

## PORE FLOW-THROUGH CATALYTIC MEMBRANE REACTOR FOR STEAM METHANE REFORMING: CHARACTERIZATION AND PERFORMANCE

M. Angulo<sup>1</sup>, I. Agirre<sup>1</sup>, A. Arratibel<sup>2</sup>, M.A. Llosa Tanco<sup>2</sup>, D.A.  
Pacheco Tanaka<sup>2</sup>, V.L. Barrio<sup>1</sup>

<sup>1</sup>Engineering Faculty of Bilbao, University of the Basque Country (UPV/EHU. Plaza  
Ingeniero Torres Quevedo 1, 48013 Bilbao, Spain, maria.angulo@ehu.eus

<sup>2</sup>TECNALIA, Basque Research and Technology Alliance (BRTA), Mikeletegi Pasealekua 2,  
20009 Donostia-San Sebastián, Spain

### Experimental procedure

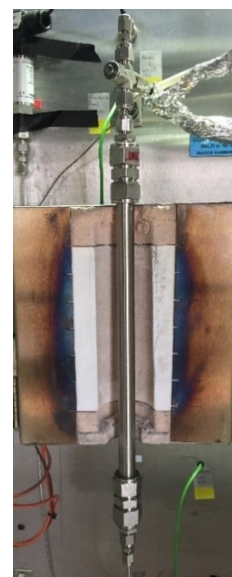
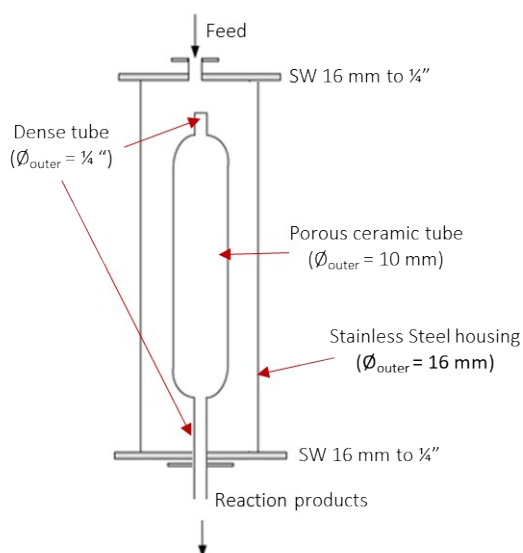
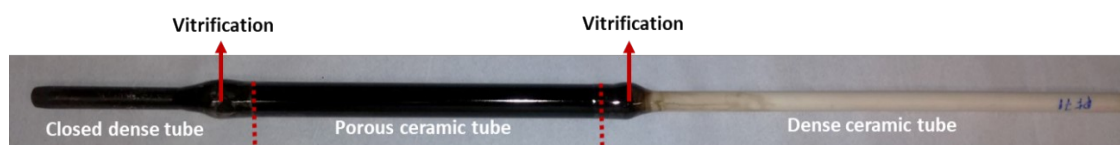


Figure S1. Non-tested ceramic reactor and configuration of "Pore Through Catalytic unselective Membrane Reactors (PTCMR)"

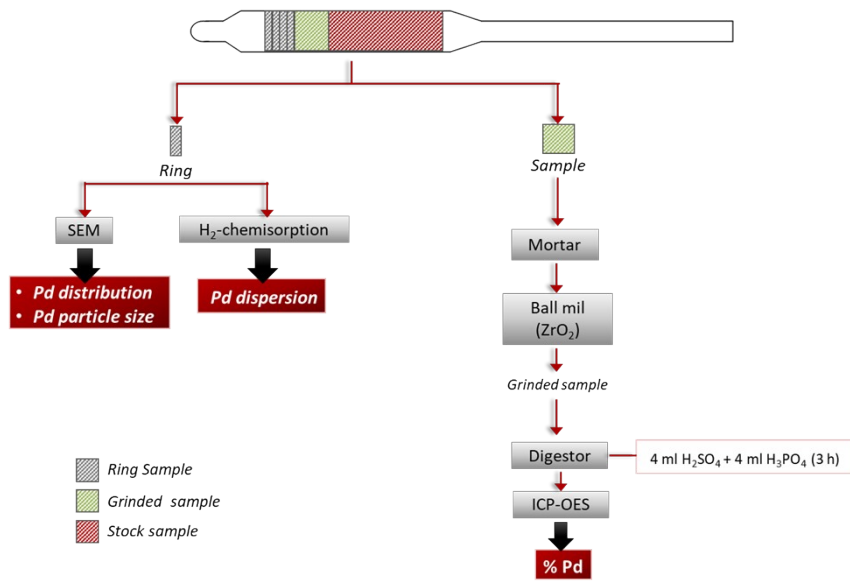


Figure S2. Characterization of ceramic tubular reactors.

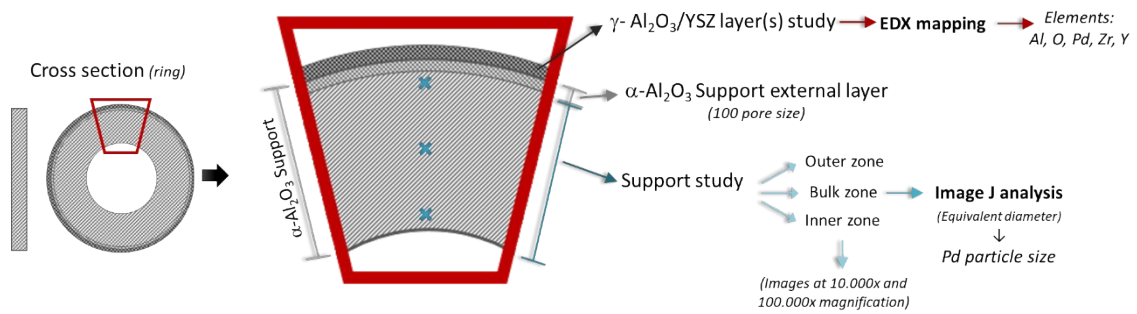


Figure S3. SEM study of the transversal section of the reactor

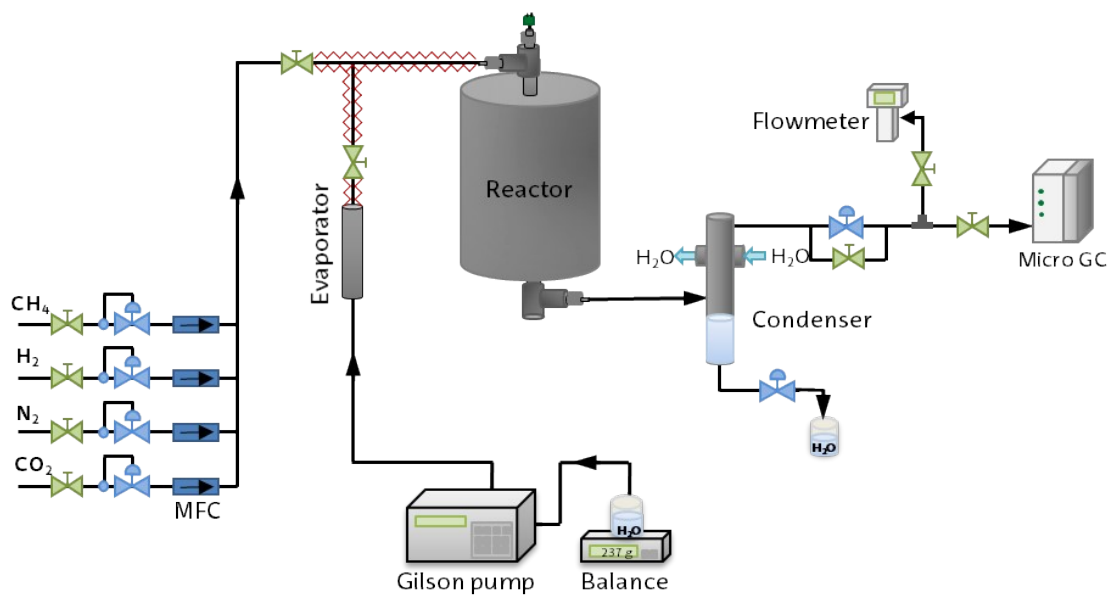


Figure S4. Scheme of the lab plant

## Results and Discussion

### 1.1. Characterization tests

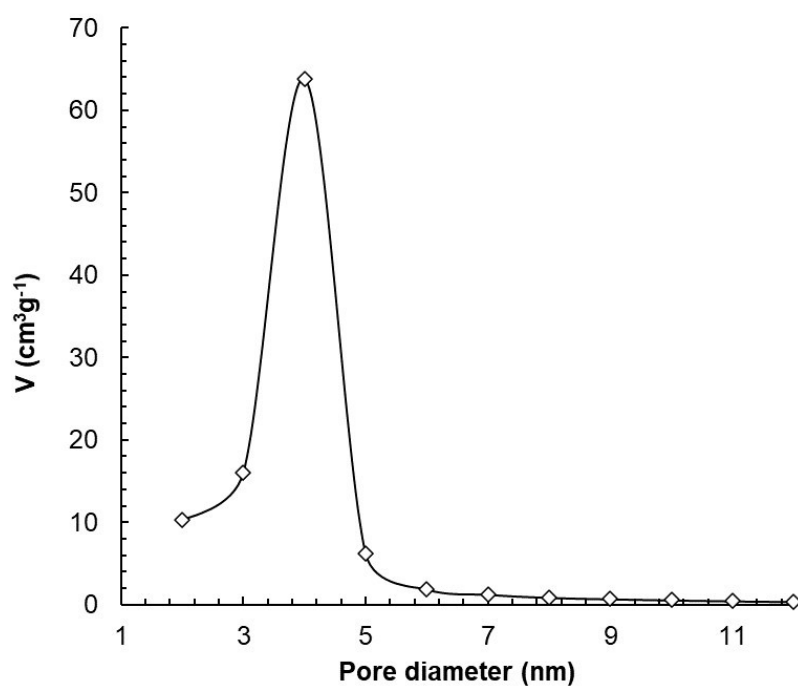


Figure S5. Pore size distribution of  $\gamma\text{-Al}_2\text{O}_3/\text{YSZ}$  obtained by BJH method.

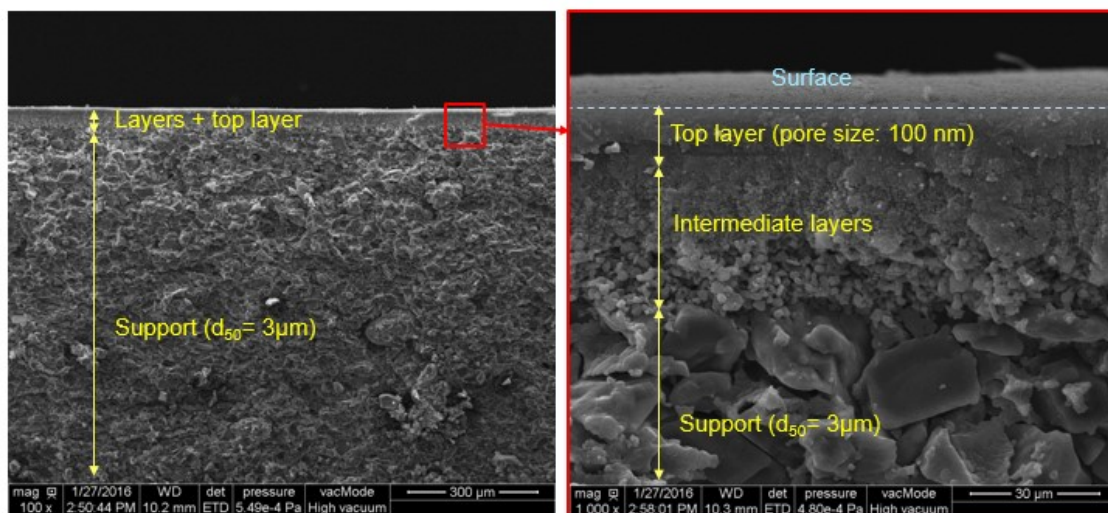


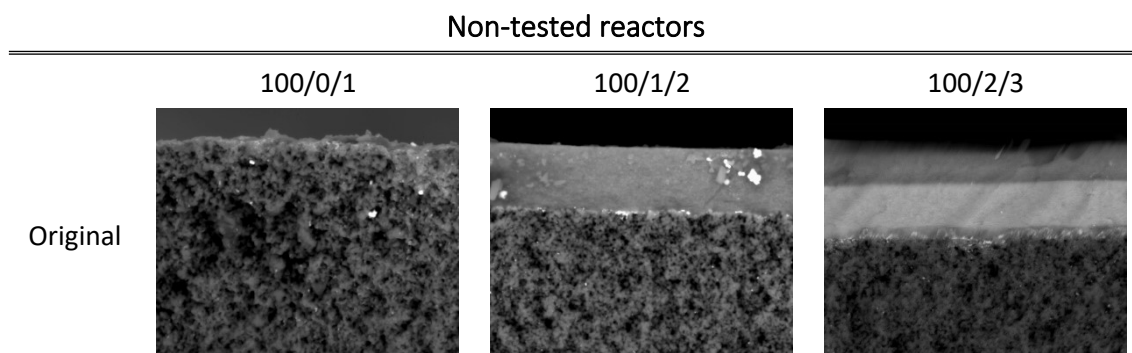
Figure S6. SEM images of a membrane reactor

Table S1. Thickness measurements of each layer

Reactor	Layer 1 ( $\mu\text{m}$ )	Layer 2 ( $\mu\text{m}$ )	Layer 3 ( $\mu\text{m}$ )
100/1/1	$2.65 \pm 0.07$	-	-
100/1/2	$2.56 \pm 0.11$	-	-
100/2/2	$4.09 \pm 0.05$	$1.50 \pm 0.01$	-
100/2/3	$2.21 \pm 0.15$	$1.31 \pm 0.03$	-
100/3/3	$4.19 \pm 0.16$	$1.53 \pm 0.02$	$1.43 \pm 0.01$
200/1/1	$2.23 \pm 0.04$	-	-

### 1.1.1. Effect of the addition of $\gamma$ - $\text{Al}_2\text{O}_3$ /YSZ layers on 100 nm pore size supports

A) Reactors with Pd deposition in both the  $\alpha$ -alumina support and the  $\gamma$  -  $\text{Al}_2\text{O}_3$ /YSZ nanoporous layers (100/0/1, 100/1/2 and 100/2/3)



*Figure S7. SEM micrographs of external zone of reactor 100/0/1, 100/1/2 and 100/2/3*

The length of the Boxplot graphs bars gives information about the homogeneity of the data. Consequently, the homogeneity of the palladium particles in a particular region of the support (outer, bulk or inner) could be evaluated as well as the particles size uniformity along the cross section of the reactor. If the mean particle size of the three regions is similar, palladium particle size along the cross section of the support is homogeneous.

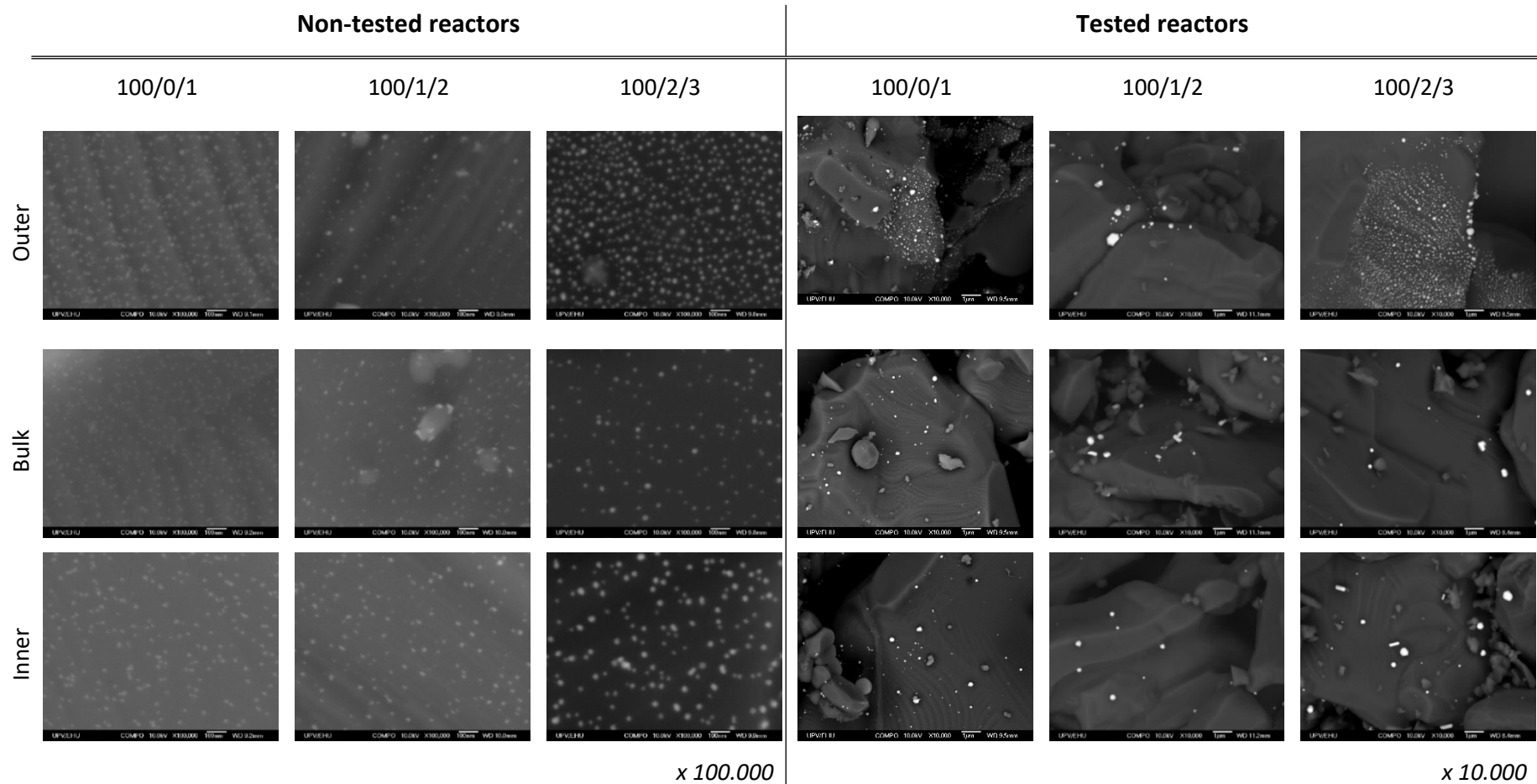


Figure S8. SEM micrographs along cross section of reactor 100/0/1, 100/1/2 and 100/2/3 (non-tested and tested reactors)

As shows Figure S9, reactor 100/0/1 presents significant uniformity among palladium particle size in the three zones of the support, due to the short length of the boxplot bars. In the case of reactor 100/1/2 particles are more heterogeneous ranging between 5 nm to 15 nm equivalent diameter in the case of the outer and bulk zones of the support and from 8 nm to 18 nm in the inner part. Reactor 100/2/3 presents more homogeneous particle size in the outer and bulk zones compared with the inner zone where the boxplot bar is longer.

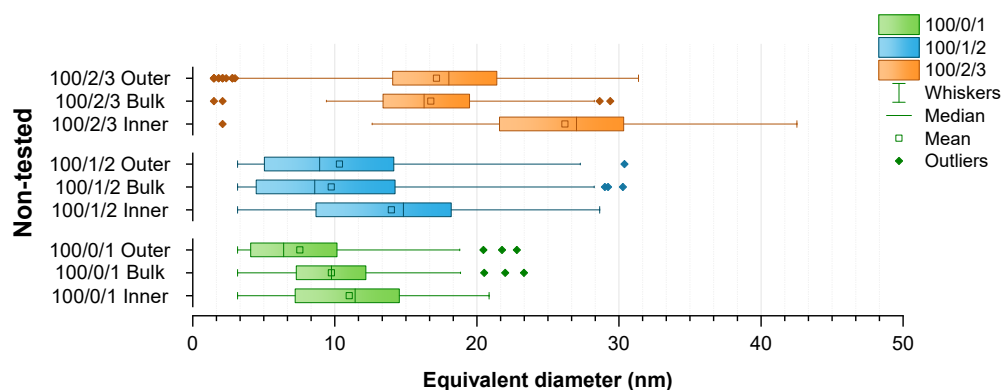


Figure S9. Boxplot graphs of palladium particles of 100/0/1, 100/1/2 and 100/2/3 non-tested reactors

B) Reactors with Pd particles in  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>/YSZ layers but not on the support (100/1/1, 100/2/2 and 100/3/3)



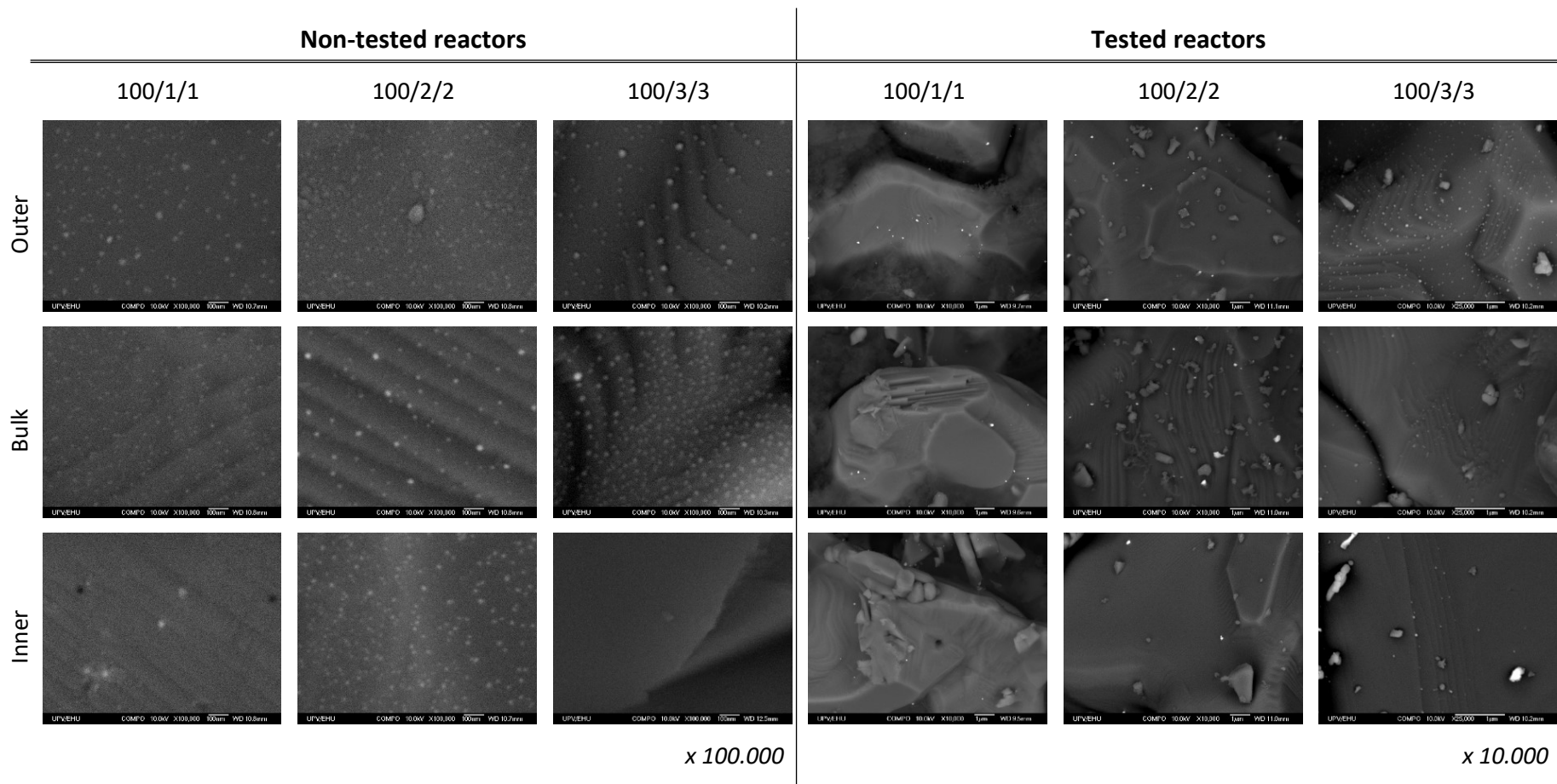


Figure S10. SEM micrographs along cross section of reactor 100/1/1, 100/2/2 and 100/3/3 (non-tested and tested reactors)

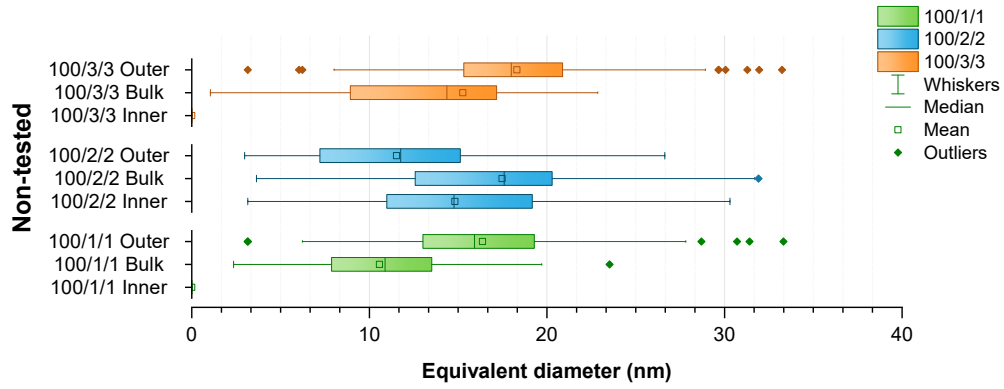


Figure S11. Boxplot graphs of palladium particles of 100/1/1, 100/2/2 and 100/3/3 non-tested reactors

100/1/1

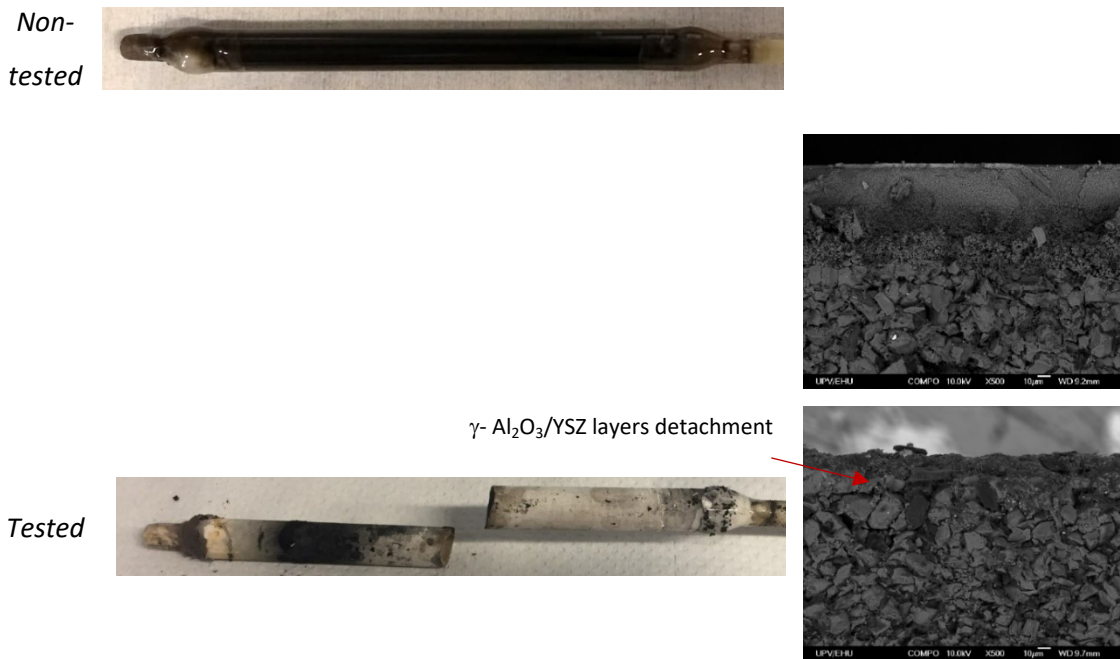


Figure S12. Images of reactor 100/1/1 (non-tested and tested) and SEM micrographs of the outer zone of the reactor