THE CONCEPTUAL DESIGN FOR LIQUID EFFLUENT TREATMENT OF UO₂ KERNEL FABRICATION

Erilia Yusnitha, Agoeng Kadarjono, Agus Sartono, Pertiwi Diah Winastri
Center for Nuclear Fuel Technology – National Nuclear Energy Agency
Kawasan PUSPIPTEK Serpong Gd.20, Tangerang Selatan 15314
e-mail: erilia@batan.go.id

ABSTRACT

THE CONCEPTUAL DESIGN FOR LIQUID EFFLUENT TREATMENT OF UO₂ KERNEL FABRICATION. The pebble fuel for HTGR is prepared through fabrication of UO$_2$ kernel, coated particle and spherical element fuel. In the fabrication of UO$_2$ kernel by external gelation method, a multicomponent of liquid effluent is generated. Therefore, the liquid effluent is required to be treated for safety reason before disposed to waste storage. In this paper, the conceptual design for the liquid effluent treatment of UO$_2$ kernel fabrication is performed with the simulation process using CHEMCAD software. CHEMCAD is a software that can be utilized for chemical process design. The results showed that the proposed conceptual design is able to separate valuable components: isopropyl alcohol (IPA) and tetrahydrofurfuryl alcohol (THFA). The flowrate of IPA product is 5.28 kg/h with purity of 0.99 in mass fraction and the flowrate of THFA product is 1.01 kg/h with purity of 0.99 in mass fraction.

Keywords: liquid effluent, UO$_2$ kernel, CHEMCAD, HTGR.

ABSTRACT

DESAIN KONSEP UNTUK PENGOLAHAN EFLUEN CAIR DARI FABRIKASI KERNEL UO$_2$.
Bahan bakar HTGR bentuk pebble difabrikasi melalui tahapan fabrikasi kernel UO$_2$, partikel berlapis dan elemen pebble. Pada fabrikasi kernel UO$_2$ melalui metode gelasi eksternal, menghasilkan multikomponen efluen cair. Sehingga, efluen cair ini perlu untuk dilakukan pengolahan demi alasan keselamatan sebelum dibuang ke penampungan limbah. Pada makalah ini, desain konsep untuk pengolahan efluen cair dari fabrikasi kernel UO$_2$ disimulasikan dengan software CHEMCAD. CHEMCAD adalah sebuah software yang dapat dipergunakan untuk desain proses kimia. Hasil simulasi menunjukkan bahwa desain konsep yang dirancang mampu memisahkan komponen penting: isopropil alkohol (IPA) dan tetrahydrofurfuryl alcohol (THFA). Kecepatan aliran produk IPA sebesar 5,28 kg/h dengan kemurnian 0.99 fraksi massa dan kecepatan aliran produk THFA sebesar 1,01 kg/h dengan kemurnian 0.99 fraksi massa.

Keywords: efluen cair, kernel UO$_2$, CHEMCAD, HTGR.

INTRODUCTION

High Temperature Gas-Cooled Reactor (HTGR) is one of the advanced nuclear reactors based on tri-structural isotropic (TRISO) particle [1] which has two types of fuel structure: prismatic block and pebble bed [2,3]. The progress of utilizing fuel coated technology have been reported as pebble fuel form are utilized for High Temperature Gas-cooled Reactor Pebble-bed Module (HTR-PM) [4] and the 10 MW High Temperature Gas-Cooled Reactor-Test Module (HTR-10) in China [5]. Meanwhile, prismatic fuel is used for GT-MHR in USA and the High-Temperature Engineering Test Reactor (HTTR) in Japan [5].

Research of kernel have been conducted to prepare kernel based on uranium dioxide (UO_2) [1,2,3,6,7] and other materials beside UO_2 such as uranium carbide (UC) [8], UCO [1,2,3,9,10], and uranium nitride (UN) [1,6]. The solution gelation technology is commonly applied for kernel fabrication [6] and consists of external gelation [11], internal gelation [11,12], and total gelation [13]. In the production of UO_2 kernel based on the external gelation method, the sequence of process is established consists of uranyl nitrate preparation, broth preparation, casting, aging, washing, drying, calcining, reducing and sintering process [13]. During the aging and washing process in kernel fabrication, liquid effluent is generated along with the process.

These liquid effluents are should be treated to minimize the toxicity of chemical compounds, reduce the volume of liquid waste, improve the process economically by recycling and reusing valuable component that can be used again in the kernel fabrication. Therefore, it is necessary to propose the conceptual design for liquid effluent treatment. The liquid effluent was a multicomponent mixture mainly consists of water contain ammonia solution (NH_4OH), ammonium nitrate (NH_4NO_3), tetrahydrofurfuryl alcohol (THFA), and isopropyl alcohol (IPA), which was generated from aging and washing process. The purpose of this work is to propose the conceptual design which is consists of few unit operations to recovery the valuable components: IPA and THFA. In this work, the conceptual designs are proposed for liquid effluent treatment of UO_2 kernel fabrication by the simulation process using CHEMCAD software. Besides CHEMCAD, Aspen Plus, Pro II and HYSYS are common software computer for chemical process design [14].

METHODOLOGY

The separation process based on different volatilities of the component in the multicomponent mixture. The operating condition of unit operations e.g. pressure and temperature are selected based on the optimum performance of separation process.

RESULTS AND DISCUSSION

The liquid effluent from UO_2 kernel fabrication was estimated as a multicomponent mixture as listed in Table 1.

Table 1. The quantity of component in the liquid effluent with total capacity of 28.67 kg/h

Component	Mass flowrate in kg/h
Water	19.65
NH_4OH	1.20
NH_4NO_3	0.34
THFA	1.58
IPA	5.91
Total	28.67

In this paper, the conceptual design is built to process the liquid effluent as described in Table 1. The selection of unit operations based on the advantage and the limitation of each separation equipment. The advantage and the limitation of equipment are assessed based on the complexity of separation process and the maturity of technology. The conceptual design is shown in Figure 1.
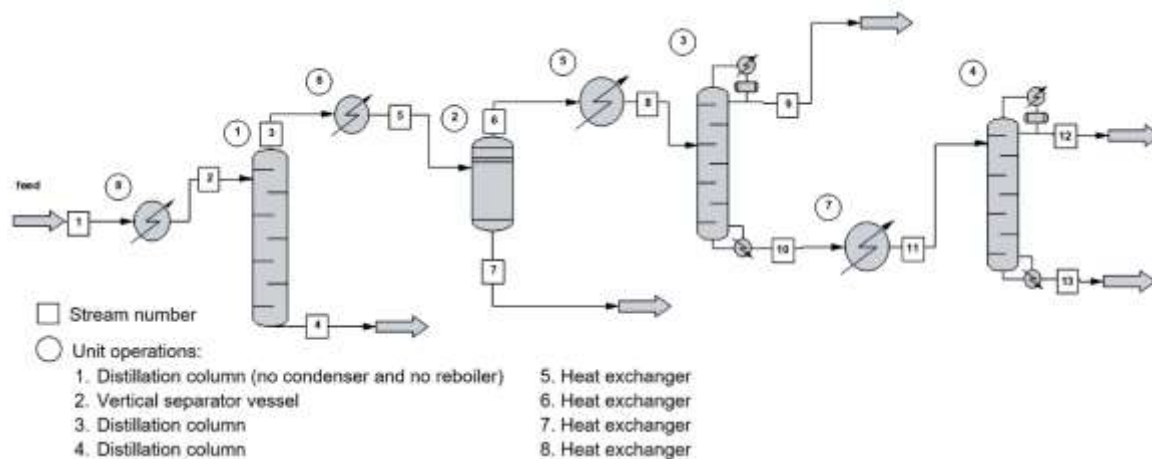
Figure 1. Conceptual design for liquid effluent treatment of UO₂ kernel fabrication

The distillation column is selected as the first main unit operation to remove the NH₄NO₃ and NH₄OH from the multicomponent mixture. The liquid effluent is a multicomponent mixture consists of water containing IPA, THFA and electrolyte species (NH₄OH and NH₄NO₃). The CHEMCAD software is able to accommodate the simulation that involved electrolyte species. Electrolyte species are not evaporated and ended at bottom stream of the first main unit operations. Therefore, it easier to purify the valuable component at the next unit operation.

The second main unit operation is vertical vessel. The determination of the vertical vessel is based on the simple separation process which is aimed to remove the water from the multicomponent mixture. The top exit stream of the vertical vessel contains multicomponent mixture rich of IPA and THFA.

The third and the fourth main unit operations are distillation columns which has duty to separate and purify the IPA and THFA respectively from the multicomponent mixture. The distillation column is chosen because distillation is common method for liquid-liquid separation to separate miscible multicomponent mixture which is the driving force derived by difference in volatility and it can obtain high purity product. The IPA product is collected from rectifying section as top product of distillation column, meanwhile the stream from stripping section which is the bottom part of distillation column was sent to the next distillation column to purify the THFA. In this work, the operation pressure of distillation column is set at 1 atm to economically the cooling water required for the distillate reach the dew point at the condenser of distillation column.

The results from simulation process using CHEMCAD software for conceptual design in Figure 1 are listed in Table 2. The IPA is collected from stream no. 9, the rectifying section of the third main unit operation, mass flowrate is 5.28 kg/h and purity of IPA at 0.99 in mass fraction. Although, in binary mixture of IPA-water has azeotrope at 0.874 in mass fraction of IPA and boiling point of mixture at 80.31°C [15]. The IPA product from stream no. 9 in this conceptual design has purity above the azeotrope of IPA-water mixture because in this case the ternary mixture of IPA-THFA-water is generated so it will overcome the limitation of IPA purity.

Table 2. Stream composition in the conceptual design

Stream No.	1	2	3	4	5	6	7	8	9	10	11	12	13
		Feed			Waste output			Waste output		IPA product			Waste output	THFA product
Temp °C	25	200	200	199.8	40	40	40	120	80.98	96.84	100	102	163.9
Pres bar	1	1	1	1	1	1	1	1	1	1	1	1	1
Flowrates in kg/h													
Water	19.65	19.65	19.62	0.02	19.62	0.16	19.47	0.16	0.03	0.12	0.12	0.11	0.01
NH₄OH	1.20	1.20	0.00	1.20	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
THFA	1.58	1.58	1.55	0.03	1.55	1.07	0.48	1.07	0.02	1.05	1.05	0.04	1.00
NH₄NO₃	0.34	0.34	0.00	0.34	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
IPA	5.91	5.91	5.91	0.01	5.91	5.88	0.02	5.88	5.22	0.66	0.66	0.66	0.00
Total kg/h	28.67	28.67	27.08	1.59	27.08	7.10	19.98	7.10	5.28	1.83	1.83	0.81	1.01

CONCLUSION

A proposed conceptual design is consisted with four main unit operations for separation process and four heat exchangers. The IPA product has mass flowrate of 5.28 kg/h and purity of 0.99 in mass fraction. The THFA product has mass flowrate of 1.01 kg/h and purity of 0.99 in mass fraction. In conclusion, the proposed conceptual design can be used for liquid effluent treatment from UO₂ kernel fabrication.

REFERENCES

[1] D. Schappel, K. Terrani, L. L. Snead, and B. D. Wirth, "Modeling radionuclide release of TRISO bearing fuel compacts during post-irradiation annealing tests," *Nucl. Eng. Des.*, vol. 357, p. 110428, 2020.

[2] P. A. Demkowicz, B. Liu, and J. D. Hunn, "Coated particle fuel: Historical perspectives and current progress," *J. Nucl. Mater.*, vol. 515, pp. 434–450, 2019.

[3] H. D. Gougar *et al.*, "The US Department of Energy's high temperature reactor research and development program – Progress as of 2019," *Nucl. Eng. Des.*, vol. 358, p. 110397, 2020.

[4] C. G. du Toit and H. J. van Antwerpen, "Effect of reactor vessel cooling insulation and reflector heat pipes on the temperatures of a pebble-bed reactor using a system CFD approach," *Nucl. Eng. Des.*, vol. 357, p. 110421, 2020.

[5] D. Schappel, N. R. Brown, and K. A. Terrani, "Modeling reactivity insertion experiments of TRISO particles in NSRR using BISON," *J. Nucl. Mater.*, vol. 530, p. 151965, 2020.

[6] R. L. Seibert, B. C. Jolly, M. Balooch, D. P. Schappel, and K. A. Terrani, "Production and characterization of TRISO fuel particles with multilayered SiC," *J. Nucl. Mater.*, vol. 515, pp. 215–226, 2019.

[7] M. Liu, Z. Chen, M. Chen, Y. Shao, B. Liu, and Y. Tang, "Scale-up strategy study of coating furnace for TRISO particle fabrication based on numerical simulations," *Nucl. Eng. Des.*, vol. 357, p. 110413, 2020.

[8] W. von Lensa, G. Brinkmann, J. Lillington, and F. Shahrokhi, "The status quo on HTGR decommissioning," *Nucl. Eng. Des.*, vol. 359, p. 110456, 2020.

[9] G. R. Bower, S. A. Ploger, P. A. Demkowicz, and J. D. Hunn, "Measurement of kernel swelling and buffer densification in irradiated UCO-TRISO particles," *J. Nucl. Mater.*, vol. 486, no. Supplement C, pp. 339–349, 2017.

[10] S. Meher, I. J. van Rooyen, and C. Jiang, "Understanding of fission

products transport in SiC layer of TRISO fuels by nanoscale characterization and modeling," *J. Nucl. Mater.*, vol. 527, p. 151793, 2019.

[11] C. Schreinemachers, G. Leinders, G. Modolo, M. Verwerft, K. Binnemans, and T. Cardinaels, "The conversion of ammonium uranate prepared via sol-gel synthesis into uranium oxides," *Nucl. Eng. Technol.*, 2019.

[12] R. D. Hunt, J. L. Collins, T. J. Reif, B. S. Cowell, and J. A. Johnson, "Key process parameters to modify the porosity of cerium dioxide microspheres formed in the internal gelation process," *J. Nucl. Mater.*, vol. 495, pp. 33–37, 2017.

[13] Z. Xiangwen *et al.*, "Preparation of ammonium diuranate particles by external gelation process of uranium in INET," *Nucl. Eng. Des.*, vol. 250, no. Supplement C, pp. 192–196, 2012.

[14] A. Argoti, A. Orjuela, and P. C. Narváez, "Challenges and opportunities in assessing sustainability during chemical process design," *Curr. Opin. Chem. Eng.*, vol. 26, pp. 96–103, 2019.

[15] S. Ma, X. Shang, M. Zhu, J. Li, and L. Sun, "Design, optimization and control of extractive distillation for the separation of isopropanol-water using ionic liquids," *Sep. Purif. Technol.*, vol. 209, pp. 833–850, 2019.