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Liquid Si-rich Si-Zr alloys in contact with C and SiC: Wettability and Interaction phenomena

About

GOAL

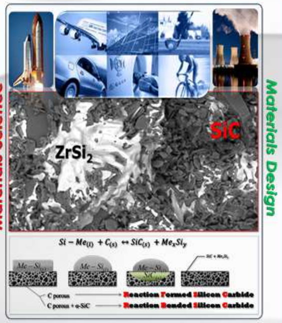
Collecting knowledge on the occurring **interfacial phenomena** between liquid Si-rich Si-Zr alloys in contact with C and SiC substrates.

WHY?

Targeted **wettability** and **reactivity studies** can easily provide useful indications for solving many technological problems affecting the **reactive infiltration** mechanisms, such as **pore closure/narrowing phenomena**.

HOW

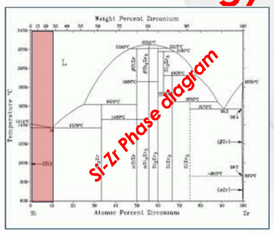
The contact heating sessile drop method (CH-SD) was applied for better understanding the interfacial phenomena occurring between 3 liquid **Si-rich Si-Zr** alloys in contact with **Glassy Carbon** (GC) and **SiC** substrates. Specifically, the **contact angles** behaviors as a function of time were obtained over the temperature range of **T = 1354-1500°C** under an **Ar atmosphere**.



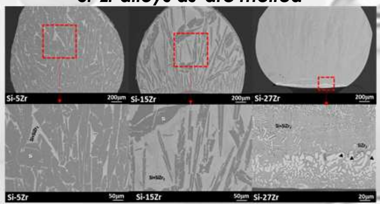
IMPACT

- **Computational models** are enabling in the design of **advanced refractory materials** such as SiC-based composites for highly demanding applications (thermal barriers, structural materials and chemically stable components for assembling re-entry space vehicles and fission/fusion nuclear, etc.).
- The fabrication of **tailored SiC/ZrSi$_2$-based composites** via cost-less **reactive infiltration** of **Si-rich Si-Zr alloys** into **C- and SiC-based preforms**, is currently one of the main goals of **materials science** and **design**.

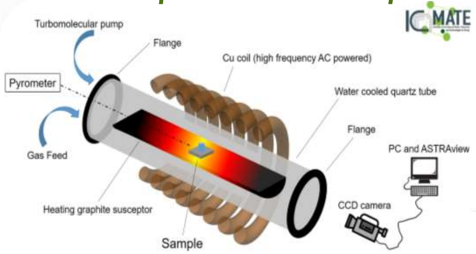
Methodology



Si-Zr alloys as-arc melted

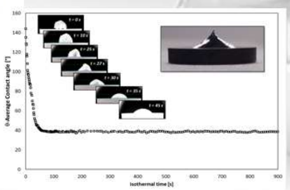


Experimental set-up



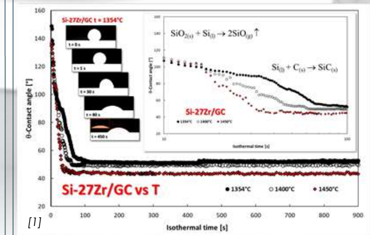
T ≤ 1700°C, 1 < P_{tot} < 10^{-6} mbar
C-heater acting as **O_2-getter**

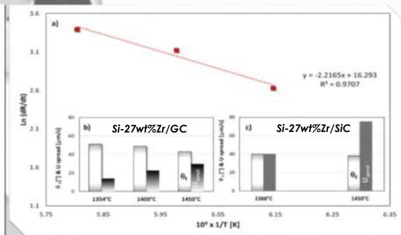
Follow-up



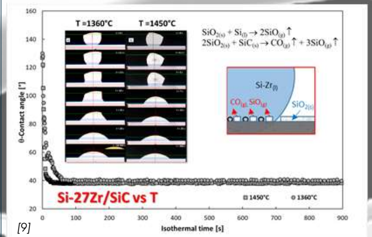
Heating rate ≤ 10°/s, Cooling rate = 20°/s
Thermocouples/Pyrometer
Real-time residual gas analysis and acquisition data (ASTRAView®)

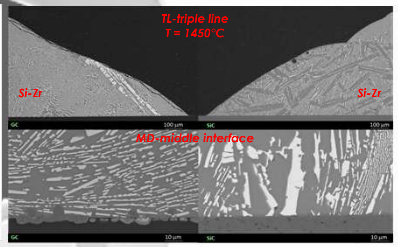
Results



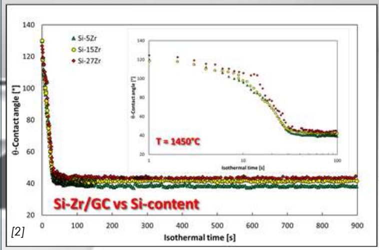


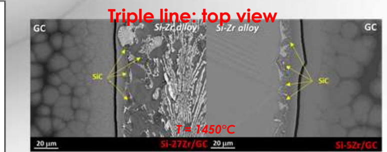
- U_{spread} (1354 °C) < U_{spread} (1400 °C) < U_{spread} (1450 °C)
- θ_f (1354 °C) > θ_f (1400 °C) > θ_f (1450 °C)
- Equilibrium contact angle values in good agreement with the literature [3-8]
- Reactive wetting at the Si-27Zr/GC triple line (E_a= 222 kJ/mol)
- Early stage of spreading affected by SiO_2-native oxide at the alloy surface

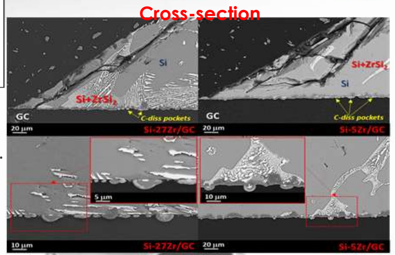




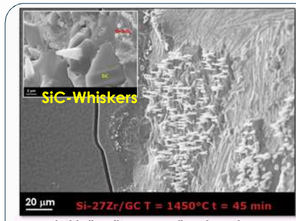
- U_{spread} ($SiC_{1360°C}$) < U_{spread} ($SiC_{1450°C}$)
- θ_f ($SiC_{1450°C}$) < θ_f ($GC_{1450°C}$)
- Equilibrium contact angle values in good agreement with the literature [1,2,3].
- Spreading kinetics affected by the presence of SiO_2-native oxide at the surface of both SiC substrate and Si-Zr alloy.



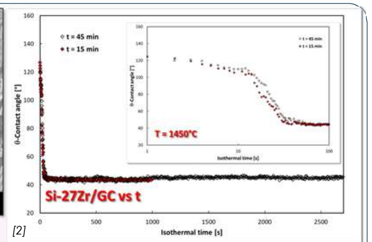




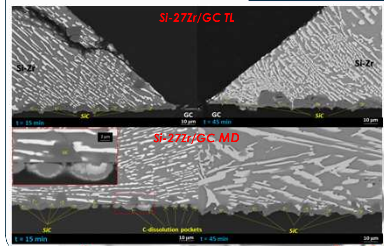
- θ_f (Si-27Zr) > θ_f (Si-15Zr) > θ_f (Si-5Zr)
- More compact SiC-layer at the interface by increasing the Si-content.
- The presence of ZrSi2 precipitates affects the melting of Si-27Zr alloy.
- Si-evaporation/condensation phenomena beyond the triple line.



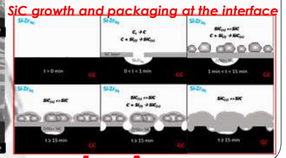
Inside the alloy surrounding atmosphere
$2C_{(s)} + O_{2(g)} \rightarrow 2CO_{(g)}$
At the alloy surface under oxide-removal conditions
$SiO_{(g)} + CO_{(g)} \rightarrow SiC_{(s)} + O_{2(g)}$
$SiO_{(g)} + 3CO_{(g)} \rightarrow SiC_{(s)} + 2CO_{2(g)}$
VLS-Process [10]



- High reproducibility
- In agreement with [11], the SiC-reaction layer growth and packaging of is time dependent phenomenon



SiC growth and packaging at the interface



Summary and Conclusions

- For the first time, a comprehensive study of the interaction phenomena occurring when a liquid **Si-rich Si-Zr alloy** is in contact with **amorphous C** and **SiC** was performed.

System	T [°C]	t [min]	$\theta_f \pm 2$ [°]	U_{spread} [µm/s]
Si-27Zr/GC	1354	15	51÷52	13.9
	1400	15	49	22.7
	1450	15	43	29.7
	1450	45	45	28.6
Si-27Zr/SiC	1360	15	40	40
	1450	15	38	75
Si-15Zr/GC	1450	15	41	36.4
Si-5Zr/GC	1450	15	38	37.7

- Careful analyses of the ***θ*-behaviors**, **spreading kinetics**, **reactivity** and **interfacial developed microstructures** as a function of the **Si-content** at **T = 1450°C**, were done.
- **Wettability of GC by Si-rich Si-Zr alloys** is controlled by the **reactive mechanism**.
- Despite the pronounced reactivity, the wetting characteristics are **slightly composition-dependent**.
- The **wetting characteristics** and **spreading kinetics** observed at the Si-rich Si-Zr alloys/GC triple lines at T = 1450°C are in a **very good agreement** with the results reported in **literature**.
- In view to provide knowledge for optimizing the infiltration process used to fabricate SiC/ZrSi$_2$ composites, except Si-27Zr alloy, the liquid Si-Zr alloys more enriched in Si, **may not enhance the pore closure phenomenon** for **limited time** of contact.
- The use of Si-Zr alloys with **Si exceeding** respect to the eutectic composition **should be avoided for preserving the overall thermo-mechanical response** of the produced composite.

Ref.

[1] D. Giuranno et al. J All Comps, 822 (5) 2020, n. 153643.
[2] D. Giuranno et al. Materials MDPI (2020), 13 (5) (2020) n.1194.
[3] M. Naikadea, et al. J Eur Ceram Soc 39 (2019) 735-742.
[4] G.W. Liu et al. Ceram Int 36 (2010) 1177–1188.
[5] Z. He et al. Ceram Int 46 (8) (2020) 10495-10502.
[6] Z. He et al. Carbon (2020) https://doi.org/10.1016/j.carbon.2020.05.109
[7] D. Giuranno et al. Met Mat Trans A 50 (10) (2019) 4814-4826.
[8] D. Giuranno et al. J Materi Eng Perf 28 (6) (2019) 3460-3467.
[9] D. Giuranno et al. J Eur Ceram Soc 40 (4) (2020) 953-960.
[10] F.J. Narciso et al. J Mater Sci 1996, 31, 779-784.
[11] R. Voytovych et al. J Eur Ceram Soc 32 (14) (2012) 3825-3835.

Acknowledgements

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