Supporting Information

Co-production of graphene sheets and hydrogen by decomposition of methane using cobalt based catalysts

Prabhas Jana,^a Víctor A. de la Peña O'Shea,^a Juan M. Coronado^a and David P. Serrano^{*ab}

^a Thermochemical Processes Unit, IMDEA Energy Institute, c/Tulipán s/n 28933, Móstoles, Madrid, Spain

^b Department of Chemical and Energy Technology, ESCET, Rey Juan Carlos University, c/Tulipán s/n, 28933 Móstoles, Madrid, Spain.

*Corresponding author: e-mail: david.serrano@imdea.org

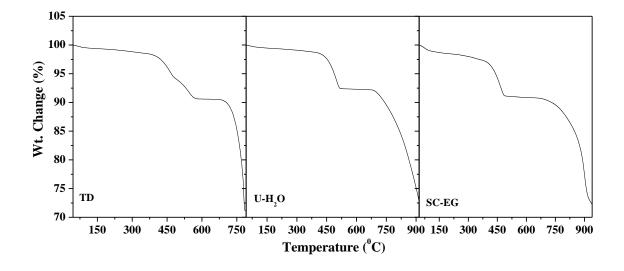


Fig. S1 Weight change of the catalysts during their reduction in presence of methane prior to the decomposition reaction.

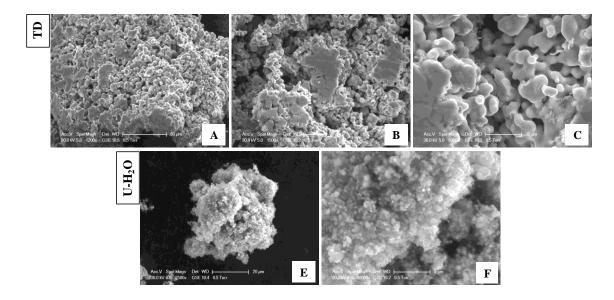


Fig. S2 SEM image of the used catalyst after the decomposition reaction with prereduction by methane.

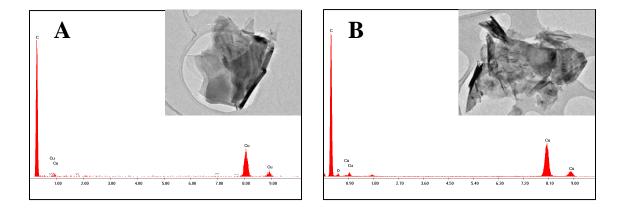


Fig. S3 EDX analyses of the TEM samples (inset) of used catalysts prepared with methane in the reduction step: A) U-H₂O and B) SC-EG (Cu peak is from the grid).

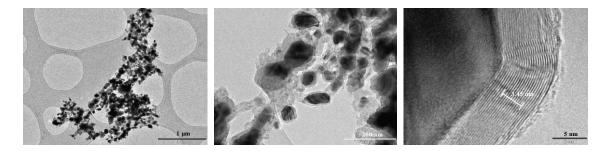


Fig. S4 HRTEM photograph of the used U- H_2O catalyst prepared using hydrogen in the reduction step.

Figure S4 presents the HRTEM image of the used U-H₂O catalyst after the decomposition reaction with pre-reduction with H₂. The catalyst shows only the formation of graphitic layers all over the catalysts which covering the cobalt metallic particles. The spacing between the neighboring sheets was measured to be 0.34 nm which is consistent with that between the (002) planes of graphite. Similar observation is also observed with the SC-EG catalyst with its pre-reduction with H₂. Therefore, the pre-treatment of the catalyst with H₂ does not facilitate the graphene formation over the catalysts.

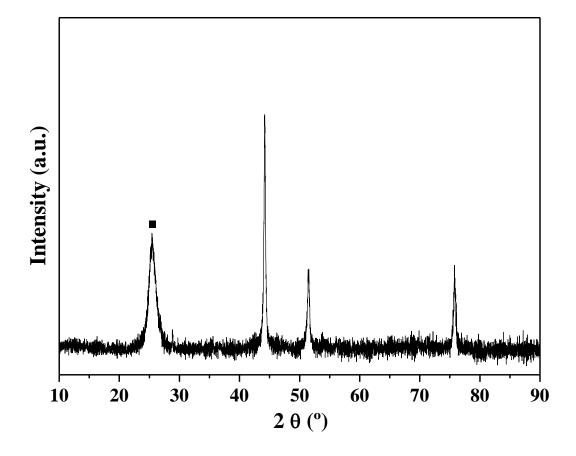


Fig. S5 XRD pattern of the used $U-H_2O$ catalyst prepared using hydrogen in the reduction step. (marked peak is due to the carbon formed in the reaction).

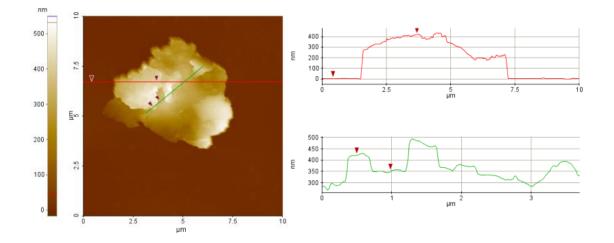


Fig. S6 AFM image of used U- H_2O catalyst, prepared using methane in the reduction step, along with the height histogram of the marked red and green lines.

Fig. S6 shows the topographic heights of the graphene layers over the used $U-H_2O$ catalysts after the decomposition reaction, obtained with in presence of CH_4 during the reduction treatment. The thickness of the graphene layer is about 300-400 nm. This value indicates a quite higher edge roughness for this catalyst compared to that of SC-EG, which is only 2.5-10 nm.