Description of the First Observed Sodium-CO₂ Reactions in the Sodium- CO₂ Interaction Experiment (SNAKE)

Nuclear Engineering Division

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ANL-SMR-7

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September 30, 2013

ABSTRACT

One appealing feature of the supercritical carbon dioxide Brayton cycle energy conversion system is the small footprint that the hardware requires, which is in part due to the use of Printed Circuit Heat Exchangers (PCHEs) as the heat source heat exchanger (sodium-to-CO₂) as well as the recuperator and cooler modules. Although PCHEs have a high degree of structural integrity, the potential for leaks to develop between the sodium and CO₂ coolant channels in the secondary heat exchanger cannot be ruled out, and this would lead to discharge of high pressure CO₂ into the secondary coolant circuit. Due to the robustness of the PCHE design, catastrophic failure leading to CO₂ jet blowdown into the secondary sodium loop is not deemed likely. Rather, small cracks (or micro-leaks) may develop in which CO₂ will bleed into the secondary system at a relatively low rate and chemically react with the sodium.

The goal of the sodium-CO₂ interaction tests is to gain a fundamental understanding of sodium-CO₂ interactions under prototypical conditions of compact diffusion-bonded heat exchanger failure, a fundamental understanding of self-plugging if it occurs, and the development of one-dimensional phenomenological models for the interactions between high-pressure CO₂ issuing into liquid sodium from a micro-leak across a stainless steel pressure boundary. These models will be validated using experiment data.

Therefore, an experiment program at Argonne was initiated in Fiscal Year 2010 to investigate the reaction characteristics between sodium and CO₂ under micro-leak conditions. Several reports have described the facility scaling rationale and design. Assembly of the SNAKE (S-CO₂, Na Kinetics Experiment) began in Fiscal Year 2011 and was completed in July 2012. Approximately 44 lbs (20 kg; ~21 L/5.5 gal) of sodium was transferred into the SNAKE dump tank from a drum of clean sodium in July 2012.

The first sodium-CO₂ interaction experiment was carried out at SNAKE in September 2012. This test was successful in that supercritical carbon dioxide was sparged into a pool of sodium through a 64 μ m diameter nozzle.

A series of sodium-CO₂ interaction experiments were carried out in Fiscal Year 2013 in the SNAKE experiment. These tests successfully injected supercritical carbon dioxide into a pool of sodium through a 64 μ m diameter nozzle. A reaction between the CO₂ and sodium was detected. The extent of this reaction was unexpected since the initial sodium temperature was 145 °C, a temperature range where previous researchers have detected little or no chemical reaction between these species. The important difference between the SNAKE experiment and previous research is that the SNAKE geometry and conditions promote high-interfacial area and mixing between the CO₂ and sodium. These characteristics could be very important in promoting accelerated chemical reactions and will be studied further as the SNAKE test matrix is carried out.

Approximately 325 standard liters of CO₂ were injected into a 45 cm (15 inch) high column of sodium at a nominal temperature of 150 °C over the course of 3 hours. The inlet CO₂

pressure was gradually increased from 3 MPa to 11 MPa over these three hours in order to study the impact of pressure and flow rate on the sodium-CO₂ interactions. For the first time in SNAKE, chemical reactions between the sodium and CO₂ were observed. Sodium temperature increased from 150 °C to nearly 270 °C over the course of the experiments. Production of carbon monoxide was clearly detected, with up to 0.3 percent of the injected CO₂ converted to CO over the course of the experiments. A significant amount of CO₂ was converted into solids, approximately 37 percent. This reaction percentage is considered to be high given the sodium temperature of this experiment. It is possible that one or more mass flow meters are out of calibration which would explain the surprisingly high conversion ratio. However, significant quantities of solid materials have been generated inside the test vessel and were observed using an inspection video camera (see Figure 1). The reaction product particulates will be removed, sampled, mounted, and sent to Analytical Chemistry in early Fiscal Year 2014. Substantial work will be required to remove these particulates from the test vessel and restart sodium-CO₂ interaction experiments using clean sodium.

Figure 1: Images of solids matrix in test vessel produced via sodium-CO₂ chemical reaction. (left) Stainless lance that is penetrating several inches into the solids matrix. (right) Close-up of hole that remains after lance is removed.

No plugging of the micro-nozzle was detected during this experiment. Data from these experiments was reported and initial analyses of the data were completed and discussed. Additional analysis of this data and improvements of the data quality and facility control will take place in early Fiscal Year 2014.

Fiscal Year 2014 will involve cleaning the test vessel of the reaction products generated by the experiments detailed in this report, carrying out additional sodium-CO₂ and sodium-CO reaction experiments, making additional hardware improvements to improve data quality, and preparing a detailed report to be delivered in September 2014. Model development and validation are planned to begin in Fiscal Year 2014, with initial modeling results included in the September 2014 report.

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1 Introduction

The Department of Energy's Office of Nuclear Energy (DOE-NE) Small Modular Reactor (SMR) program is focused on the development of clean, affordable nuclear power. SMRs have the benefit of lower initial capital investment, scalability, and siting flexibility while also having the potential for enhanced safety and security. The Advanced SMR (aSMR) program contains a spectrum of experimental facilities to support R&D on advanced SMR concepts. One such experimental is the ANL Na-CO₂ Interaction facility, SNAKE (S-CO₂ Na Kinetics Experiment).

The supercritical carbon dioxide (S-CO₂) Brayton cycle, coupled with a sodium-cooled SMR, has been identified as a new and innovative energy conversion technology that could contribute to meeting the SMR objectives especially in terms of improved economics. One appealing feature of this energy conversion system is the smaller footprint that the hardware requires relative to the traditional superheated steam cycle, which is in part due to the use of Printed Circuit Heat ExchangerTM (PCHETM) or Hybrid Heat Exchanger (H²X) compact diffusion-bonded heat exchangers as the heat source heat exchanger (sodium-to-CO₂) as well as the recuperator and cooler modules. Although PCHEs have a high degree of structural integrity, the potential for leaks to develop between the sodium and CO₂ coolant channels in the secondary heat exchanger cannot be ruled out, leading to discharge of high pressure CO₂ into the secondary coolant circuit. Due to the robustness of the PCHE design, catastrophic failure leading to CO₂ jet blowdown into the intermediate sodium loop is not deemed likely. Rather, small cracks (or micro-leaks) may develop in which CO₂ will bleed into the secondary system at a relatively low rate and chemically react with the sodium.

In recognition of the anticipated failure mode for a PCHE sodium-to-CO₂ heat exchanger, an Argonne experiment program, called SNAKE, was initiated in Fiscal Year 2010 (see Farmer et al., 2010; Gerardi et al. 2011, 2012a&b, 2013) to investigate the reaction characteristics between sodium and CO₂ under micro-leak conditions. The goal of the SNAKE facility is to:

1. Determine the fundamental nature and extent of the chemical reactions that occur when high-pressure CO₂ is injected into liquid sodium from a micro-leak across a stainless steel pressure boundary as a function of the sodium pool temperature and inlet CO₂ flowrate,
2. Examine the potential for the micro-leak to seal itself up as a result of blockage formation from the chemical reaction byproducts of the Na-CO₂ reaction, or as a result of oxide layer buildup on the crack faces,
3. Develop one-dimensional phenomenological models for the interactions between high-pressure CO₂ as it issues into liquid sodium from a micro-leak across a stainless steel pressure boundary. These models will be validated using the experiment data obtained during the completion of the previous two objectives.

An initial test matrix for fundamental sodium (Na)-carbon dioxide (CO₂) interactions was formulated to provide the data needed to understand sodium-CO₂ interactions and their consequences for Sodium-Cooled Fast Reactors (SFRs) incorporating supercritical CO₂ Brayton cycle power converters with compact diffusion-bonded sodium-to-CO₂ heat

exchangers (Gerardi et al., 2013). The detailed test matrix recognizes that with the ongoing incorporation of improvements to the facility in Fiscal Year 2013, testing in SNAKE in Fiscal Year 2013 and early Fiscal Year 2014 may largely be characterized as shakedown testing, such that testing will need to be extended into following years. This report provides an update on the improvements made to SNAKE in Fiscal Year 2013 and on shakedown testing that included sodium-CO₂ interaction experiments where chemical reactions were observed.

2 Fiscal Year 2013 Improvements to SNAKE

A summary of the improvements that were made to SNAKE over the course of Fiscal Year 2013 are included in this section. A recent photo of the entire experiment is shown in Figure 2 where improvements and modification to the test vessel are visible. Particularly, the test vessel has been heavily insulated and a yellow & red gamma-level meter was installed directly beside the vessel.

Figure 2: Recent photograph of the SNAKE experiment at Argonne.

2.1 Thermal acoustic gas analyzer

The mass spectrometer has the ability to detect a broad range of molecules across a large spectrum of concentrations; however, it is not well suited to reliably detect molecule concentrations below several hundred ppm without very rigorous calibration and maintenance. Since the mass spectrometer for SNAKE is attached to the apparatus itself in a large high-bay space and is not stored in a laboratory environment, rigorous calibration and maintenance was not deemed possible. Therefore, a dedicated carbon monoxide gas monitor was purchased, the MSA Chemgard Infrared Thermal Acoustic gas analyzer (see Figure 3).

Figure 3: Chemgard Gas Monitor installed connected to the SNAKE experiment exhaust line.

The Chemgard Gas Analyzer has a sensitivity in the range of 0-1000 ppm of CO with an accuracy of $\pm 10\%$ of the reading. This instrument does not have cross-sensitivity to water vapor. In fact, humidity is added to the gas flow and subtracted from the gas reading in order for the sensor to work optimally. However, there is some CO cross sensitivity to CO₂; up to 15 ppm of CO is detected for 1% CO₂ according to the instrument manual (MSA, 2006).

2.2 Effluent gas flow meter

A digital mass flowmeter was added to the exhaust stream in order to enable mass balance calculations to be performed for each experiment. This is a Teledyne Hastings HFM-D-300(H) flow meter (see Figure 4) with a range of 0-30 SLM of argon. The mass spectrometer was used to measure the exhaust gas ratio. The exact mass flow was then determined through the Teledyne HFM-D-300. Known gas conversion factors from Teledyne were used to calculate the actual mass flow of the argon, carbon dioxide and carbon monoxide mixture from the measured equivalent argon mass flow.

Figure 4: Teledyne exhaust gas flow meter.

2.3 Gamma level-meter

Modeling of the sodium-CO₂ reaction in SNAKE required that quality measurements of sodium level be obtained prior to and during experiments. There are a number of methods to determine the sodium level with a wide range of accuracy. Nearly all of them require that some object be immersed or in contact with sodium (*Sodium Technology*, 1970; *Liquid-Metals Handbook*, 1950). Sodium contact is not desired in the SNAKE experiments because these objects could alter the chemical reaction and flow dynamics of the CO₂ in the sodium which would then complicate modeling. One method of non-contact sodium level measurement is via gamma ray absorption. This type of level measurement is routinely used in challenging environments including subsea separator level and composition measurements.

The Tracerco LevelFinderPlus was selected to measure sodium level in SNAKE. It consists of four small cesium-137 sealed-sources (each 0.2 mCi). These sealed-sources are placed in shielding containers that are equipped with a shutter to enable a collimated gamma beam to shine through the SNAKE test vessel toward an array of Geiger-Müller (GM) tubes. The radiation detected by the GM array can be linearly related to the sodium level in the test vessel. The four sealed radioactive sources (Cs-137 sources, each < 0.2 mCi) each consist of a 0.2 mCi Cs-137 ceramic plug double-encapsulated in 8x6 mm stainless steel containers which are classified as “DOT special form containers.” The sealed radioactive sources are then installed and padlocked inside lead-shielded blocks which have ports with shutters to allow opening and closing of the radiation beam. Dose rates from the installed system were very low. Subsequently with the ports open, there was no detectable radiation using standard ion chamber dose rate instruments. The system was installed by the test vessel (see Figure 5) and was calibrated by Tracerco technicians. The system performs with a resolution of approximately 10 cm for the sodium level.

Ten GM tubes are contained inside the long stainless tube. This detector array receives the radiation beam from the cesium sealed-sources which is linearly proportional to the sodium level.

Decomposition of CO₂ into O₂, CO, O+, etc. due to gamma irradiation has been considered as a possible source of the chemical reactions observed during the experiments described in this report. During the experiments, the shutters to the sources remained open in order to measure the sodium level throughout an experiment. However, these sources release extremely low levels of radiation. They are designed to emit just enough radiation to be detected above background, but radiation monitors are not required to be worn while working around them since the dose rate is well below all DOE and Argonne thresholds. A simple bounding analysis (included in Section 3.1) clearly showed that negligible decomposition of CO₂ will occur during the course of an experiment since the radiation levels are so small.

Figure 5: Several views of the sodium gamma-level meter. (Left) Red support frame without cesium sealed-source containers installed. (Middle) Four cesium sealed-sources are installed inside the yellow containers. (Right) Ten GM tubes are contained inside the long stainless tube. This detector array receives the radiation beam from the cesium sealed-sources which is linearly proportional to the sodium level.

2.4 Automatic Safety Shutdown System

An automatic safety shutdown system was custom built at Argonne to ensure that the system returns to a safe state in the event of a low-probability system failure or sodium fire. This system is designed to drain sodium from the test vessel back into the dump tank and put the heaters and valves in a safe state. It will turn off the three trace heater cabinets, turn off the CO₂ heater, and open or close the appropriate valves to allow the sodium to drain down to the

dump tank. If the smoke detector alarms or the manual dump button is pressed it will also start the exhaust fan in the enclosure and start the scrubber. All systems will remain in the off condition and the scrubber will remain on until operator intervention. The event triggers and resulting actions the SNAKE automatic safety system takes are listed in Table 1.

Table 1: Description of SNAKE Automatic Safety System Event/Action Sequences

SNAKE: Automatic Safety Systems	
Event	Action taken
Manual control	All valves
	Trace heaters (3 racks)
	CO ₂ pre-heater
	Scrubber
Labview control	All valves
	Trace heaters (3 racks)
	CO ₂ pre-heater
Smoke alarm	Opens PV-08
	Closes/opens other valves (NC/NO)
	Shutdown trace heaters (3 racks)
	Shutdown CO ₂ pre-heater
	Turns on scrubber
480V power loss/ 120 V power loss to heaters	Opens PV-08
	Closes/opens other valves (NC/NO)
	Shutdown trace heaters (3 racks)
	Shutdown CO ₂ pre-heater
120 V power loss to solenoids	Opens PV-08
	Closes/opens other valves (NC/NO)
	Shutdown trace heaters (3 racks)
	Shutdown CO ₂ pre-heater
Labview program crashes (watchdog timer times out)	Opens PV-08
	Closes/opens other valves (NC/NO)
	Shutdown trace heaters (3 racks)
	Shutdown CO ₂ pre-heater
Manual emergency button	Opens PV-08
	Closes/opens other valves (NC/NO)
	Shutdown trace heaters (3 racks)
	Shutdown CO ₂ pre-heater

The system has two tiers of operation. The first one monitors the trace heater power (120V/480V), power to the valve control chassis, and LabView. If any of these indicates a power loss the system will shut off the trace heaters, the valve power, and the CO₂ heater allowing the sodium to drain into the dump tank. The second tier also monitors the smoke alarm and manual dump button. If either of these trip, the exhaust fan and the scrubber also start. The wiring for the SNAKE automatic safety system is detailed in Figure 6 and a picture of the front panel is in Figure 7.

Figure 6: Schematic of the SNAKE Automatic Safety Shutdown System.

Figure 7: Picture of the front panel of the SNAKE Automatic Safety Shutdown System.

2.5 Data acquisition system improvements

Over the course of Fiscal Year 2013 many improvements were made to the SNAKE data acquisition system (DAS). The major improvements are listed below:

- Addition of an unattended alert system that emails and sends a text message to the operators if an abnormal event such as a temperature over-limit or sodium fire occurs.

- Addition of a watchdog timer control routine to ensure that the SNAKE Safety Shutdown System operates if Labview freezes or the DAS computer fails.
- Addition of temperature limits settings and control.
- Improvements to the data recording and display routines and control.
- Addition of exhaust flow meter, ChemGard carbon monoxide concentration, and gamma sodium-level routines and displays.
- Integrated valve and heater controls.

3 Fundamental Sodium-CO₂ interaction experiments

A continuous set of sodium-CO₂ experiments were run over the course of approximately three hours with the nominal sodium temperature set to 150 °C and sodium height of 45 cm above the injection nozzle. The inlet CO₂ pressure was gradually increased from 3 MPa to 11 MPa over these three hours in order to study the impact of pressure and flow rate on the sodium-CO₂ interactions. For the first time in SNAKE, chemical reactions between the sodium and CO₂ were observed. Sodium temperature increased from 150 °C to nearly 270 °C over the course of the experiments. Production of carbon monoxide was clearly detected, with up to 0.3 percent of the injected CO₂ converted to CO over the course of the experiments. A larger amount of CO₂ was converted into solids, approximately 37 percent. This reaction percentage is considered to be high given the sodium temperature of this experiment. It is possible that one or more mass flow meters are out of calibration which would explain the surprisingly high conversion ratio. The mass flow meters will be sent to the factory for recalibration prior to additional experiments. Significant quantities of solid materials have been generated inside the test vessel and were observed using an inspection video camera. These particulates will be removed, sampled, mounted, and sent to analytical chemistry in early Fiscal Year 2014. Substantial work will be required to remove these particulates from the test vessel and restart sodium-CO₂ interaction experiments using clean sodium.

This section first gives a brief overview of the entire three-hour string of experiments to describe the overall experimental run and the corresponding connectivity of the individual experiments. The individual experiments are then discussed in detail and compared.

3.1 Description of full experiment run

For all data plotted in this section, the initiation of CO₂ injection begins at time zero and stops at time 10,650 seconds. The measured flowrates of the CO₂, argon cover gas, and exhaust are shown in Figure 8. Argon cover gas flowrate was raised from 533 g/hr to 748 g/hr at time 1,800 seconds. The flowrate of CO₂ was surprisingly irregular given that the CO₂ pressure did not exhibit similar fluctuations (see Figure 9). The CO₂ flowrate exhibits a number of peaks throughout the testing, eventually reaching the maximum flowrate observable with the Brooks QMBM3 Coriolis flowmeter of 630 g/hr. The average flow rate for the first 6,000 seconds

was 300 g/hr, while the average flowrate from 6,000 seconds until the experiment end was 120 g/hr. The exhaust flow rate is adjusted for the presence of a mixture of gases by using the gas conversion ratios and the measured concentrations at the mass spectrometer.

Figure 8: Gas flowrates for the CO₂ injection nozzle inlet, argon cover gas in, and the exhaust out.

Figure 9: Pressure history of the CO₂ reservoir which connects to the CO₂ injection nozzle and pressure history of the test vessel itself.

Temperatures of the sodium column are shown in Figure 10 as observed by the thermocouple rake that has ten thermocouples spaced in 5 inch (12.7 cm) intervals starting 1.7 inches (4 cm) above the nozzle. The thermocouple rake is also radially offset from the centralized nozzle by 1.37 inches (3.5 cm) as shown in Figure 11.

Figure 10: Temperature history of the sodium column as measured by a thermocouple rake at several elevations above the injection nozzle.

The temperature at the top of the sodium column, 55 cm begins increasing 1,200 seconds after injection eventually climbing to 270 °C at 6,300 seconds.

Figure 11: Drawing of the test vessel with a cutout showing the CO₂ injection nozzle.

The thermocouple immediately above, at 67 cm, also climbs in temperature up to 250 °C at 8,200 seconds. This temperature increase is likely due to the sodium height rising somewhat

due to the CO₂ flow pushing sodium up. The sodium height increase seen in Figure 12 from ~45 cm to ~60 cm over the course of the experiment could also be due to particulate generation from the reaction increasing the overall volume of solids in the vessel. Particulate generation has been shown to expand sodium volume by five times in previous experiments (Sherman et al., 2002). Note that the measured sodium level is not consistent with the post-experiment solids height discussed later (Section 3.5). The ending solids height was approximately 77 cm. This discrepancy is being investigated but could be due to the density changes of the solids altering the relationship between measured level and mass.

Figure 12: Sodium level as measured by the gamma level meter.

In Figure 10, the sodium column temperature increases at 42 cm above the nozzle (to 260 °C at 9,400 seconds) and at 17 cm above the nozzle (to 233 °C at 9,400 seconds). However, just above the nozzle, at 4 cm height, the sodium temperature does not begin increasing until 5,300 seconds and has a maximum temperature of 200 °C. This data shows that the chemical reaction intensifies as the CO₂ rises through the sodium column since the first temperature increase observed occurs 55 cm above the nozzle while the sodium temperature increase is delayed at lower elevations. Also, the maximum temperature observed also decreased with elevation.

As mentioned previously, the gas flowrates plotted in Figure 8 have significant variability and a mass balance would be challenging to obtain since each inlet stream has a different time lag before reaching the exhaust flowmeter. Instead, the cumulative mass flows for each are shown in Figure 13. These cumulative flows represent the integral flow through each mass flowmeter. This plot is useful in clearly identifying various trends and, importantly, accounts for the time it takes for the gases to flow from the inlets to the outlet. A total of 650 g of CO₂ were injected, while 2,400 g of argon was used as the cover gas.

Figure 13: Cumulative gas mass flows at the CO₂ injection nozzle inlet, argon cover gas in, and the exhaust outlet. Also plotted is the sum of the two inlet mass flows (CO₂ plus argon).

Also shown in this figure is the total inlet mass flow of the CO₂ plus the argon cover gas. If no reaction took place the exhaust mass flow should exactly match the total inlet mass flow. This mass deficit or the mass flow in minus the mass flow out (via the exhaust) is plotted in Figure 14. A total mass deficit of approximately 240 g is observed over the experiment duration which should correspond to 240 g of CO₂ reacting with the sodium, or approximately 37% of the injected CO₂.

Figure 14: Cumulative mass deficit of CO₂ calculated by subtracting the cumulative exhaust mass flow from the sum of the cumulative inlet mass flows at any given time.

Based on the mass deficit of the CO₂, it is possible to estimate the total masses of the solids that would be produced should the reaction be stoichiometric. Since the sodium and CO₂ temperatures are well below the high temperature reaction threshold of ~450 °C, the following reaction pathway is hypothesized (Gicquel et al., 2010):

Thus, for every mole of CO₂ reacted, one quarter mole of sodium carbonate, Na₂CO₃, will be produced. The oxalate is expected to react immediately with sodium at all temperatures, so for every mole of CO₂ reacted, one quarter mole of elemental carbon and three-quarters of a mole of sodium oxide, Na₂O, will be produced. In order to calculate the masses produced using the above formulations, the molar masses of each molecule must be used appropriately. The formulas used to obtain the quantities of each product are shown below, with m_{XXXX} being the mass of molecule XXXX.

$$m_{Na_2CO_3} = \frac{1}{4} m_{CO_2} \left(\frac{1}{\frac{44g}{mol CO_2}} \right) \left(\frac{104g}{mol Na_2CO_3} \right) \tag{2}$$

$$m_C = \frac{1}{4} m_{CO_2} \left(\frac{1}{\frac{44g}{mol CO_2}} \right) \left(\frac{12g}{mol Na_2CO_3} \right) \tag{3}$$

$$m_{Na_2O} = \frac{3}{4} m_{CO_2} \left(\frac{1}{\frac{44g}{mol CO_2}} \right) \left(\frac{62g}{mol Na_2CO_3} \right) \tag{4}$$

Estimates of the masses of each reaction product produced during the course of the experiment are plotted in Figure 15. Approximately 406 g of total solids were produced. The solids product composition includes approximately 250 g of sodium oxide, 140 g of sodium carbonate, and 16 g of elemental carbon.

Note that it is possible that the oxylate did not substantially react during the relatively short duration of these experiments. The reaction between the oxylate and residual sodium could have slowly continued over several days after the experiment. This is because the sodium was kept in the test vessel at 150 °C and no gas was purged through the system for one week after the conclusion of the experiments described above. All gas valves were kept closed to prevent air ingress. The CO that could be produced by the oxylate reaction with sodium would remain in the test vessel and could further react with sodium as: $2Na + CO \rightarrow Na_2 + C$.

Figure 15: Estimate of solids produced throughout experiments.

Unlike in the experiment run in Fiscal Year 2012 (Gerardi et al., 2012), carbon monoxide was clearly detected in the exhaust gasses during the Fiscal Year 2013 tests described in this report. This was an additional indicator that a reaction between the sodium and CO₂ occurred. Two instruments were used to simultaneously measure the concentration of carbon monoxide in the effluent gas. The most sensitive instrument, the thermal-acoustic gas analyzer was newly installed in Fiscal Year 2013 and detects carbon monoxide from 1 ppm up to 1,000 ppm (gas fraction of 1×10^{-3}). The mass spectrometer is linear above 100 ppm. The evolution of the gas fractions over the course of the experiments is shown in Figure 16. There is some disagreement between the mass spectrometer and gas analyzer, especially considering that the gas analyzer pegs to its maximum of 1,000 ppm CO for a significant portion of the experiment, while the mass spectrometer reads below 1,000 ppm CO for much of the experiment run. It is possible that the instruments need further calibration which will be examined further in Fiscal Year 2014. An effort will be made to calibrate both instruments using gasses that approximate the makeup of this experiment. A peak CO₂ concentration of 10% is reached by 2,000 seconds into the experiment and then maintained between 1 and 10% for the majority of the rest of the experiment.

Figure 16: Gas fractions of CO₂ and CO as measured by the mass spectrometer at the exhaust line. The CO gas analyzer simultaneously measured CO, which was also plotted.

The carbon monoxide mass flow detected by the mass spectrometer can be compared with the theoretical stoichiometric reaction CO production. The CO mass measured can be calculated by multiplying the gas fraction at the mass spectrometer by the exhaust flow rate.

The theoretical CO mass that could be produced if all of the CO₂ that reacted, estimated from the mass flow deficit, can be calculated using Equation 5. In this equation, it is assumed that no oxylate reacts with sodium in order to put a lower bound on the CO production.

$$m_{CO, nooxylate} = \frac{1}{4} m_{CO_2, deficit} \left(\frac{1}{\frac{44g}{mol CO_2}} \right) \left(\frac{28g}{mol CO} \right) \quad (5)$$

The theoretical CO mass that could be produced if all of the CO₂ that was injected into the system completely reacted can be calculated using Equation 6. In this equation, it is assumed that 100% of the oxylate reacts with sodium in order to put an upper bound on the CO production.

$$m_{CO, oxylate} = \frac{1}{2} m_{CO_2, injected} \left(\frac{1}{\frac{44g}{mol CO_2}} \right) \left(\frac{28g}{mol CO} \right) \quad (6)$$

The measured and estimated CO production masses are shown in Figure 17. The measured CO production is at least an order of magnitude lower than either of the estimates, which suggests that less CO was produced than predicted by the stoichiometry or the CO further reacted with Na to form Na₂O and C.

Figure 17: Estimated total mass CO produced.

As mentioned above, decomposition of CO₂ into O₂, CO, O⁺, etc. due to gamma irradiation from the gamma-level meter has been considered as a possible source of the chemical reactions observed during the experiments described in this report. During the experiments, the shutters to the sources remained open in order to measure the sodium level throughout an experiment. However, these sources release extremely low levels of radiation. They are designed to emit just enough radiation to be detected above background, but radiation monitors are not required to be worn while working around them since the dose rate is well below all DOE and Argonne thresholds.

Carbon dioxide is a molecule and will decompose due to irradiation. The CO₂ might largely decompose to according to the reaction,

The design of a SFR utilizing a S-CO₂ Brayton cycle power converter must assure that the CO₂ is not exposed to any significant radiation dose. Thus, it is not possible to install sodium-to-CO₂ heat exchangers inside of a pool-type reactor vessel because of the activity of the primary coolant as well as radiation streaming from the core. An intermediate sodium circuit is required and shielding must be provided to limit activation of the intermediate sodium. Decomposition can form long chain molecules having the net composition of CO that can gum up rotating turbomachinery as well as create species having a greater potential to corrode the S-CO₂ cycle turbomachinery and structural materials.

The total activity of the four Tracerco gamma level-meter sources is 0.8 mCi. One Curie equals 3.7×10^{10} decay/sec. The total experiment run lasted 10,650 seconds. In order to bound the decomposition, it is assumed that all of the emitted radiation is absorbed by the

CO₂ and each absorption results in one-half molecule of O₂ and one molecule of CO. This is an extreme overestimate for a number of reasons including the facts that most of the emitted radiation does not reach the CO₂ due to intervening shielding, and the gamma capture cross section of CO₂ is not high. However, these assumptions are very useful in providing a high-end estimate for decomposition. The molar masses of CO and O₂ are 28 g/mol and 32 g/mol, respectively. Based on these assumptions, the following two equations can be used to estimate CO and O₂ production:

$$0.8 \times 10^{-3} Ci \left(\frac{3.7 \times 10^{10} \text{ decay}}{\frac{\text{sec}}{Ci}} \right) (10,650 \text{ sec}) \left(\frac{1 \text{ molecule of CO produced}}{\text{decay}} \right) \left(\frac{1 \text{ mole}}{6.022 \times 10^{23} \text{ atoms}} \right) \left(\frac{28 \text{ g}}{\text{mol CO}} \right) = 1.4657 \times 10^{-11} \text{ g of CO}$$

$$0.8 \times 10^{-3} Ci \left(\frac{3.7 \times 10^{10} \text{ decay}}{\frac{\text{sec}}{Ci}} \right) (10,650 \text{ sec}) \left(\frac{0.5 \text{ molecule of O}_2 \text{ produced}}{\text{decay}} \right) \left(\frac{1 \text{ mole}}{6.022 \times 10^{23} \text{ atoms}} \right) \left(\frac{32 \text{ g}}{\text{mol O}_2} \right) = 8.375 \times 10^{-12} \text{ g of O}_2$$

Based on this analysis, it was expected that very little CO or O₂ would be produced due to decomposition of CO₂ via gamma irradiation. A future experiment run will include ensuring that the shutters are closed on the gamma-level meter to prevent CO₂ irradiation which is expected to confirm that negligible decomposition occurred as a result of radiation.

3.2 Description of individual sub-experiments

The experiment cluster described above was actually comprised of six individual experiments where pressure was gradually increased from 3 MPa through the critical point of CO₂ (7.38 MPa at 31.1 °C) up to 11.3 MPa. The CO₂ pressure was leveled off at the six intermediate points for at least 250 seconds. The main experiment parameters from each test are listed in Table 2.

Table 2: Summary of main experiment parameters

Experiment #	Nominal Pressure (MPa)	Minimum Na Temp (°C)	Maximum Na Temp (°C)	Maximum Nozzle Temperature (°C)	Nominal CO ₂ flowrate (g/hr)	Total CO ₂ Mass Injected (g)
1	2.8	71.0	145.7	143.1	82.9	9.2
2	6.5	70.9	183.3	143.3	199.8	22.3
3	7.4	71.4	210.7	143.4	440.8	49.2
4	9.1	72.7	248.8	146.0	383.6	42.8
5	10.1	72.3	253.3	145.1	350.0	39.1
6	11.3	72.9	259.3	145.4	363.2	40.5

Two experiments, #2 and #5 with CO₂ injection pressures of 6.5 and 10.1 MPa, respectively, are chosen to be examined in detail here.

3.3 Experiment #2: CO₂ into sodium at 6.5 MPa

The sodium temperature at the beginning of this 6.5 MPa experiment was not axially uniform at time zero since previous experiments (see discussion above) had already resulted in some chemical reaction that heated portions of the sodium. The temperature rose at some heights of the sodium during this experiment but not significantly as shown in Figure 18(a). The sodium temperature at 55 cm above the nozzle began and ended at 181 °C. However, below and above this level, the sodium temperature began at around 145 °C. Between 4 and 17 cm above the nozzle, this temperature did not change. However, the temperature rose to 153 °C and 158 °C, at 67 cm and 42 cm above the nozzle respectively. The nozzle temperature remained at 143 °C throughout this test as shown in Figure 18(b).

Figure 18: Temperatures of the sodium (a) and CO₂ injection nozzle (b).

The flowrates of the argon cover gas, injected CO₂, and exhaust flow are shown in Figure 19(a), while the cumulative mass flow measured by each flow meter are shown in Figure 19(b). The exhaust flow rate is adjusted for the presence of a mixture of gases by using the gas conversion ratios and the measured concentrations at the mass spectrometer. The sum of the argon cover gas and inlet CO₂ flows are also plotted on the cumulative mass flow graph. The difference between this sum and the total exhaust flow is plotted in Figure 19(c). There was approximately 9 g of CO₂ estimated to have reacted based on the mass deficit. Utilizing the same analysis technique as described in Section 3.1, the theoretical mass produced of each possible solids product is shown in Figure 19(d).

Figure 19: Mass flow data from 6.5 MPa test. (a) Flowrates for inlet and exhaust gases, (b) cumulative mass flows, (c) net mass deficit at exhaust, and (d) estimated solids production based on mass deficit.

The CO₂ pressure, test vessel pressure, and level were constant throughout this experiment at 6.50 MPa, 159 kPa, and 46.3 cm, respectively. These parameters are plotted in Figure 20.

Figure 20: CO₂ and test vessel pressure (left) and sodium level (right) for the 6.5 MPa test.

3.4 Experiment #6: CO₂ into sodium at 11.3 MPa

The sodium temperature at the beginning of this 11.3 MPa experiment was not axially uniform at time zero since previous experiments (see discussion above) had already resulted in some chemical reaction that had heated portions of the sodium. The temperature rose in some parts of the sodium during this experiment but not significantly as shown in Figure 21(a). The sodium temperature at 55 cm above the nozzle started at 255 °C and ended at 259 °C. Above and below this point, the temperature ranged from 255 °C to 145 °C. The greatest

change during the course of this 250 second run was observed at the 67 cm point which suggests that the CO₂ bursting from the sodium pool was reacting further with the sodium vapor in the air. The nozzle temperature remained at 146 °C throughout this test as shown in Figure 21(b).

Figure 21: Temperatures of the sodium (a) and CO₂ injection nozzle (b).

The flowrates of the argon cover gas, injected CO₂ and exhaust flow are shown in Figure 22(a), while the cumulative mass flow measured by each flowmeter is shown in Figure 22 (b). The exhaust flow rate is adjusted for the presence of a mixture of gases by using the gas conversion ratios and the measured concentrations at the mass spectrometer. The sum of the argon cover gas and inlet CO₂ flows are also plotted on the cumulative mass flow graph. The difference between this sum and the total exhaust flow is plotted in Figure 22 (c). There was approximately 7 g of CO₂ estimated to have reacted based on the mass deficit. Utilizing the same analysis technique as described in Section 3.1, the theoretical mass produced of each possible solids product is shown in Figure 22(d).

Figure 22: Mass flow data from 11.3 MPa test. (a) Flowrates for inlet and exhaust gases, (b) cumulative mass flows, (c) net mass deficit at exhaust, and (d) estimated solids production based on mass deficit.

The CO₂ pressure, test vessel pressure, and level were constant throughout this experiment at 11.3 MPa, 134 kPa, and 53.7 cm, respectively. These parameters are plotted in Figure 23.

Figure 23: CO₂ and test vessel pressure (left) and sodium level (right) of the 11.3 MPa test.

3.5 Discussion of Sodium-CO₂ Experiments

The six experiments discussed above can be summarized by examining key data like maximum sodium temperature, gas flowrates, solids production estimates, etc. This section parses the key data and describes how the controlled change in CO₂ injection pressure impacted these key pieces of data.

The maximum and minimum sodium temperatures are plotted in Figure 24 for each nominal CO₂ injection pressure. The minimum sodium temperature, always at the bottom of the test vessel, stays nearly constant at 145 °C, while the maximum sodium temperature gradually increases with pressure. Of course, since the experiments took place in succession, the sodium was exposed to CO₂ flow for an increasingly longer total duration as pressure was increased. Thus, the temperature increase could have been solely due to time that the sodium was exposed to CO₂ rather than a function of pressure.

Figure 24: Sodium temperatures vs. CO₂ injection pressure.

Flowrate also increases with pressure, as seen in Figure 25, since the flow is only “controlled” by the principle of choked flow. Along with the measured flowrates, the predicted flowrates are plotted that use the measured CO₂ inlet temperature of 145 °C to calculate the CO₂ properties at a given pressure. Choked flow is assumed and the standard critical mass flux for a perfect gas from Moody (1990) is used:

$$\frac{P_c}{P_o} = \left(\frac{2}{k+1} \right)^{\frac{k}{k-1}} \quad (7)$$

$$\frac{G_c}{\sqrt{k P_o \rho_o}} = \left(\frac{2}{k+1} \right)^{\frac{k+1}{2(k-1)}} \quad (8)$$

where the gas constant, k , and the density of the fluid, ρ_o , are calculated using the CO₂ properties and the Span and Wagner (1996) equation of state for CO₂. The critical pressure and critical mass flux are represented by P_c and G_c , respectively. The nozzle diameter is assumed to be 64.0 μm. Thermal expansion in the nozzle is also taken into account by using the thermal expansion coefficient of 316 stainless steel of 16 m/m-K, and a temperature

increase from room temperature of 120 K. The total expansion of the nozzle hole is estimated to be 0.123 μm . The predicted mass flow rate and measured mass flow rates track remarkably well in Figure 25. There is some variability around the critical point of 7 MPa which could be real, due to dynamic flow instabilities between the gas booster pump and the micro nozzle, or large errors in the flow measurement. Next fiscal year, a large range of pressures will be tested in air to help isolate the phenomena responsible for the observed behavior.

Figure 25: Measured and predicted CO₂ flow rates through the micro-nozzle as a function of pressure.

As CO₂ reacts in the column of sodium, carbon monoxide and solid reaction products are produced. Since these solids are not passed through the exhaust gas line, there is a net loss between the outlet and inlet flow rates. This mass deficit was discussed above and the total mass deficit over each 250 second experiment is plotted versus CO₂ injection pressure in Figure 26. The mass deficit tended to go up with pressure, with a spike occurring from 7 to 9 MPa which corresponds to the spike in flowrate shown in Figure 25. It will be useful to replicate this data and go to higher pressures in the future.

Figure 26: Gas mass deficit, which is the total mass of gas input to the system minus the exhaust gas mass, vs. CO₂ injection pressure.

As the pressure and flowrate increased through the nozzle, the nominal sodium column height rose as shown in Figure 27.

The gas holdup model by Brockman et al. (1989) and Renjun et al. (1988) was used to estimate the maximum void fraction. This study focused on modeling the sparging of high temperature (1823 K) molten steel with nitrogen, which is more akin to the current work than previous models that were developed predominately on the basis of water voiding experiments. Brockman et al. reviewed several models, and on this basis developed their own correlation to match the data they obtained with molten steel and oleic acid. Their correlation is based on the Morton Number:

$$M = \frac{g\mu_l^4(\rho_l - \rho_v)}{\rho_l^2 \sigma_l^3} \quad (9)$$

and is given as:

$$\alpha = cM^a j_g^{*b} \quad (10)$$

where the values of the coefficients a, b, and c come from data fit by Brockman et al. (1989) and Renjun et al. (1988) in Table 3. The dimensionless gas phase velocity is given as

$$j_g^* = \frac{U_\infty}{\left[\frac{\sigma_l g}{\rho_l} \right]^{1/4}} \quad (11)$$

while the bubble terminal rise velocity, U_∞ , can be approximated from (Todreas and Kazimi, 1990):

$$U_\infty = 1.53 \left(\frac{g \sigma (\rho_l - \rho_g)}{\rho_l^2} \right)^{1/4} \quad (12)$$

Table 3: Values of a, b, and c for Equation Eq. (10).

Source	a	b	c
Brockman et al. (1989)	0.0207	0.584	0.128
Renjun et al. (1988)	-0.0070	0.5897	0.1966

The measured sodium height does rise to approximately the level predicted by Brockmann et al. (1989), but not close to the height predicted by Renjun et al. (1988).

Figure 27: Measured and predicted sodium column heights. Uncertainty is approximately ± 10 cm.

The predicted total solids production at a given pressure is shown in Figure 28. There does not appear to be substantial correlation between pressure and solids production.

Figure 28: Estimated solids produced (left) vs. pressure and (right) vs. CO₂ injection flowrate.

Figure 29: Drawing of test vessel with estimated height of solid particulates generated by chemical reaction of sodium and CO₂ denoted by gray hash marks. Solid height was estimated to be 0.77 m above nozzle.

Significant quantities of solids were produced in the course of these experiments due to the chemical reaction between sodium and CO₂. Once the experiments were completed, draining was extremely difficult. Gravity drainage did not work and a few pounds of positive pressure on the test vessel were required to force sodium into the dump tank. Sodium was cycled between the dump tank and test vessel and back prior to allowing the apparatus to cool down

in order to ensure that there was no substantial blockage near the valves and small-diameter tubing. After the experiment cooled, borescope cameras were used to examine the interior of the test vessel while an argon gas purge was maintained. The results were surprising. Solids in the cold test vessel accumulated to at least 77 cm above the nozzle as depicted in Figure 29.

The observable solids at the top of the test vessel were composed of loose granular particulates (Figure 30 and Figure 31). These particulates easily broke up when a stainless lance was pushed into the top of the solids matrix (see Figure 30(left)). The strength of this solids matrix was not quantitatively measured but was observed to have the consistency of peanut butter. Solid sodium would be significantly more difficult to breach.

Figure 30: Images of solids matrix in test vessel produced via sodium-CO₂ chemical reaction. (left) Stainless lance that is penetrating several inches into the solids matrix. (right) close-up of hole that remains after lance is removed.

Figure 31: Images of solids matrix in test vessel produced via sodium-CO₂ chemical reaction. (left) shows the thermocouple rake (bottom left denoted by red arrow) is covered by the solids matrix and small spherical white particulates cling to the rake's surface. (right) Close-up of different zones of the solids matrix.

Based on the literature review of sodium – CO₂ chemical reactions, it is expected that the solids are composed up of a mixture of Na₂O, Na₂CO₃, C, and Na₂C₂O₄ (Gicquel et al, 2009-2013; Eoh et al., 2010; Cordfunke Ouweltjes, 1969). Since the literature is sparse, there could be other potential solid reaction products present. Samples are being prepared to be sent to

Argonne's Analytical Chemistry group to be analyzed with XRD and potentially additional diagnostics.

4 Updated near-term detailed test matrix and near-term improvements

The general test matrix for SNAKE described in Gerardi et al. (2013) included a near-term test matrix to guide experiments for Fiscal Year 2013 into Fiscal Year 2014. The experiments listed in the near-term test matrix were considered shakedown tests and were designed to obtain useful data; however, data collection was not their primary purpose and would not be considered 'failures' should little data arise from the first few tests. The experiments described in the current report were among those shakedown tests. Surprisingly, these experiments did yield very useful data. These experiments also will be used to update the near-term test matrix to obtain high-quality data for modeling purposes with the smallest number of experiments.

A summary of the first completed experiments and the target conditions of the next several experiments are listed in Table 4. All of these tests will be direct injection of CO₂ or CO/CO₂ mixtures into a static sodium pool. For the first few tests, the sodium will be maintained at the lowest temperature it can safely be maintained at without any risk of freezing, around 140-180 °C. Over the course of several experiments the sodium temperature will gradually be increased, but it is not anticipated to reach the maximum facility operating temperature (510 °C) in the initial test series. The sodium column height will be kept low initially, and gradually increased, up to a maximum of approximately 80 cm. This height can be increased after enough experiments verify that gas-holdup is not pushing sodium high enough up to spill into the cover gas exhaust line. Pressure will be gradually increased from just over the critical point up to 20 MPa. Low pressure (sub-critical) CO₂ tests will also be carried out to add to the simulation verification database. Most tests will last approximately 1-30 minutes, or until steady state is reached on all primary instruments.

One test will be carried out to fully shake down the new sodium level-detector (Test 8). This experiment will study gas-holdup where argon at varying pressures will be injected into the sodium column and the resulting height of the sodium will be measured. This experiment is useful to make the safety case for the maximum sodium height allowed, and also could be useful in providing novel data for gas hold-up in liquid metals.

A detailed CO monitor test will be run utilizing both the mass spectrometer and new dedicated thermal-acoustic CO gas monitor. Pure CO₂ gas and mixtures of argon, CO₂, and CO will be injected through the nozzle into an empty test vessel at various pressures. These will simulate the time it takes for the gas mixtures to travel from the nozzle to the exhaust system where the gas monitors are. These types of tests act as baselines to compare with the sodium experiments that may generate some CO via the sodium-CO₂ reaction.

The first test listed was carried out in September 2012 and summarized in Gerardi et al. (2012b). The next six tests were carried out in Fiscal Year 2013 and detailed in the current report. Most of the remaining near-term tests listed in Table 4 do correspond to tests recommended in the general test matrix detailed in Gerardi et al. (2013). It is expected that

slight deviations from what is listed in Table 4 will occur as the experimental reality on a given day and the results of previous tests are taken into account. Additional experiments discussed in Gerardi et al. (2012) that involve gas blowdown into air and water will be carried out in Fiscal Year 2014.

Table 4: Updated detailed near-term test matrix. First seven experiments (in light green) have been completed

CO ₂ /CO injection from micro-nozzle into open sodium pool							
Test #	Test Gas	Gas stagnation pressure, MPa	Gas stagnation temperature, °C	Sodium column temperature, °C	Sodium column height, cm	Injected gas mass, g	Test duration, min
1	CO ₂	9	170	180	38.1	247	30
2	CO ₂	2.8	143	141-145	46.8	7.8	4.2
3	CO ₂	6.1	146	141-184	48.4	48.5	4.2
4	CO ₂	7.6	144	142-216	53.6	61.4	4.2
5	CO ₂	8.6	146	143-242	55.0	87.9	4.2
6	CO ₂	10.1	145	145-253	54.9	39.1	4.2
7	CO ₂	11.3	145	145-259	53.7	40.5	4.2
8	Argon	Sodium level detector testing and gas hold-up experiment			10-90	-	-
9	CO ₂	8.6	145	145	40.0	-	5
10	CO ₂	11	250	250	20.0	-	5
11	CO ₂	11	250	250	60.0	-	5
12	CO ₂	11	250	250	20.0	-	5
13	CO ₂	11	250	250	80.0	-	5
14	CO ₂	11	330	330	20.0	-	5
15	CO ₂	20	330	330	20.0	-	5
16	CO ₂	20	330	330	60.0	-	5
17	CO ₂	20	330	330	80.0	-	5
18	CO	11	150	150	40.0	-	5
19	CO	11	150	150	80.0	-	5
20	50% CO/50% CO ₂	11	150	150	40.0	-	5
21	CO ₂	11	450	450	20.0	-	5

4.1 Near-term improvements and other non-experiment work

Near-term improvements that are anticipated for Fiscal Year 2014 and other non-experiment work are summarized here in list form.

- Add a flow cleaning loop which includes a sodium electromagnetic pump and sodium electromagnetic flow meter. These will be integrated with the current cold trap and filter, and possibly an additional filtering device such as a settling tank to handle large amounts of solids. A plugging meter may be included in order to assess the cleanliness of the sodium after cleaning. An additional option to be considered will be to forego the cleaning loop and simply begin each experiment with new clean sodium after the test vessel has been thoroughly cleaned in place or in a scrubber.
- Send all mass flowmeters to an external vendor for calibration. An 'as-received' calibration will be requested to determine how far the flow meters were out of calibration during this year's tests.
- Calibrate the mass spectrometer and gas analyzer using gases that approximate the makeup of the most recent experiments. While these instruments were calibrated this year, there was no operational history to suggest the ratios of argon, carbon dioxide, and carbon monoxide they would need to detect. Now that some operational experience has been gained, the instruments will be evaluated using known calibration gases with ratios that mimic the most recent experiments.

- Continue to make incremental improvements to the data acquisition system and control hardware.
- The hardware must be cleaned to remove all of the solid reaction products produced during the test discussed in this report. Unfortunately, this is not trivial. The experiment was designed to clean sodium of small amounts of solid products generated during short duration tests. A filter and cold trap are part of the system design. However, since no substantial reaction was expected to occur during this low temperature test, no time limit was placed on the test duration. Significant quantities of solids were generated and make it impossible to clean via the cold trap and filter. It is likely that parts of the system will need to be removed and emptied of the particulates, then cleaned. Ideally, the system will simply be cleaned using sodium. However, if it is too challenging to remove most of the particulates manually the system may need to be cleaned using an ethanol and water wash. Plans are being drawn up now to complete this cleaning as soon as possible.

5 Summary

A series of sodium-CO₂ interaction experiments were carried out in Fiscal Year 2013 in the SNAKE (S-CO₂ Na Kinetics Experiment) experiment. These tests successfully injected supercritical carbon dioxide into a pool of sodium through a 64 μ m diameter nozzle. A reaction between the CO₂ and sodium was detected. The extent of this reaction was unexpected since the initial sodium temperature was 145 °C, a temperature range where previous researchers have detected little or no chemical reaction between these species. The important difference between the SNAKE experiment and previous research is that the SNAKE geometry and conditions promote high-interfacial area and mixing between the CO₂ and sodium. These characteristics could be very important in promoting accelerated chemical reactions and will be studied further as the SNAKE test matrix is carried out.

Approximately 325 standard liters of CO₂ were injected into a 45 cm (15 inch) high column of sodium at a nominal temperature of 150 °C over the course of 3 hours. The inlet CO₂ pressure was gradually increased from 3 MPa to 11 MPa over these three hours in order to study the impact of pressure and flowrate on the sodium-CO₂ interactions. For the first time in SNAKE, chemical reactions between the sodium and CO₂ were observed. Sodium temperature increased from 150 °C to nearly 270 °C over the course of the experiments. Production of carbon monoxide was clearly detected, with up to 0.3 percent of the injected CO₂ converted to CO over the course of the experiments. A significant amount of CO₂ was converted into solids, approximately 37 percent. This reaction percentage is considered to be high given the sodium temperature of this experiment. It is possible that one or more mass flowmeters are out of calibration which would explain the surprisingly high conversion ratio. However, significant quantities of solid materials have been generated inside the test vessel and were observed using an inspection video camera. The reaction product particulates will be removed, sampled, mounted, and sent to Analytical Chemistry in early Fiscal Year 2014. Substantial work will be required to remove these particulates from the test vessel and restart sodium-CO₂ interaction experiments using clean sodium.

Gamma radiation from the gamma level meter was ruled out as a possible source of CO₂ decomposition and formation of decomposition products since the radioactive source activity of the level meter was extremely low.

No plugging of the micro-nozzle was detected during this experiment. Data from these experiments was reported and initial analyses of the data were completed and discussed. Additional analysis of this data and improvements of the data quality and facility control will take place in early Fiscal Year 2014.

Fiscal Year 2014 will involve cleaning the test vessel of the reaction products generated by the experiments detailed in this report, carrying out additional sodium-CO₂ and sodium-CO reaction experiments, making additional hardware improvements to improve data quality, and preparing a detailed report to be delivered in September 2014. Model development and validation are planned to begin in Fiscal Year 2014, with initial modeling results included in the September 2014 report.

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