

## Supplementary File

Monetite/ amorphous silica complex for long term dentine hypersensitivity treatment through the acid stability and mineralization promoting effect of silica

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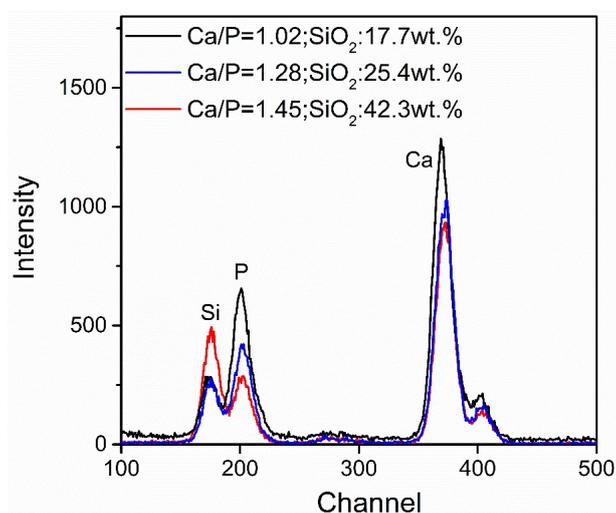


Figure S1 XRF of MMSi with different content of silica. It showed that the atomic ratio of Ca/P increased as the increasing of the content of silica.

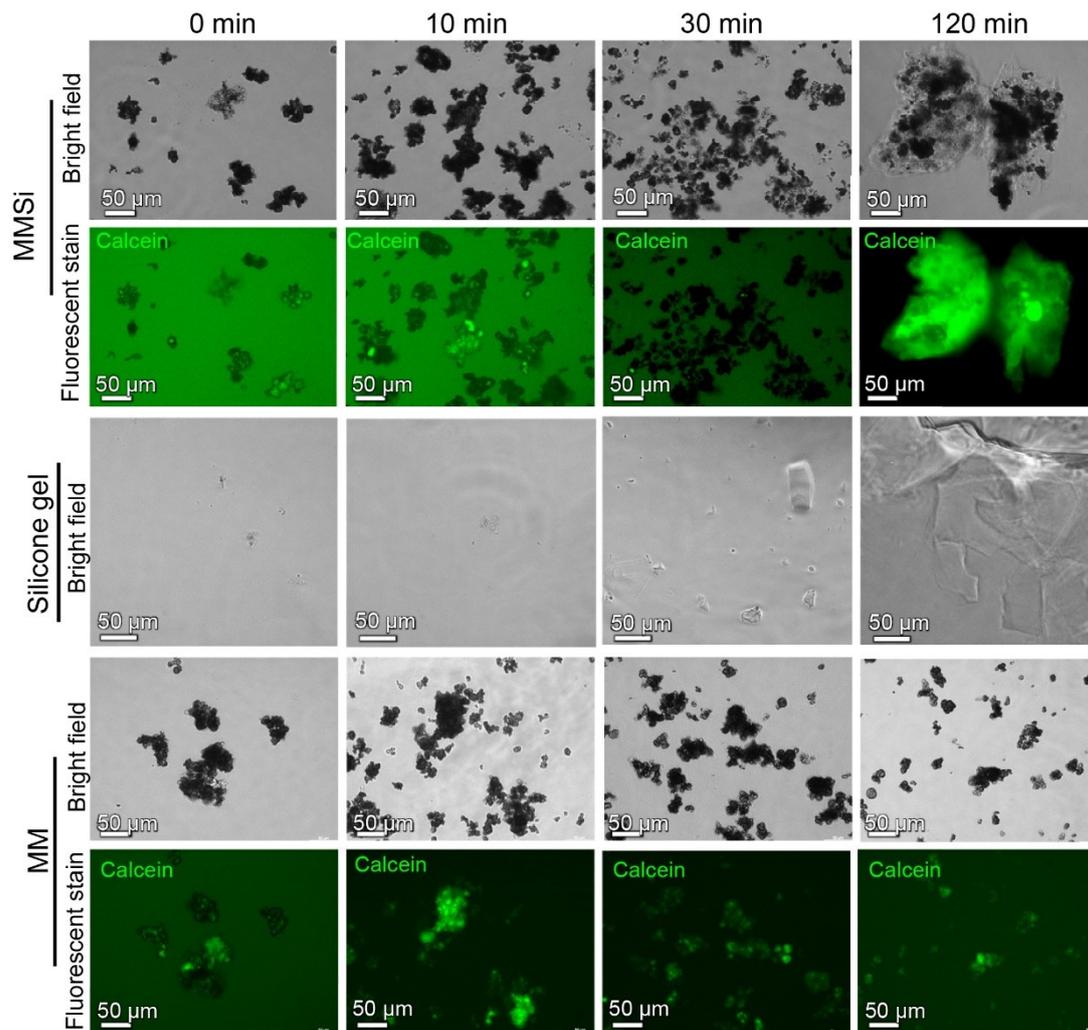


Figure S2 Bright field and Fluorescent stain of MMSi, Silicone gel and MM at different fabricating time. Samples were collected and soaked into anhydrous ethanol to stop reaction.

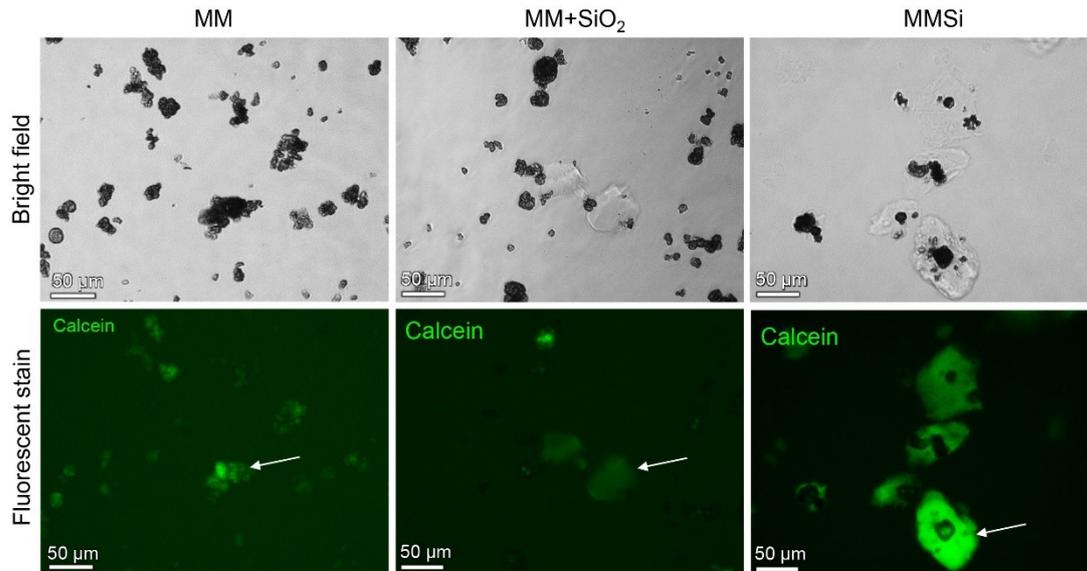


Figure S3 Bright field and Fluorescent stain of MM、mixture of MM and Silicone gel(MM+SiO<sub>2</sub>) and MMSi. White arrow: Calcium phosphate minerals labeled with calcein

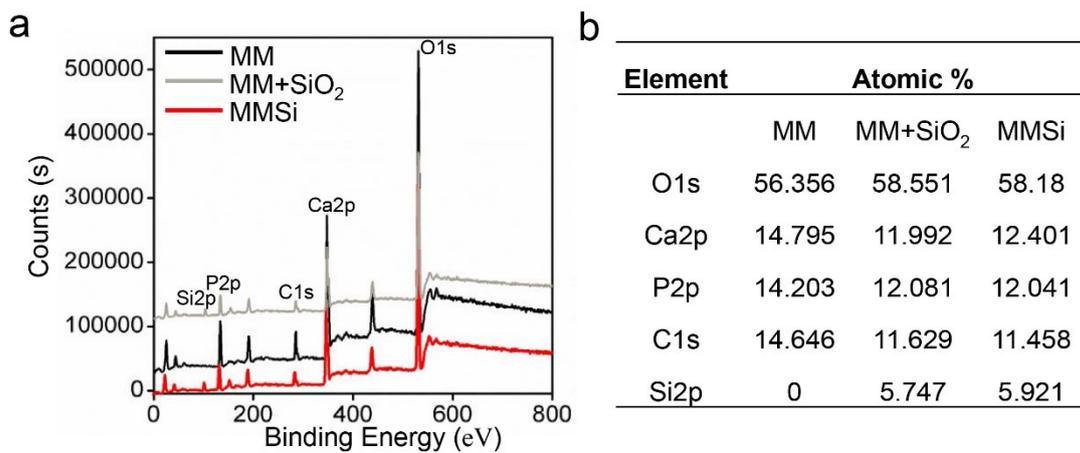


Figure S4 XPS of MM、mixture of MM and Silicone gel(MM+SiO<sub>2</sub>) and MMSi. (a) XPS spectrum, (b) Atomic percentage of O、Ca、P、C and Si in different mineral.

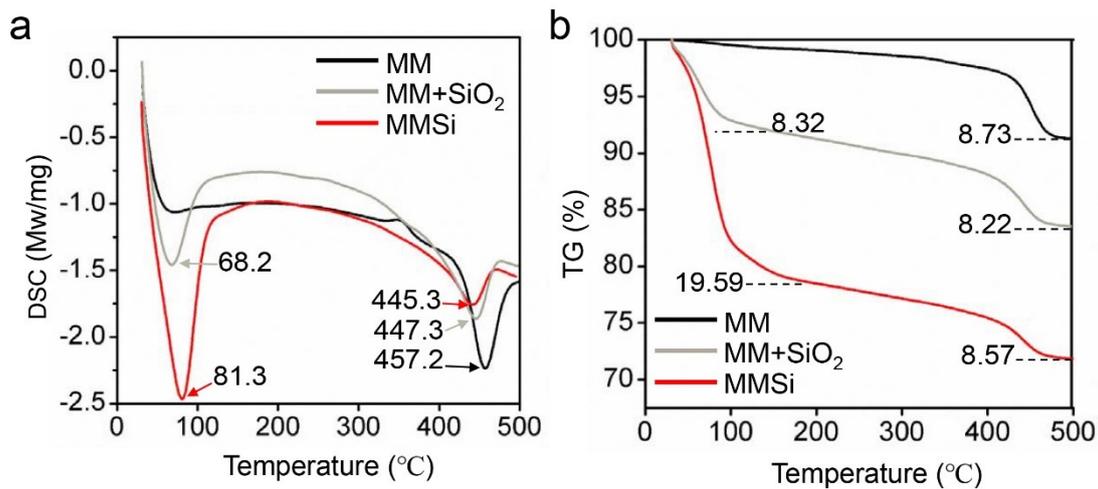


Figure S5 DSC and TG analysis of MM, mixture of MM and Silicone gel(MM+SiO<sub>2</sub>) and MMSi. (a) DCS spectrum, (b) TG spectrum. TGA was performed in nitrogen at a heating rate of 10°C/min.

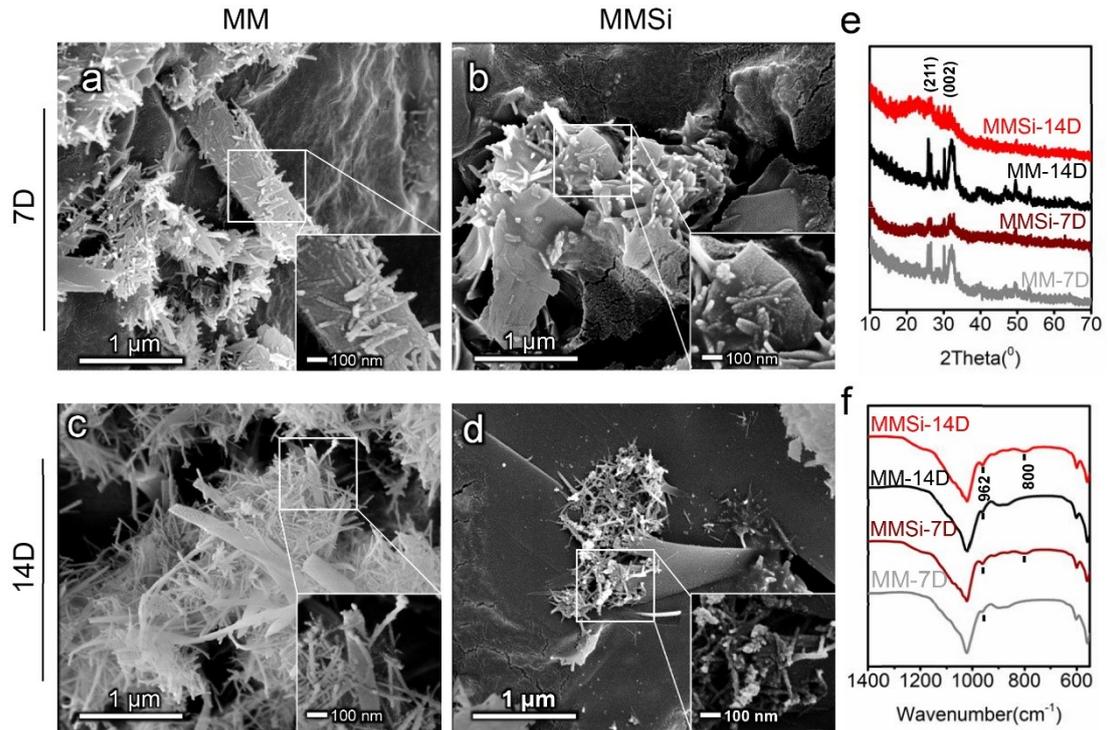


Figure S6 The surface morphology of MM and MMSi after mineralized for 7 days and 14 days in artificial saliva. (a, b, c, d) As the mineralization time increased, Rod-shaped particles formed and gradually grew up in MM; while small round particles formed in MMSi except for rod-shaped particles. (e) XRD showed the phase composites of mineralized minerals. As the extension of the mineralization time, the characteristic peak of hydroxyapatite (JCPDS 09-0432) (211 and 002) gradually became sharper for MM, while shorter for MMSi, indicating the size of hydroxyapatite particle became larger in MM and smaller in MMSi. (f) FTIR displayed that for MMSi, Si-O-Si vibration absorption peaks and Si-OH absorption peaks still appear at 800  $\text{cm}^{-1}$  and 962  $\text{cm}^{-1}$  with time.

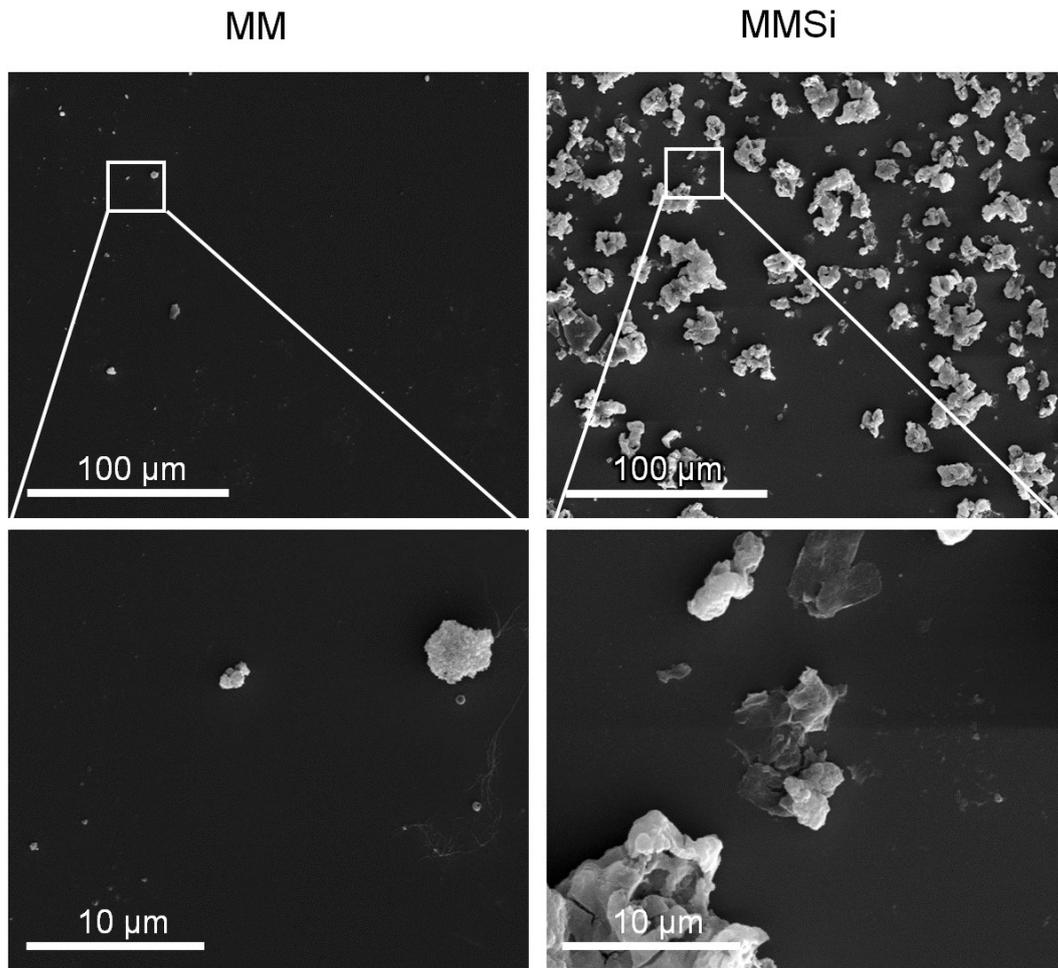


Figure S7 Acid resistance of MM and MMSi in 1% citric acid solution. SEM images showed that almost all the particles MM were dissolved, while many MMSi particles retained their structural integrity.

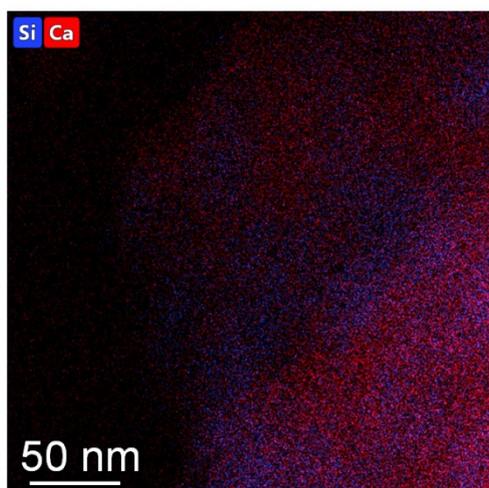


Figure S8 TEM of MMSi after 7 days' de/remineralization. One de/remineralization per day included remineralization in artificial saliva for 24 h and demineralization in 1.0 wt.% citric acid for 5min. Element surface analysis showed that the crystalline phase in this region is calcium, and the amorphous phase is silicon.

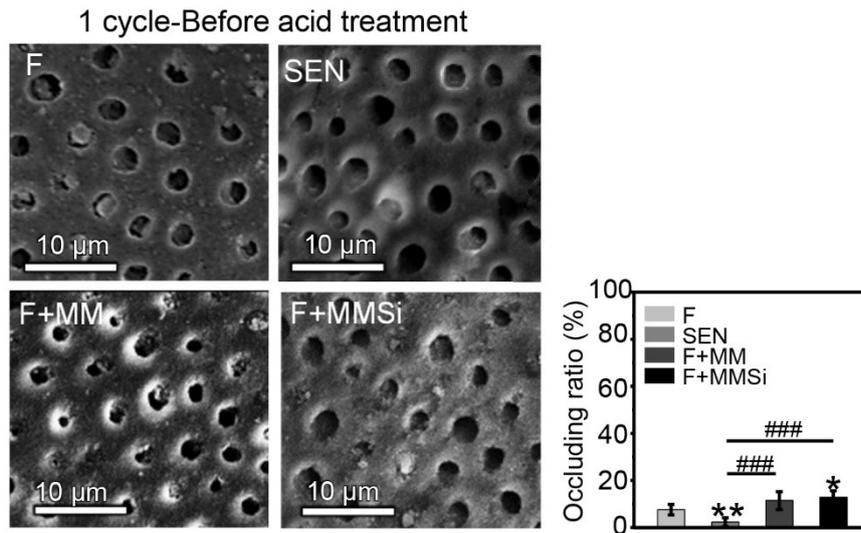


Figure S9 Assessment of dentin tubule occluding ratio after 1 de/remineralization cycles. \*Significant difference compared with the F group, \* $p < 0.05$ , \*\* $p < 0.01$ , \*\*\* $p < 0.001$ ; # significant difference between groups, # $p < 0.05$ , ## $p < 0.01$ , ### $p < 0.001$ ).

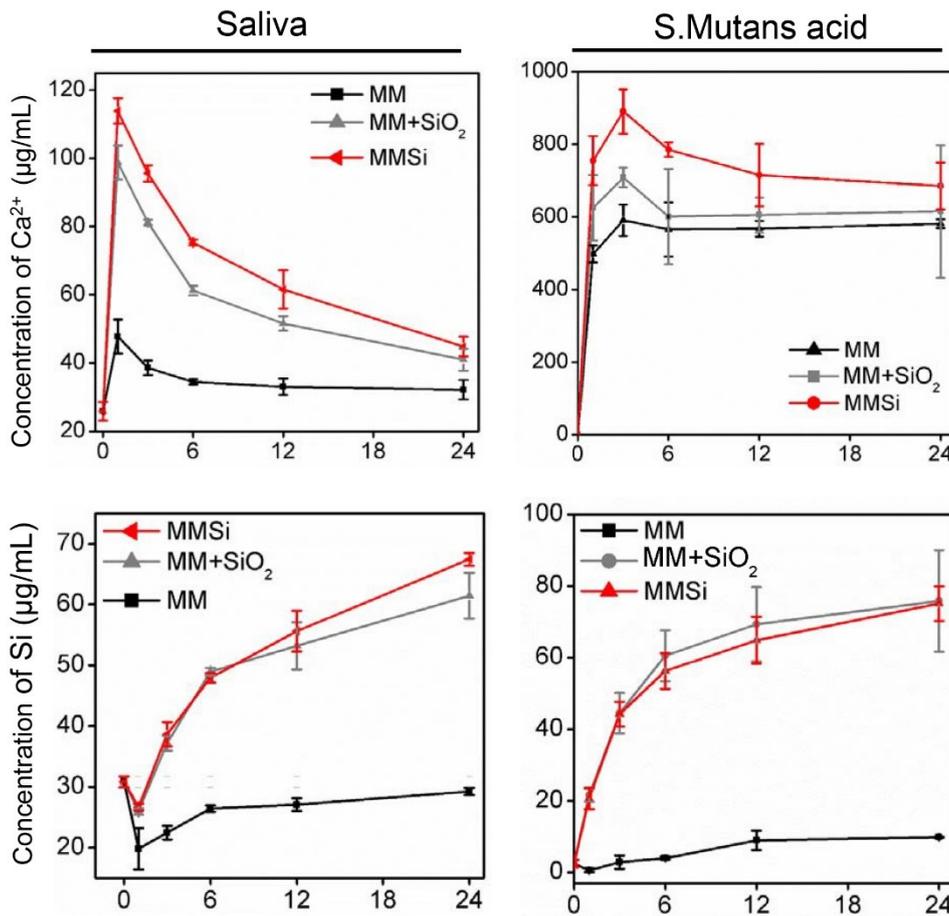


Figure S10 Accumulated ions release of MM, Mixture of MM and Silicone gel(MM+SiO<sub>2</sub>) and MMSi in saliva and S.Mutans acid