Wettability and infiltration of molten Si on SiO_2 substrate containing porous Si_3N_4 coating: Influence of α-Si_3N_4 coating and β-Si_3N_4 coating

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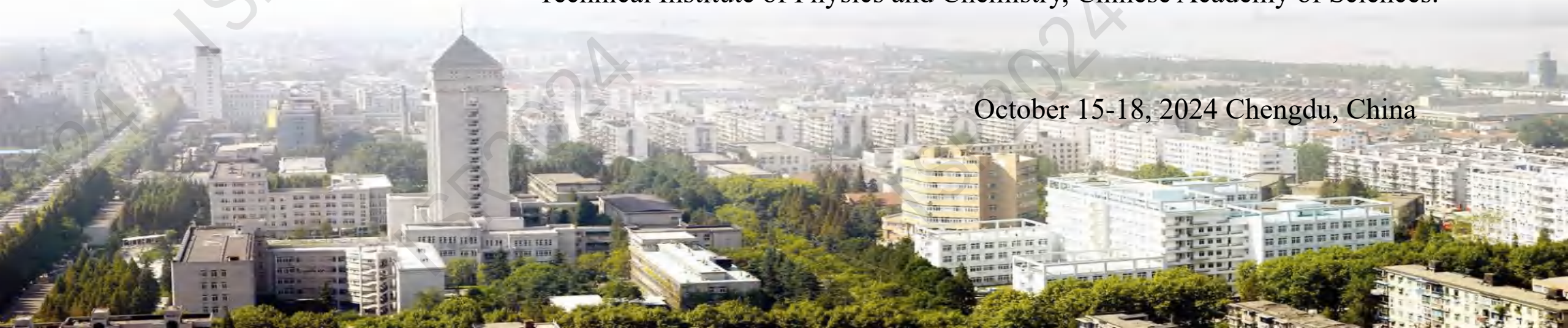

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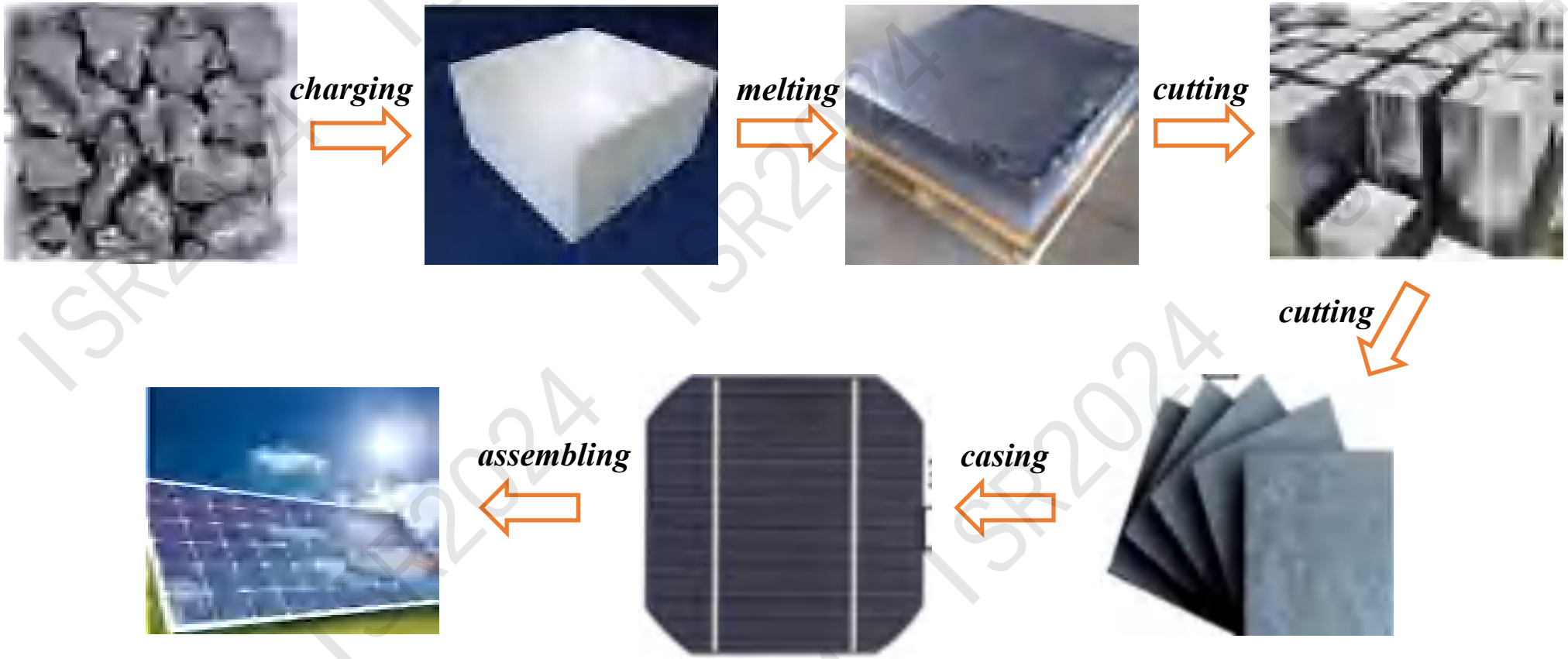
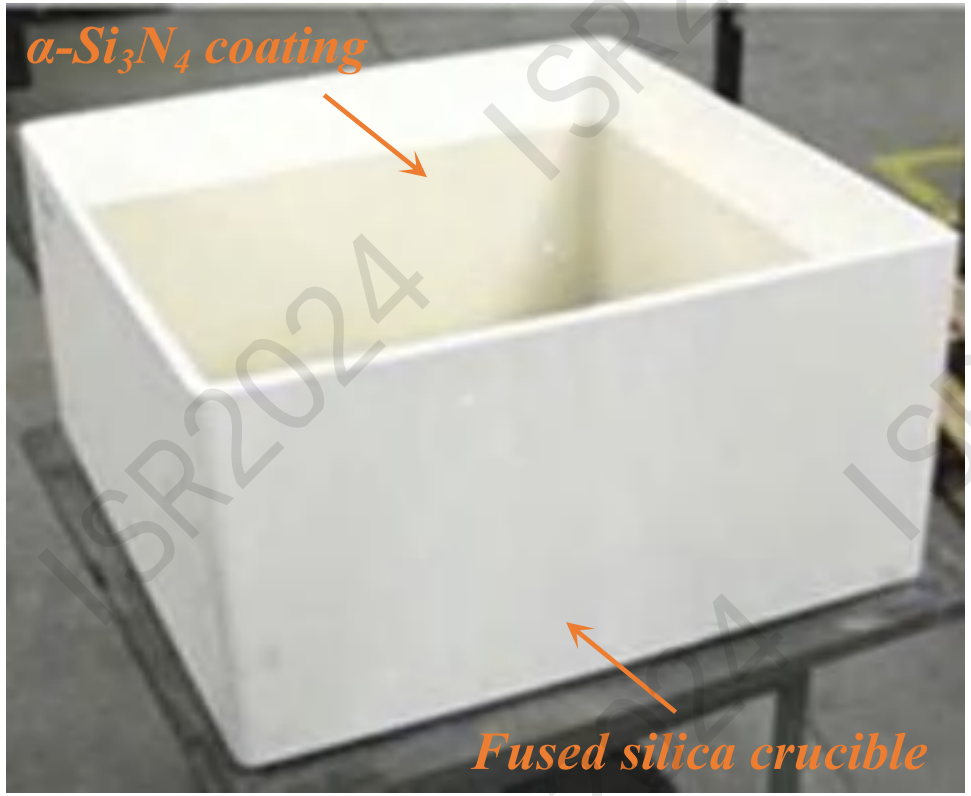
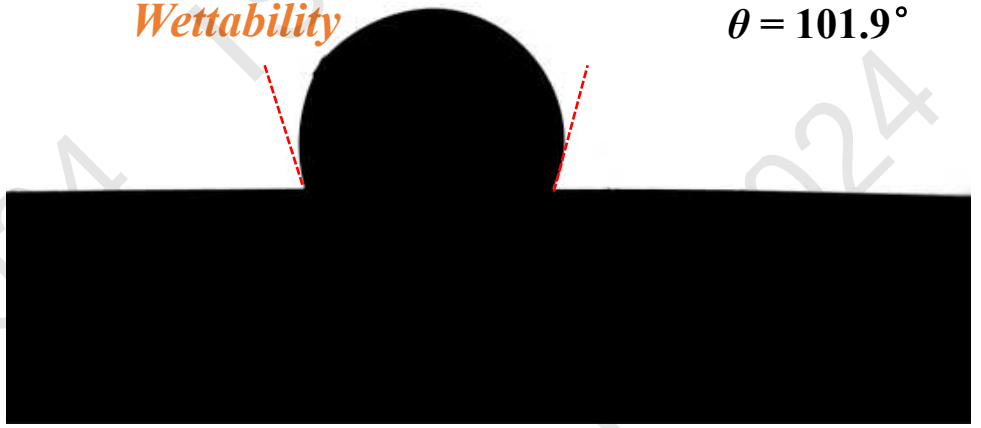
➢ **Fused silica crucible is the key melting vessel for the growth of PV silicon**

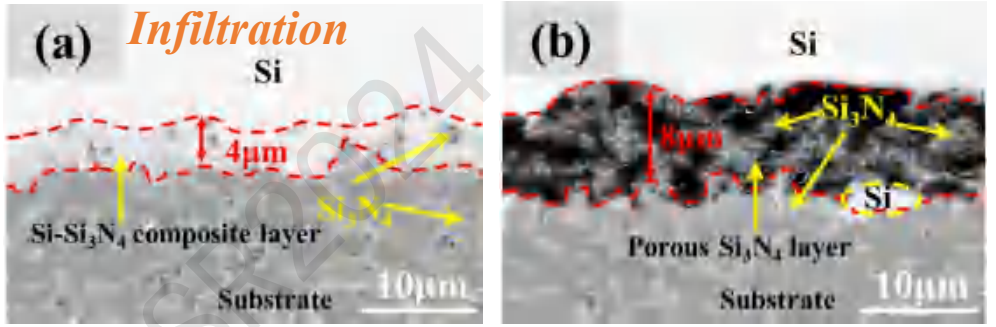
Directional solidification



- The α-Si_3N_4 coating effectively improve the **wettability** and **infiltration** behaviors of Si/SiO_2 binary system.

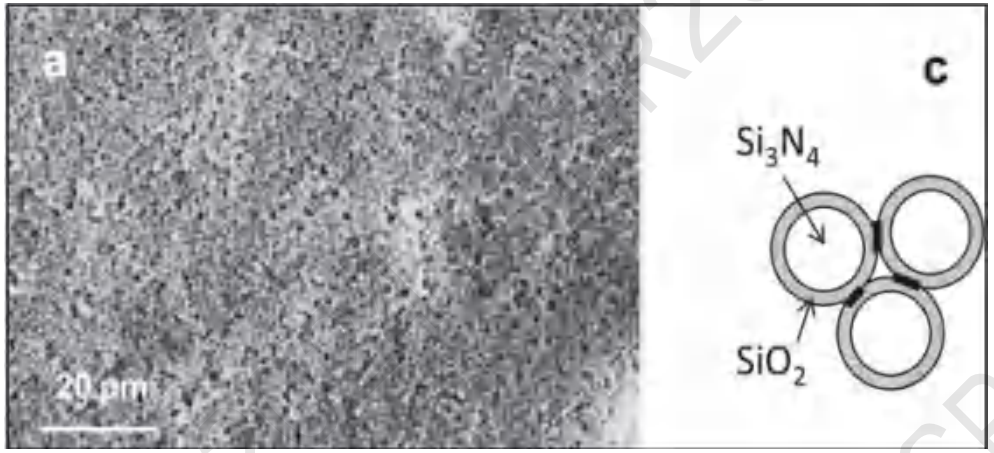






Why can α-Si_3N_4 coating improve the contact angle and infiltration behavior between Si and SiO_2 ?

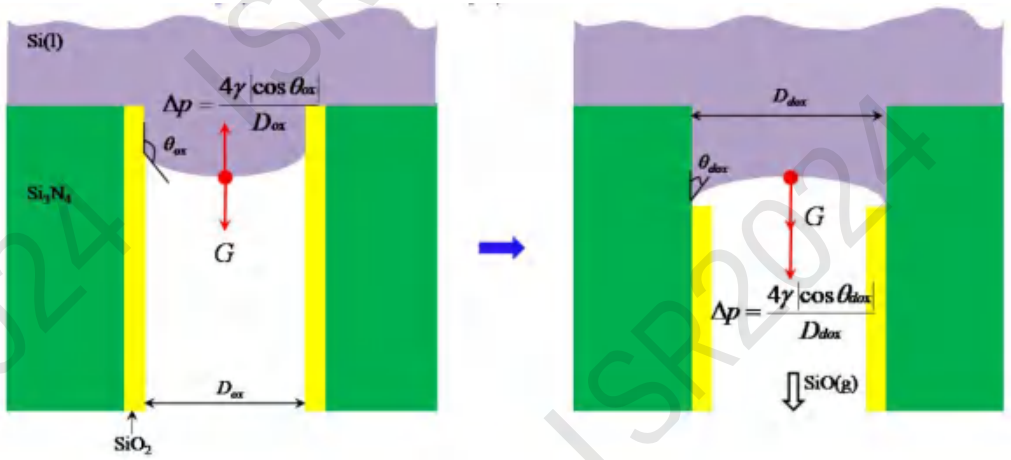
➢ The mechanism of α-Si_3N_4 as coating material



Si/SiO_2 = 83~ 88° *near non-wetting*

Si/Pore = 180° *non-wetting*

Si/(SiO_2+porous coating) ＞90° *non-wetting*



prede-oxidation

Convexity→θ_{OX}＞90°

- Δp and G keep balance with opposite vector direction
- lower the infiltration

postde-oxygenation

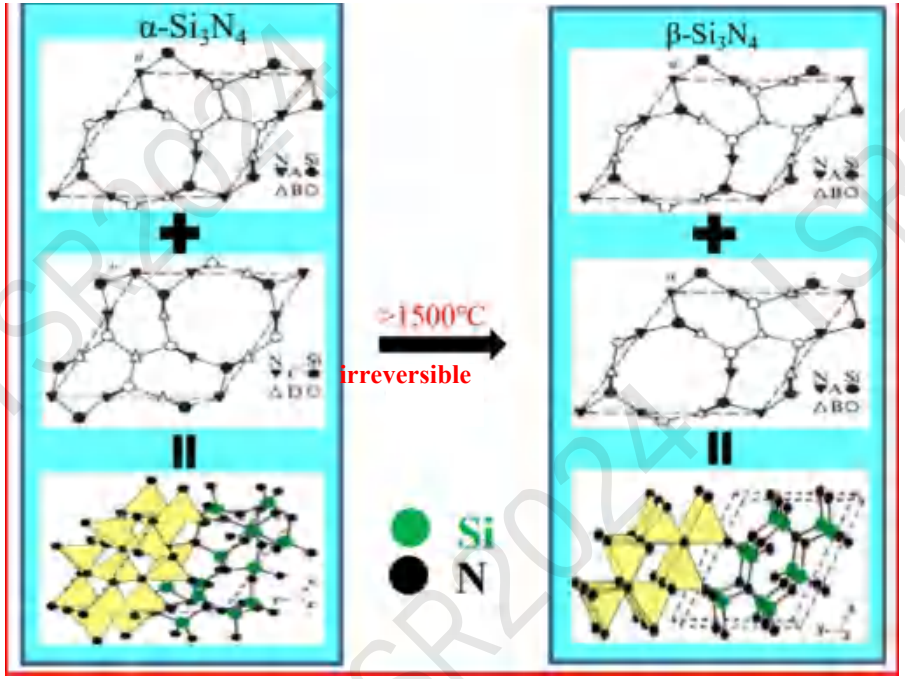
Concavity→θ_{OX}＜90°

- Δp and G with the same vector direction
- faster the infiltration

The SiO_2 film on the surface of α-Si_3N_4 particles in porous coating is the key to achieve non-wetting and no infiltration.

➢ **The β-Si_3N_4 has potential as the coating material**

× ***deficiencies of α-Si_3N_4*** { import; high cost } **Why not use β-Si_3N_4** → ***Unclearly*** { similar structure; crystal transformation; lower cost } ***advantages of β-Si_3N_4*** √



the price of α-Si_3N_4:

＞1000 yuan/Kg (UBE, Japan)

＞800 yuan/Kg (Stack, Germany)

＞800 yuan/Kg (ALZ, Germany)

＞600 yuan/Kg (Alticera, China)

the price of β-Si_3N_4:

≈300 yuan/Kg

However, it is of importance to develop a low cost coating material of β-Si_3N_4.

➢ The wetting and infiltration behavior of β-Si_3N_4 coating on Si/Si_3N_4/SiO_2 system

Three batches of Si_3N_4 coating
- α-Si_3N_4 coating (S-α)
- β-Si_3N_4 coating (S-β)
- silica sol modified β-Si_3N_4 coating (S-βO)

→ spay on SiO_2 substrate → sessile drop experiments → interface behavior

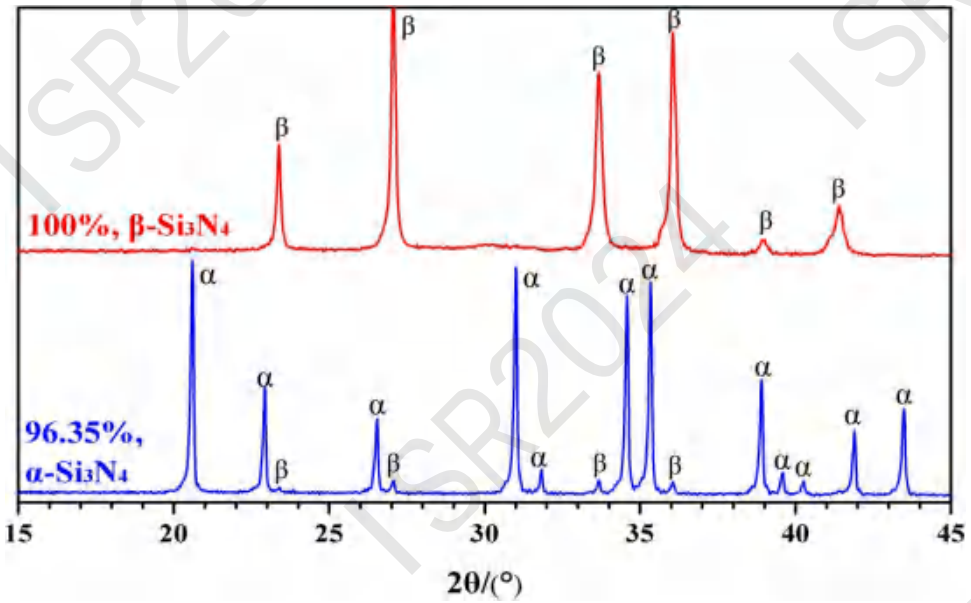
increase O content

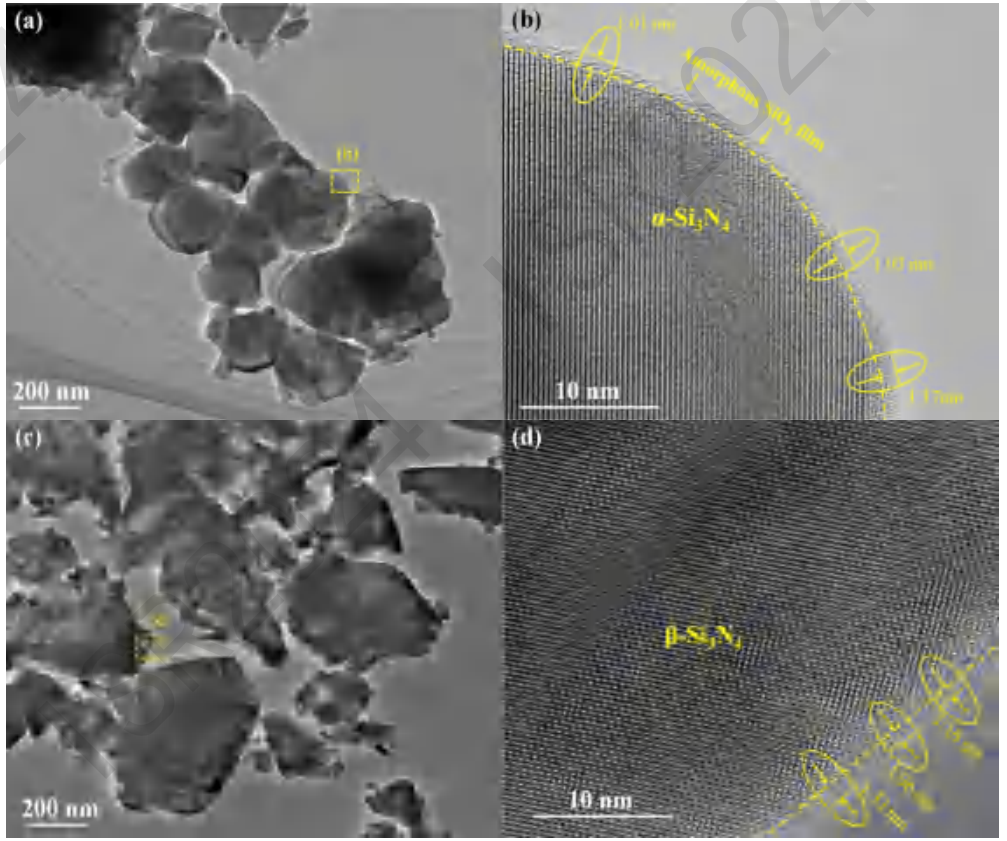
Ingredient (wt%)	S-α	S-β	S-βO
PVA	5	5	5
α-Si_3N_4	10	0	0
β-Si_3N_4	0	10	10
SiO_2 sol	0	0	10
Deionized water	85	85	75

➢ The characterization of Si_3N_4 powders

- The O content of both Si_3N_4 powders are similar.

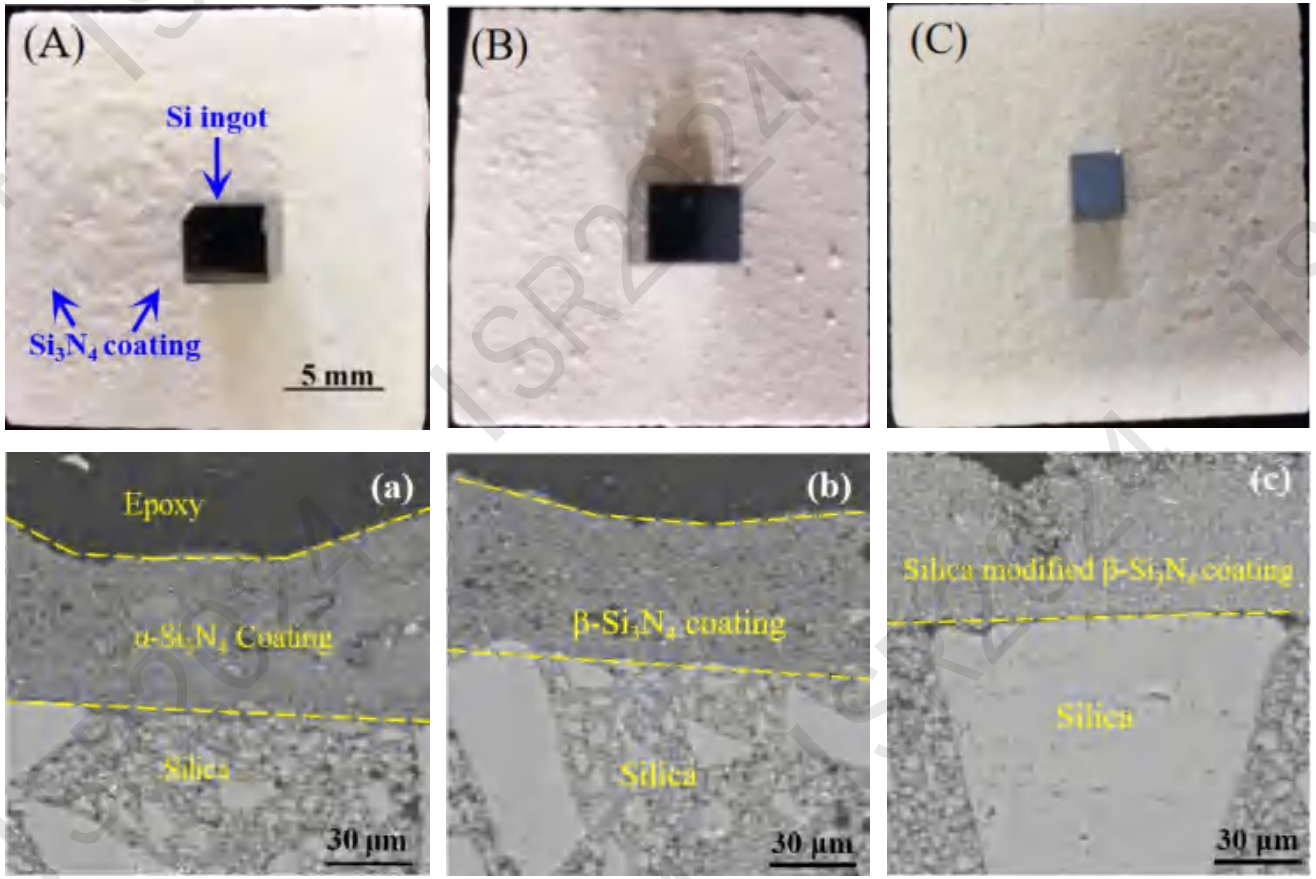
Si_3N_4 powder	α phase content/%	O content/wt%	Size/μm		
			D10%	D50%	D90%
α-Si_3N_4	96.35	1.04	0.63	0.86	1.21
β-Si_3N_4	0	1.18	0.62	0.87	1.19





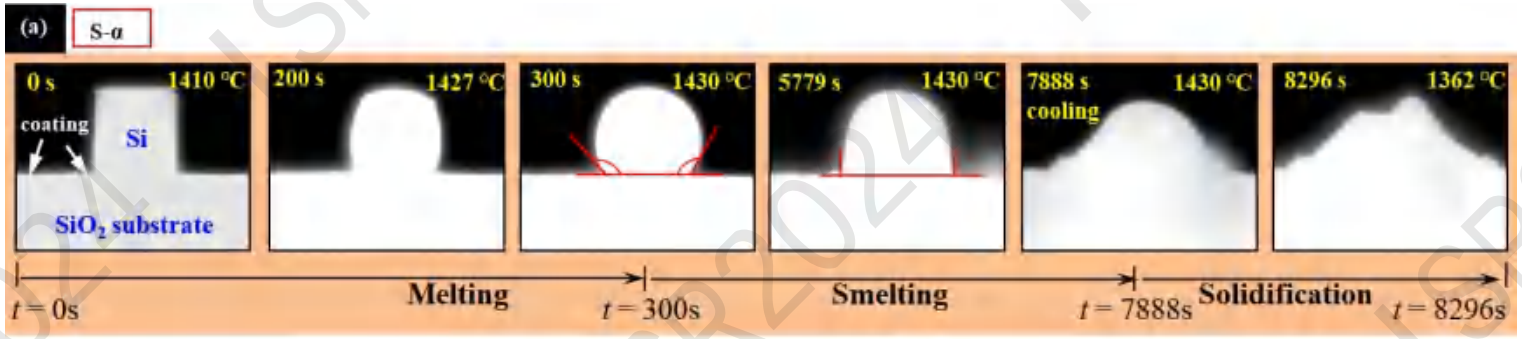
➢ The characterization of coatings

- The three porous Si_3N_4 coatings with similar thickness were prepared by pre-oxidizing at 900℃ for 1 h.

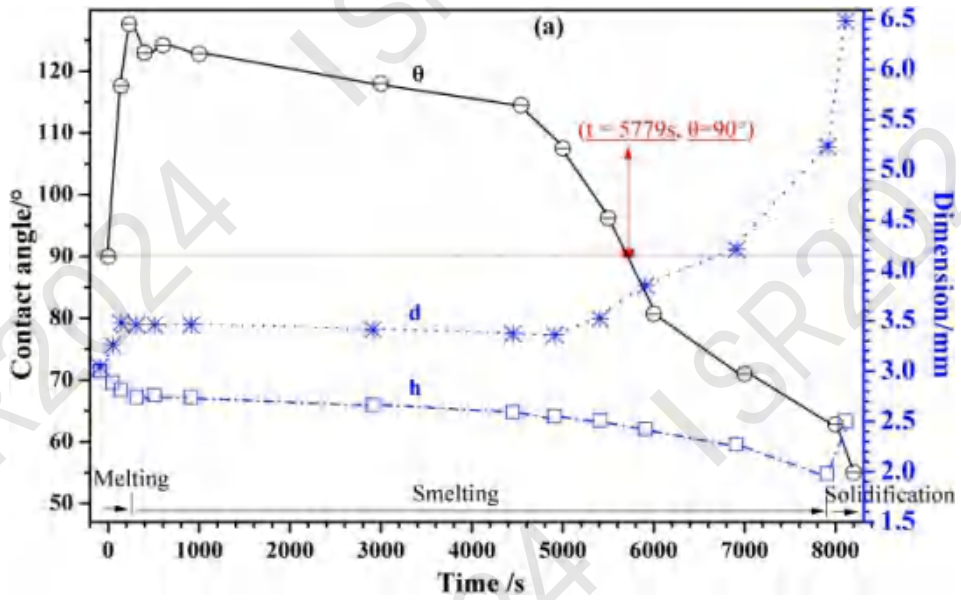


➢ The wetting behavior of α-Si_3N_4 coating

- The Si/α-Si_3N_4 kept non-wetting at the end of mlting process.
- The α-Si_3N_4 coating exhibited the longest non-wetting duration of 5779s.

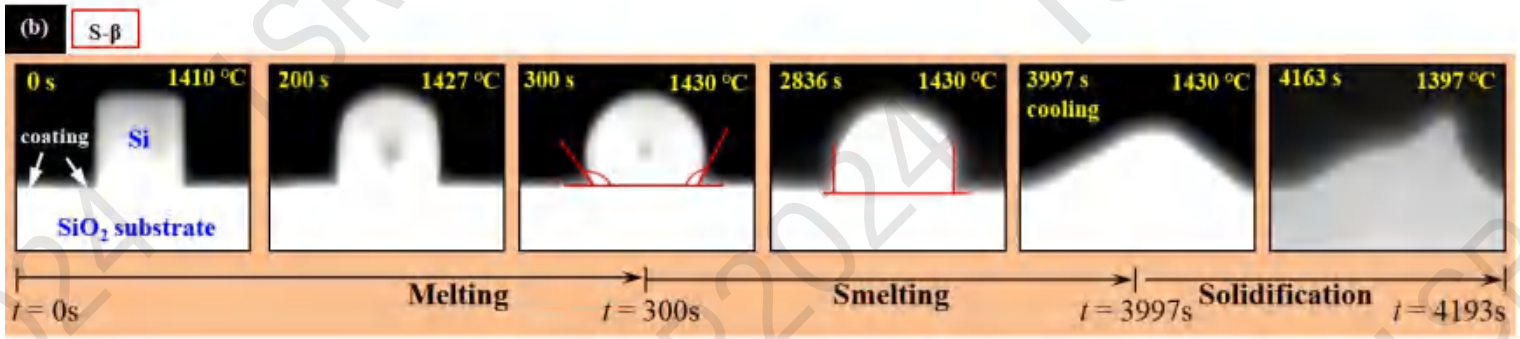


t = **5779 s**

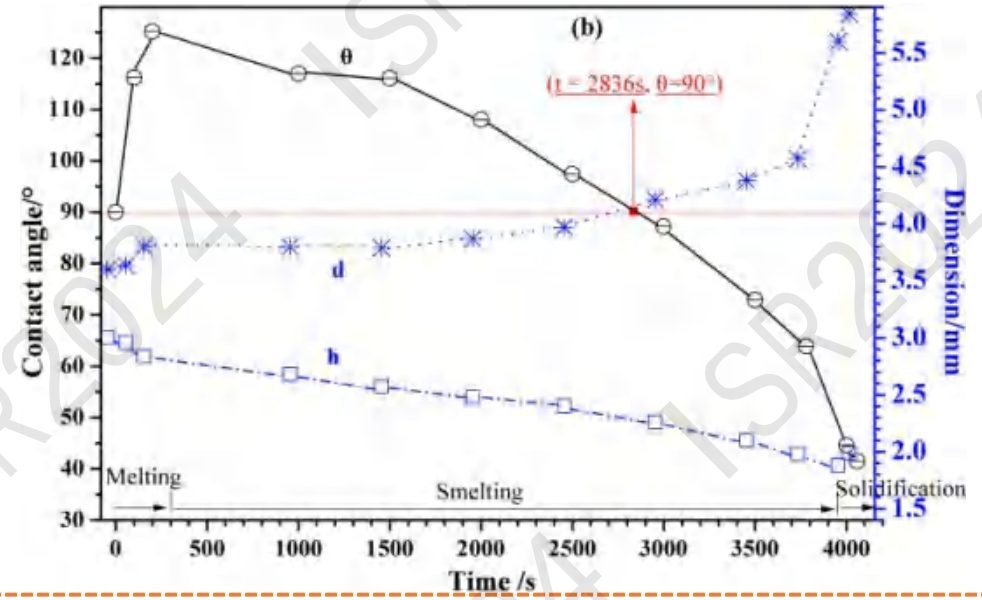


➢ The wetting behavior of β-Si_3N_4 coating

- The β-Si_3N_4 coating exhibited the shortest non-wetting duration of 2836s.

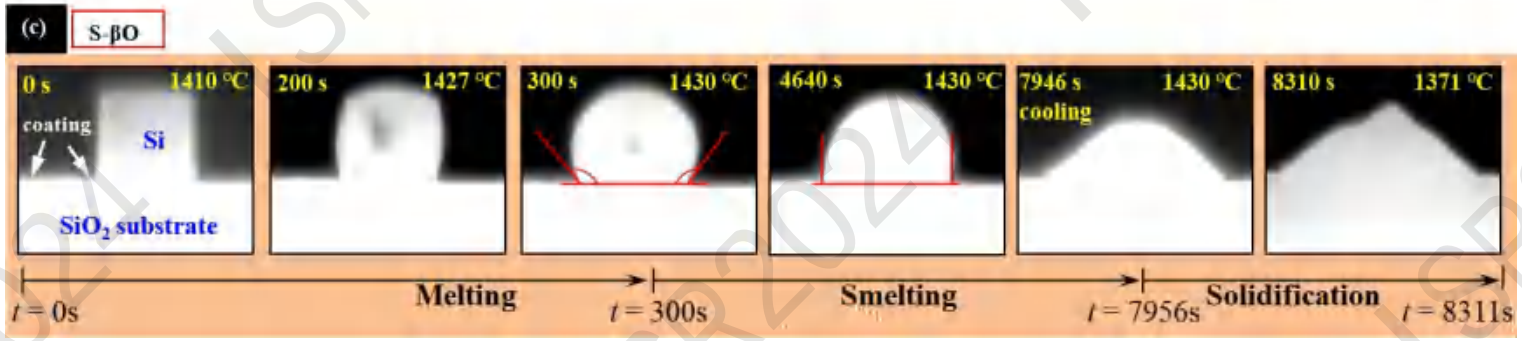


t = **2836 s**

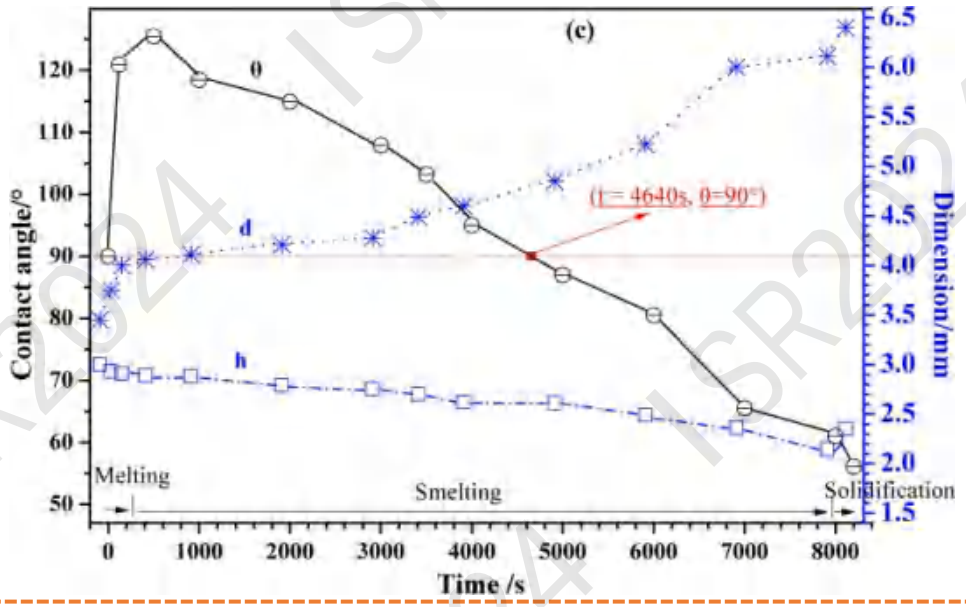


➢ The wetting behavior of silica sol modified β-Si_3N_4 coating

- The silica sol modified β-Si_3N_4 coating exhibited the non-wetting duration of 4640s.
- Compared with β-Si_3N_4 coating, the non-wetting duration of silica sol modified β-Si_3N_4 coating extended effectively.



t = **4640 s**



➢ The infiltration behavior of Si on various coating surfaces (beyond Si drop)

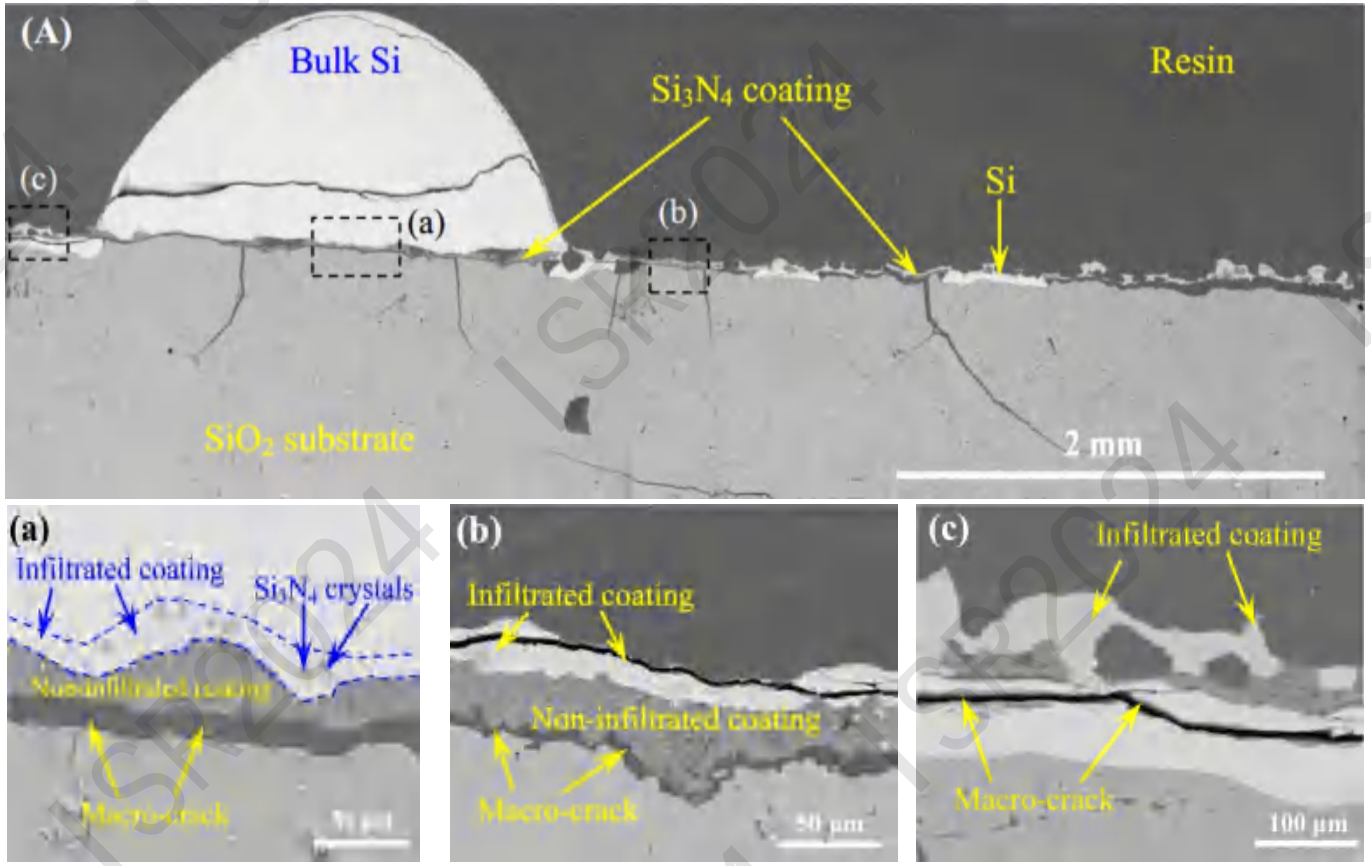
- The infiltration area of Si on the substrate coated different Si_3N_4 coatings is various.
- The C-α and C-βO have the silimar infiltration aeras, which is smaller than that of C-β.

Infiltrated region ratios on coating surface

Index	C-α	C-β	C-βO
Ratio/%	66.2	92.0	53.2

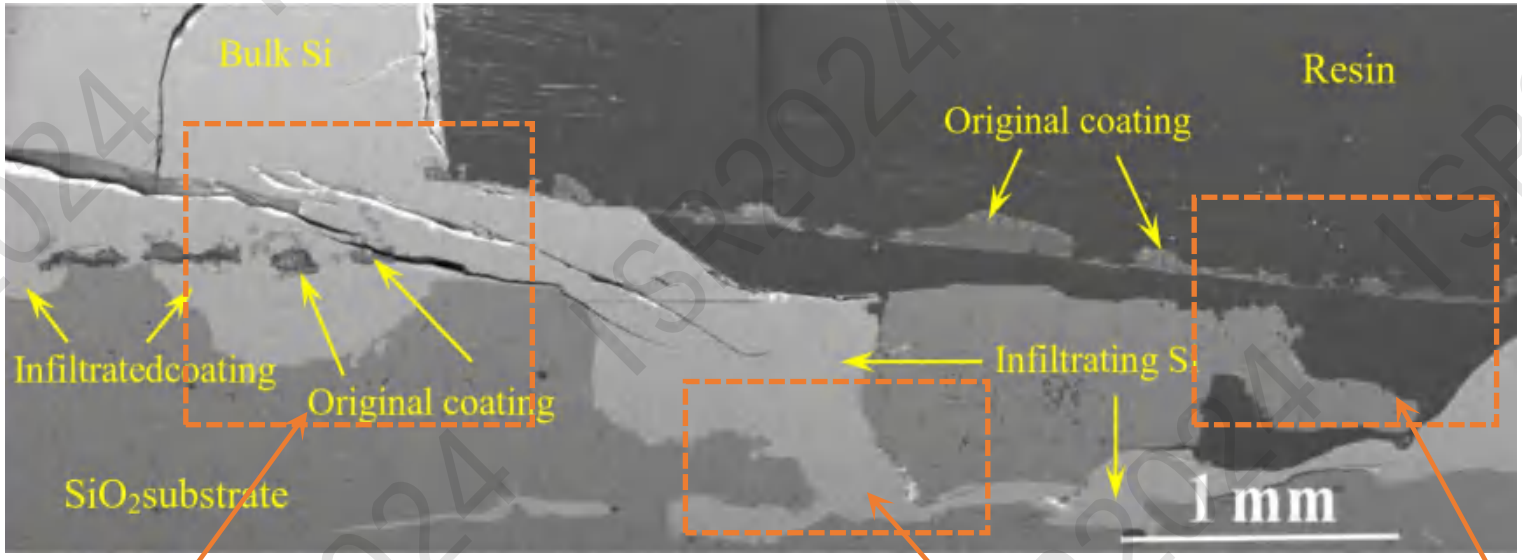
➢ The longitudinal infiltration behavior of Si on α-Si_3N_4/SiO_2 substrate

- The α-Si_3N_4 coating inhibited the infiltration of Si, only part of Si infiltration on coating.
- The detachment of Si and SiO_2 was separated with marco-cracks.



➢ The longitudinal infiltration behavior of Si on β-Si_3N_4/SiO_2 substrates

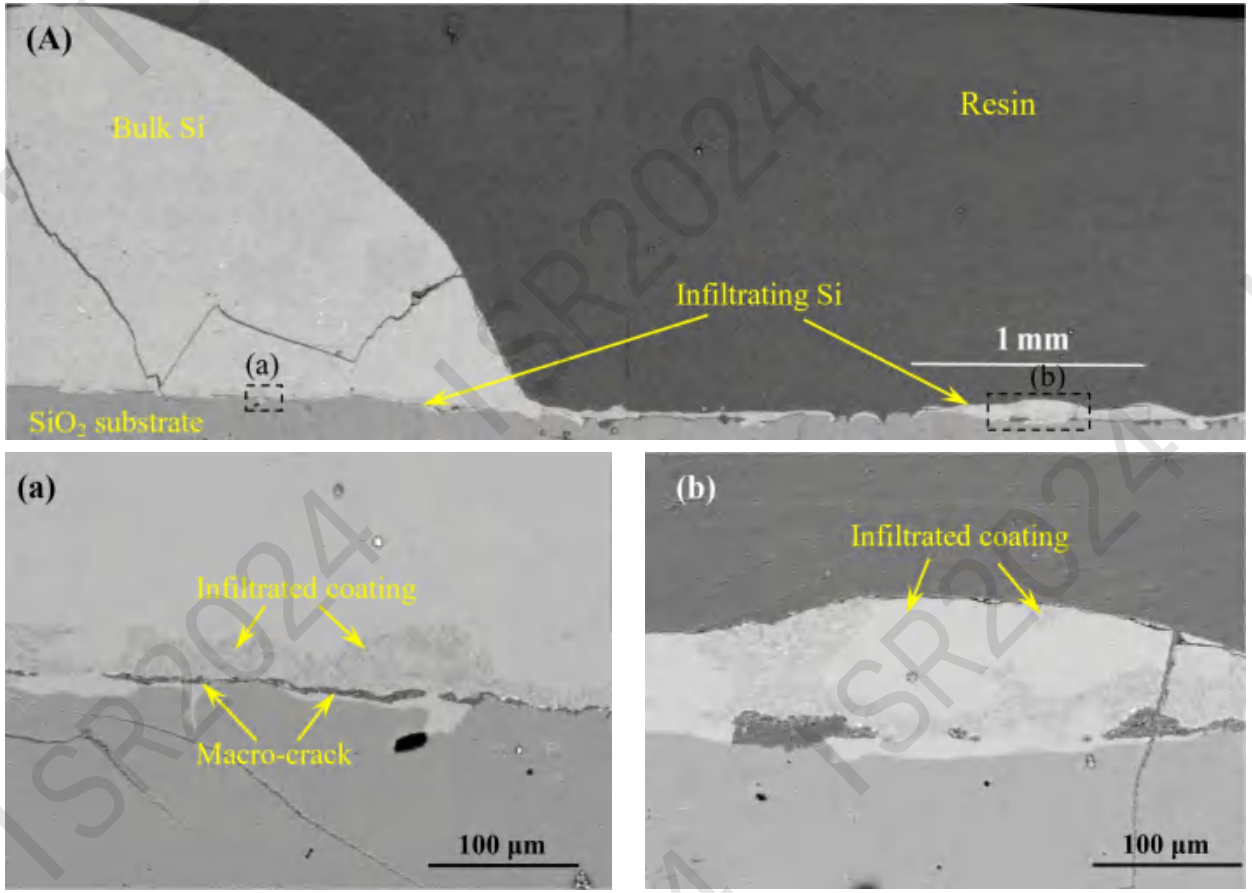
- The β-Si_3N_4 coating was completely infiltrated by molten Si.
- Si and SiO_2 substrate could not be separated due to interlock structure at Si/SiO_2 interface.



infiltrated coating of molten Si *interlock structure* *original and infiltrated coating*

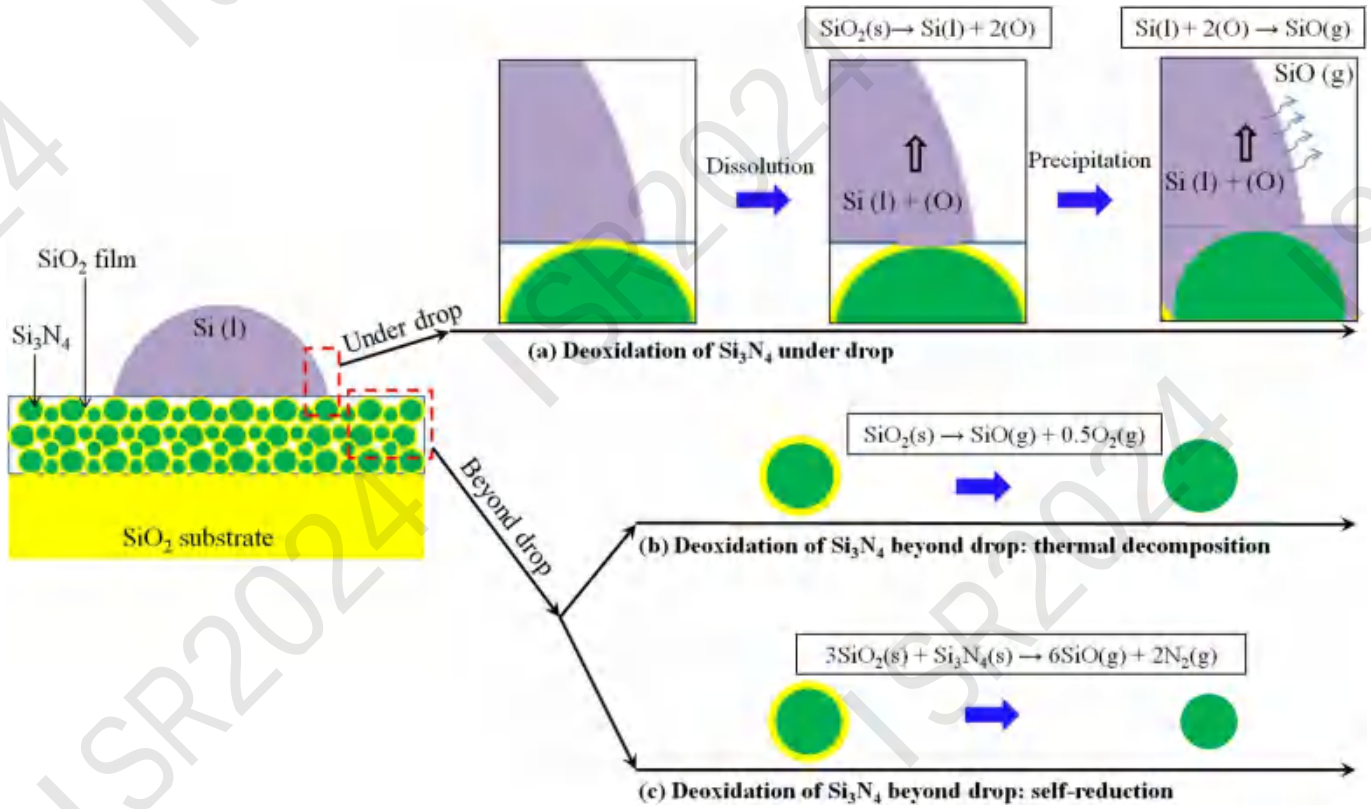
➢ The longitudinal infiltration behavior of Si on silica sol modified β-Si_3N_4/SiO_2 substrates

- Compared with S-β coating, the infiltration behavior of S-βO coating were improved effectively.
- The coating was partially infiltrated by molten Si. The detchment of Si and SiO_2 can be achieved with macro-cracks.



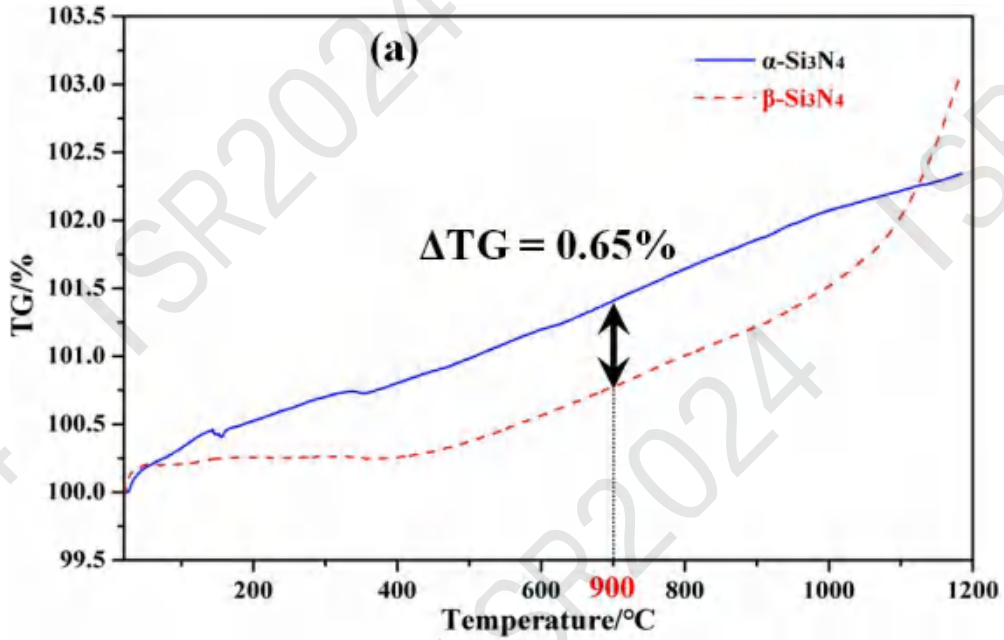
➢ The mechanism of de-oxidation process during wetting experiment

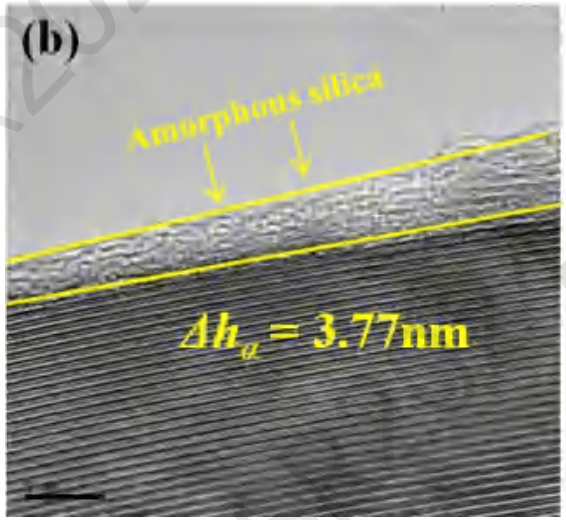
- The differences of wetting and infiltration behaviors can be attributed to the de-oxidation process of reactions.
- The interface behavior of Si/β-Si_3N_4 coating can be improved by adding the O content.

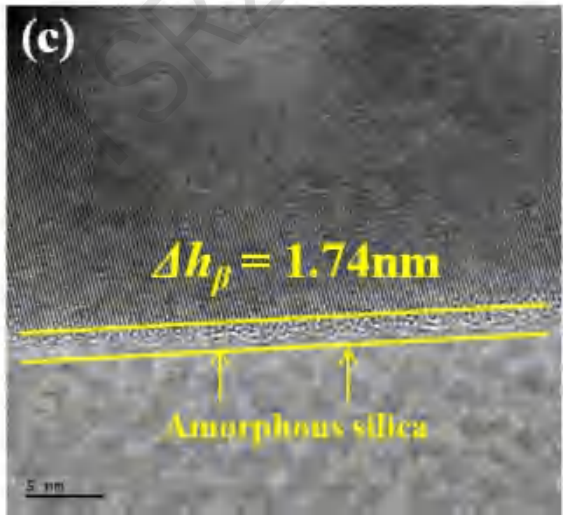


➢ The O content of Si_3N_4 coating is the key factor of interface behavior with molten Si.

- The mass gain gap between α-Si_3N_4 and β-Si_3N_4 is 0.65% due to the different oxidizability during the pre-oxidation process.
- The thickness of SiO_2 film of α-Si_3N_4 is 3.77nm, which is more than twice that of β-Si_3N_4 (1.74nm).







- Compared with α-Si_3N_4 coating, Original β-Si_3N_4 coating presents much shorter wettability transformation duration (from non-wetting to wetting) and severe infiltration by Si melt, due to its lower O content.

- By increasing O content in β-Si_3N_4 coating with SiO_2 sol, wettability transformation and infiltration are delayed significantly because of increased O evacuation time..

- β-Si_3N_4 can be applied as novel coating material for photovoltaic Si preparation by reasonable O modification.

It's of great importance for reducing the cost of coating and realizing the application of β-Si_3N_4.

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Thank you for your attention