the uranium oxide (UOX) fuel utilized in traditional pressurized water reactors, the elevated plutonium content in mixed oxide (MOX) fuel presents a range of challenges in post-processing. As of now, a comprehensive post-processing pathway for MOX fuel has

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not been fully established, and there are currently no stable operational facilities dedicated to fast reactor post-processing.

At present, two primary technological approaches are employed for the reprocessing of mixed oxide (MOX) fuel. The non-aqueous processes [16–18] constitute a high-temperature chemical method that leverages the distinct electrochemical properties of various nuclides for separation within a molten inorganic salt medium [19–21]. On the other hand, the aqueous processes [22,23] involve dissolution in nitric acid, followed by extraction and separation [24–26]. While non-aqueous processes offer advantages such as reduced insoluble residue, there remain several challenges that must be addressed before their industrialization becomes feasible. Conversely, the aqueous process, particularly the PUREX process for hot reactor spent fuel, has demonstrated relative maturity, exemplified by France's extensive treatment of MOX spent fuel utilizing existing facilities [27]. Given the rapid advancements in fast reactor technology, the aqueous-based reprocessing of MOX fuel currently stands as the most suitable approach for the Chinese reprocessing strategy [28–30].

The reprocessing of MOX fuel introduces significant differences and challenges compared to uranium oxide (UOX) fuel, particularly in the cutting and dissolution stages [31,32]. The presence of stainless steel casing in fast reactor assemblies significantly impacts the cutting process. The dissolution process encounters two main difficulties: the high plutonium (Pu) content in MOX fuel gives rise to issues such as criticality during nitric acid dissolution, increased insoluble residue, and slow dissolution rates [33–35]. Additionally, caution must be exercised to avoid major modifications in the front-end process, as the intricate nature of nuclear fuel reprocessing necessitates careful consideration and repeated evaluation of potential impacts on subsequent processes [36–38].

Ideally, the dissolution process for MOX fuel should involve minimal or no addition of new elements and utilize nitric acid as the solvent [39–41]. This approach not only minimizes the impact on subsequent processes but also facilitates faster progress towards engineering-scale implementation.

This article provides a comprehensive overview of the dissolution chemistry of MOX fuel, delving into the related research on factors influencing the dissolution of MOX fuel. Additionally, it examines practical experiences gained from MOX dissolution projects conducted in various countries. Drawing upon the insights derived from the research content and accumulated experiences, the article proposes a potentially viable future industrial reprocessing route for MOX fuel. Furthermore, it offers a forward-looking perspective on the key areas of focus for future work.

2. Chemical dissolution of MOX fuel with nitric acid

Following the dissolution of a significant portion of the spent fuel in nitric acid, the resultant dissolution products typically comprise 3 mol/L of nitric acid, 1.0–1.3 mol/L of uranium (U), and a specific concentration of plutonium (Pu) [42]. The dissolution process is typically carried out at elevated temperatures, ranging from 70 to 100 °C, during which the UO_2 undergoes oxidation to form uranyl ions [43,44]. Depending on the specific dissolution conditions, plutonium can exist in either tetravalent or hexavalent ion states [45–47].

$$3\ UO_2 + 4(2+x)\ HNO_3 \rightarrow 3UO_2(NO_3)_2 + 2(1-x)\ NO + 6xNO_2 + 2(2+x)\ H_2O\ (0 < x < 1) \tag{1}$$

The process of dissolving spent fuel represents a solid-liquid reaction process and the reaction is illustrated by Equation (1), encompassing surface oxidation and dissolution. This intricate process unfolds through a self-catalytic reaction mechanism, wherein the reaction products play a role in enhancing the overall reaction rate. Essentially, after overcoming the initial sluggish period, known as the induction period, the dissolution rate experiences a substantial increase. Researchers have delved into the effects of nitrogen oxides and nitric acid concentration on this dissolution process, revealing that the concentrations of nitric acid and nitrogen oxides exert a notable influence [48]. Notably, under typical dissolution conditions, the oxidation process involving nitrous acid outpaces that involving nitric acid [49], providing a mechanistic explanation for the inherently self-catalytic nature of the nitric acid dissolution process. The reaction equations for the reaction of uranium dioxide with nitric acid and nitrous acid are shown in Equations (2) and (3).

$$UO_2 + NO_3^- + 3\ H^+ \rightarrow UO_2^{2+} + HNO_2 + H_2 \tag{2}$$

$$UO_2 + 2\ HNO_2 + 2\ H^+ \rightarrow UO_2^{2+} + 2\ NO + 2\ H_2O \tag{3}$$

Unlike uranium dioxide, plutonium dioxide has a comparably lower solubility in nitric acid. However, if the amount of plutonium (Pu) in MOX fuel is relatively small, it can still dissolve into nitric acid and the reactions are shown in Equations (4)–(6). On the contrary, if the Pu content is excessively high, the solubility significantly decreases [50].

$$PuO_2 + 4\ H^+ \rightarrow Pu^{4+} + 2\ H_2O \tag{4}$$

$$PuO_2 + NO^{3-} + 2H^+ \rightarrow PuO_2^+ + NO_2 + H_2O \tag{5}$$

$$PuO_2 + NO^{3-} + 3H^+ \rightarrow PuO_2^{2+} + HNO_2 + H_2O \tag{6}$$

Both UO_2 and PuO_2, as well as MOX fuel, are amenable to dissolution in nitric acid. However, their dissolution rates exhibit notable differences, primarily attributed to the PuO_2 content. The observed slower dissolution rate at higher PuO_2 content finds its explanation in the examination of the crystal structure. Both UO_2 and PuO_2 predominantly possess a face-centered cubic structure [51–55] and the lattice structure is shown in Fig. 1. Notably, uranium (U) readily undergoes oxidation by nitric acid, resulting in the formation of UO_2^{2+} ions and leading to the transfer of the uranyl ion oxide lattice into the solution.

In contrast, plutonium (Pu) is less prone to oxidation into plutonyl ions by nitric acid. When the PuO_2 content is relatively low, Pu occupies lattice positions originally held by U. As U undergoes oxidation by nitric acid to form UO_2^{2+} and enters the solution, it disrupts the lattice, facilitating the entry of Pu as Pu_4^+ into the nitric acid solution. However, when the adjacent atoms within the lattice are also Pu atoms, the crystal structure remains more stable in the presence of nitric acid. This stability consequently leads to the formation of insoluble residues rich in Pu within MOX fuel [56].

3. Factors affecting the dissolution of MOX fuel

The dissolution of MOX fuel is a highly intricate process influenced by various factors. Key factors include fuel manufacturing processes, the addition of high-valence metal ion oxidants, plutonium content in the fuel, dissolution temperature, acidity, and other factors.

3.1. Effect of fuel manufacturing processes on the dissolution of MOX fuel

The solubility of MOX fuel is intricately tied to its production process. Specifically, elevating the sintering temperature of mixed oxide fuel from 1400 °C to 1700 °C results in a notable increase in fuel solubility, ranging from 3% to 5% (mass fraction). This correlation is visually represented in Fig. 2, which delineates the influence of PuO_2 content and sintering temperature on MOX dissolution. The graphical representation underscores that the solubility of UO_2-25%PuO_2 fuel pellets experiences the most significant enhancement with rising temperature [57].

The solubility of PuO_2 exhibits variation based on its source, with the order of increasing solubility being calcined metal $>$ calcined oxalate $>$ calcined nitrate. PuO_2 derived from higher temperature calcination displays lower reactivity attributed to its intact lattice structure. Notably, research by Bjorklund and Staritzky [51] revealed that the refractive index of PuO_2 derived from Pu(IV) oxalate increased from 1.9 to 2.4 as the calcination temperature rose from 150 °C to 1000 °C. In contrast, no change in refractive index was observed for oxides derived from metal calcination at 170 °C and higher temperatures. X-ray diffraction patterns for the oxide at 150 °C appeared weak and diffuse, while at 1000 °C, they became sharp. With an increase in the calcination temperature, the dissolution rate of PuO_2 in HCl-KI decreased significantly. These findings suggest that the originally distorted PuO_2 structure becomes more complete at higher calcination temperatures, impacting the optical properties of the high-temperature oxide.

3.2. The effect of high-valence metal ion oxidants and HF on the dissolution of MOX

In summary, to enhance the solubility of the PuO_2 lattice in aqueous solutions, two distinct approaches can be considered: oxidizing Pu to form plutonyl ions or forming complexes to increase its solubility. For instance, the addition of high-valence metal ion oxidants can be employed to oxidize Pu, or the introduction of hydrofluoric acid can facilitate the formation of complexes in aqueous environments.

3.2.1. The effect of Ce(IV) on the dissolution of MOX

According to reports from the US Exxon Nuclear Fuels Recovery and Recycle Center, irradiated mixed oxide residues can be

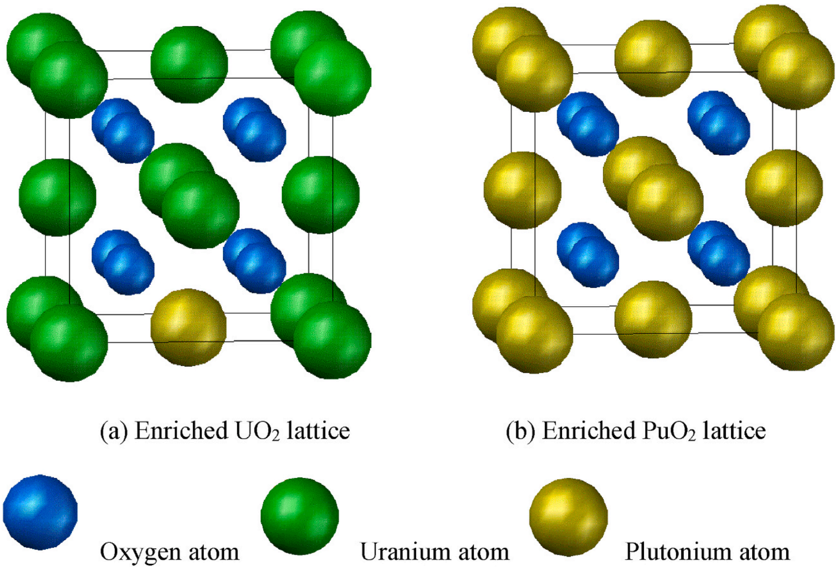

Fig. 1. Lattice of enriched UO_2 and enriched PuO_2 in MOX fuel after irradiation (a) Enriched UO_2 lattice, (b) Enriched PuO_2 lattice.

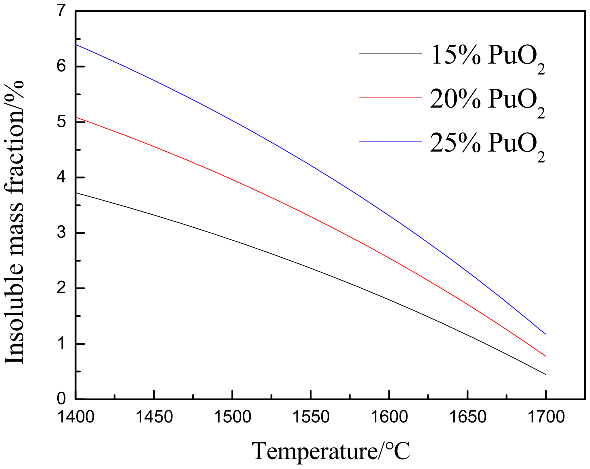

Fig. 2. Influence of PuO_2 content and sintering temperature on the solubility of PuO_2-UO_2 fuel containing PuO_2 (prepared from calcined oxalate) [57].

effectively dissolved using a solution of 4 mol/L HNO_3-0.05 mol/L Ce(IV). Within a period of 2 h, 88% of PuO_2 is dissolved, and this dissolution rate increases to 99.84% after 4 h. The addition of Ce(IV) ions at concentrations ranging from 0.03 to 0.1 mol/L significantly enhances the dissolution rate of PuO_2, demonstrating a notable improvement by a factor of 10^3-10^4 [57]. These findings highlight the substantial positive impact of including Ce(IV) in the nitric acid solution, leading to enhanced solubility of PuO_2. Notably, Ce(IV) proves highly effective in improving the dissolution of insoluble residues containing a substantial amount of plutonium. However, it's crucial to note that directly introducing Ce(IV) during the dissolution of spent fuel could result in the complete conversion of plutonium to the hexavalent state. This outcome is undesirable for subsequent extraction processes and may lead to the volatilization of a significant amount of ruthenium in the form of RuO_4 [58].

3.2.2. The effect of Ag(II) on the dissolution of MOX

The concept of mediated electrochemical oxidation (MEO) was first proposed in the 1980s and has since been utilized for the dissolution of PuO_2 [56,59]. PuO_2 presents challenges in terms of dissolution in nitric acid systems, particularly in the absence of fluorine. In the MEO process, Ag(II) acts as a catalyst, and the anodic oxidation process is employed to generate Ag(II). The highly oxidizing Ag(II) then oxidizes PuO_2 to PuO_2^{2+}, facilitating its dissolution.

In an electrolytic cell, separated by a porous membrane (such as porous ceramics), the anode chamber contains PuO_2 fuel, catalysts (such as Ag^+ or Ce^{3+}), and nitric acid. The cathode chamber exclusively contains nitric acid (at concentrations ranging from 10 to 16 mol/L). The anode is typically composed of platinum, with a layer of platinum metal deposited on the surface of titanium metal. The titanium metal serves as the cathode. Within the anode region, Ag^+ (or Ce^{3+}) undergoes electrolytic oxidation at a high anodic potential. The chemical reaction equations for the entire process are shown in Equations (7)–(11).

$$Ag^+ \rightarrow Ag^{2+}\left(\text{或}Ce^{3+} \rightarrow Ce^{4+}\right) \tag{7}$$

$$2Ag^{2+} + PuO_2 \rightleftarrows PuO_2{}^{2+} + 2Ag^+ \tag{8}$$

reduction reaction occurring at the cathode

$$2H^+ + NO_3^- + e^- \rightleftarrows NO_2 + H_2O \tag{9}$$

$$4H^+ + NO_3^- + 3e^- \rightleftarrows NO_2 + 2H_2O \tag{10}$$

The overall reaction is:

$$3PuO_2 + 2NO_3^- + 8H^+ \rightleftarrows 3PuO_2^{2+} + 2NO + 4H_2O \tag{11}$$

The catalytic electrolysis method for dissolving PuO_2 can be conducted at near room temperature, offering several advantages. These include low acid requirements, fast dissolution rate (twice as fast as the HNO_3–HF method), and complete dissolution of PuO_2. This method is suitable for dissolving poorly soluble PuO_2 and $(U, Pu)O_2$ spent fuel. The dissolution rate of PuO_2 is not significantly influenced by its specific surface area or calcination temperature. Maintaining the necessary concentration of Ag^{2+} (or Ce^{4+}) ions through electrolytic oxidation in the anode chamber is sufficient. Experimental evidence indicates that electron transfer within the Ag^{2+}/Ag^+ ion pair is easily achieved. However, there are some drawbacks to this method, including complex equipment, higher corrosiveness, and uncertainties regarding the potential impact of Ag^{2+}/Ag^+ on subsequent processing steps.

3.2.3. The effect of HF on the dissolution of MOX

Plutonium dioxide (PuO_2), especially when produced at high temperatures, presents a significant challenge due to its high insolubility using conventional dissolution methods [60]. Holley et al. [51] evaluated the effectiveness of various solvents, ranking them as follows: 85–100% H_3PO_4 (200 °C) > 10 M HNO_3-0.05 M HF > 5 M HF. Although boiling HNO_3–HF is commonly employed for PuO_2 dissolution, the process is slow when dealing with oxide compounds formed at high temperatures.

In the 1980s, France conducted dissolution experiments on MOX fuel at the Fontenay-aux-Roses hot cell, achieving enhanced dissolution of insoluble residues using 11 mol/L HNO_3+0.05 mol/L HF [61]. For French fuel with a plutonium content of 5% (mass fraction) before irradiation, 3.9% after irradiation, a burnup of 30,000 MWd/t, and short fuel segments measuring 30 mm in length, dissolution was carried out using 6.2 mol/L HNO_3. After 6 h of boiling nitric acid dissolution, 92% of the fuel was dissolved. The remaining undissolved portion underwent filtration, washing, and drying before being calcined at 400 °C for 8 h. The resulting solid residue was then dissolved in 11 mol/L HNO_3+0.05 mol/L HF, with the dissolution process lasting 12 h at boiling temperature. Mass spectrometry was employed to determine the uranium and plutonium content in the dissolution solution. The overall relative loss of plutonium was found to be 0.017%, while the loss of uranium was 0.009%. These results highlight the capability of 11 mol/L HNO_3+0.05 mol/L HF to effectively dissolve insoluble plutonium-rich residues in nitric acid, showcasing its potential in addressing challenges associated with PuO_2 dissolution.

3.3. The effect of the plutonium content in the fuel on the dissolution of MOX

Experiments conducted by Vollath [48] to investigate the dissolution of MOX fuel in nitric acid with varying plutonium contents yielded insightful results, as illustrated in Fig. 3. The findings distinctly reveal a substantial decrease in the solubility of MOX fuel in nitric acid when the plutonium content surpasses 35%. Notably, at a plutonium content of 70%, the solubility of MOX fuel becomes almost negligible, rendering it practically insoluble.

Due to the high cost associated with Pu materials, researchers frequently opt to use Ce as a substitute in their studies [62]. Desigan [63] undertook a study to examine the impact of Pu content on the nitric acid dissolution process, utilizing Ce as a surrogate. Dissolution experiments were conducted using an HNO_3 solution with an initial concentration of 8 mol/L at a temperature of 80 °C. The experimental findings are presented in Fig. 4.

The observations from Fig. 4 clearly indicate that the dissolution curve follows an S-shaped pattern, indicative of a self-catalytic reaction. As the proportion of Ce increases, the dissolution time required to achieve the desired solubility also increases. However, even with a 35% Ce content in the MOX fuel, complete dissolution can still be attained. The time needed for complete dissolution approximately doubles when 35% Ce is introduced, underscoring that MOX fuel dissolution presents a significantly more challenging process compared to UO_2 fuel.

3.4. The effect of the dissolution temperature on the dissolution of MOX

Temperature plays a crucial role in the dissolution of MOX fuel. Desigan [63] conducted a comprehensive investigation to explore the impact of temperature on the dissolution of $(U_{0.78}Ce_{0.22})O_2$ MOX in an 8 mol.L-1 nitric acid solution. The findings of this study are visually presented in Fig. 5.

Upon analysis of Fig. 5, it is evident that the dissolution curve of nitric acid exhibits an S-shaped pattern, indicative of a self-catalytic reaction. A comparison of the curves at 343K and 373K reveals that the time required for complete dissolution at 373K is nearly half of that at 343K, despite the relatively modest temperature difference of 30 °C. This observation underscores the significant impact of temperature on the dissolution of MOX fuel.

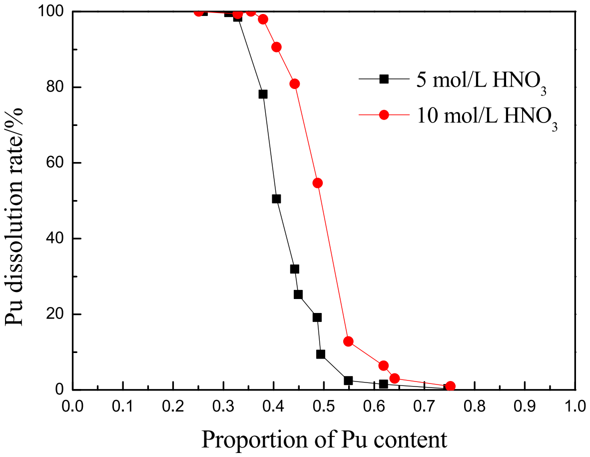

Fig. 3. Solubility of MOX fuel at different plutonium contents in 5/10 mol.L-1 nitric acid solution [48].

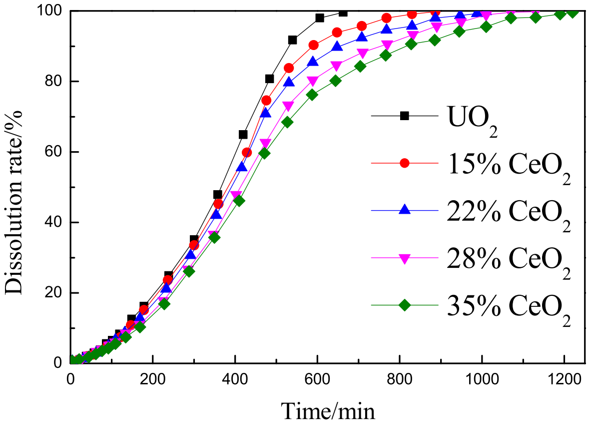

Fig. 4. The influence of Ce content on the nitric acid dissolution of MOX (U, Ce) fuel [63].

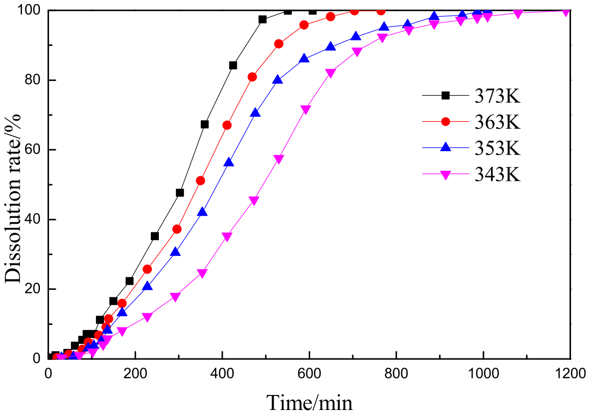

Fig. 5. The effect of initial acid concentration on the dissolution of $(U_{0.78}Ce_{0.22})O_2$ MOX [63].

3.5. *The effect of ultrasonic on the dissolution of MOX*

The Marcoule Ultrasonic Chemistry Separation Laboratory in France conducted a series of fundamental studies on the application of ultrasonic technology in the reprocessing of spent fuel, including the dissolution of CeO_2 and PuO_2 through ultrasonic waves [64]. They used CeO_2 as a surrogate for unirradiated PuO_2 and dissolved CeO_2 powder in a 4 mol/L nitric acid solution using ultrasonic waves at frequencies of 20,500 Hz and 1700 kHz [65]. The experimental results revealed that under the influence of ultrasonic waves at 20,500 Hz, the dissolution rate of CeO_2 powder was 3–4 times faster compared to traditional mechanical stirring. However, under the high-frequency ultrasonic waves at 1700 kHz, the dissolution rate was even lower than that at 20,500 Hz. Subsequently, they conducted dissolution experiments using PuO_2 powder, studying the dissolution of 100 mg of PuO_2 powder in a 4 mol/L HNO_3 solution with the addition of 0.5 g AgO under a 20 kHz ultrasonic wave frequency [65]. The research findings demonstrated that the dissolution rate under the influence of ultrasonic waves was three times higher than that without ultrasonic waves.

4. Engineering practices related to the MOX dissolution in various countries

4.1. *France*

As of now, France has provided reports on the MOX fuel treatment process, and various research studies have been conducted by prominent French organizations, including CEA, COGEMA, and SGF, to validate the feasibility of MOX fuel reprocessing. The initial stage involved a comprehensive investigation into the dissolution behavior of MOX fuel at the Marcoule Laboratory. Following this, semi-industrial scale hot experiments were conducted at the Marcoule pilot plant. In 2004, COGEMA successfully executed the first full-scale industrial operation of MOX fuel reprocessing at the UP2 plant [66–69].

In 1967, France initiated laboratory research on the dissolution of MOX fuel in fast reactors. Subsequently, in 1987, comprehensive studies were conducted during the reprocessing of pressurized aqueous reactor MOX fuel at the Fontenay-aux-Roses hot cell. These studies covered various aspects, including dissolution performance, MOX fuel manufacturing processes, burnup, and dissolution conditions. Additional research explored supplementary dissolution methods for insoluble materials, and the composition of dissolution residues was identified [57,61].

The fuel utilized in these French experiments had a burnup of 30,000 MWd/tU, featuring a plutonium content of 5% (by weight) before irradiation, which decreased to 3.9% after irradiation. The fuel was in the form of short segments, measuring 30 mm in length. Batch dissolution commenced using a 6.2 mol/L HNO_3 solution, starting at an initial temperature of 45 °C. Within 35 min of introducing the feed, the dissolution temperature was raised to 70 °C, and boiling of the solution began after 85 min. After 3 h of dissolution under boiling conditions, 80% of the fuel was dissolved. Following 6 h of boiling dissolution, 92% of the fuel was dissolved. Upon completion of the dissolution process, the solution underwent filtration, and the solid residues were washed, calcined, and further dissolved for 12 h using an 11 mol/L HNO_3 + 0.05 mol/L HF solution under boiling conditions. Table 1 provides information on the relative losses of uranium and plutonium under conventional dissolution conditions, determined by measuring their concentrations in the solution.

In November and December 2004, a significant milestone was achieved with the successful processing of MOX spent fuel at the UP2-800 plant in La Hague [27,66,69,70]. Specifically, 10 tons of Grafenreinfeld MOX spent fuel were dissolved at the R1 front-end facility of the UP2-800 plant. Over the course of four batches between 2004 and 2008, approximately 50 tons of MOX spent fuel with varying burnup levels and plutonium content (ranging from 4% to 8% before irradiation) were processed at the UP2-800 plant. This accomplishment affirmed the feasibility of conducting industrial-scale reprocessing of MOX spent fuel. The fuel used in these French experiments was manufactured using the MELOX process. It's crucial to note that during the dissolution experiments, non-irradiated fuel produced through the MELOX process demonstrated insolubility with 0.5–1% Pu content. Dissolution under boiling conditions with 10 mol/L nitric acid was performed for a duration of 10 h. Consequently, when dissolving one ton of fuel containing 5% Pu, an estimated 250–500 g of insoluble Pu would remain.

The normal operating conditions for the R1 front-end facility in processing depleted fuel in a hot reactor involve using an acidity level of 3 mol/L, maintaining a temperature of approximately 90 °C, and ensuring a fuel residence time of 2 h. However, it has been observed that under these conditions, MOX fuel, especially components with a high plutonium (Pu) concentration, cannot dissolve completely.

In 1992, France conducted two reprocessing campaigns at the UP2-400 plant for MOX fuel with a burnup of around 35,000 MWd/t and a Pu content of approximately 4%. The batch dissolution process was conducted under specific conditions, including an initial acidity level of 6 mol/L, a final acidity level of 3 mol/L, and a dissolution temperature of 90–92 °C. It was found that the final amount of insoluble plutonium was less than 0.1% [69]. These results led France to conclude that satisfactory dissolution of MOX fuel could be achieved by improving the operating conditions of the R1 facility without major modifications to the UP2-800 equipment.

The new operating conditions introduced for improved dissolution of MOX fuel are as follows: an acidity level of 5 mol/L, a temperature of approximately 90–92 °C, and a dissolution time of around 7 h. These conditions were chosen for specific reasons. Limiting the acidity to 5 mol/L helps prevent excessive corrosion of the dissolution wheel, thereby extending the overall lifespan of the facility. Similarly, maintaining the temperature at 90 °C helps avoid the formation of foam and MoZr fouling, ensuring smoother operations.

Between 2004 and 2008, approximately 50 tons of MOX fuel were processed in four batches. The operational results demonstrated that by increasing the acidity of the dissolution solution from 3 mol/L to 5 mol/L, elevating the temperature to 92 °C, and extending the residence time to 7 h, effective dissolution of MOX fuel could be achieved. The characteristics of the fuel are summarized in Table 2, while Table 3 presents the results of each experiment carried out at the front-end and back-end plants. The experiments were conducted progressively, with increasing burnup, plutonium content, fuel tonnage, and processing flow rate.

It is worth noting that by 2013, the UP2-800 facility had successfully treated and processed approximately 60 tons of irradiated MOX fuel.

4.2. Japan

In 2007, the Japan Atomic Energy Agency initiated the FaCT (Fast Reactor Cycle Technology) program as part of the "NEXT" process (New Extraction System for Transuranium Elements) for advanced aqueous reprocessing [71]. Japan considered adopting a powder feed method to enhance dissolution and obtain a high-concentration heavy metal solution of 400-500gHM/L required for subsequent processes. Therefore, they believed it was necessary to shear the spent fuel into short segments or even powderize it during the shearing process [72].

To improve the dissolution performance, experiments were conducted using JOYO fast reactor-irradiated MOX fuel (MK-2 core fuel with an average burnup of 55,000 MWd/t, cooled for 6800 days, and an initial PuO_2 content of 23%–29%). The relationship between the dissolution rate and fuel size was studied under conditions of 95 °C and 8 mol/L HNO_3. The experimental parameters included the

Table 1
Relative losses of uranium and plutonium [57].

Dissolution time under boiling conditions (hours)	Relative losses/%	
	U	Pu
3	0.009	0.028
6	0.009	0.017

Table 2
Experimental results 2004–2008: MOX fuel characteristics [66].

test	D_1	D_2	D_3	D_4
date	November–december 2004	September–November 2006	November–december 2007	September–October 2008
Fuel source	Grafenreinfeld	Graf.+ Grohnde		Goesgen
Manufacturing process	OCOM	OCOM	OCOM + MIMAS	MIMAS
Plutonium content before irradiation	4–4.25 %	4–4.26 %	3.9–4.4 %	7.2 %
Plutonium content at disposal	~3 %	~3 %	~3 %	~5 %
Initial amount of insoluble plutonium	0.13 %	0.20 %	0.30 %	0.7 %
Burn-up (MWj/)	30,500–35,500	33,500–43,000	36,000–45,000	51,000–53,500
Cooling time (years)	>10	>10	>10	~6

Table 3
Experimental results 2004–2008: Dissolution Parameters and results [66].

test	D_1	D_2	D_3	D_4
Fuel tonnage	10.6	16.5	31.3	5.1
Flow rate (tHM/day)	0.97	1.6	1.75	2.3
Dissolved acidity and temperature (M,°C)	5.1/92	5.1/92	5.1/92	5.1/92
Insoluble particles (kg/tHM)	~3.5	~4.5	~4.6	*
Plutonium content in insoluble particles	~0.13 %	~0.16 %	~0.16 %	*
Ratio of insoluble plutonium to total plutonium at disposal	~0.014 %	~0.03 %	~0.02 %	*

size of the fuel segments or powder, with a cutting method called "Short Stroke" used to efficiently cut the fuel rods into shorter segments [73–75]. The results showed that the dissolution rates of fuel with a size of 2 mm and fuel powder (<850 μm) shorter than 2 mm were almost the same and achieved complete dissolution within three and a half hours [76,77].

Subsequently, dissolution experiments were carried out using fuel segments with a length of 10 mm and a dissolved Pu + U concentration of 510 g/L. The initial acidity of the dissolution solution was 12 mol/L, and the dissolution temperature was 95 °C. The experimental results indicated that even under such high solution concentrations, complete reactions could be achieved within 3 h [78]. This suggests that using a high acid concentration of 12 mol/L is capable of dissolving MOX fuel with a PuO_2 content of 23%–29%.

At the Tokai Reprocessing Plant (TRP) in Japan, they conducted the dissolution of ATR-MOX spent fuel, which contained approximately 2% Pu. Following the dissolution, the composition of the insoluble residue was analyzed [79]. The results revealed that the primary components of the insoluble residue included platinum group elements (Ru, Rh, Pd), molybdate-zirconate compounds (Mo, Zr), nuclear materials (U, Pu), and fission products (Fe, Te, Na, Tc). The majority of the residue consisted of small particles measuring equal to or less than 1.0 mm in size.

4.3. India

In 2003, India established the Compact Reprocessing facility for Advanced fuels in Lead shielded cell (CORAL) at the Indira Gandhi Atomic Research Centre [80]. This facility was specifically designed to handle the reprocessing of spent fuel from the Prototype Fast Breeder Reactor (PFBR), which comprises two composition components: $(Pu_{0.7},U_{0.3})C$ and $(Pu_{0.55},U_{0.45})C$. Additionally, it was intended to process future mixed oxide spent fuel from the PFBR, with an inner zone composition of 21% PuO_2 and 79% UO_2 MOX, and an outer zone composition of 28% PuO_2 and 72% UO_2 MOX. However, there is currently no specific report available regarding the dissolution process for the mixed oxide spent fuel at CORAL. Further information on the dissolution and reprocessing of the mentioned mixed oxide spent fuel may be obtained from future research publications or official reports from the Indira Gandhi Atomic Research Centre.

4.4. China

China Institute of Atomic Energy conducted a study on the impact of acidity on the dissolution of MOX fuel, using Ce as a surrogate for plutonium [81]. The study involved testing simulated MOX pellets containing a mixture of 10% CeO_2 and UO_2 in nitric acid. In the presence of 8 mol/L nitric acid, the dissolution rate of Ce was relatively rapid, reaching approximately 70% after immersing the pellets for 10 min. Subsequently, there was a minimal increase in the dissolution rate of Ce over time, reaching approximately 90% after 5 h of dissolution. On the other hand, when dissolved in 4 mol/L nitric acid, the dissolution rate of Ce was slower, gradually increasing and reaching a level similar to that observed in 8 mol/L nitric acid after about 3 h. During the dissolution process of uranium-cerium mixed oxide, uranium demonstrated a clear preference for dissolution, exhibiting a higher dissolution rate compared to cerium under the same conditions. The dissolution rate of uranium in the simulated pellets was higher than that reported in the literature for UO_2 pellets, which could be attributed to variations in pellet size and fabrication processes.

China Nuclear Four Zero Four Co., LTD conducted a study on the dissolution of irradiated PuO_2-UO_2 fuel in nitric acid [82]. The research involved intermediate-scale hot tests, which demonstrated that post-irradiation PuO_2-UO_2 fuel exhibited improved dissolution performance. It could be largely dissolved within 6 h in nitric acid concentrations above 8 mol/L. However, complete dissolution of the remaining small fraction was slow, sometimes requiring the use of 11 mol/L HNO_3 for complete dissolution. The study also revealed the presence of significant amounts of insoluble residues during the dissolution of spent fuel from fast reactors. These residues were thoroughly inspected and analyzed, and they were identified as a multi-metal alloy (containing Mo, Ru, Rh, Pd, Tc) along with small amounts of U, Pu, and corrosion products. These residues had a high density and particle sizes of around 0.5 μm, making them challenging to separate and prone to causing filter clogging.

4.5. *America*

In the 1970s, the Oak Ridge National Laboratory in the United States established a comprehensive experimental facility for the reprocessing of fast reactor fuel [83]. The process flowchart for this facility is depicted in Fig. 6. Initially, the fast reactor fuel underwent oxidation and volatilization treatment. Following this treatment, the spent fuel transformed into small particles and was introduced into a continuous dissolver with a rotary drum for dissolution. Any remaining undissolved components underwent a powerful dissolution process with the addition of hydrofluoric acid (HF).

4.6. *Other countries*

In the late 1950s, the United Kingdom established a comprehensive fast reactor fuel reprocessing facility in Dounreay, Scotland, with a primary focus on the fuel cycle of the Dounreay Fast Reactor (DFR). In August 1980, the facility successfully processed DFR mixed oxide fuel for the first time.

In 1971, Germany constructed the MILLI experimental facility at the Institute for Applied Chemistry. This facility processed Dounreay Fast Reactor fuel (consisting of 15% PuO_2 and burnt to 60 GWd/t) in the first half of 1974. The process involved dissolving 500 g of spent fast reactor fuel in 2.3 L of 8 mol/L nitric acid. The insoluble materials, including Mo, Ru, Zr, Pd, and Sn, accounted for 0.4–0.8% of the total weight, while the insoluble plutonium made up 0.02–0.04% of the total weight.

In the 1990s, Russia conducted hot experiments on spent fast reactor fuel [84]. The experiments employed BR-10, BOR-60, and

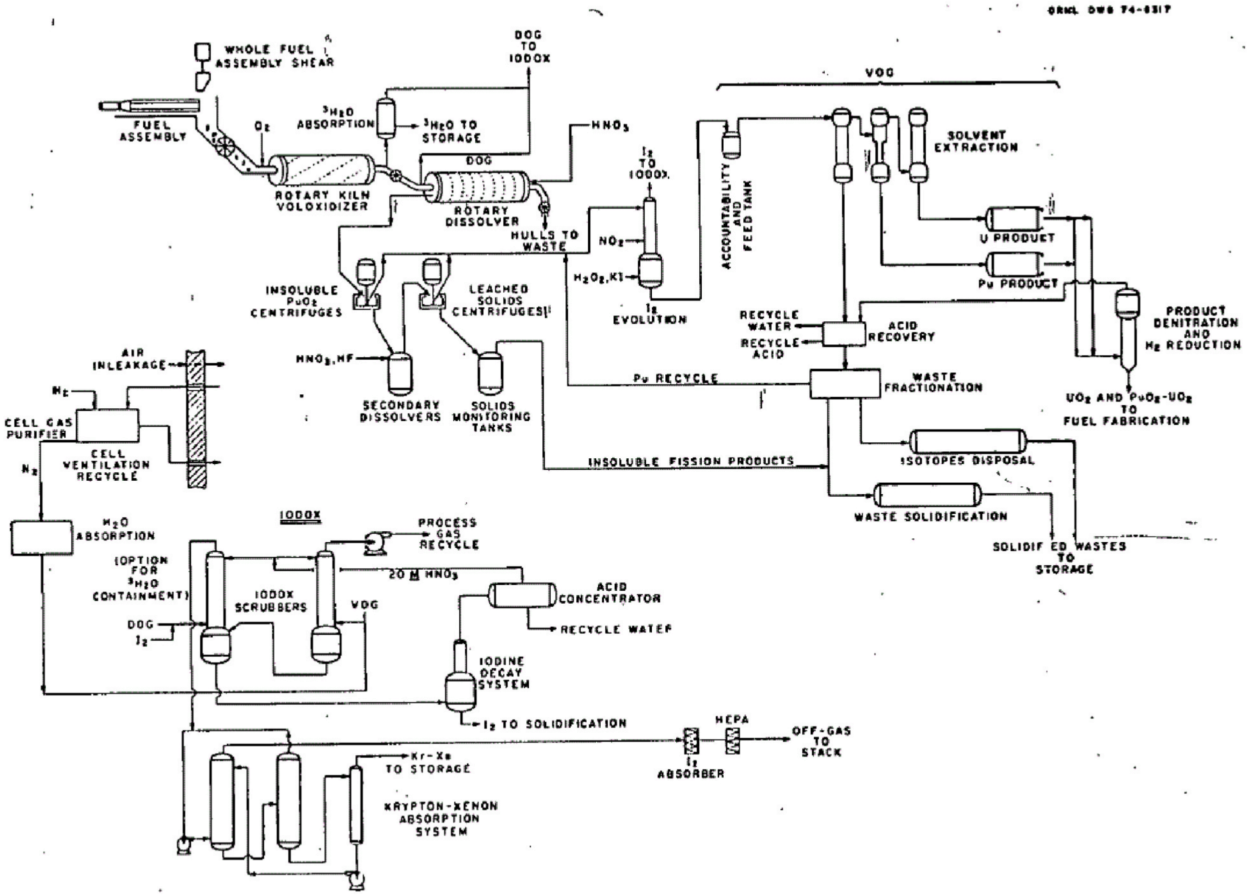

Fig. 6. Flowchart of the post-processing process at Oak Ridge National Laboratory [83].

BN-350 fuels, which contained plutonium ranging from 17% to 27% and had a burnup of 100 GWd/tHM. The cooling time varied between 8 months and 3 years.

5. Conceptualization of front-end processing in future MOX fuel reprocessing

While there is presently no globally operational stable fast breeder MOX fuel reprocessing plant within the scope of our research, numerous countries have engaged in experimental studies, with some even exploring engineering-scale processing. The research findings from various countries are summarized in Table 4. For instance, France has undertaken multiple industrial-scale dissolutions of MOX fuel, utilizing 6.2 mol/L HNO_3 for dissolution, resulting in approximately 8% undissolved residues. These residues can be dissolved under the conditions of 11 mol/L HNO_3 + 0.05 mol/L HF. Additionally, in the 2000s, France employed continuous dissolution technology, processing around 50 tons of MOX fuel under conditions of 5 mol/L HNO_3, a temperature of approximately 90–92 °C, and a dissolution time of about 7 h. China and Japan have also experimented with high acidity methods for MOX fuel dissolution, with results indicating that increasing acidity has a significant promoting effect on the dissolution of MOX fuel.

In global research and engineering experiments, MOX fuel dissolution is closely tied to component fabrication processes. While direct comparisons are challenging due to component design variations, a consistent trend emerges: MOX fuel poses greater dissolution challenges than pressurized water reactor fuels, often requiring higher acidity and longer dissolution times. Notably, MOX fuel dissolution may generate plutonium-rich undissolved residues.

Drawing upon research outcomes and our comprehension of MOX fuel reprocessing, we propose the following process as a potentially viable approach in the future. The outlined procedure is depicted in Fig. 7.

Initially, MOX fuel undergoes shearing and high-acid dissolution (with an initial concentration exceeding 8 mol/L). The resulting solution is then mixed with pressurized water reactor solution to readjust the acid concentration to around 3 M. This aims to maximize MOX component dissolution under specified temperature and acidity conditions. For portions that don't dissolve within the stipulated time, enhanced dissolution is applied, including undissolved residues and plutonium-rich insoluble materials. Three methods for enhanced dissolution are considered: introducing HF, catalytic oxidation with Ce^{4+} and Ag^{2+}, or high-temperature and high-pressure enhanced dissolution. However, current research on these methods is insufficient, and each has drawbacks.

The addition of HF introduces F- ions, raising corrosion resistance requirements for equipment. F- ions may also react with Pu, affecting subsequent extraction and separation. The strong oxidative nature of Ce^{4+} and Ag^{2+} increases equipment demands and metal ion content in high-level waste solutions. High-temperature and high-pressure enhanced dissolution demand high equipment reliability and entail certain accident risks, presently deemed unacceptable in nuclear fuel reprocessing engineering.

In the final step, the solutions from the pressurized water reactor short segment, MOX fuel, and enhanced dissolution of residues are combined. However, due to critical safety constraints on subsequent equipment, the Pu concentration in the mixed solution must be carefully controlled. The specific concentration is determined based on both the processing requirements and comprehensive considerations of equipment critical safety design. Following the mixing, the solution undergoes filtration, and the acidity is adjusted to 3 mol/L, while the uranium concentration is set to 225–250 g/L. The subsequent processes can then proceed using the PUREX process.

6. Conclusion

This article provides an overview of the ongoing research on the nitric acid dissolution process of MOX (Mixed Oxide) fuel. MOX fuel stands out due to its elevated plutonium content, potentially leading to the creation of stubbornly insoluble plutonium residues. Researchers have employed diverse methods to delve into the dissolution dynamics of MOX fuel, culminating in several noteworthy findings.

1) The oxidation process with nitric acid is more favorable for uranium, leading to the formation of UO_2^{2+} ions, allowing the uranyl ion oxide lattice to seamlessly enter the solution. In contrast, plutonium exhibits a lower tendency for oxidation, and the formation of plutonyl ions is not as readily achieved. This resistance to oxidation contributes to the stability of the lattice, particularly when the plutonium content in MOX fuel surpasses 35%. At plutonium content levels between 55% and 65%, plutonium becomes virtually insoluble, presenting a notable challenge in the dissolution process.
2) The exploration of MOX fuel dissolution has been a collaborative effort involving numerous countries. French engineers conducted post-irradiation treatment of MOX fuel characterized by high plutonium content (approximately 5%) within hot reactors at the UP2-800 plant. This involved the strategic use of high dissolution acidity and an extended dissolution time. Similarly, in Japan, MOX fuel with a fuel length of 10 mm and plutonium content ranging from 23% to 29% was dissolved using a 12 mol/L nitric acid solution. Other countries have also engaged in reprocessing initiatives for MOX fuel, resulting in varying degrees of enhancement to the dissolution process.
3) Currently, various methods are employed for MOX dissolution, encompassing strategies such as increasing temperature, acidity adjustment, crushing fuel pellets, and utilizing ultrasound-assisted dissolution. The incorporation of metal ion oxidants or hydrogen fluoride has been recognized for its potential to augment the dissolution rate of MOX fuel. However, it is crucial to acknowledge that the introduction of these substances imposes considerable demands on equipment materials and may introduce new ions, potentially impacting subsequent processes. The process of crushing fuel pellets introduces an additional step, thereby amplifying the overall process risk. While ultrasound-assisted dissolution displays promise, it remains in the experimental stage. Importantly, the escalation of temperature and acidity heightens the risks associated with the dissolution stage and concurrently curtails the lifespan of pertinent equipment.

Table 4
Dissolution conditions of various countries.

Time	Country	Dissolved object	Initial nitric acid concentration(mol/L)	Dissolution time and temperature	Residue dissolution condition	references
1980s	France	Burnup: 30,000 MWd/tU, Plutonium content:	6.2 mol/L	6 h of boiling	11 mol/L HNO_3 + 0.05 mol/L HF	[57,61]
2000s	France	Pre-irradiation plutonium content ranging from 4% to 8%)	5 mol/L HNO_3;	around 7 h temperature approximately 90–92 °C;	–	[66]
2010s	Japan	Burnup of 55,000 MWd/t, initial PuO_2 content in the fuel ranging from 23% to 29%).	12 mol/L HNO_3,	3 h temperature is 95 °C	–	[78]
2010s	China	Post-irradiation PuO_2-UO_2 fuel.	8 mol/L HNO_3 or higher	6 h of boiling	11 mol/L HNO_3.	[82]
1950s–2000s	Other countries	There have been reports on the dissolution or experimental attempts of spent fuel from fast reactors. However, within the scope of our research, these processes lack sufficient detail.				[80,83,84]

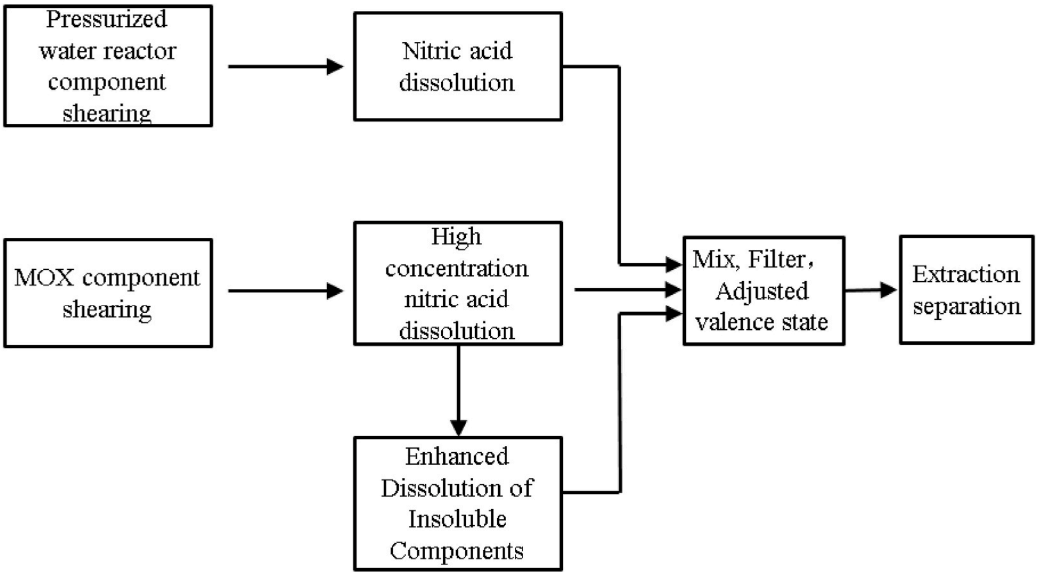

Fig. 7. A possible front-end processing in future MOX fuel reprocessing.

Future outlook

With the ongoing advancements in fast breeder technology, the demand for MOX fuel reprocessing is set to rise. Presently, water-based reprocessing emerges as the most suitable approach, given the evolving reprocessing landscape. Achieving engineering-scale conditions for nuclear fuel reprocessing demands years of refinement and validation. To minimize the risk of significant modifications in subsequent stages, it is advisable to limit or avoid introducing new substances during the initial MOX fuel dissolution process. Future efforts should focus on optimizing dissolution processes, exploring lower concentrations of nitric acid, and minimizing the addition of substances like HF. Additionally, there's potential for developing new techniques such as high-temperature and high-pressure dissolution processes. We will also persist in researching the MOX fuel dissolution process.

Data availability statement

No data was used for the research described in the article.

CRediT authorship contribution statement

Tianchi Li: Writing – original draft, Methodology, Data curation. **Fang Liu:** Writing – review & editing, Conceptualization. **Zhou Jia:** Investigation, Formal analysis. **Fangxiang Luo:** Supervision, Conceptualization. **Taihong Yan:** Project administration, Conceptualization. **Weifang Zheng:** Software, Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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