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### ACCEPTED MANUSCRIPT

# Graphene supported NiO/Ni nanoparticles as efficient photocatalyst for gas phase $CO_2$ reduction with hydrogen

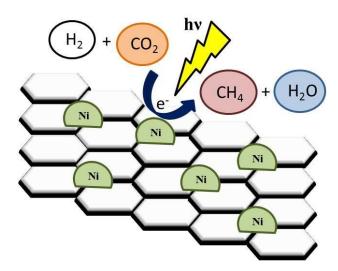
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Graphical abstract



Highlights

- Ni nanoparticles supported on graphene form spontaneously a thin layer of NIO
- NiO/Ni on graphene promotes photoassisted CO<sub>2</sub> methanation at 642 μmol/g·h at 200 °C
- The process can be carried out under continuous flow at rates 244.8  $\mu$ L · h<sup>-1</sup>
- The presence electron donors increases the rate of the photoassisted methanation
- Nitrobenzene quenches methanation, supporting charge separation

#### **Abstract**

The photocatalytic activity of NiO/Ni nanoparticles (NPs) supported on defective graphene (NiO/Ni-G) has been tested for the photoassisted  $CO_2$  reduction with  $H_2$ . NiO/Ni-G was prepared by  $H_2$  reduction of NiCl<sub>2</sub> adsorbed on few-layers defective G and storage under air. An optimal Ni loading of 23 wt% was found, reaching the maximum specific  $CH_4$  formation rate (642  $\mu$ mol  $CH_4 \cdot g_{Ni}^{-1} \cdot h^{-1}$  at 200 °C) and quantum yield of 1.98 %. Under the same conditions Ni NPs supported on silica-alumina or NiO NPs exhibit notably lower specific  $CH_4$  production rates than NiO/Ni-G. It was found that  $H_2O$  formed in the reaction has a detrimental influence on the photocatalytic activity and evidence supports that  $H_2O$ 

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