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ASSESMENT OF PARTICLE CANDIDATES FOR FALLING PARTICLE RECEIVER APPLICATIONS THROUGH IRRADIANCE AND THERMAL CYCLING

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ABSTRACT

Falling particle receiver (FPR) systems are a rapidly developing technology for concentrating solar power applications. Solid particles are used as both the heat transfer fluid and system thermal energy storage media. Through the direct irradiation of the solid particles, flux and temperature limitations of tube-bundle receives can be overcome, leading to higher operating temperatures and energy conversion efficiencies. Candidate particles for FPR systems must be resistant to changes in optical properties during long term exposure to high temperatures and thermal cycling using highly concentrated solar irradiance. Five candidate particles, CARBOBEAD HSP 40/70, CARBOBEAD CP 40/100, including three novel particles, CARBOBEAD MAX HD 35, CARBOBEAD HD 350, and WanLi Diamond Black, were tested using simulated solar flux cycling and tube furnace thermal aging. Each particle candidate was exposed for 10 000 cycles (simulating the exposure of a 30-year lifetime) using a shutter to attenuate the solar simulator flux. Feedback from a pyrometer temperature measurement of the irradiated particle surface was used to control the maximum temperatures of 775 °C and 975 °C. Particle solar-weighted absorptivity and emissivity were measured at 2000 cycle intervals. Particle thermal degradation was also studied by heating particles to 800 °C, 900 °C, and 1000 °C for 300 hours in a tube furnace purged with bottled unpurified air. Here particle absorptivity and emissivity were measured at 100-hour intervals. Measurements taken after irradiance cycling and thermal aging were compared to measurements taken from as-received particles. WanLi Diamond Black particles had the highest initial value for solar weighted absorptance, 96%, but degraded up to 4% in irradiance cycling and 6% in thermal aging. CARBOBEAD HSP 40/70 particles currently in use in the prototype FPR at the National Solar Thermal Test Facility had an initial value of 95% solar absorptance with up to a 1% drop after irradiance cycling and 4% drop after 1000 °C thermal aging.

Keywords: Falling Particle Receiver, Particle Stability, Particle Absorptance, Particle Thermal Aging

1. INTRODUCTION

Falling particle receiver (FRP) systems use a solid particle heat transfer fluid to achieve higher temperatures (>700 °C) for use in concentrating solar power (CSP) to enable higher efficiency power cycles. Particles are used as the system's heat transfer fluid (HTF) and thermal energy storage (TES) [1]. Direct heating of particles with solar flux reduces peak flux limitations associated with tube bank receiver systems that utilize steam or molten salt as the HTF and TES. Particles descend through the system's cavity receiver, thermal storage, and heat exchanger before being elevated to the top of the system to continue the cycle. To be considered a viable candidate for FPR systems, particles must maintain their absorptivity after numerous exposures to concentrated solar flux and long durations in high temperature environments (>700 °C).

In collaboration with the University of Tulsa, five particle candidates have been tested for stability at high sustained temperatures and 10,000 cycles of concentrated solar irradiance (representing a 30-year system lifetime).

2. BACKGROUND

Ceramic proppants made of sintered bauxite are used by the oil and gas industry in hydraulic fracturing. These particles have many advantageous properties for FPR systems. The proppants are durable, highly absorptive, and are considered inert at temperatures exceeding 1200°C [2]. CARBO Ceramics' CARBOBEAD CP 40/100 (ACCUCAST) and CARBOBEAD HSP 40/70 particles have been tested in a prototype 1 MW_{th} FPR at the National Solar Thermal Test Facility (NSTTF). Over 150 hours of tests utilizing the HSP 40/70 particles were conducted with flux values as high as 150 W/cm^2 [4].

A previous study conducted by Siegel et al [5]. documented changes in particle bed solar weighted hemispherical absorptance for CARBOBEAD HSP and ACCUCAST particles. The particle median diameter was 697 μm for the HSP particles

and 300 µm for the ACCUCAST particles. Two tests were conducted using the HSP particles in which the particle samples were heated to 700 °C and 1000°C for 192 hours. The ACCUCAST particles were heated to 700 °C for 192 hours. Particle absorptivity was then measured to determine the stability of the particle candidates. The HSP particles decreased from an initial absorptance of 93% to 92% after being held at 700 °C for 192 hours and 84% after being held at 1000 °C for 192 hours. The ACCUCAST particles decreased from an initial absorptance of 91% to 90% after being held at 700 °C.

Decreases in solar absorptance for sintered bauxite particles such as CARBOBEAD HSP have been attributed to the reaction of the species FeAl-TiO_5, Fe_2TiO_5, Al_2TiO_5. The color change at the particle surface was seen to change from dark brown to light brown to orange during isothermal holds at 700 °C lasting for 500 hours. [6]

The absorptivity of a particle curtain can be greater than that of the packed bed of particles. Given a low solids volume fraction and high thickness, the curtain absorptance for HSP particles can be as high as 97%. A method of calculating individual particle absorptivity was outlined by Gonzalez-Portillo et al. Individual particle surface absorptivity of HSP particles was calculated to be 87% [7].

The five particle candidates shown in Figure 1 were considered for this study. The candidates consisted of the two previously tested particles (CARBOBEAD HSP and CP) and three novel particles including a CSP specific, highly absorptive, CARBO HD 350 particle, a high sphericity CARBOBEAD MAX HD 35 particle, and a highly absorptive, low sphericity, WanLi Diamond Black particle.

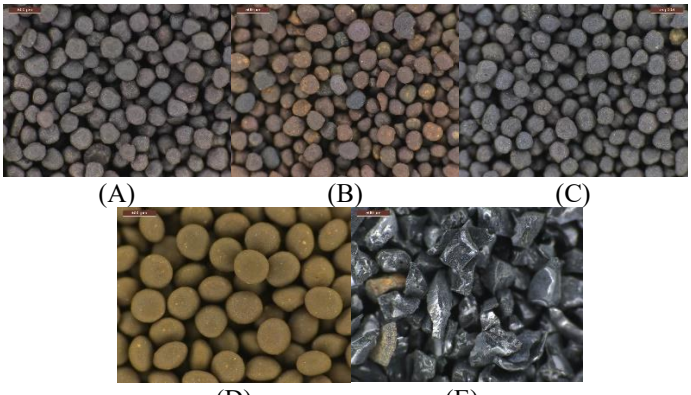


FIGURE 1: PARTICLE CANDIDATES HSP 40/70 (A), CP 40/100 (B), HD 350 (C), HD35 (D), AND WANLI DIAMONDBLACK (E) AT 2X MAGNIFICATION

3. METHOD

Particle samples are held a 1”x1” stainless steel crucible forming a ~1 mm thick packed bed, shown in Figure 2. Particle irradiance cycling, thermal aging, and measurement are conducted on the particles while they are contained in the crucible.

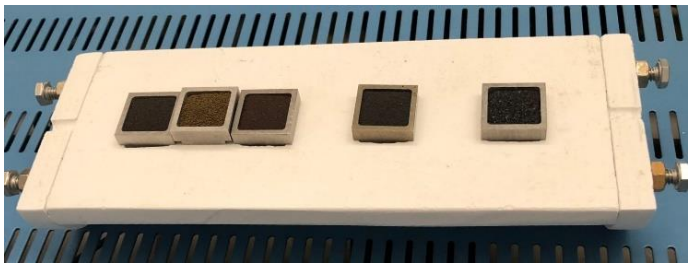


FIGURE 2: TUBE FURNACE SAMPLE TRAY CONTAINING THE 5 PARICLE CANDIDATES IN CRUCIBLES

Particle absorptivity and emissivity is measured using a Solar 410 reflectometer and an ET100 emissometer. The Solar 410 reflectometer has a spectral range between 300-2500 nm and measures total material reflectance at from incident light at an angle of 20° from normal. Solar absorptance is obtained by subtracting the measured solar hemispherical reflectance from one (reflected light between 0.2-7 µm was investigated). The ET100 emissometer measures material reflectance in six bands of the infrared spectrum at incident angles 20° and 60° from normal. The device measures the hemispherical total emissivity (HTE) at room temperature which it then uses to calculate the HTE of the material at 970 °C. Both devices, shown in Figure 3, are handheld with a circular measurement aperture of .5” in diameter. Particle reflectivity and emissivity values were measured above the particle bed allowing the particle to remain in the crucible. The resulting measurement is representative of a packed bed.

FIGURE 3: SOLAR 410 REFLECTOMETER AND ET 100 EMISSOMETER

Particle surface discoloration and visible damage is documented through microscopy. The Leica DMS 1000 microscope is used to image particle samples at 0.75x, 2x, and 4x magnification.

Irradiance cycling is conducted with a 7.2 kW_e solar simulator. The simulator is composed of 4 lamps with silver coated reflectors concentrating the light onto a 1”x1” spot [8]. Each lamp is capable of producing a peak flux of 175-350 W/cm^2. A top mounted pyrometer is used to measure the sample temperature. The measurement spectrum of the pyrometer is outside solar spectrum produced by the lamps

ensuring the accurate measurement of sample temperature unpolluted by the reflected solar spectrum. A high-speed shutter covers the sample after a given time interval or sample temperature before uncovering the sample to start the next cycle.

An OTS Programmable Tube Furnace is used for isothermal temperature holds over hundreds of hours. The tube furnace uses three thermocouples to maintain a setpoint temperature in 3 individually controlled zones. The endcaps of the quartz tube feature valves that allow for flow of a purge or cover gas over samples while they are heated.

4. PROCEDURE

The solar simulator was used to cycle the particle candidates through concentrated flux for a total of 10,000 cycles representing a 30-year operating lifetime. Three experiments were conducted with the solar simulator with varying cycle ending criteria. Two experiments featured a 775 °C and a 975 °C peak cycle temperature achieved with 35 W/cm^2, and one experiment featuring a constant exposure time to 70 W/cm^2 after the sample reached a prescribed temperature. The constant exposure experiment simulates the particle residence time inside the receiver in a 100 MW power plant. Particle samples were exposed to the flux until the particle temperature reached the proposed inlet temperature of the receiver, 615 °C, after which the particles were exposed for an additional 1.7 seconds. Particle absorptivity and emissivity was measured in 2,000 cycle intervals. Microscope images were taken during each measurement interval.

Table 1 summarizes the cycle ending criterion and measurement interval for the particles exposed to 10,000 cycles of solar flux.

TABLE 1: MEASUREMENT INTERVALS FOR EACH SOLAR SIMULATOR CYCLE END CRITERION

Cycle End Criterion	**615 °C + 1.7 seconds**	**Final Temp. 775 °C**	**Final Temp. 975 °C**
Measurement Intervals (Cycles)	0, 2k, 4k, 6k, 8k, 10k	0, 2k, 4k, 6k, 8k, 10k	0, 2k, 4k, 6k, 8k, 10k

During the irradiance cycling experiments it was found that the particles did not remain at high temperatures as particles would cool rapidly between each cycle. An isothermal temperature experiment was considered to understand particles stability at high sustained temperatures present in proposed FPR designs.

The OTS programmable quartz tube furnace was used to heat the particle samples to a constant temperature for up to 400 hours. The time at temperature does not reflect a plant 30-year lifetime, instead the goal was to gain insight into the maximum particle degradation caused by continuous exposure to high temperature. Three temperatures were considered, 800 °C, 900 °C, and 1000 °C. During each high temperature aging cycle, an air cover gas flowed over the particles at a flowrate of 200 cm^3/min. Table 2: summarizes the tests conducted with the tube furnace.

TABLE 2: MEASUREMENT INTERVAL FOR A GIVEN SUSTAINED TEMPERATURE.

Tube Furnace Temperature	**800 °C**	**900 °C**	**1000 °C**
Measurement Intervals (Hours)	100, 200, 300, 400	100, 200, 300	100, 200, 300

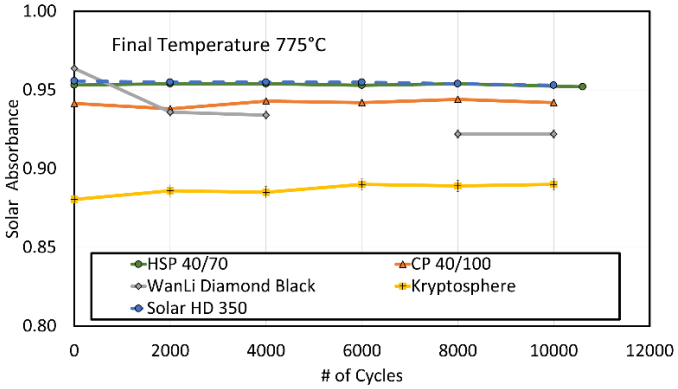
Particle absorptivity and emissivity was determined by averaging three individual measurements. The error between these measurements in each case was between 0%-.1%. Error between samples of the same candidate particle was determined by measuring five independent as-received particle samples three times for a total of fifteen measurements. The error between samples was more significant, .1%-.4%. This error is plotted but not visible in the subsequent figures as the error bars are approximately the same size as the datapoints.

In summary each of the particle candidates were tested in 6 scenarios all starting in an as-received state. Measurements were taken at regular intervals to determine the change in absorptivity and emissivity of each particle candidate.

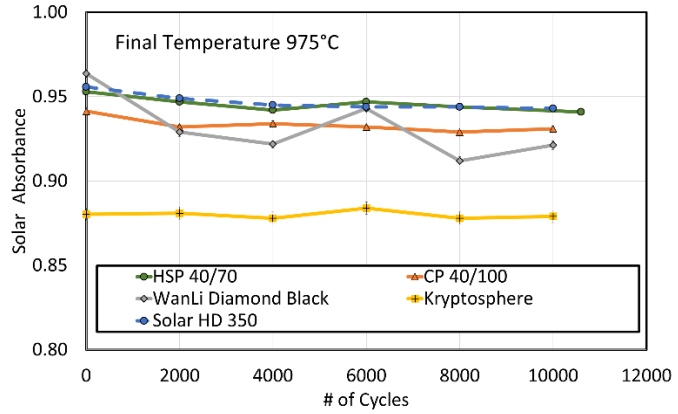
5. RESULTS AND DISCUSSION

Changes in particle absorptivity are primarily discussed for each scenario. Particle absorptivity is more influential in net energy absorption at the temperatures tested given the cavity receiver design.

Particles were cycled in the solar simulator using each of the cycle end criterion. Figure 4 shows the particles absorptivity as a function of cycle number for each particle candidate. Figure 4(A and B) illustrate the particle absorptivity as a function of irradiance cycling with a peak flux of 35 W/cm^2 and peak cycle temperatures of 775 °C and 975 °C respectively. Figure 4(C) illustrates the scenario with the most substantial drop in particle absorptivity as 70 W/cm^2 was applied inducing peak particle temperatures between 1100 °C and 1200 °C.



(A)



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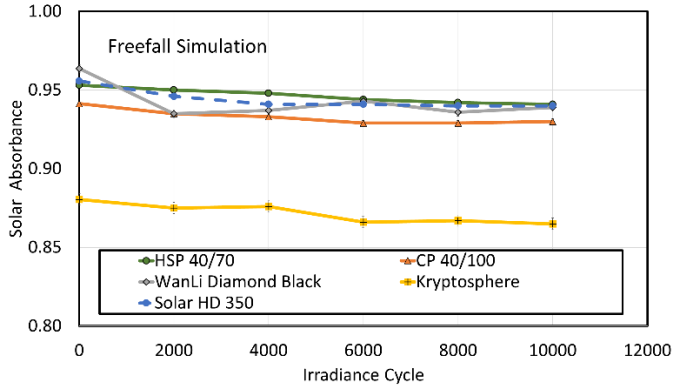

(C)

FIGURE 4: PARTICLE ABSORPTANCE VS IRRADIANCE CYCLES FOR 775°C (A), 975°C (B), AND SIMULATED FREEFALL (C) CASES. ERROR BARS ARE PRESENT BUT NOT VISIBLE

TABLE 3: AS RECEIVED PARTICLE ABSORPTIVITY WITH PERCENT CHANGE AFTER 10,000 CYCLES FOR EACH CYCLE ENDING CRITERION

Particle	As Received Particle Absorptance	615 °C + 1.7 seconds	Final Temp. 775 °C	Final Temp. 975 °C
HSP 40/70	95.3%	-1.3%	-0.12%	-1.3%
CP 40/100	94.1%	-1.2%	0.06%	-1.1%
MAX HD 35	88.0%	-1.7%	1.1%	-0.16%
HD 350	95.6%	-1.7%	-.29%	-1.3%
WanLi Diamond Black	96.4%	-2.6%	-4.3%	-4.4%

The change in particle absorptance after 10,000 irradiance cycles is listed in Table 3:. CP 40/100 featured the smallest change in absorptivity for the 975 °C scenario (-1%), MAX HD35 increased in absorptivity for the 775 °C scenario (+1), and HSP 40/70 featured the smallest change for the freefall simulated scenario (-1%).

The initial absorptivity of the HSP 40/70 and HD 350 particles was 95% and 96% respectively. The HSP 40/70 and HD 350 particles experienced a similar change in absorptivity for each scenario, 0.12% – 1.3% decrease, with final particle absorptivity values between 94% and 95%. Microscopy did not show signs of particle degradation.

At a peak temperature of 775 °C, CP 40/100 particle absorptivity increased by 0.06% after 10,000 cycles, while at a peak temperature of 975 °C, particle absorptivity decreased by 1.1% after the same cycle time. The orange/yellow color of the particles became more pronounced after the 975 °C peak temperature cycling.

FIGURE 5: WANLI DIAMOND BLACK PARTICLES AS-RECEIVED (LEFT) AND AFTER 10,000 CYCLE WITH A PEAK CYCLE TEMPERATURE OF 975 °C (RIGHT)

The WanLi Diamond Black particles experienced the largest decrease in absorptivity for each of the cycle ending criterion. The absorptivity decreased by 4.4% after 10,000 cycles with a peak particle temperature of 975 °C and 4.3% after 10,000 cycles with a peak particle temperature of 775 °C. Images taken of the cycled particles revealed visible degradation and cracks in the particle surface. Figure 5 shows the Diamond Black particles after 10,000 cycles with a peak cycle temperature of 975 °C. Visible fractures can be seen in the upper left corner of the image on the right. The top layer of the particle bed fused together in

each of the experiments which caused the expansion of the particles in the crucible. As measurements were taken, the top layer of particle material cracked and shifted resulting in the inconsistent measurement shown in Figure 4(B) at 6000 cycles. The absorptivity of the particles decreased by 2.6% following the freefall simulation despite featuring a higher peak cycle temperature of >1200 °C. At the higher temperatures measured during the freefall simulation the particles melted completely before fusing back together. It is assumed that the complete melting and re-solidifying of the particles had a restorative effect on the particle absorptivity.

HD 35 particles featured the lowest initial absorptivity of the five particle candidates. After cycling the particle with a peak temperature of 775 °C the particles increased in absorptivity by 1.1%. At a peak cycle temperature of 975 °C the particle absorptivity decreased by .16%. The particles did not show significant signs of deterioration.

Particle absorptivity was then measured following isothermal holds in a quartz tube furnace. Particle absorptivity as a function of hours at temperature is illustrated in Figure 6 with the change in particle absorptivity outlined in Table 2. Particles were heated to 800 °C for 400 hours compared to 300 hours for the 900 °C and 1000 °C scenarios due to a downward trend in particle absorptivity for the first 300 hours. To confirm the particle absorptivity would not decrease further, an additional 100-hour interval was considered. The absorptivity for all particle candidates remained constant between 300 and 400 hours at 800 °C. WanLi Diamond Black particles experienced the largest decrease in absorptivity for the 800 °C and 1000 °C scenarios. HD 35 had the largest decrease in absorptivity for the 900 °C scenario.

HSP 40/70 and HD 350 experienced similar changes in absorptivity for each temperature setpoint. For both particles the absorptance decreased to 94%, 93%, and 91% after being heated at 800 °C, 900 °C, and 1000 °C respectively.

CP 40/100 particle absorptivity was stable at 800 °C and 900 °C with a maximum decrease in particle absorptivity of 2%. After being heated to 1000 °C the particle absorptivity decreased by 6%.

WanLi particles decreased by 3.4%-5.9% of initial particle absorptivity during the isothermal holds. The top layer of particles fused after exposure to each of the temperature setpoints. While cracks in the particles surface were not seen after the sustained heating, particle discoloration was apparent in the microscope images. A green discoloration formed on the crucible holding the Diamond Black particles, and a yellow scaling was also seen on the particles surface.

HD 35 particle absorptivity decrease by 1%, 5.2%, and 3.8% from initial absorptivity in the 800 °C, 900 °C, and 1000 °C temperature setpoints respectively. Particle deterioration was not seen in microscopy.

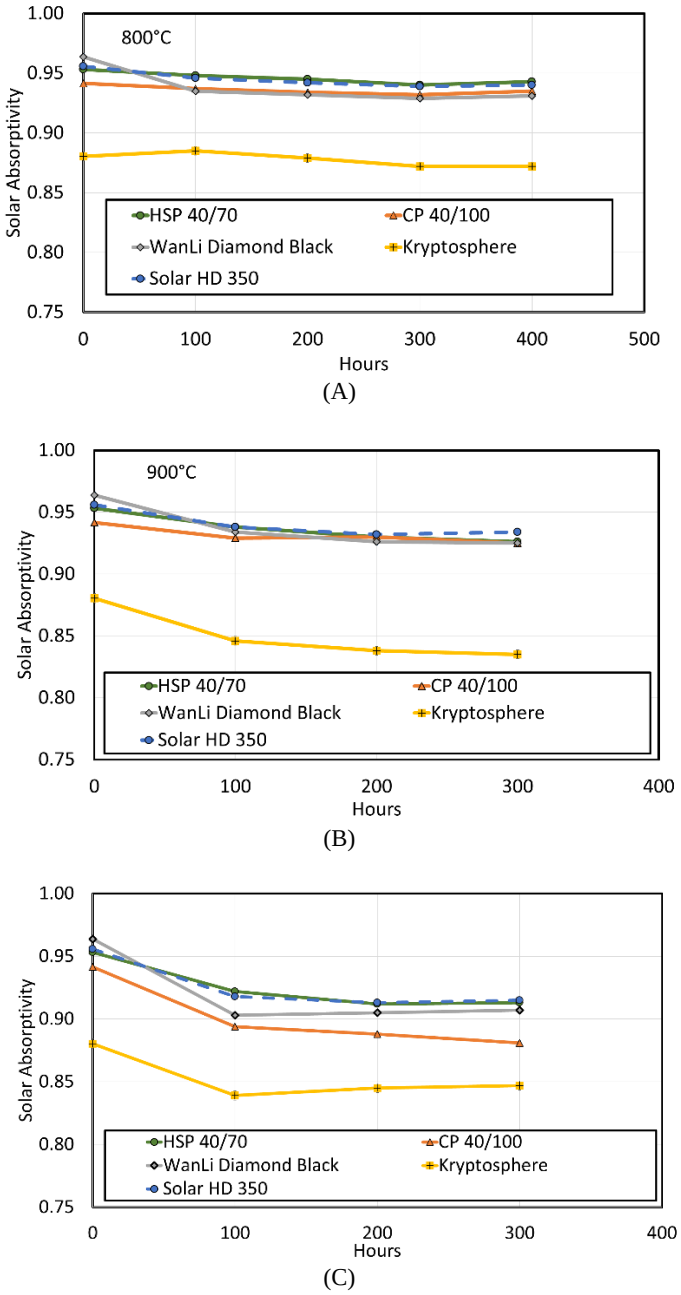

Figure 6: PARTICLE CANDIDATE ABSORPTIVITY VS HOURS AT 800 °C (A), 900 °C (B), AND 1000 °C (C). ERROR BARS ARE PRESENT BUT NOT VISIBLE.

TABLE 4: AS RECEIVED PARTICLE ABSORPTIVITY WITH PERCENT CHANGE AFTER HUNDREDS OF HOURS AT 800 °C, 900 °C, AND 1000 °C

Particle	As-Received Particle Absorptance	400 hours @ 800 °C	300 hours @ 900 °C	300 hours @ 1000 °C
HSP 40/70	95.3%	-1.1%	-2.9%	-4.2%
CP 40/100	94.1%	-0.7%	-1.8%	-6.4%
HD35	88.0%	-1.0%	-5.2%	-3.8%
HD 350	95.6%	-1.7%	-2.8%	-3.8%
WanLi Diamond Black	96.4%	-3.4%	-4.0%	-5.9%

Values for particle emissivity and absorptivity after each experiment are shown in Table 5 through
Table 8. Apart from the WanLi particles, particle emissivity remained relatively constant after flux cycling for each cycle end criterion with a maximum change of less than 1%. Diamond Black particle emissivity decreased by 17% following the freefall simulated flux cycling. Diamond Black particles also experienced the greatest decrease in emissivity for the sustained temperature experiments with a decrease of 32% for the 1000 °C isothermal hold. HSP 40/70, CP 40/100, MAX HD 35, and HD 350 particles emissivity was relatively stable following the 800 °C and 900 °C scenario with a maximum change in emissivity of 5%. Following the 1000 °C scenario all particle candidates had a decrease in emissivity of 7%-11%. MAX HD 35 emissivity decreased the least, 7%, while HD 350 decrease the most, 11%, out of the particle candidates.

TABLE 5: PARTICLE ABSORPTIVITY AFTER 10,000 FLUX CYCLES

Particle	As Received Particle Absorptance	615 °C + 1.7 seconds	Final Temp. 775 °C	Final Temp. 975 °C
HSP 40/70	95.3%	94.1%	95.2%	94.1%
CP 40/100	94.1%	94.0%	94.2%	93.1%
MAX HD35	88.0%	86.5%	89.0%	87.9%
HD 350	95.6%	93.0%	95.3%	94.3%
WanLi Diamond Black	96.4%	93.9%	92.2%	92.1%

TABLE 6: PARTICLE HTE AFTER 10,000 FLUX CYCLES

Particle	As Received Particle Absorptance	615 °C + 1.7 seconds	Final Temp. 775 °C	Final Temp. 975 °C
HSP 40/70	90.8%	88.4%	91.2%	90.5%
CP 40/100	86.8%	84.3%	88.5%	85.9%
MAX HD35	69.4%	66.3%	70.1%	69.5%
HD 350	90.6%	87.4%	90.5%	89.7%
WanLi Diamond Black	94.4%	77.5%	83.2%	80.2%

TABLE 7: PARTICLE ABSORPTIVITY AFTER SUSTAINED HIGH TEMPERATURE

Particle	As- Received Particle Absorptance	400 hours @ 800 °C	300 hours @ 900 °C	300 hours @ 1000 °C
HSP 40/70	95.3%	94.3%	92.6%	91.3%
CP 40/100	94.1%	93.5%	92.5%	88.1%
MAX HD35	88.0%	87.2%	83.5%	84.7%
HD 350	95.6%	94.0%	93.7%	91.5%
WanLi Diamond Black	96.4%	93.1%	92.5%	90.7%

TABLE 8: PARTICLE HTE AFTER SUSTAINED HIGH TEMPERATURE

Particle	As- Received Particle Absorptance	400 hours @ 800 °C	300 hours @ 900 °C	300 hours @ 1000 °C
HSP 40/70	90.8%	90.3%	87.3%	80.9%
CP 40/100	86.8%	87.5%	87.2%	77.6%
MAX HD35	69.4%	69.4%	65.9%	63.9%
HD 350	90.6%	89.5%	88.0%	79.9%
WanLi Diamond Black	94.4%	87.4%	79.9%	62.4%

5. CONCLUSION

Five particle candidates, CARBOBEAD HSP 40/70, CP 40/100, MAX HD 35, HD 350, and WinLi Diamond Black, were tested for stability at high temperatures through irradiance cycling and sustained elevated temperatures. HSP 40/70 and HD 350 particles had a similar decease in absorptivity for each of the tests conducted with a maximum decrease of 4% after being

heated to 1000 °C for 300 hours. HSP 40/70 and HD 350 particles experience the smallest change in absorptivity of the particle candidates. WanLi Diamond Black particles experience the greatest change in absorptivity for all the irradiance cycling experiments and the majority of the sustained heating experiments with a decrease in absorptivity up to 33% after being heated to 1000 °C for 300 hours. CP 40/100 and HD 35 particles had maximum decrease of 6% and 5% respectively. Particle emissivity following each test was listed.

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