Trial of Pilot Scale Nanofiltration Unit for Improvement of Precipitation Circuit at Tummalapalle Mill

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Abstract: Alkaline pressure leaching is used in the Tummalapalle Uranium Ore processing plant due to the ore's high carbonate content (85%). About 70% of the uranium-laden liquor from the leached slurry filter is recycled to repulp the pre-leach filter cake, increasing the mother liquor concentration. This creates a large inventory of concentrated slurry, affecting precipitation efficiency and settling characteristics. Higher mother liquor concentration may improve settling after precipitation. A laboratory nanofiltration unit at UCIL Tummalapalle was tested to enhance clarified mother liquor concentration. Preliminary experiments showed reproducible results, suggesting an increase in uranium concentration from 0.5 gpl to 0.9 gpl. This process reduces leach liquor recycling, improves wash liquor flexibility, and operates with lower leach liquor values. This paper presents pilot-scale nanofiltration trials at Tummalapalle Mill, which could enhance U_3O_8 recovery. Further study is needed to assess water balance for full-scale implementation.

Keywords: Uranium; Tummalapalle Mill; Nano Filtration Skid; Ultra Filtration; Precipitation; Alkali Leaching.

Nomenclature:

AERB	Atomic Energy Regulatory Board
CNSC	Canadian Nuclear Safety Commission
CPL	Clarified Pregnant Liquor
ED	Electrodialysis
EPA	Environmental Protection Agency
EU	European Union
HP	High Pressure
IAEA	International Atomic Energy Agency
LP	Low Pressure
LPD	Liters per Day
NF	Nanofiltration
NRC	Nuclear Regulatory Commission
NTU	Number of Transfer Units
OSHA	Occupational Safety and Health Administration
REEs	Rare Earth Elements
RO	Reverse Osmosis
SDI	Silt Density Index
SDU	Sodium Diuranate
SS	Stainless Steel
TDS	Total Dissolved Solids
TSS	Total Suspended Solid
UCIL	Uranium Corporation of India Limited
UF	Ultrafiltration

1. Introduction and Literature Survey:

Inorganic membranes have been explored extensively for their potential to reject uranium from both fresh and saline waters (Lin et al., 2019a). The feasibility of using nanofiltration (NF) to remove uranium from groundwater under environmentally relevant conditions has been evaluated, highlighting its effectiveness in contaminated environments (Verma & Loganathan, 2024). Scaling up NF processes through pilot trials has been proposed as a critical step towards wider industrial adoption (Guerra et al., 2023). From an economic perspective, electrodialysis (ED) emerges as a more cost-effective solution due to its lower energy requirements and high membrane durability, which minimizes replacement expenses (Guerra et al., 2024). Existing active mine water treatment technologies have been comprehensively reviewed, offering valuable insights into current practices (Wolkersdorfer, 2022). Furthermore, there is a growing trend among mining companies to adopt technologies to reuse water within internal circuits (Witecki et al., 2022). The potential recovery of rare earth elements (REEs) from acid mine drainage has also been investigated, with advancements in precipitation strategies yielding an 8% improvement in REE recovery and a 2.8-fold increase in purity (Mwewa et al., 2022); (Liu et al., 2024). A review by (Yadav et al., 2022a) discusses prospects in this domain, emphasizing ongoing innovations. In the fabrication of nanofiltration membranes, techniques such as phase inversion and interfacial polymerization have been utilized to enhance performance (Mahmoud & Mostafa, 2023). For uranium and thorium separation, chitosan–polypropylene membranes (C–PHF–M) have demonstrated precise selectivity, with thorium dioxide retained almost entirely. At the same time, aluminum is recovered as sodium aluminate during filtration (Man et al., 2024). The behavior of extractable alkali metal complexes with specific ligand mixtures has also been characterized (Bezdomnikov et al., 2024). While some promising approaches have been proposed, further validation at industrial scales remains necessary (Pola et al., 2022). Membrane technologies are being increasingly studied for lithium recovery, with lithium phosphate precipitation yielding an impressive 84% recovery from salt solutions containing around 200 ppm lithium (Annunzi et al., 2023). Finally, uranium removal using NF membranes has achieved remarkable performance, with a rejection rate of 99.83% and a permeate flux of 71.1 L/(m^2·h) (Meng et al., 2023). The economic and operational trade-offs between nanofiltration (NF) and electrodialysis (ED) in uranium extraction from alkaline leach liquor primarily stem from differences in energy consumption, selectivity, scalability, and cost-effectiveness. Both technologies are used to concentrate and separate valuable elements such as uranium from leachate, but they operate on different principles and have varying impacts on industry operations. Below is an in-depth comparison:

1. Principle of Operation: The nanofiltration (NF) membrane filtration process works by using semi-permeable membranes to separate ions, molecules, and particles based on their size and charge. In uranium extraction, NF is typically employed to separate uranium and other monovalent ions from the alkaline leach liquor. Electrodialysis (ED) is an electrochemical process, that uses an electric field to drive the migration of ions through selective ion exchange membranes. In ED, uranium and other ions are separated based on their charge, with cations (positively charged ions) moving through cation-exchange membranes and anions (negatively charged ions) passing through anion-exchange membranes.

2. Energy Consumption: Energy consumption in NF primarily depends on the transmembrane pressure required to push the liquid through the membrane. The pressure can be significant but is often lower than electrodialysis. For uranium extraction, NF tends to have moderate energy requirements, especially when compared to ED. Electrodialysis, on the other hand, typically consumes more energy due to the need for an electric current to drive ion migration. The energy requirement is proportional to the ion concentration and the number of ion-exchange membranes used. While ED can be highly efficient for specific ion separations, it generally has a higher operational energy cost compared to NF. From an energy perspective, NF tends to be more cost-effective due to lower energy consumption, especially for large-scale operations where energy costs are significant.

3. Selective Separation: NF membranes can be designed to selectively separate uranium and other ions based on their size and charge. However, NF membranes may have lower selectivity compared to ED, as they might not effectively differentiate between ions with similar charge or size. Electrodialysis has high selectivity, especially for ions with distinct charges. ED membranes can selectively separate cations like uranium ions from the solution. This is particularly useful for industries where purity or specific separation of ions is important. If high purity of uranium or specific ion separation is critical, ED might be the better choice despite higher energy costs. However, for more general applications where perfect selectivity is not as crucial, NF can be more cost-effective.

4. Scalability and Flexibility: Nanofiltration systems are generally more scalable, easier to operate, and more flexible for handling large volumes of alkaline leach liquor. They can be operated continuously, and membrane fouling (a major issue in many filtration processes) can be mitigated with regular cleaning procedures. While ED systems can also be scaled, they tend to be more complex to operate and maintain. The ion-exchange membranes in ED can be susceptible to scaling and fouling, requiring more frequent maintenance and specialized cleaning, which can lead to higher operational downtime and costs. NF is often more attractive for large-scale operations due to its ease of scaling and lower maintenance needs, making it a more stable option in terms of long-term costs.

5. Capital and Operational Costs: The capital investment for NF is typically lower than for electrodialysis, especially in terms of membrane costs and system complexity. Operating costs are lower due to lower energy usage and reduced maintenance requirements. The capital investment for ED systems can be higher due to the need for multiple ion exchange membranes, electrodes, and power supplies. Moreover, operating costs can be higher because of the greater energy consumption and more frequent maintenance due to membrane fouling. NF usually has a lower upfront capital cost and lower ongoing operational costs compared to ED, making it more economically viable for many industries that need to extract uranium in bulk or where cost efficiency is a priority.

6. Environmental Impact and Waste Management: NF systems produce a concentrate that contains the uranium and other solutes. Proper disposal or further treatment of this concentrate is necessary to prevent environmental contamination. However, the system itself has a smaller environmental footprint due to its lower energy consumption. Electrodialysis systems generate waste in the form of brines or concentrated waste streams. This requires effective waste management practices to ensure minimal environmental impact. The higher energy use can also result in a larger carbon footprint, especially if the energy is sourced from non-renewable sources. Environmental considerations and waste disposal costs are factors that can affect the overall economics of the operation. NF may have the edge in terms of lower energy consumption, but both processes need to manage waste effectively.

7. Product Recovery and Purity: While NF is effective at concentrating uranium, the purity of the recovered uranium can sometimes be lower compared to ED, as some unwanted ions may coalesce with the uranium in the permeate. Electrodialysis offers the potential for higher-purity uranium recovery due to its more selective ion separation capabilities, ensuring that the final product is enriched to the desired level. Industries that prioritize the high purity of uranium may find ED to be more economically advantageous in the long term, despite its higher operational costs. NF is likely to be more economically favorable for large-scale uranium extraction, especially when the operation focuses on minimizing energy costs, simplifying maintenance, and maintaining operational flexibility. Its lower capital and operational expenses make it an attractive option for cost-conscious industries. ED may be more appropriate when very high selectivity, purity, and effective ion separation are necessary, despite its higher initial investment, energy consumption, and maintenance needs. In summary, the choice between NF and ED depends on the specific requirements of the uranium extraction process, including factors such as desired product purity, scalability, energy costs, and long-term operational feasibility. For most industries, NF tends to offer better economic efficiency, while ED excels in processes where purity and ion selectivity are critical.

1.1 Alkali leaching process & chemical reactions: Sodium bicarbonate generation takes place as per exothermic reactions (1,2,3,4) inside a pressurized autoclave which helps in the completion of chemical reactions in the presence of oxygen (S. T. , S. V. K. Reddy B.N.K., 2024). For autoclave operations, low concentration $NaHCO_3$ is sufficient, and it benefits product precipitation.

$$2UO_2 + O_2 \rightarrow 2UO_3 \quad (1)$$
$$UO_3+3Na_2CO_3+H_2O \rightarrow Na_4UO_2(CO_3)_3+2NaOH \quad (2)$$
$$NaOH+NaHCO_3 \rightarrow Na_2CO_3+H_2O \quad (3)$$
$$2FeS_2 + 7O_2 + 8Na_2CO_3 + 6H_2O \rightarrow 2Fe(OH)_2 + 4Na_2SO_4 + 8NaHCO_3 \quad (4)$$
$$2NaHCO_3+\Delta(80\ ^{o}C) \rightarrow Na_2CO_3+H_2O+CO_2 \quad (5)$$

Filtration of autoclave discharge slurry takes place after alkali leaching for solid-liquid separation. Filtrate goes to the clarification unit for removal of remaining TSS (Thamida S.K., 2023). In the precipitation stage, bicarbonate is first neutralized with caustic soda (reaction 6). Precipitation of clarified liquor takes place for precipitating sodium diuranate in the presence of 48% conc. NaOH (reaction 7) (Rajesh L., 2018).

$$NaHCO_3+NaOH \rightarrow Na_2CO_3+H_2O \quad (6)$$
$$2Na_4UO_2(CO_3)_3 + 6NaOH \rightarrow Na_2U_2O_7 + 6Na_2CO_3 + 3H_2O \quad (7)$$

1.2 R&D activities at Tummalapalle Mill: Tummalapalle project processes low-grade uranium ore. Although it is one of the largest ore reserves, still it is the main challenge for the R&D team to improve concentration using various existing technologies (Sharma V. K., 2023). A team of engineers conducted several experiments for the enhancement of U_3O_8 concentrations which are as follows:

1. Candle Filtration
2. Centrifugation
3. Ultra Filtration
4. Re-Dissolution
5. Ion Exchange
6. Nano Filtration

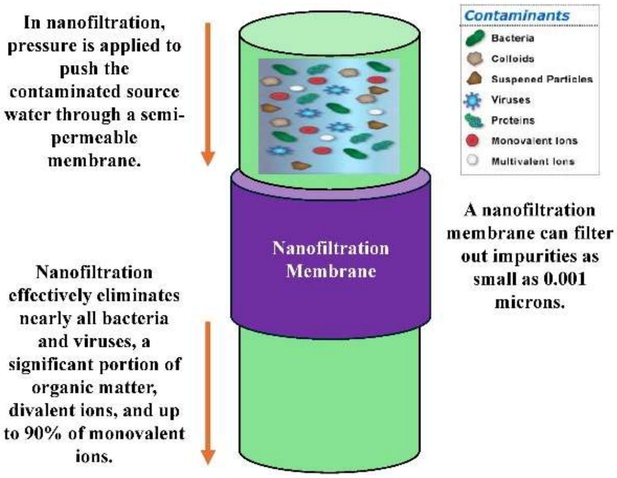

Figure 1. Working function of nanofiltration membrane

1.3 Membrane Separation: Membrane separation processes operate without heating and therefore use less energy than conventional thermal separation processes such as distillation, sublimation, or crystallization (Rani N., 2009). The separation process is purely physical, and both fractions (permeate and retentate) can be used. Cold separation using membrane technology is widely used in food technology, biotechnology, and pharmaceutical industries (Tiwari R., 2008). Furthermore, using membranes enables separations to take place that would be impossible using thermal separation methods. The challenges for the membrane system for such applications are as under (Rajesh L., 2019):

3.1 The membrane system does not handle high salt concentration due to osmotic pressure limitation.
3.2 Membrane systems need highly clarified liquor with a very low NTU/SDI index.
3.3 High temperature is not conducive to membrane life.

1.4 SaltOut Technology: It was noticed that the membrane process can help in achieving the desired concentration of product starting from low concentrations (Sharma V.K., 2008b). Membranes can also assist in the separation of salt, purification of process stream, and recycling of liquor. A comprehensive study of the current process was made to apply technology at other points (A. Ghaddar, 2008). A new process scheme with SaltOut Technology which may avoid the recycling of liquors to achieve high concentration in CPL and facilitate recycling of salts into the process was used in Tummalapalle Mill (Sharma V.K., 2008a).

1.5 Objectives of nanofiltration at Tummalapalle Mill: The main objectives of nanofiltration at Tummalapalle Mill are as follows (Sharma V.K., 2022):

1. The recycling loop can be avoided.
2. Increase in concentration of process liquor.
3. U_3O_8 Recovery Enhancement.
4. Improved floc generation inside product thickener.
5. Improved settling characteristics at product thickener.
6. Control on TSS present in process liquor.

Figure 2. (a) Top view of nanofiltration membrane used for the trial, (b) Side view of nanofiltration membrane used for the trial.

1.6 Technical Justification of Nanofiltration Selection: The selection of nanofiltration (NF) over other membrane technologies like reverse osmosis (RO) and ultrafiltration (UF) for uranium extraction from alkaline leached liquor is based on several factors related to the specific properties of these membranes, their performance in processing uranium-laden streams, and the operational requirements of the extraction process. Below is a detailed discussion covering the technical, economic, and operational considerations in selecting nanofiltration for uranium extraction:

1. Membrane Characteristics

a. Nanofiltration (NF)

- **Pore Size**: Nanofiltration membranes have pore sizes in the range of 1-10 nanometers. This size allows NF membranes to selectively reject divalent ions (such as uranium) while allowing monovalent ions (e.g., sodium and potassium) to pass through. This is a key feature for uranium extraction from alkaline-leached liquors.
- **Ion Selectivity**: NF membranes exhibit selective permeability to ions based on their charge and size. Uranium ions, which are typically in their divalent form (UO_2^{2+} or $UO_2(OH)_2$), are effectively rejected by NF membranes, making them ideal for concentrating uranium while allowing the passage of less valuable ions.

- **Pressure Requirement**: NF membranes typically operate at lower pressures than reverse osmosis membranes, making them more energy-efficient in certain applications.

b. Reverse Osmosis (RO)

- **Pore Size**: RO membranes have a much smaller pore size (typically around 0.0001 microns) compared to NF. While they are highly effective at removing almost all ions, including both monovalent and divalent ions, the rejection of monovalent ions (like sodium) is not desirable in some cases.
- **Energy Requirement**: RO membranes require high pressure (up to 60-80 bar) to overcome osmotic pressure and achieve effective separation. The high energy demand makes RO less economical for certain applications like uranium extraction, where the goal is the selective removal of divalent ions.
- **Water Recovery**: RO typically operates with a lower water recovery rate than NF membranes. Since uranium extraction often involves the handling of large volumes of solution, lower water recovery can lead to significant waste generation.

c. Ultrafiltration (UF)

- **Pore Size**: UF membranes have larger pores (typically in the range of 10-100 nanometers), making them suitable for separating larger particles, such as suspended solids, colloids, and larger molecules. However, UF is not selective enough to remove dissolved ions like uranium, especially in its divalent form.
- **Limited Ion Rejection**: UF does not effectively reject dissolved species like uranium ions, which makes it unsuitable for uranium extraction in leached liquor where divalent metal ions need to be separated from the solution.

2. Uranium Extraction Process

- **Alkaline Leaching of Uranium**: In uranium extraction, the ore is treated with an alkaline solution (such as sodium carbonate or sodium hydroxide) to leach uranium into solution as soluble complexes. This results in a liquor containing uranium in the form of uranyl ions (UO_2^{2+}) and other alkali metal ions like sodium or potassium.
- **Selective Removal of Uranium**: The goal of using a membrane process is to selectively remove or concentrate uranium from the leachate while allowing other components to pass through. Since uranium in leach liquor exists predominantly as divalent uranyl ions, a membrane that is selective for divalent ions but passes monovalent ions (e.g., sodium) would be ideal.

3. Performance Considerations

a. Ion Rejection

- **Nanofiltration**: NF membranes provide selective rejection of divalent ions like uranium while allowing monovalent ions such as sodium to pass. This selective rejection ensures that uranium can be concentrated without excessive removal of beneficial monovalent salts, which is crucial for maintaining the chemical balance in the leach liquor.
- **Reverse Osmosis**: RO membranes would reject almost all ions, including the monovalent ones, which would result in a very concentrated stream of uranium but also waste a significant amount of the alkali salts (such as sodium) that are essential for the leaching process. The rejection of monovalent ions could also result in operational challenges, such as scaling and fouling.
- **Ultrafiltration**: UF does not effectively reject dissolved ions such as uranium. It is primarily used for separating larger particles and suspended solids, making it unsuitable for applications requiring the removal of dissolved metal ions.

b. Operational Efficiency

- **Energy Consumption**: NF membranes operate at much lower pressures (typically 5-20 bar) compared to RO membranes, making them significantly more energy-efficient for processes like uranium extraction from leachate. The reduced energy demand is especially important in large-scale industrial applications, where energy costs can significantly impact the overall economy.

- **Water Recovery**: NF systems tend to have higher water recovery rates compared to RO systems, meaning they produce more permeate (clean water) relative to waste (concentrated uranium solution). This higher recovery is beneficial in reducing water consumption and minimizing waste generation.

c. Membrane Fouling

- **Nanofiltration**: While NF membranes are generally less prone to fouling than RO membranes due to their larger pore size, fouling can still occur due to the presence of organics, suspended solids, or high concentrations of certain salts in the leachate. However, the fouling tendency is typically lower for NF than for RO, and periodic cleaning can restore membrane performance.
- **Reverse Osmosis**: RO membranes are more prone to fouling, especially in complex solutions like alkaline leachate that may contain organic matter, salts, and other impurities. The need for frequent cleaning and replacement of RO membranes can increase operational costs.
- **Ultrafiltration**: UF membranes may suffer from fouling due to the accumulation of colloidal particles or other macromolecules. However, since they do not offer significant ion separation, they are not suitable for uranium extraction where ion selectivity is crucial.

4. Economic Considerations

- **Capital and Operational Costs**: The capital cost for NF systems is generally lower than for RO systems due to the lower pressure requirements and less complex infrastructure. Additionally, the operational costs for NF systems tend to be lower due to the reduced energy consumption, which is a critical consideration for large-scale uranium extraction.
- **Membrane Lifespan and Maintenance**: NF membranes, while still requiring maintenance, tend to have a longer lifespan than RO membranes because they are exposed to lower operating pressures. This results in reduced frequency of replacement and lower maintenance costs in the long run.
- **Waste Generation**: RO systems tend to produce a high volume of brine or concentrated waste, which can be difficult to dispose of, particularly in regions with strict environmental regulations. NF membranes produce less waste, making them a more environmentally friendly option.

5. Suitability for Uranium Extraction

Nanofiltration is particularly well-suited for uranium extraction from alkaline leached liquors due to:

- Its ability to selectively reject divalent uranium ions while allowing essential monovalent ions (like sodium and potassium) to pass through.
- It lowers energy requirements compared to reverse osmosis.
- Higher water recovery and lower brine production, which reduces waste handling.
- Its economic advantages, including lower capital and operational costs, especially in large-scale operations.
- Its relative resistance to fouling compared to RO membranes, reduces maintenance costs.

In conclusion, nanofiltration (NF) is an ideal membrane technology for uranium extraction from alkaline leached liquor due to its selective rejection of uranium while permitting the passage of monovalent ions. NF membranes operate at lower pressures, consume less energy, and offer higher water recovery rates compared to reverse osmosis, making them more cost-effective for large-scale operations. Furthermore, NF systems generate less waste and are less prone to fouling, ensuring a more efficient and sustainable process. On the other hand, reverse osmosis is energy-intensive and rejects both monovalent and divalent ions, which can disrupt the chemical balance in the leachate, while ultrafiltration is not effective for ion separation. Therefore, nanofiltration stands out as the most suitable membrane technology for uranium extraction in this context.

2. Material & Methods: Three SS316 tanks of capacity 1000 liters and one tank of capacity 200 liters are present in the nanofiltration skid (fig. 3). Tank 1 filled with clarified pregnant liquor (CPL) received after filtration of leached liquor received from autoclaves. Ultra filtration takes place initially inside tank 1 with the help of a ceramic filtration unit (Lin et al., 2019b)**.** This ultra-filtration helps with the removal of foreign objects that may be present in CPL. It decreases the load on the main nanofiltration membrane (Sharma V.K., 2020). This ultra-filtered liquor goes to tank 2. Liquor of tank 2 is used as feed for the main nanofiltration skid. All pumping operation takes place by HP pump which may operate at a maximum 8 m^3/hr flow and 60 bar pressure (Sharma et al., 2024). Initially, the LP pump starts manually which is designed for a maximum 8 m^3/hr flow and 2 bar pressure. The air may be released by opening drain valves at the suction and discharge side (K. T. S. , S. V. K. Reddy B.N.K., 2022). LP pump requires proper suction to the HP pump to avoid starvation/liquid hammering. The HP pump starts after starting the LP pump (Sharma et al., 2023). Feed from tank 2 passes through the nanofiltration membrane and the concentrate goes to tank 3 after filtration. Permeate goes to tank 4 after nanofiltration. This cycle continues until the HP pump automatically stops when tank 3 reaches its maximum capacity of 200 liters. The heat exchanger is available in a skid for heating/cooling of process liquor based on requirements (fig. 3). Approximately 4 hours take the completion of one complete cycle (Sriharsha P., 2023). A sampling of feed, concentration, and permeate for 21 cycles was completely taken, and concentrations of sodium carbonate, sodium bicarbonate, and uranium oxide were analyzed in the CR&D lab (table 1,2&3).

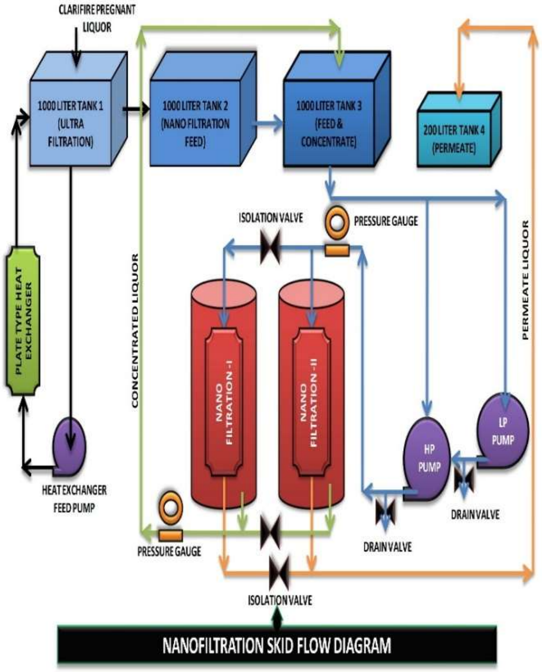

Figure 3. Flow diagram of nanofiltration skid.

Table 1. Design parameters of nanofiltration skid

Parameter	Input	Output	Side Stream
Flow	[illegible]td>	>100 LPD	Corresponding
Product	0.1-0.3 gpl	4 gpl+	<0.1 gpl
TSS	2-10 ppm	NA	<10 ppm
Na_2CO_3	5-10 gpl	<1	<1
$NaHCO_3$	10-20 gpl	<2	>20-40 gpl
Na_2SO_4	30-40 gpl	NA	NA
pH	8-10	8-10	8-10
Temperature	25-60 °C	25-60 °C	25-60 °C

3. Results & Discussions:

3.1 Operating parameters of nanofiltration skid: The control panel board displayed various parameters (Rajesh N.V., 2023). Below mentioned parameters were noted down in various time intervals (table 1). The average pH of the liquor was observed at 9.5

Table 2. Process Parameters of Nanofiltration Skid Trial

Sl. No.	Process Parameters	Unit	11:00 am	11:30 am	12:00 pm	12.30 pm	Avg. Value
1	Feed/Concentrate Tank-3 Volume	liter	581	475	419	350	-
2	Nanofiltration Pressure	bar	7.5	7.6	7.6	7.6	7.6
3	Permeate Flow	lit/min	1.59	1.60	1.58	1.58	1.59
4	Feed/ Concentrate Tank-3 Temp.	°C	37	37	38	39	38
5	Permeate Tank-4 Temp.	°C	27	28	28	28	28

3.2 Nanofiltration skid trial results: Sodium carbonate and sodium bicarbonate are the main reagents used for alkali pressurized leaching-based Tummlapalle uranium processing plants. U_3O_8 is the main content of the Tummalapalle Mill product (sodium diuranate). The concentration of these parameters was analyzed for various trials of nanofiltration skid (table 2,3&4). Figures 4,5&6 represent the compositions for various trials (Sarkar Suman, 2022). Initial values of U_3O_8 were in the range of 0.5 to 0.6 gpl in feed (Rao M.S., 2019b). After several trials, it rose to 0.8 to 0.9 gpl in concentrate. It is satisfactory results on the initial level. However, it was observed that the concentration of U_3O_8 in permeate was not as desired. It creates further selectivity scope of the nanofiltration membrane (Sreenivas T., 2010).

Table 3. Chemical compositions of Trial – I

Contents	Unit	Feed	Concentrate	Permeate
Na_2CO_3	**gpl**	4.810	8.020	6.420
$NaHCO_3$	**gpl**	15.260	13.980	13.980
U_3O_8	**gpl**	0.583	0.892	0.390

Table 4. Chemical Compositions of Trial - II

Contents	Unit	Feed	Concentrate	Permeate
Na_2CO_3	**gpl**	6.420	4.810	6.420
$NaHCO_3$	**gpl**	15.260	15.260	15.260
U_3O_8	**gpl**	0.614	0.867	0.431

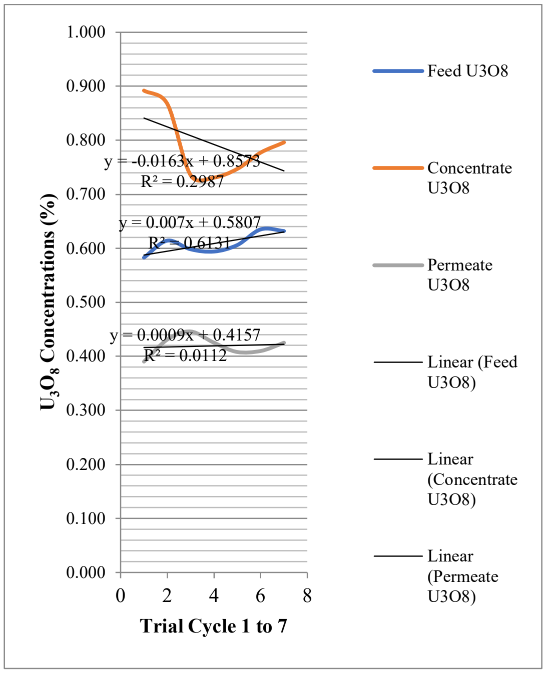

Figure 4: U3O8 analysis graph for samples of feed, concentrate, and permeate received from nanofiltration pilot scale trial cycles no. 1 to 7

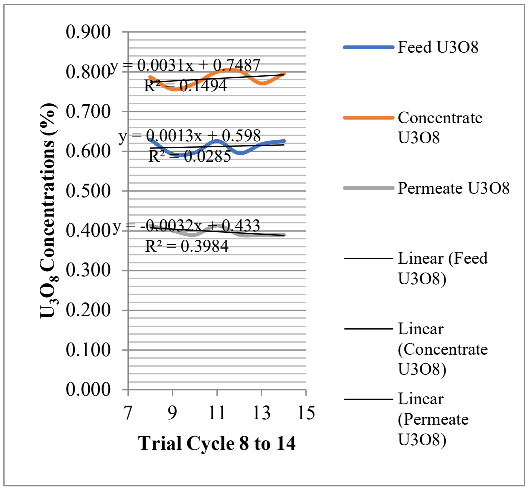

Figure 5. U3O8 analysis graph for samples of feed, concentrate and permeate received from nanofiltration pilot scale trial cycles no. 8 to 14

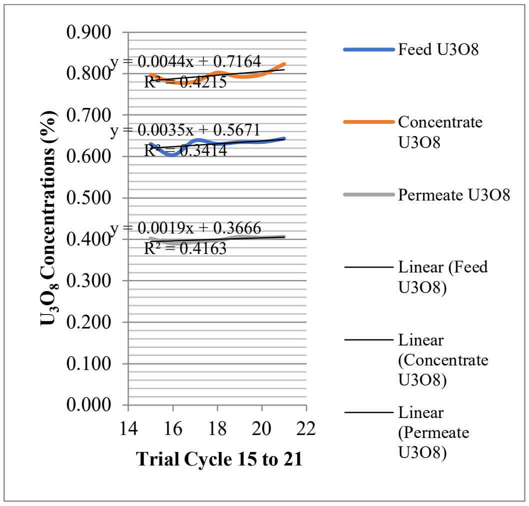

Figure 6. U_3O_8 analysis graph for samples of feed, concentrate and permeate received from nanofiltration pilot scale trial cycles no. 15 to 21

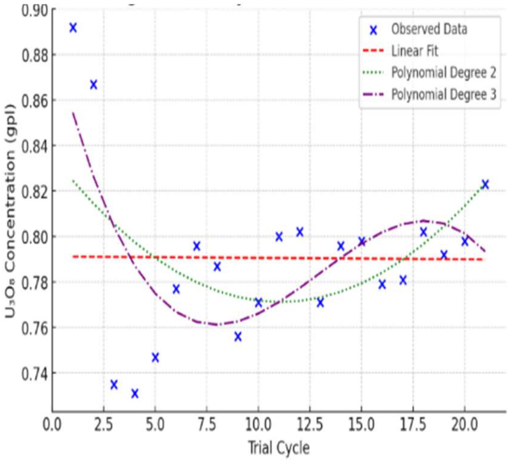

Figure 6. Regression analysis of U_3O_8 concentrations

3.3 Validation of U_3O_8 Concentration Increase: The study originally claimed that U_3O_8 concentration varied from 0.5-0.6 gpl in the feed and 0.8-0.9 gpl in the concentrate. To statistically validate this claim, a paired t-test was conducted to compare feed and concentrate U_3O_8 concentrations across multiple trials. The results showed a t-statistic of -19.59 and a p-value of 1.61×10^{-14}, which is well below the 0.05 threshold, indicating that the increase in U_3O_8 concentration is statistically significant. These findings support the conclusion that the nanofiltration process enhances uranium concentration.

3.4 Reproducibility and Standard Deviation Analysis: To assess the reproducibility of the results, the standard deviation of U_3O_8 concentration in the concentrate was calculated across multiple trials. The mean U_3O_8 concentration in the concentrate was found to be 0.791 gpl, with a standard deviation of 0.038 gpl. This relatively low standard deviation suggests a consistent increase in concentration across trials, confirming the reliability of the process.

3.5 Regression Analysis and Data Representation: Regression modeling was performed to evaluate the trends in U_3O_8 concentration changes. The results of different regression techniques are as follows:

- **Linear Regression**: R^2 = **0.0001**, indicating a very poor fit with no clear trend.

- **Polynomial Regression (Degree 2)**: R^2 = **0.219**, showing weak correlation.

- **Polynomial Regression (Degree 3)**: R^2 = **0.385**, providing a slightly better fit but still indicating significant variability. Given these results, the low R^2 values suggest that additional factors may be influencing U_3O_8 concentration trends, raising concerns about data reliability. Further investigation into operational conditions (e.g., temperature, pressure fluctuations) is recommended to improve predictive modeling.

4. Conclusions:

4.1 Process modification scope after nanofiltration installation:

The nanofiltration skid trial demonstrated its potential for enhancing the separation efficiency in uranium processing at the Tummalapalle plant. The average operational parameters, including pH, temperature, and pressure, remained stable across multiple trials, ensuring consistent process conditions. Key findings include the increased concentration of U_3O_8 in the concentrate from 0.5–0.6 gpl to 0.8–0.9 gpl, marking a significant improvement at the initial stages of implementation (Yadav et al., 2022b). The results revealed that while the permeate U_3O_8 concentration did not meet the desired specifications, the findings provide valuable insights for optimizing the nanofiltration membrane's selectivity. Additionally, the chemical composition analyses indicated that sodium carbonate and sodium bicarbonate concentrations varied across the feed, concentrate, and permeate streams, which could influence overall separation efficiency (Mahmoud & Mostafa, 2023b). This study highlights the efficacy of nanofiltration technology in improving uranium recovery while emphasizing the need for further refinement of membrane properties to achieve better selectivity. Future work should focus on membrane customization and parameter optimization to address the observed challenges, paving the way for more efficient and sustainable operations in uranium processing plants. Redissolution of SDU cake in liquor takes place before precipitation of process liquor (Rao M.S., 2019a). The nanofiltration unit may be installed after clarification. It may help in increasing liquor concentration and finally effective addition of caustic lye in a series of tanks (fig.7).

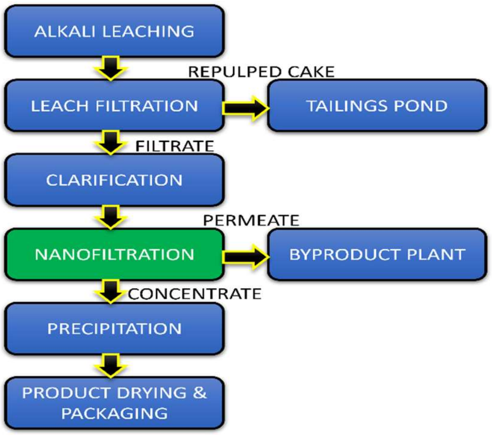

Figure 7. Scope of process modification at Tummalapalle Mill after nanofiltration

4.2 Limitations of nanofiltration: Nanofiltration is the least used method of membrane filtration in industry as the membrane pores sizes are limited to only a few nanometers. A main disadvantage associated with nanotechnology, as with all membrane filter technology, is the cost and maintenance of the membranes used. Repairs and replacement of membranes are dependent on total dissolved solids, flow rate, and components of the feed. Tests were conducted to check the performance of membranes for repeated use to establish membrane life, efficiency, and amenability for regeneration. Sensitivity analysis and reproducibility tests were also conducted for the performance of the membrane at various concentration levels of salts. This pilot unit is also required to generate higher concentration liquor to determine the threshold concentration for better precipitation and settling of SDU precipitate. The flux also needs to be optimized for the given input liquor. The performance of a nanofiltration (NF) membrane for uranium extraction in an alkaline leach liquor environment at a uranium ore processing unit is influenced by variations in water composition. Here's how different factors can impact NF membrane performance:

1. **Effect of Total Dissolved Solids (TDS) and Ionic Strength:** Higher TDS and ionic strength increased ionic concentration (Na^+, CO_3^{2-}, HCO_3^-) can lead to greater osmotic pressure, reducing the permeate flux and affecting uranium rejection efficiency. Lower TDS reduces osmotic pressure but may lead to lower uranium concentration in the concentrate, affecting overall recovery.
2. **Influence of Carbonate and Bicarbonate Levels:** Uranium in alkaline leach liquor primarily exists as uranyl carbonate complexes ($UO_2(CO_3)_3^{4-}$, $UO_2(CO_3)_2^{2-}$). Higher Na_2CO_3 / $NaHCO_3$

Concentrations can enhance uranium solubility but might reduce membrane selectivity by increasing the passage of uranium into the permeate. Lower carbonate levels may cause lower uranium solubility, potentially leading to membrane fouling due to the precipitation of uranium or other salts.

3. Presence of Competing Ions (Ca^{2+}, Mg^{2+}, SO_4^{2-}): Calcium and Magnesium can form insoluble carbonates or hydroxides, leading to membrane scaling and reduced efficiency. Sulphate (SO_4^{2-}), if present in significant amounts, can interfere with uranium rejection due to complexation or precipitation effects.

4. pH Variations: Higher pH (>10) enhances uranium rejection but can lead to carbonate scaling on the membrane surface. Lower pH (<9) may reduce uranium rejection efficiency as uranium complexes become less stable and can dissociate.

5. Temperature Effects: Higher temperatures can increase membrane permeability but may also degrade membrane material over time. Lower temperatures reduce diffusion rates, lowering uranium flux through the membrane.

6. Organic and Colloidal Contaminants: Organic matter (humic/fulvic acids) and fine colloids can lead to membrane fouling, requiring frequent cleaning. Pre-treatment methods such as filtration or chemical dosing (e.g., antiscalants) might be necessary to maintain performance.

4.3 Regulatory Challenges in Implementing Nanofiltration in an Operational Uranium Mill: Implementing nanofiltration (NF) technology in an operational uranium mill for improving U_3O_8 concentration in alkaline leach liquor presents several regulatory challenges. These challenges primarily arise due to the strict environmental, safety, and radiological controls governing uranium processing facilities. Below are key regulatory considerations:

1. Radiation Safety and Worker Protection Regulations: Regulatory agencies (e.g., AERB, IAEA, EPA, NRC, CNSC) impose strict radiation dose limits for workers handling uranium-containing solutions. The NF membrane system must comply with radiation shielding and dose monitoring standards. NF processes can lead to aerosolization of uranium during pressure-based filtration, requiring adequate ventilation and containment measures.

2. Environmental Compliance and Waste Management: NF generates concentrate and permeate streams, with the concentrate containing higher uranium levels. Proper usage and recycling must comply with radioactive regulations. Alkaline leach liquors contain sodium carbonate, bicarbonate, and trace metals. Zero liquid discharge can be followed by the recycling of entire liquor into the plant.

3. Regulatory Approval for Process Modification: Modifying an existing uranium mill to include NF technology requires approval from regulatory bodies such as the Nuclear Regulatory Commission (NRC) in the U.S., or equivalent authorities in other countries. Detailed risk assessments, including radiation impact studies, are required. Pilot studies may be mandated to prove that NF improves U_3O_8 concentration while meeting safety and performance standards. Data must be submitted to regulators, demonstrating uranium recovery rates, membrane life expectancy, and potential scaling/fouling issues.

4. Chemical Handling and Transportation Regulations: Alkaline leach solutions involve Na_2CO_3 and $NaHCO_3$, requiring adherence to chemical storage and handling standards set by OSHA (U.S.) or REACH (EU). Nanofiltration offers a promising method for improving U_3O_8 concentration in alkaline leach liquor, but its implementation in uranium mills faces stringent regulatory challenges. Compliance with radiation safety, environmental laws, chemical handling regulations, and public engagement requirements is essential. A proactive approach involving pilot studies, risk assessments, and regulatory engagement can help facilitate successful adoption.

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