

THE ADSORBER COMPONENT ADDITION IN CHEMCAD SOFTWARE PALETTE FOR HELIUM PURIFICATION MODELLING AND SIMULATION

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ABSTRAK

PENAMBAHAN KOMPONEN ADSORBER PADA PALETTE SOFTWARE CHEMCAD UNTUK PEMODELAN DAN SIMULASI PEMURNIAN HELIUM. Sistem pemurnian helium (SPH) adalah sistem yang sangat penting dalam reaktor RDE. Sistem ini berfungsi menghilangkan pengotor pada pendinginnya berbentuk partikel debu dan gas-gas. Pengotor tersebut antara lain debu karbon, N₂, O₂, H₂, O₂, CH₄, CO, dan CO₂. Salah satu komponen yang dibutuhkan pada SPH adalah komponen adsorber. Pada palette software ChemCAD, komponen adsorber belum tersedia, sehingga perlu ditambahkan. Penambahan dilakukan melalui fasilitas icon Excel pada palettenya dengan membuat coding proses terlebih dahulu menggunakan program Visual Basic. Tujuan penambahan komponen adsorber pada software ChemCAD adalah agar software ini dapat digunakan untuk memodelkan dan mensimulasikan proses pemurnian helium pada pendingin primer RDE. Metode yang digunakan pada pemodelan adsorber adalah prinsip penyerapan dengan metoda Langmuir. Setelah coding untuk komponen adsorber selesai dibuat dalam Visual Basic kemudian diintegrasikan ke software ChemCAD, dan digunakan untuk memodelkan proses penyerapan gas pengotor. Pada penelitian ini dilakukan simulasi untuk penyerapan pengotor oksigen pada kondisi kriogenik. Proses ini merupakan tahapan terakhir pada SPH RDE. Validasi hasil penambahan komponen adsorber pada ChemCAD dilakukan dengan membandingkan hasil simulasi untuk kasus yang sama dengan software SuperPro Designer. Berdasarkan hasil kedua simulasi diketahui bahwa perbedaaan rata-rata hasil running ChemCAD dan SuperPro Designer tidak lebih dari 0,02% (0,0001), sehingga komponen adsorber yang ditambahkan pada software ChemCAD valid dan dapat digunakan untuk pemodelan dan mensimulasikan sistem pemurnian helium pada reaktor RDE.

Kata kunci : penambahan, komponen adsorber, ChemCAD, pengotor, purifikasi, RDE

ABSTRAK

THE ADSORBER COMPONENT ADDITION IN CHEMCAD SOFTWARE FOR HELIUM PURIFICATION MODELLING AND SIMULATION. Helium purification system (HPS) is an important system in RDE. This system removed dust particles and gas impurities in primary coolant of RDE. The impurities are dust carbon, N₂, O₂, H₂, O₂, CH₄, CO, and CO₂. One of the components which required in HPS is an adsorber. In ChemCAD software palette, adsorber component is not yet available and should be added. This paper discussed the adsorber component addition in ChemCAD. The addition is made by using Excel icon facility, while coding is made by using Visual Basic. The purpose of adding adsorber component to ChemCAD palette is it can be used for helium purification modeling. The adsorption process principle used in adsorber modeling is Langmuir method. After finishing the coding, adsorber component will be integrated into ChemCAD. This component then used for modeling and simulated the helium purification of primary coolant of RDE especially for oxygen adsorption. The simulation results are compared to SuperPro Designer modeled which is also demonstrated the same problem. The differences results of adsorber component simulation between ChemCAD and SuperPro Designer did not more than 0.02%. It can be concluded that the adsorber component which is added to the ChemCAD is valid so it can be used to simulate the helium purification system in RDE.

Keywords : adsorber component, addition, ChemCAD, impurities, RDE.

1. INTRODUCTION

Reaktor Daya Eksperimental (RDE) is developed in BATAN. This reactor is designed base on HTGR technology^[1]. RDE has indirect cycle power conversion system and uses pebble fuel type. The primary coolant of RDE is helium and secondary is water. Helium purification system (HPS) is an important system in RDE. This system removes impurities contained in primary helium cooling system^[2, 3]. There are two kinds of impurities i.e. particulates and gaseous form^[4]. The particulates impurities are graphite dust and fission products releases from the reactor core, and the gaseous impurities are N₂, O₂, H₂, O₂, CH₄, CO, H₂O and CO₂^[5]. Based on HTGR operating experiences graphite dust is very small, but it must be cleaned from the reactor coolant due to negative impact to system, structure and component (SCC) of reactor. The carbon dust will adhere to SCC reduces heat removal capacity^[6, 7]. Carbon dust is generally derived from the pebble fuel friction in the core. The gaseous impurities are derived from the interaction between water/air ingress with fuel/reflector in the core^[2].

ChemCAD (Chemical Computer Aided Design) is a software that can be used for process modeling in various industries^[8]. This software will be used to simulate the helium purification system of RDE. HPS has several main components including cyclone separator, filter, chemical reactors, condenser, and adsorber^[9]. Cyclone and filter used to remove carbon dust and other particulates impurities. Chemical reactors used to simulate the CuO oxidizer column bed. CuO is a catalyst in converting molecules of CO to CO₂ also converting CH₄ and H₂ into CO₂ and H₂O respectively^[10]. The H₂O output from column bed separated by water trap and CO₂ is captured by the molecular sieve adsorber. Other impurities gases such as N₂ and O₂ will be adsorbed in the last stage of activated carbon. The N₂ and O₂ molecular size is very small to capture them it required cryogenic conditions in the temperature of -180 °C^[11, 12].

Two important components in the HPS are molecular sieve and activated carbon adsorber^[13]. In process modeling usually adsorber component is used to simulate them. Both of these components are not available in ChemCAD therefore it must be created to simulate the HPS. This paper discusses the built of adsorber components to ChemCAD palette and integrate it into the HPS system. The purpose of creating adsorber component is to simulate the HPS in RDE. At first, adsorber coding is made based on Langmuir method by using Visual Basic. After this coding finished, it will integrated into ChemCAD with Excel icon facility in its palette. The component addition in ChemCAD will be validated and compared by using SuperPro Designer computer code with the same case of adsorption process in HPS. The comparing results expected very low differences between of both simulation executions so the adsorber component in ChemCAD can be used for HPS simulation.

2. THEORY

2.1 The Langmuir Adsorption in Isothermal Condition.

Adsorption is the adhesion of atoms, ions or molecules from a gas, liquid or dissolved solid to a surface^[8]. This process creates a film of the *adsorbate* on the surface of the *adsorbent*. This process differs from absorption, in which a fluid (the *absorbate*) is dissolved by or permeates a liquid or solid (the *absorbent*), respectively^[14]. Adsorption is a surface-based process while absorption involves the whole volume of the material. The term sorption encompasses both processes, while desorption is the reverse of it. Adsorption is a surface phenomenon.

Adsorption capacity is the ability of an adsorbent attracts a number of adsorbate. The adsorption process depends on, among others: a specific area of solids (adsorbent surface area), the concentration of solute or pressure equilibrium gas adsorption, the temperature during the process and the nature of the adsorbate or adsorbent itself. The

greater the surface area of the adsorbent, the adsorption power will grow stronger. Adsorption on the surface properties of solids are highly selective means that the mixture of substances is only one component adsorbed by certain solids. By its nature, there are two types of adsorption i.e. physical and chemical adsorption. Differences between both types are shown in Table 1. There are some methods in adsorption process i.e. Henry method, the Freundlich method and Langmuir method.

Table 1. The differences between chemisorption and physisorption^[11].

Physically adsorption (<i>physisorption</i>)	Chemically adsorption (<i>chemisorption</i>)
It arises due to van der Waals forces	It arises due to chemical bond formation
Enthalpy of adsorption is low (20-40 kJ/mol)	Enthalpy of adsorption is high (80-240 kJ/mol)
It is reversible	It is irreversible
It is not specific in nature	It is highly specific in nature
No appreciable activation energy is required	High activation energy is sometimes required
It depends on the nature of the gas. More easily liquitable gases are adsorbed reactly	It also depends on the nature of the gas. Gases which can react with the adsorbent, show chemisorption

2.2 Adsorber Unit and Adsorption Process Modeling

Unit operation (component) adsorber can be used to simulate the process of adsorption on a molecular sieve and activated carbon. The mass transfer process of adsorbate gas to the adsorbent using equation (1), and the solid-gas equilibrium described by Henry balance equation (2), Freundlich equation (3) and Langmuir equations in isothermal (4).

$$F_A = k_y a \frac{\rho_g}{M r_g} (y_A - y_A^*) S \Delta z \varepsilon \dots\dots\dots (1)$$

$$y_A^* = H_A \cdot x_A \dots\dots\dots (2)$$

$$y_A^* = K_f \cdot x_A^{n_f} \dots\dots\dots (3)$$

$$y_A^* = \frac{ax_A}{1+bx_A} \dots\dots\dots (4)$$

where F_A is the adsorbate mole transferred to the adsorbent per time (mol/s), k_y is the mass transfer coefficient in the gas phase (m^2/s), a is the area per unit volume (m^2/m^3), y_A is the mole fraction of adsorbate in gas, y_A is the adsorbent mole fraction on the surface of the adsorbent, S is the cross-sectional area of the column (m^2), Δz is the length increment (m), ε is the porosity of the bed, H_A is the Henry constant, K_f and n_f is the Freundlich equilibrium constant, A and b are Langmuir equilibrium constants, x_A is the adsorbate mole fraction per mass of adsorbent.

2.3 Mass balance adsorbate in the adsorption process increment of volume $S \cdot \Delta z$

In the adsorption process of adsorber having an adsorbent of S volume, with increment of $z+\Delta z$, can be solved by equation of (5), (6) and (7). The volume adjustment model of adsorber is shown in Figure 1.

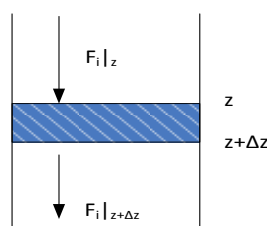


Figure 1. The increment model of adsorber volume.

Mass flow rate of input – mass flow rate of output – adsorption rate =
rate of gas accumulation (5)

$$\begin{aligned} & (Gy_A|_z - Gy_A|_{z+\Delta z}) - \left(D_{e,ax} \varepsilon S \frac{\rho_g}{Mr_g} \frac{\partial y_A}{\partial z} \Big|_z - D_{e,ax} \varepsilon S \frac{\rho_g}{Mr_g} \frac{\partial y_A}{\partial z} \Big|_{z+\Delta z} \right) - k_y (y_A - y_A^*) a \frac{\rho_g}{Mr_g} S \Delta z \varepsilon = S \Delta z \varepsilon \frac{\rho_g}{Mr_g} \frac{\partial y_A}{\partial t} \\ \lim_{\Delta z \rightarrow 0} & \left[\frac{(Gy_A)|_{z+\Delta z} - (Gy_A)|_z}{\Delta z} \right] + \lim_{\Delta z \rightarrow 0} \left[\frac{\left(D_{e,ax} \varepsilon S \frac{\rho_g}{Mr_g} \frac{\partial y_A}{\partial z} \Big|_{z+\Delta z} - D_{e,ax} \varepsilon S \frac{\rho_g}{Mr_g} \frac{\partial y_A}{\partial z} \Big|_z \right)}{\Delta z} \right] - k_y a (y_A - y_A^*) \frac{\rho_g}{Mr_g} S \varepsilon = S \varepsilon \frac{\rho_g}{Mr_g} \frac{\partial y_A}{\partial t} \\ & -G \frac{\partial y_A}{\partial z} + D_{e,ax} \varepsilon S \frac{\rho_g}{Mr_g} \frac{\partial}{\partial z} \left(\frac{\partial y_A}{\partial z} \right) - k_y a (y_A - y_A^*) \frac{\rho_g}{Mr_g} S \varepsilon = S \varepsilon \frac{\rho_g}{Mr_g} \frac{\partial y_A}{\partial t} \end{aligned} \quad \dots\dots\dots (6)$$

2.4 Mass balance adsorbate in the solid volume element

$$\begin{aligned} & \left(k_y (y_A - y_A^*) a S \Delta z \varepsilon \frac{\rho_g}{Mr_g} \right) - 0 = S \Delta z (1 - \varepsilon) \rho_p \frac{\partial x_A}{\partial t} \\ & \left(k_y (y_A - y_A^*) a S \Delta z \varepsilon \frac{\rho_g}{Mr_g} \right) - 0 = S \Delta z \rho_b \frac{\partial x_A}{\partial t} \text{ and} \\ & \frac{\partial x_A}{\partial t} = \frac{k_y (y_A - y_A^*) a \varepsilon \frac{\rho_g}{Mr_g}}{\rho_b} \dots\dots\dots (7) \end{aligned}$$

Where the initial condition of $x_A(z, 0) = 0$, $y_A(z, 0) = 0$, and boundary condition of $y_A(0, t) = y_{Ain}$. The value of G is the mass flow rate or G = mol flow (gmol/s), and ρ_b = bulk density (g/m3). Determination of the physical parameter of the Diffusivity property (Dm) can be determined by equation (8). (Coulson and Richarson, 1983)

$$D_m = 1.10 \cdot 10^{-7} T^{1.75} \frac{\left(\frac{BM_{H_2} + BM_{CO_2}}{BM_{H_2} BM_{CO_2}} \right)^{1/2}}{P} \frac{1}{101325 \left(v_{H_2}^{1/3} + v_{CO_2}^{1/3} \right)^2} \dots\dots\dots (8)$$

Where BM_i is the molecular weight (g/gmol), the u_i is the diffusion volume. The axial dispersion coefficient value is shown in equation (9).

$$D_{e,ax} = 0,37 D_m + \frac{0,5 v_s d_p}{1 + \frac{9,7 D_m}{v_s d_p}} \dots\dots\dots (9)$$

Where v_s is superficial velocity, the mass transfer coefficient is calculated using equation (10) and the Reynold number in equation (11)

$$k_y = \frac{D_m}{d_p} (2 + 1,1 Re^{0,6} Sc^{1/3}) \dots\dots\dots (10)$$

$$Re = \frac{d_p v_s}{\nu} \dots\dots\dots (11)$$

$$Sc = \frac{\mu}{D_m} \dots\dots\dots (12)$$

where d_p is the particle diameter (m), v is the superficial velocity (m/s), kinematics viscosity (m^2/s), and μ is the viscosity (Pa.s). To calculate cross

sectional area is using the equation: $S = \pi R^2$, with R = radius of bed (m), specific surface specification calculated by equation (13) and superficial velocity equation calculated by equation (14).

$$a = \frac{6(1-\varepsilon)}{d} \dots\dots\dots(13)$$

$$v_s = \frac{G}{\frac{\rho_g}{Mr_g} S} \dots\dots\dots(14)$$

Where G is mass flow rate or mol flow (gmol/s), with the equilibrium constant is determined by literature or experimental research.

3. METHODOLOGY

The existing of components in ChemCAD palette has its limitations. The adsorber component did not available in palette. However, in ChemCAD software there is an Excel icon feature to make some components addition. With this feature, we can create program coding of component with its function by using Visual Basic and add it as a new component in ChemCAD palette . The icon of the Excel unit operation in the ChemCAD palette is shown in Figure 2. In this study the adsorber component will be added, the adsorber component will be used to model the molecular sieve and activated carbon in the HPS of RDE. Both of these components conduct to adsorbed the helium gas impurities i.e. CO, H₂O, CO₂, N₂ and O₂.

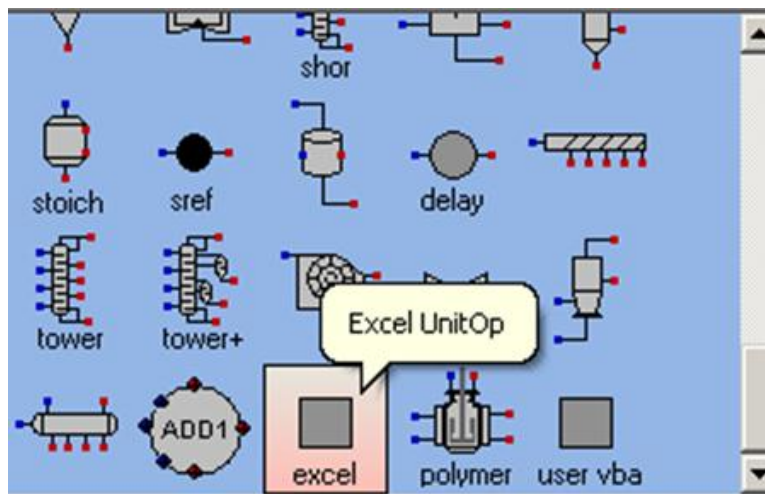


Figure 2. Icon Excel of Unit operation in ChemCAD.

Table 2. The input value for adsorber modeled and simulation

Helium mass flow rate	:	10.5 kg/hour
Helium temperature	:	-180 C
Helium pressure	:	30 bar
Oxygen concentration (as impurity)	:	10

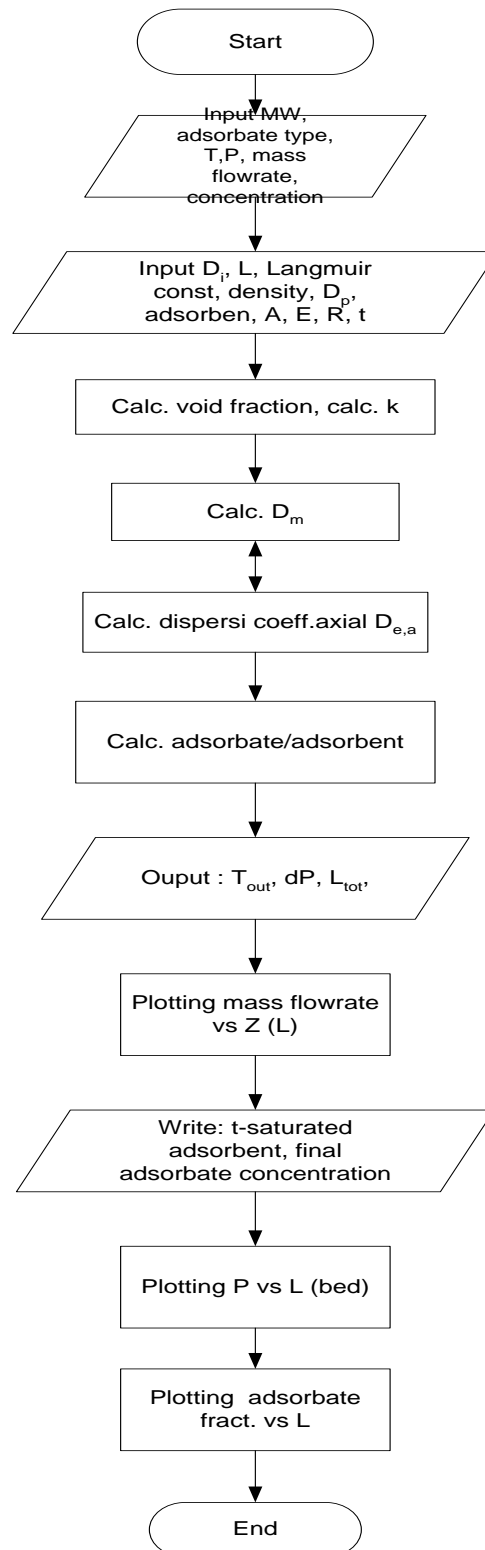


Figure 3. The flowchart for the adsorber coding in Langmuir method.

The main primary coolant stream of RDE is 4.3 kg per second, and the HPS bypass flow rate is designed of 5% or about 0.215 kg per second. The RDE parameters used in this model are shown in Table 2. The results from the ChemCAD is compared with SuperPro Designer adsorber model which is shown in Figure 4 and Figure 5.

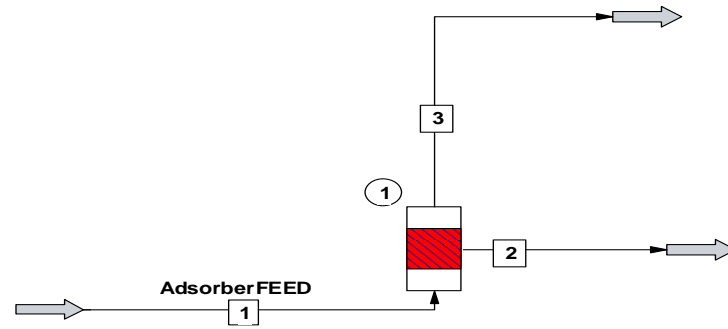


Figure 4. The adsorber modelling by using ChemCAD.

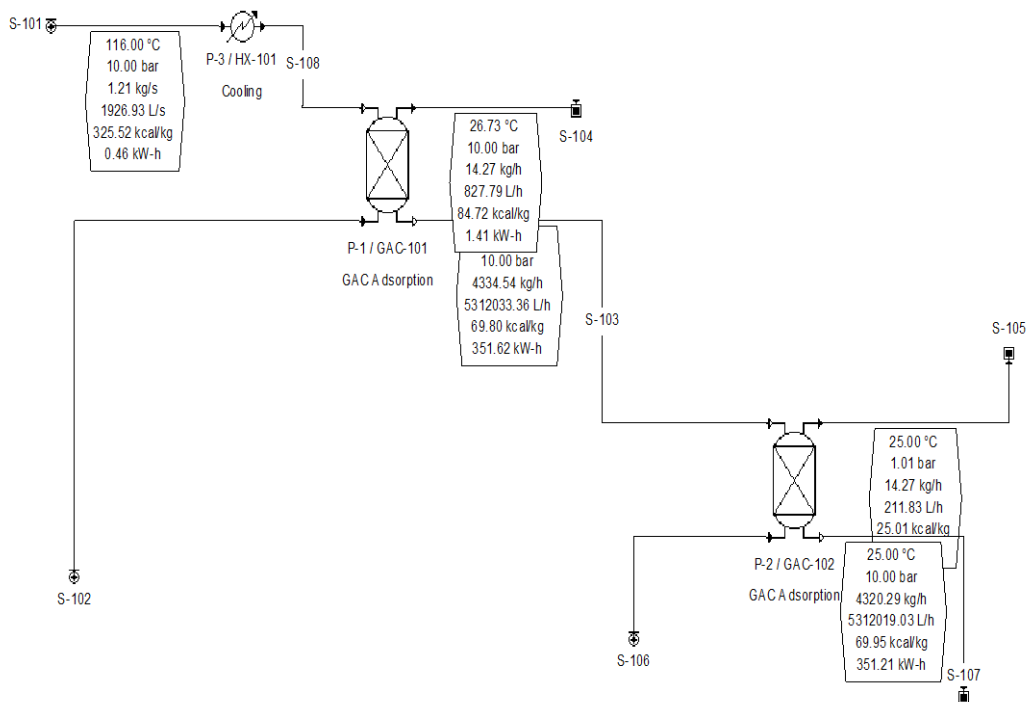


Figure 5. The adsorber modeling by using SuperPro Designer.

4. RESULT AND DISCUSSION

The adsorber component has already integrated with the ChemCAD and used to create an HPS model of RDE especially for adsorption process. Simulation is done to validate the adsorber process function. The results from the ChemCAD is compared with SuperPro Designer adsorber model. Both of this software is built for industrial process modeling. In SuperPro, there are three optional mode choice for gas adsorption. First is Langmuir, second is Henry and the third is Freundlich method. Adsorption modeling in ChemCAD is used Langmuir method so it can be compare by using SuperPro by choosing the same method (first mode option).

The last stage of HPS is the adsorption of N_2 and O_2 uses activated carbon. This process operates at very low temperatures (cryogenic), i.e. $-180\text{ }^\circ\text{C}$. In HPS train, the outside carbon column is circulated by liquid nitrogen to maintain very low temperature. The N_2 and O_2 gases will be adsorbed into the activated carbon, when reached it saturation the activated carbon will be regenerated. One of the specifications of activated carbon used as shown in Table 3.

Table 3. Active carbon spesification in cryogenic condition.

Parameter	Value/unit
Spesific volume	0,6 cm ³ /g
Average porosity diameter	24 Å
Particle size	2 mm or 5 mm
Specific surface area	1100 m ² /g
Internal porosity	55% - 75%
Zeolithe density	450 kg/m ³

The comparison results of simulation both of those two softwares are shown in Table 4.

Table 4. The effect of active carbon adsorption to the O₂ gas.

ChemCAD Adsorber		SuperPro Designer Adsorber		Differences (%)
Duration (hour)	O ₂ remaining (ppm)	Duration (hour)	O ₂ remaining (ppm)	
0.5	0.0018	0.5	0.0019	5.2632
0.75	0.0022	0.75	0.0021	-4.7619
1.0	0.0026	1.0	0.0025	-4.0000
2.0	0.0055	2.0	0.0054	-1.8519
3.0	0.0114	3.0	0.0115	0.8696
3.5	0.0164	3.5	0.0165	0.6061
4.0	0.0237	4.0	0.0238	0.4202
4.5	0.0342	4.5	0.0340	-0.5882
5.0	0.0494	5.0	0.0493	-0.2028
5.5	0.0712	5.5	0.0713	0.1403
6.0	0.1028	6.0	0.1029	0.0972
6.5	0.1483	6.5	0.1482	-0.0675
7.0	0.2139	7.0	0.2138	-0.0468
7.5	0.3088	7.5	0.3089	0.0324
8.0	0.4455	8.0	0.4454	-0.0225
8.5	0.6428	8.5	0.6429	0.0156
9.0	0.9275	9.0	0.9276	0.0108
9.5	1.3384	9.5	1.3382	-0.0149
10.0	1.9311	10.0	1.9313	0.0104
10.5	2.7862	10.5	2.7863	0.0036
11.0	4.0212	11.0	4.0214	0.0050
11.5	5.7969	11.5	5.7968	-0.0017
11.8	6.9710	11.8	6.9712	0.0029
11.8	7.1097	11.8	7.1098	0.0014

The differences between SuperPro and ChemCAD simulation results is not more than 0.02%. The oxygen concentration adsorbed is shown in Figure 6. After going through the process of absorption with Activated Carbon is expected to have pure helium. The purified helium will be stored into storage tanks. Cryogenic conditions are conditions at very low temperatures. In this simulation the cryogenic condition that

applied to the Activated Carbon is $-180\text{ }^{\circ}\text{C}$ of temperature. At that condition the absorption capacity of activated carbon becomes larger because the gas impurities moved more slow and it makes easier to be absorbed^[13].

Except temperature, the ability of active carbon adsorption to N_2 and O_2 impurities is determined by many factors, including contact time and the amount of activated carbons. Sufficient contact time allows the adsorbate to be adsorbed into the surface of the adsorbent, and less contact time results is not achieving the adsorption equilibrium so that impurities taken are not optimal. Therefore the active carbon bed is simulated with a height of 15 m, it is expected that the adsorbate has sufficient contact time to be absorbed by the adsorbent. In this study, to determine the time of regeneration of activated carbon is done by observing the concentration of impurities N_2 and O_2 on the clean output stream of activated carbon bed, i.e. stream number 3.

Based on the maximum limit of O_2 impurity gas allowed in helium coolant is 0.02 ppm^[14], if it is using the active carbon bed design as much as 654.16 kg as above bed design then after operating for 3.5 hours active carbon must be regenerated. From Table 2, it is known that at time of 3.5 hours, the remaining O_2 impurity concentration is 0.0164 ppm, and at time of 4 hours, the impurity concentration has exceeded 0.02 ppm.

The O_2 impurity conditions used in this simulation is 1018 ppm or equivalent to 50,900 times the normal operating conditions of RDE, so if it is implemented in HPS, and the RDE operates under normal conditions it can be concluded that the active carbon bed will be outage $(3.5 \times 50,900)\text{ hr} = 178,150$ hours or equivalent to 7422 days.

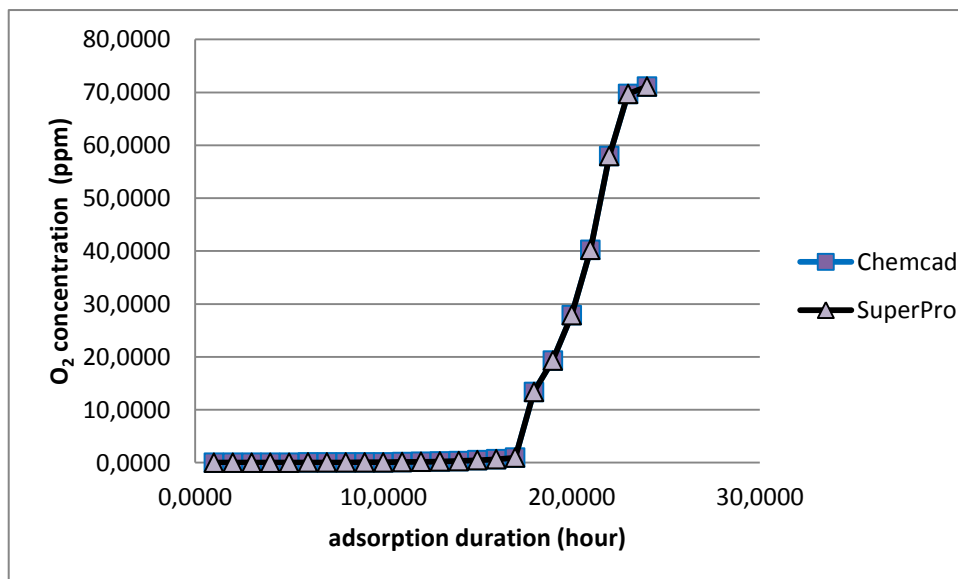


Figure 6. The correlation between adsorption duration vs remaining O_2 in the output stream of carbon active column bed.

5. CONCLUSION

One of the components which is required in HPS is an adsorber. At ChemCAD software, adsorber component is not yet available and should be added. The adsorption principle used in adsorber components is Langmuir method. The additions will used excel icon feature in ChemCAD palette, and the coding is created using by Visual Basic. After finishing the coding step, adsorber component integrated into ChemCAD. This component then used to modeled and simulated the helium gas purification of RDE. The simulation results are compared to SuperPro Designer which is also simulated the same problem. The results shown that ChemCAD and SuperPro

Designer differences did not more than 0.02%. It can be concluded that the adsorber component which is added to the ChemCAD software can be used to simulate the helium purification system in RDE reactors.

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