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Cover photo: Foam catalyst during reaction in plasma.
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Regeneration and repetitive use of deactivated catalysts in VOC/PAH removal and CO₂ conversion

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ABSTRACT

The packed-bed dielectric barrier discharges (DBDs) represent one of the most often used geometries that allows integration of nonthermal plasma (NTP) with a solid base heterogeneous solid catalyst to be used for a plasma catalysis. The packing material of a plasma discharge reactor, which is either the catalyst or catalyst support, usually takes a form of pellets, beads or granulates [1]. It has significant influence on discharge properties, mechanism of discharge formation and propagation as well as on processes ongoing on its surface, etc. [2,3]. This complex interaction of processes in NTP and on catalyst surface determines an overall chemical efficiency of plasma catalysis [4]. In this contribution, we present two distinct applications of packed-bed DBDs.

The first one is devoted to regeneration (i.e., reactivation) of deactivated catalysts that were previously used for plasma-catalytic removal of model volatile organic compound (toluene) or polycyclic aromatic hydrocarbon (naphthalene) in synthetic air as a carrier gas. The packed-bed DBD reactors had a cylindrical coaxial geometry and were packed with various catalytic materials (Pt/ γ -Al₂O₃, TiO₂, BaTiO₃). Decomposition of these model compounds resulted in formation of various gaseous and solid compounds/products often found as carbon containing deposits on surface of catalysts. These deposits were responsible for deactivation of catalysts and decrease of their activity over time.

The catalyst regeneration was performed in the packed-bed DBD reactors of the same geometry by NTP in oxygen as a carrier gas at ambient (25°C) and elevated temperature (100°C) for 2 hours. The gaseous products (CO₂, CO and HCOOH) of oxidative regeneration and temporal evolution of their concentrations was monitored by FTIR. The concentrations rapidly increased, reached a maxima and then gradually decreased, while concentration of O₃ generated by the discharge gradually increased. The temporal evolutions of gaseous products in Pt/ γ -Al₂O₃ and TiO₂ showed similar trends and high concentrations compared to BaTiO₃. Surface of the catalysts was analysed by SEM and optical microscopy and proved oxidation and partial removal of carbon deposits. In general, the results showed that efficiency of the catalyst regeneration depends on the catalytic material that determines its interaction with the plasma as well as plasma discharge modes occurring in various packed-bed DBD reactors. It is either a surface discharge or localised filamentary discharges depending on the dielectric constant of the packing material.



Figure 1. Photographs of fresh, used, and regenerated catalysts (TiO₂).

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Subsequently, regenerated catalysts were reused and tested for the removal of toluene by NTP in three repetitive cycles. The results showed that regenerated catalysts exhibited higher efficiency than non-regenerated ones. This effect was quite evident for Pt/ γ -Al₂O₃, and less for TiO₂.

The second application focused on plasma-catalytic CO₂ conversion. The DBD reactors had the same geometry as those used in the first application. They were either empty or packed with various catalytic materials (TiO₂, MgO, BaTiO₃). CO₂ conversion was investigated in a gas mixture of N₂ + CO₂ with an initial CO₂ concentration of 2.5 vol.%. Production of CO, as the only product of CO₂ conversion under the investigated conditions, was analysed by FTIR. The electrical characteristics of the reactors (voltage and current waveforms, discharge power) were examined and CO₂ conversion efficiency (in %), energy efficiency (kWh/g) and energy cost (g/kWh, or mmol/kJ) were also evaluated as a function of discharge power. The results showed that a presence of the catalytic material in the reactors significantly affected discharge characteristics what was clearly reflected in the results of CO₂ conversion. The obtained conversion efficiencies in various reactors followed a sequence: BaTiO₃ > MgO > TiO₂ > plasma alone. Thus, plasma-catalytic CO₂ conversion was more efficient than plasma alone conversion for all three catalytic materials used. Both the maximum conversion efficiency (8.4 %) and the maximum energy cost (15.37 g/kWh) were obtained with BaTiO₃, although for different values of discharge power (4 W and 1 W, respectively).

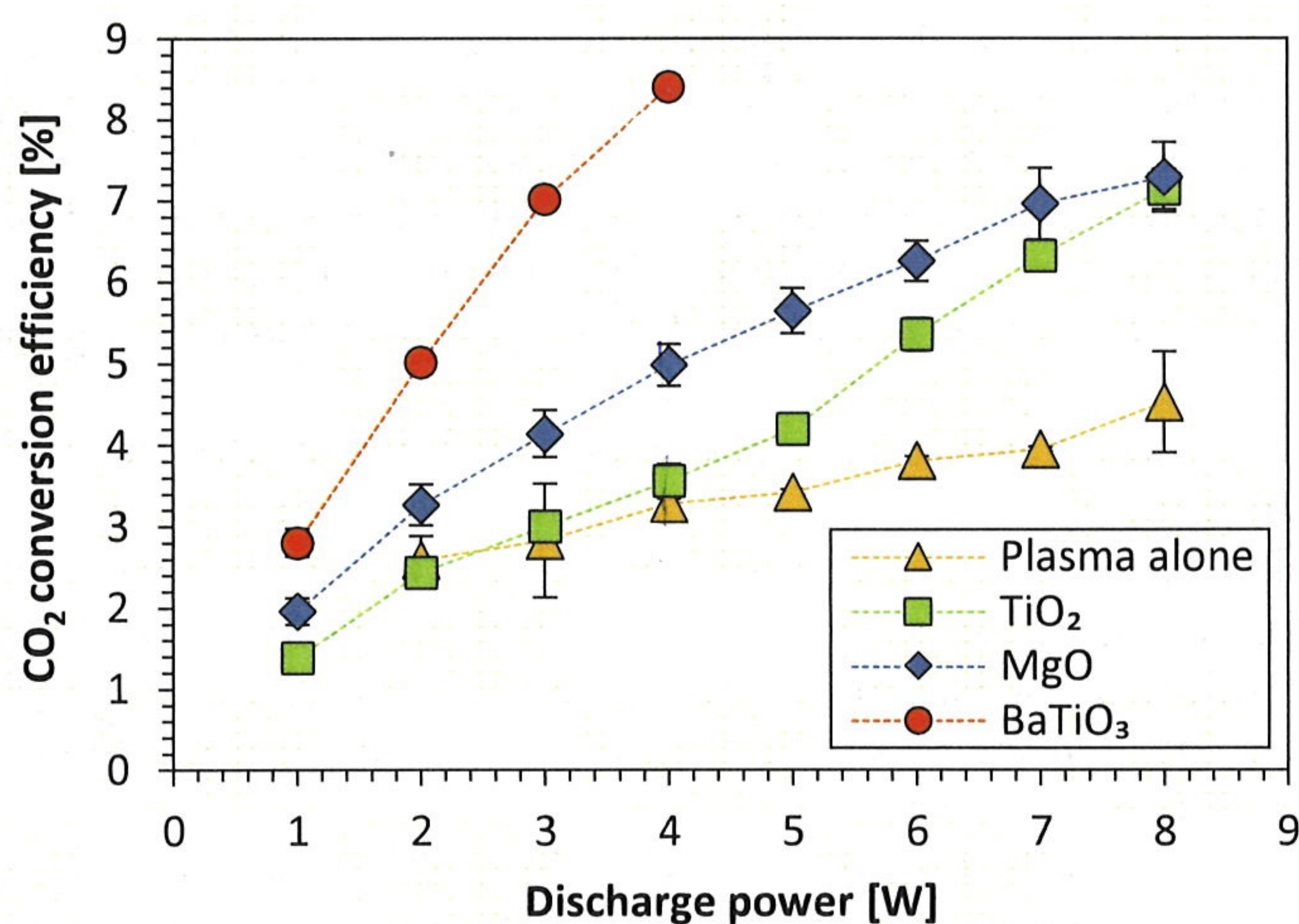


Figure 2. CO₂ conversion efficiency as a function of discharge power.

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