Preliminary Studies of Two-Phase Reactive Process of Sodium-CO₂ in S-CO₂ Power Conversion Cycle Coupled to SFR System

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1. Introduction

One of the most promising Generation IV Nuclear Energy Systems under development is the sodiumcooled fast reactor (SFR). Yet, the issues surrounding sodium opacity and its vigorous chemical reactivity with air or water/steam are critical technical challenges for the SFR. In the steam Rankine cycle, a traditional power conversion system of SFR, the potential sodium-water reaction (SWR) has been considered as one of the most critical issues. As a competing alternative to the steam Rankine cycle, the supercritical CO₂ (S-CO₂) Brayton cycle has been highlighted due to its high thermal efficiency, compact turbomachinery and heat exchangers sizes, and the reduced risk of SWRs. While the reduced risk of an SWR is considered as the one of most pronounced benefits of S-CO₂ Brayton cycle, there is still an interaction problem between liquid sodium and CO_2 .

Although the chemical interaction between liquid sodium and CO₂ demonstrates less serious potential risks than those of a SWR [1], the Na/CO₂ interaction should be understood to evaluate safety and reliability of Intermediate Heat eXchanger (IHX). A noticeable characteristic of the reaction environment is that there is a large pressure difference between the liquid sodium and CO₂ side by about 1 and 200 bar, respectively. This would imply that the presence of a micro-crack in a heat exchanger tube will cause a high-pressure leak of CO₂ into liquid sodium side [2]. Although the Na/CO2 interaction may play an important role in the safety of the SFR reactor system, there has not yet been any research on understanding Na/CO₂ reaction by leakage through IHX. For this problem, the Korea Advanced Institute of Science and Technology (KAIST) research team is studying the mechanism of CO₂ leakage and Na/CO₂ interaction in more details.

The KAIST research team developed the MATLAB code, KAIST_HXD, which can be used to design and evaluate performance of a heat exchanger of an S-CO₂ cycle. The size of heat exchanger and the amount of CO_2 in the cycle are calculated from the KAIST_HXD code to estimate the amount of reaction products in Na/CO₂ interaction as well as liquid sodium.

2. Research and Results

2.1 CO₂ Leak Mechanism Research

A large pressure difference between the liquid sodium and the CO_2 side can cause an expansion of CO_2 when the leakage of CO_2 is occurred. It is certain that the volume of CO_2 will suddenly increase then a decrease of CO_2 temperature will be followed. The blowdown of CO_2 may take place in the form of two-phase reactive flow in IHX with liquid sodium. Also, this CO_2 during leakage into liquid sodium side may be chocked and chocked flow of supercritical state CO_2 is still a remaining issue.

So it is necessary to define an exact mechanism of how the leakage takes place and the behavior of CO_2 through the leakage. Specifically, it should be studied that what happens when there is a large break or a small crack in IHX and how the pressure of system and the flow of CO_2 will be changed in each case.

2.2 Na/CO₂ Interaction Modeling

The expected major chemical reactions when the $S-CO_2$ leaks into liquid sodium side in a Na/CO₂ IHX of SFR already have been proposed in the following equations from (1) to (4), by previous experimental and theoretical studies [3]. These reactions occur competitively and all four reactions generate heat.

$$4Na(l) + CO_{2}(g) \rightarrow 2Na_{2}O + C + \Delta H^{\circ}$$
$$\Delta H^{\circ} = -442.3kJ/mol \qquad (1)$$

$$4Na(l) + 3CO_2(g) \rightarrow 2Na_2CO_3 + C + \Delta H^\circ$$

$$\Delta H^{\circ} = -1081.1 kJ/mol \qquad (2)$$

$$2Na(l) + 2CO_2(g) \rightarrow Na_2CO_3 + CO(g) + \Delta H^{\circ}$$
$$\Delta H^{\circ} = -454.5kJ/mol \qquad (3)$$

$$4Na(l) + 4CO_2(g)$$

$$\rightarrow Na_2C_2O_4 + CO(g) + Na_2CO_3 + \Delta H^{\circ}$$
$$\Delta H^{\circ} = -980.1kJ/mol \qquad (4)$$

Although most of the reaction products are in solid state, toxic gaseous carbon monoxide and accumulated heat in the system can be serious problems. Hence, it is important to quantify how much reaction products and heat will be produced.

The KAIST research team calculated the amount of CO₂ from the KAIST_HXD code for a proposed system layout of Gen-IV SFR under development. The code is planned to develop the conceptual design for this

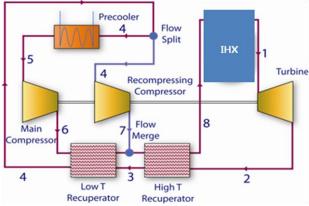


Fig 1. The Layout of $S-CO_2$ Recompressing Brayton Cycle from works of H. J. Yoon et al.

Gen-IV SFR with the optimized S-CO₂ recompressing Brayton cycle layout like in Fig 1 [4]. The S-CO₂ Brayton cycle can achieve high pressure ratio and efficiency of each component as well as cycle itself. In this cycle, to save rejected heat and increase the total thermal efficiency of the cycle, the fluid is recompressed [4]. And the cycle consists of three kinds of heat exchangers (i.e. IHX, high and low temperature recuperators and precooler). In the KAIST_HXD code, all heat exchangers were assumed to be the Printed Circuit Heat Exchanger (PCHE) type, which is one of the most widely accepted heat exchangers for the S-CO₂ power cycle applications.

The amount of CO_2 was obtained by calculating the total amount of CO_2 in above all heat exchangers of the S-CO₂ cycle. With the obtained amount of CO_2 , the amount of liquid sodium and reaction products were calculated by molar ratio of the reaction shown in Eq. (3). The total released heat was calculated by enthalpy change of Eq. (3). The reaction shown in Eq. (3) was used as a reference because it becomes the dominant reaction as temperature increases [3]. The calculated quantities of the reaction products and amount of heat released are summarized in Table I.

Table I: Quantitative Calculation of Reaction Products and Heat of Reaction

Mass (ton)				Total Heat of
CO ₂	Na	Na ₂ CO ₃	СО	Reaction (GJ)
1.00	0.52	0.60	0.32	5.16
2.00	1.05	1.20	0.64	10.33
3.00	1.57	1.81	0.96	15.49
4.00	2.09	2.41	1.27	20.65
5.00	2.61	3.01	1.59	25.82
6.00	3.13	3.61	1.91	30.98

The results presented in Table I are a rough estimation based on the thermodynamic equilibrium, which assumes that all the existing reactive molecules underwent reaction shown in Eq. (3). Nevertheless, the total heat of reaction on the order of Giga-Joule (GJ) raises concern of overheating upon the Na/CO₂ reaction. If this heat is not properly removed, the temperature of the location may continuously increase. This may imply that the sodium self-ignition which was observed in a very high temperature condition of sodium (~600 °C) as the results of Na/CO₂ interaction test performed by EOH et al. [3] will become more problematic. In addition, a drastic pressure and temperature transient can also occur [2].

3. Summary and Further Works

The amount of liquid sodium and reaction products in Na/CO₂ reaction was estimated based on the assumption that all CO₂ molecules underwent reaction. However, the obtained total heat of reaction was on the order of GJ, raising concerns of the proper removal of thermal energy release associated with the reaction. Therefore, the Na/CO₂ interaction needs to be studied in more details because it is very complex phenomenon and may play an important role in safety of S-CO₂ cycle couple to SFR system.

The KAIST research team will study for reduction of heat of reaction by literature survey and chemistry database. More studies for self-ignition of sodium and more detailed analysis of Na/CO_2 interaction will be also conducted.

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