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## A Preliminary Study of a Multifunctional DOC/Wet-Scrubber Capable to Reduce both Chemical and Acoustic Emissions in Marine Field

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**Abstract:** The reduction of ship emissions (*i.e.*, Nitrogen Oxides – NO<sub>x</sub>, Sulfur Oxides – SO<sub>x</sub> and Particulate Matter PM) are of paramount importance particularly when shipping in Emission Control Areas (ECAs) where restrictive limits on emissions are imposed by the International Maritime Organization (IMO). In such ECAs, which starting from 2025 will also include Mediterranean Sea, the use of Ultra Low Sulfur Fuel Oil (ULSFO – sulfur content  $\leq 0.1$  wt%) or, alternatively, an emission-equivalent Exhaust Gas Cleaning System (EGCS) is mandatory. The price of ULSFO is about twice that of the ordinary Heavy Fuel Oil, which heavily affects marine transport economics. Consequently, several EGCS concepts have been studied for the abatement of NO<sub>x</sub>, SO<sub>x</sub> and PM, yet they have big dimensions which preclude their installation on already existing ships and, therefore, making necessary the re-design of the whole propulsion system. In the presented study, the use of a Diesel Oxidation Catalyst (DOC) in series with a closed loop scrubber to reduce NO<sub>x</sub>, SO<sub>x</sub>, PM and acoustic emissions is studied with the aim to design a compact exhaust line, with multifunctional performances, that allow to be compliant with the IMO's regulations. The role of the DOC is that of oxidizing the emitted NO to improve the solubility of the NO<sub>x</sub> species in the scrubber, besides hydrocarbons and PM abatement. It is shown that the oxidation activity and stability of the Pt-based catalysts can be significantly improved by doping with acidic oxides in comparison to a conventional Pt/Al<sub>2</sub>O<sub>3</sub> catalyst. This allows to design a compact EGCS which, in addition to the chemical aspects, incorporates modification aimed at minimizing acoustic emissions as well, acting as a silencer. Preliminary results assessing the validity of the integrate system on a full engine-EGSC mockup are reported in the paper.

**Keywords:** Diesel oxidation catalyst, Marine pollution control, Chemical emissions, Noise emissions

### Introduction

Maritime transport is of great importance for the global economy, as it accounts for around 80% of worldwide trade, together with its related activities (e.g., shipbuilding, repairs, ports activities) (Adam et al., 2021). Due to the extensive use of HFO (S~ 2.5 wt%), maritime transport has accounted for approximately 10-15% of global SO<sub>x</sub> and NO<sub>x</sub> emissions. This led to an increasing concern about the global impact of maritime emissions. IMO, consistently, has restricted the limits imposed by MARPOL 73/78 Annex VI Regulation (Revised MARPOL Annex VI, 2008) on ships emissions. The more restrictive limits are imposed on SO<sub>x</sub> and NO<sub>x</sub> emissions (Figure 1) for ECAs (Emission Control Areas), *i.e.* SECAs (Sulfur Emission Control Areas) and NECAs (Nitrogen Emission Control Areas) (Figure 2) making TIER III NO<sub>x</sub> limits effective since January 1<sup>st</sup>, 2016 for the ships build after that date (Revised MARPOL Annex VI, 2008).

Marine engines emit various pollutants into the air: HC, CO, NO<sub>x</sub>, SO<sub>x</sub> and PM. SO<sub>x</sub> emissions depend on the sulfur content of the fuel used, while NO<sub>x</sub> emissions depend, in addition to the nitrogen content in the fuel, also on the engine speed and power rating, resulting in their strong dependency on the cruising speed. Two ways to

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comply with international regulation are represented by the reduction of the ship cruising speed and/or using fuels with low sulfur and nitrogen content, e.g., VLSFO – Very Low Sulfur Fuel Oil ( $S \leq 0.5$  wt%) or, even ULFSO (Johnsen et al., 2019). However, such solutions heavily affect the shipping economics due to the cost of refined fuels and the slowdown of naval trade. The use of alternative fuels (e.g., LNG – Liquefied Natural Gas, Hydrogen) or alternative energy sources (e.g., electric propulsion) is compliant with the regulations but require the re-design and the refitting of the entire propulsion system, especially for the ships already in navigation, besides the higher running costs of these energy sources (Johnsen et al., 2019). Thus, the use of EGCS (Exhaust Gas Cleaning Systems) represents a viable solution to make ships eco-sustainable and economically efficient.

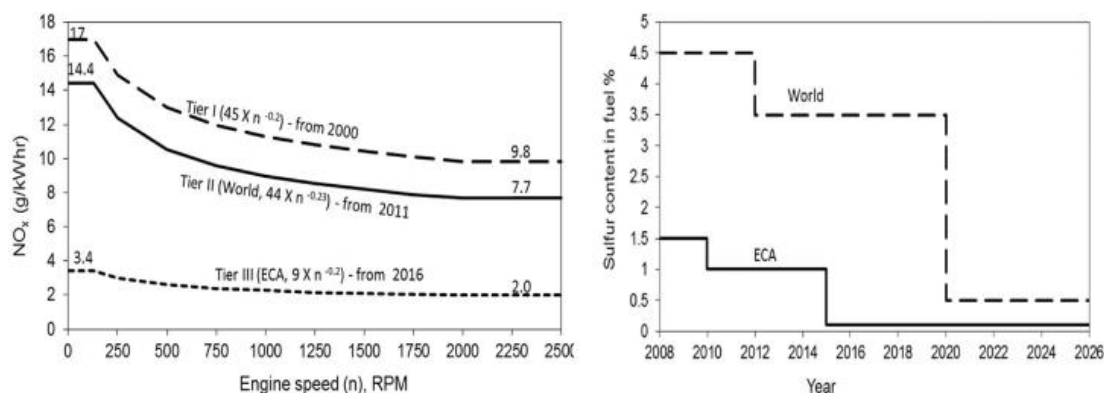


Figure 1. NO<sub>x</sub> and SO<sub>x</sub> limits.

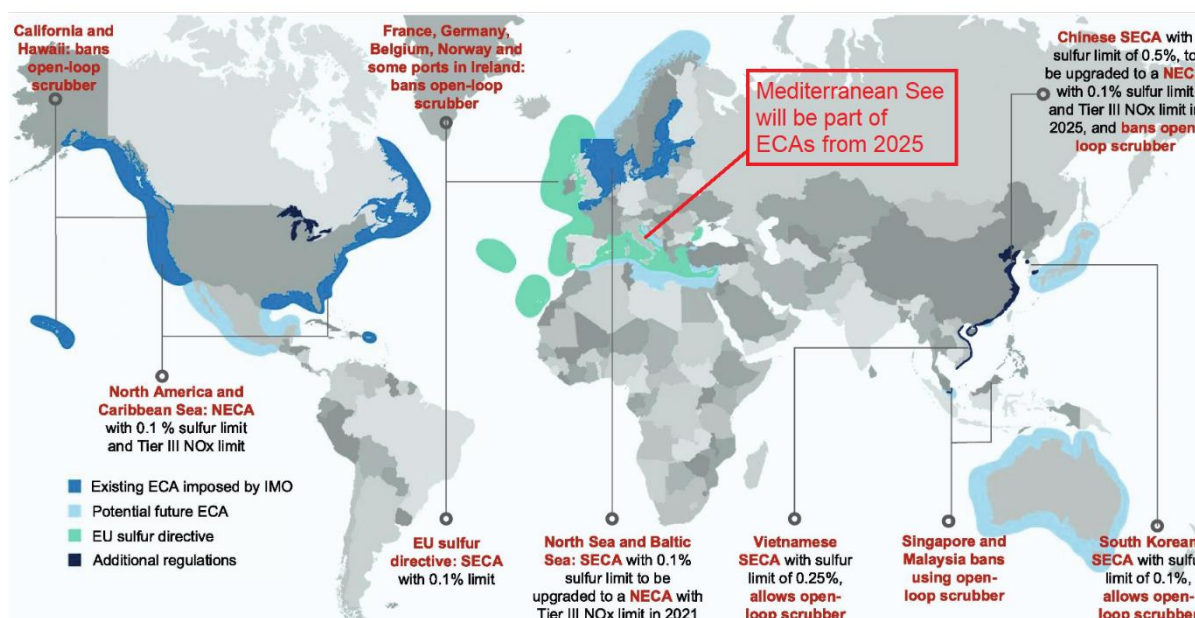


Figure 2. ECAs, adapted from (Zhao et al., 2021).

EGR (Exhaust Gas Recirculation) and SCR (Selective Catalytic Reduction) systems are effective technologies for the control of NO<sub>x</sub> emissions (Chen et al., 2015; Ushakov et al., 2020), while for SO<sub>x</sub> abatement the use of scrubber is proven to be efficient (Lindstad et al., 2017).

EGR recirculates a portion of the exhaust gas into the engine's combustion chamber, lowering its internal temperature. As a result, NO<sub>x</sub> emissions are reduced, however causing an increase in fuel consumption and engine efficiency characteristics (Huang et al., 2022; Lou et al., 2022). Therefore, it is preferred to limit the EGR ratio and use other techniques such as the SCR.

SCR systems, through the in situ production of ammonia from the decomposition of urea, allow the reduction of NO<sub>x</sub> to nitrogen (N<sub>2</sub>) (Zhu et al., 2022). This technique can be applied efficiently to a marine engine, but due to the high sulfur content of traditional fuel, poisoning of the active sites of the catalyst can occur, reducing the SCR efficiency and leading to a possible "slip" effect of the ammonia, which is a very polluting and toxic gas. Moreover, the use of SCR requires the storage of urea on board ships (Ayre et al., 2011).

The scrubbers abate SO<sub>x</sub> by absorption in a washing liquid; sea water is used as washing liquid in open loop scrubber, while in closed loop scrubber, water with chemicals additives is used (United States Environmental Protection Agency Office of Wastewater Management Washington, DC 20460, 2011; Lindstad et al., 2016). Scrubbers are able to abet also PM.

Ships emissions are not limited to chemical pollutant, but the noise radiated by the exhaust gas is also a dangerous emission for both the passengers and the environment. Several regulations limit the noise emission during the navigation and the mooring in port (Code on Noise Level On Board Ships - Res. A.468(XII), 2012; Procedure for the Determination of Airborne Noise Emissions from Marine Vessels, 2019). For the reduction of the noise emitted by the engine and radiated in to the surrounding of the ships through the exhaust gases silencers are used along the exhaust line (Bodn et al., 2007).

The current EGCS systems previously discussed represent effective solutions, but the simultaneous application on board ships of these systems is very complex and/or not economically feasible (Kyaw Oo D'Amore, Biot, et al., 2021). Based on these observations, this work aims at developing an innovative solution that allows the simultaneous abatement of both the emitted pollutants and noise, using an EGCS which consists of an oxidation catalyst followed by a scrubber. In this paper the attention is placed in particular on the converter containing a Pt-based DOC, which serves to oxidize NO to NO<sub>2</sub> to improve NO<sub>x</sub> solubility in the scrubber, further the silencing effect of the DOC converter is addressed. Figure 3 illustrates this abatement strategy which has the advantage of being very compact, allowing, in principle, the elimination of SCR, EGR and silencer.

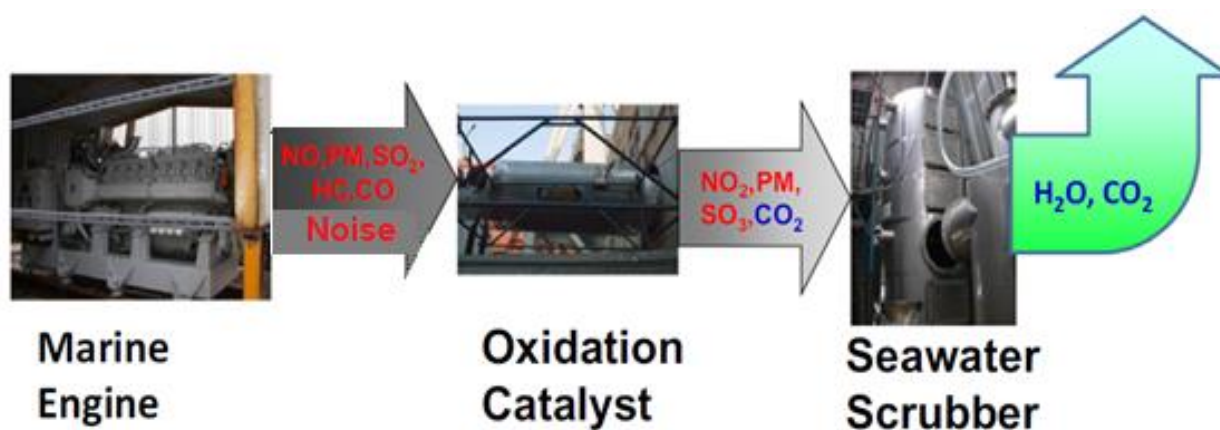
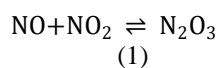


Figure 3. Abatement strategy. adapted from (Boscarato et al., 2015).

Considering that SO<sub>x</sub> and NO<sub>x</sub> emitted by the engine are mostly in the form of SO<sub>2</sub> and NO, due to the conditions in which combustion takes place in the engine and which do not allow for complete oxidation (EGCSA group, 2012), the ideal DOC converter should have the following characteristics:

- Able to achieve in operating conditions an average conversion of NO to NO<sub>2</sub> of about 50% since in the wet scrubber NO/NO<sub>2</sub> mixtures are further absorbed as N<sub>2</sub>O<sub>3</sub> according to the following equation (Thomas et al., 2000):



- To present a low oxidation activity toward SO<sub>2</sub> since its oxidation produces SO<sub>3</sub>, which is responsible for acid mist in the wet scrubber, is very corrosive and can damage pipes and machinery;
- To feature a high resistance to poisoning by sulfur, i.e., it must have a low affinity to adsorb sulfites/sulfates that can cover the active sites, reducing the activity of the catalyst;
- Present high acoustic performance in terms of Transmission Loss (TL) to reduce the noise radiated by the engine.

In this paper, first a study on the effect of different doping additives to increase the resistance to sulfur poisoning of the DOC is addressed. Then, a real DOC is designed and tested on the mock-up of a marine Genset to evaluate its efficiency in NO conversion. Finally, numerical simulations are performed to evaluate the



acoustic performance of the DOC and find a geometry that increase its TL, while keeping unaffected its chemical efficiency.

## Methods

### Experimental Measurements to Evaluate the Efficiency of NO Oxidation

The laboratory experimental setup used to test the catalysts with different doping is reported in Figure 4).

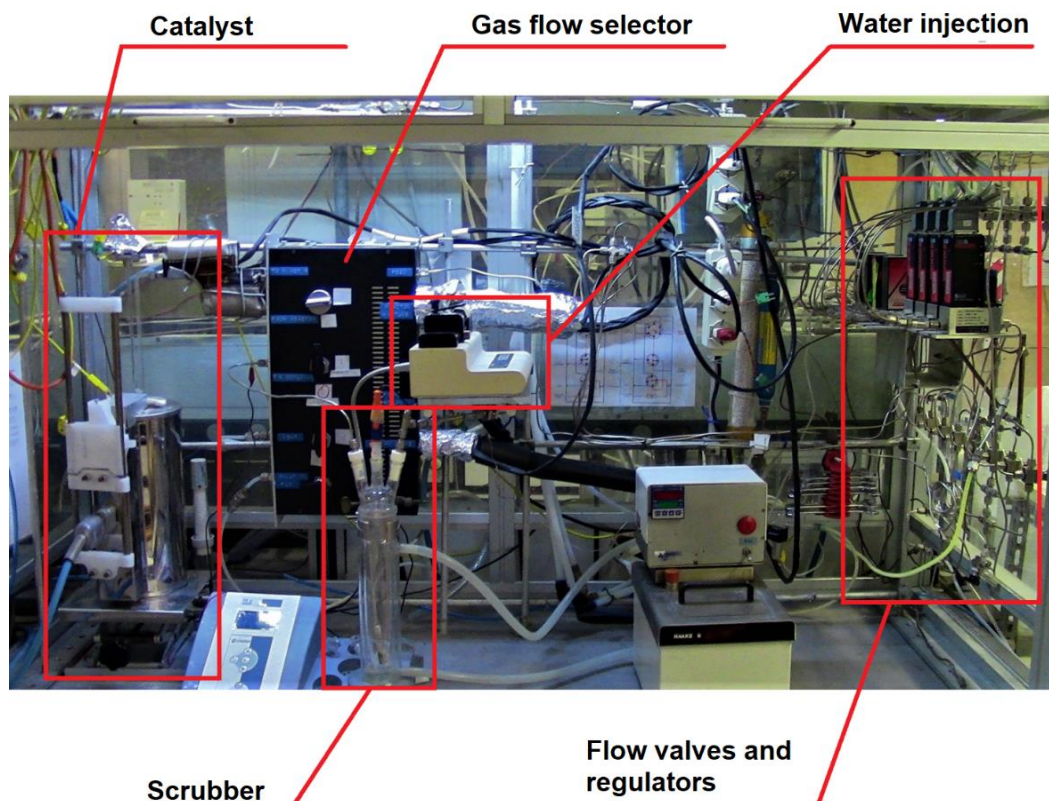


Figure 4. Laboratory experimental set-up.

The catalysts are loaded in a U-tube flow microreactor, which is connected to a gas flow selector that allows to exclude or include a saturator at the outlet of the reactor which simulates a scrubber. The inlet and outlet gas mixtures are analyzed using an on-line FTIR MKS 2000 instrument, which is provided with calibration curves for all the reactants of interest, i.e., NO, NO<sub>2</sub>, CO<sub>2</sub>, CO, SO<sub>2</sub>, N<sub>2</sub>O, H<sub>2</sub>O, except N<sub>2</sub> and O<sub>2</sub>, these species being IR inactive, whereas SO<sub>3</sub> is absorbed in the transfer line in order to avoid damage on the instrument. Therefore, the conversion of SO<sub>2</sub> is calculated as SO<sub>2</sub> disappearance whereas NO conversion is calculated using the formulae:

$$\text{SO}_2 \text{ conversion (\%)} = \left(1 - \frac{[\text{SO}_2]_{\text{reactor out}}}{[\text{SO}_2]_{\text{reactor int}}}\right) \cdot 100 \quad (2)$$

$$\text{NO conversion (\%)} = \frac{[\text{NO}_2]}{[\text{NO}] + [\text{NO}_2]} \cdot 100 \quad (3)$$

A simulated exhaust is employed with the following composition is employed (mol%): SO<sub>2</sub> 570 ppm, NO 900 ppm, O<sub>2</sub> 9%, CO<sub>2</sub> 6,6%, H<sub>2</sub>O 6%, total flowrate 100 ml min<sup>-1</sup>, using a Gas Hourly Space Velocity (GHSV) of 100,000 h<sup>-1</sup>. M<sub>x</sub>Zr<sub>1-x</sub>O<sub>2±m</sub> mixed oxide, where M is a transition metal, were synthesised by MEL Chemicals using proprietary synthesis methods aimed at incorporation of the dopant within the ZrO<sub>2</sub> structural framework. The dopant levels varied between 8 and 18 mol%.

Once the most promising catalyst composition has been identified, i.e., the one which has the greatest resistance to sulfur poisoning, a full-scale DOC is designed and tested on the exhaust line of a diesel generator mockup (Figure ).



Figure 5. Diesel Genset mockup, DOC converter rounded in red.

### Numerical Simulations to Evaluate and Optimize the Acoustic Properties of the DOC

To calculate the TL of the DOC a combined approach that uses CFD and acoustic FEM simulation is used. This methodology allows to consider the influence of the flow on the acoustic properties while keeping the computational effort reduced. The velocity, pressure and temperature fields are calculated with CFD simulations and are then imported in the FEM solver through mesh mapping to estimate the TL (Kyaw Oo D'Amore, Mauro, et al., 2022).

Settings used for CFD and FEM simulations and the characteristics of the mesh used to discretise the geometry are chosen on the basis of the results obtained in previous study (Kyaw Oo D'Amore & Mauro, 2021; Kyaw Oo D'Amore, Morgut, et al., 2022).

Having calculated the TL of the reference DOC, geometrical modifications are performed to increase the efficiency of the DOC in noise reduction. The flow resistivity and the OAR (Open Area Ratio) of the monoliths inside the DOC converter, which represent the catalytical elements, are not modified to ensure the chemical efficiency. The external steel casing geometry of the converter is changed and perforated elements are inserted to increase the TL of the DOC, also evaluating the distribution of the flow entering the monoliths (it should be as homogeneous as possible) and the generated back pressure (it must not be greater than the limit value admitted by the engine in order not to lower its efficiency) using the CFD technique.

## Results and Discussion

### Efficiency in NO Oxidation

Platinum catalysts represent the state of the art DOC used for PM abatement (Zhang et al., 2022), in addition their feature excellent activity for NO oxidation, with relatively good tolerance to SO<sub>x</sub> and resistance at high temperatures, present in diesel exhaust gases (Hong et al., 2017).

The activity of Pt catalysts depends on the type of support on which the active metal is dispersed. The most common support is  $\gamma$ -alumina (Pt/ $\gamma$ -Al<sub>2</sub>O<sub>3</sub>). The Eley-Rideal mechanism is generally accepted for NO oxidation which can occur through two paths (Figure 5): the NO can react with the oxygen adsorbed (O) on the active metal, directly giving NO<sub>2</sub> in the gaseous phase or the reaction produces nitrates (NO<sub>3</sub><sup>-</sup>) which are adsorbed on the support and may decompose giving NO<sub>2</sub> or NO, depending on the operating conditions (Li et al., 2010). This

second mechanism clearly inspired research on the effects of the support in NO oxidation, either by changing the  $\text{Al}_2\text{O}_3$  support itself for *e.g.*,  $\text{CeO}_2$ ,  $\text{ZrO}_2$ ,  $\text{TiO}_2$ , etc., or by a surface doping of the  $\text{Al}_2\text{O}_3$  carrier with transition metal oxides (Hong et al., 2017). Among the supports,  $\text{TiO}_2$  and  $\text{ZrO}_2$  have been recognized to confer sulfur-resistance to the supported Pt particles in the oxidation of NO to  $\text{NO}_2$ . At variance with  $\text{TiO}_2$  which has relatively low thermal stability, it is well known that structural doping can confer high thermal stability to  $\text{ZrO}_2$ -containing mixed oxides (Di Monte et al., 2005); accordingly, the latter systems are chosen for this study.

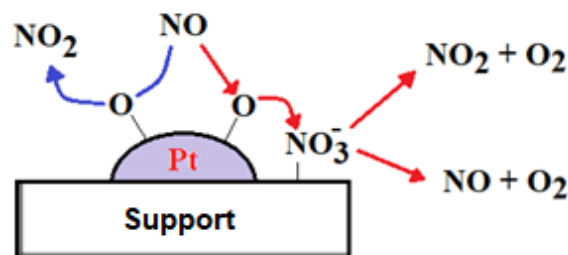


Figure 5. Eley-Rideal mechanism proposed for NO oxidation over Pt catalysts (Li et al., 2010).

Specifically,  $\text{Pt}/\text{M}_x\text{Zr}_{1-x}\text{O}_{2\pm m}$  catalysts with four different structural dopants were employed in the laboratory scale study with the aim to assess the following effects:

- Tungsten oxide ( $\text{WO}_3$ ), to investigate the effects of acidity of the reactivity;
- Cerium oxide ( $\text{CeO}_2$ ), to investigate the effects of high redox reactivity;
- Yttrium oxide ( $\text{Y}_2\text{O}_3$ ), which is a neutral element that acts as a thermal stabilizer;
- Lanthanum oxide ( $\text{La}_2\text{O}_3$ ), to investigate the effects of basicity of the reactivity.

As previously explained, the study on the influence of different doping on the conversion of both  $\text{SO}_2$  and NO is first addressed to highlight the composition with higher resistance to sulfur poisoning. To evaluate the conversion as a function of time and temperature a typical analysis cycle that includes two ramps of heating and subsequent cooling from 100 °C to 500 °C was performed. Figure 6 reports the conversion vs. temperature curves for the Pt/doped- $\text{ZrO}_2$  samples.

Lanthanum, cerium and yttrium doped Pt/ $\text{ZrO}_2$  samples feature similar behavior, the conversion of NO does not have a high efficiency and reaches its maximum around 425 °C (about 20%); at lower temperatures the reaction is kinetically controlled, while at higher temperatures it is thermodynamically disadvantaged. A perusal of the  $\text{SO}_2$  conversion curves reveals that the heat-up curves present lower conversion compared to run-down part of the cycle at comparable temperatures. This hysteresis is associated to the surface adsorption of  $\text{NO}_x$  and  $\text{SO}_x$ -derived species, mainly on the support, which actively participates to the reaction (Figure 6): during the heating ramp adsorption mainly occurs, then, at the maximum temperature reached by the cycle (500 °C) reasonably desorption of most of the compounds present on the catalyst occurs, leaving the surface clean during the ramp-down part of the cycle (Boscarato et al., 2015). However, the catalyst is not fully regenerated as the second heating curves is lower than the first one. Noticeably, when these experiments are performed in the absence of  $\text{SO}_2$ , the hysteresis phenomenon is still present and the NO conversion is higher, in line with the poisoning effect of  $\text{SO}_2$  (data not reported).

Tungsten is the doping that leads to the best results: the NO conversion reaches 30% at 375 °C. This is attributed to the effect of tungsten which reduces the affinity of  $\text{SO}_x$ -derived species with the active sites, which remain free for NO conversion.

Based on these results, a Pt/ $\gamma$ - $\text{Al}_2\text{O}_3$  catalyst supported with 20% of  $\text{WO}_3$  was prepared and wascoated on two metallic honeycombs there were inserted in a stainless steel casing to obtain the converter described in Figure 7. and preliminarily tested using above described mock-up with the exhaust line and the diesel Genset operating at a GHSV of 100.000  $\text{h}^{-1}$ .  $\gamma$ - $\text{Al}_2\text{O}_3$  is chosen as support instead of  $\text{ZrO}_2$  due to its high surface area useful to achieve a high Pt dispersion that could increase the conversion of NO in a real case scenario. Preliminary results showed a NO conversion of 14% at a temperature of about 330°C, which is well-in-line with the results obtained in the model laboratory system, thus confirming the validity of the present approach. Further measurements are on-going and will be reported in a future.

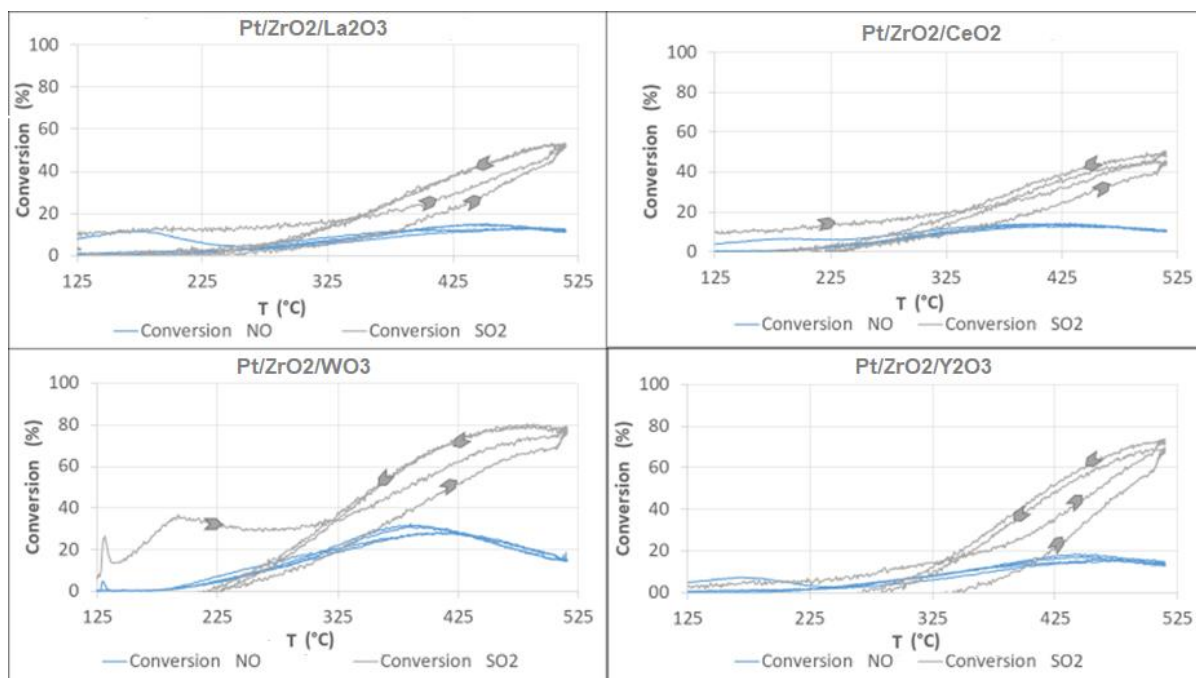


Figure 6. Comparison of conversion curves (NO to NO<sub>2</sub> and SO<sub>2</sub> to SO<sub>3</sub>) as a function of temperature over Pt/doped-ZrO<sub>2</sub> catalysts.

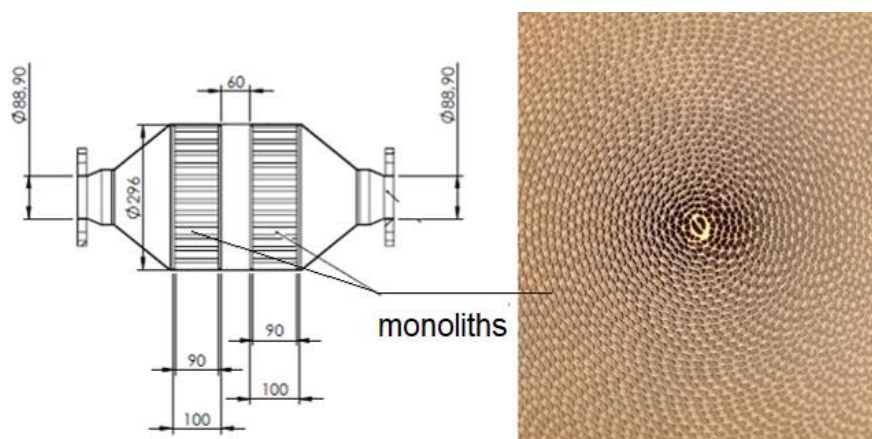


Figure 7. Scheme of the DOC converter and detail of the honeycomb structure.

### Evaluation and Optimization of the Acoustic Properties of the DOC

The TL of the reference DOC (Figure 7) is evaluated with the combined CFD-FEM methodology considering a flow with characteristics as those measured on the diesel Genset mockup, i.e., a linear flow velocity of 42.5 m/s at a temperature of 270 °C.

Figure 8 compares the TL curve calculated using the actual converted geometry (Figure 8) with the TL of a modelled DOC converter where modifications of the geometry are applied in order to improve the silencing effect of the converter, without affecting the chemicals processes occurring in the two honeycombs. Figure 9 compares the two geometrical configurations of the analyzed DOCs, whereas their main dimensions are reported in Table 1.

Remarkably, modest modifications of the converter geometry allow to increase the TL up to 10 dB in the frequency range 0-200 Hz and up to 20 Hz in the frequency range 350-650 Hz. In the frequency range 200-350 Hz a decrease of the TL curve up to 6 Hz is highlighted; for a real application this dip should not correspond with the engine frequency to maximize the overall noise reduction. Accordingly, further studied should be performed to find a configuration which allow the optimization of the TL in the whole frequency range.



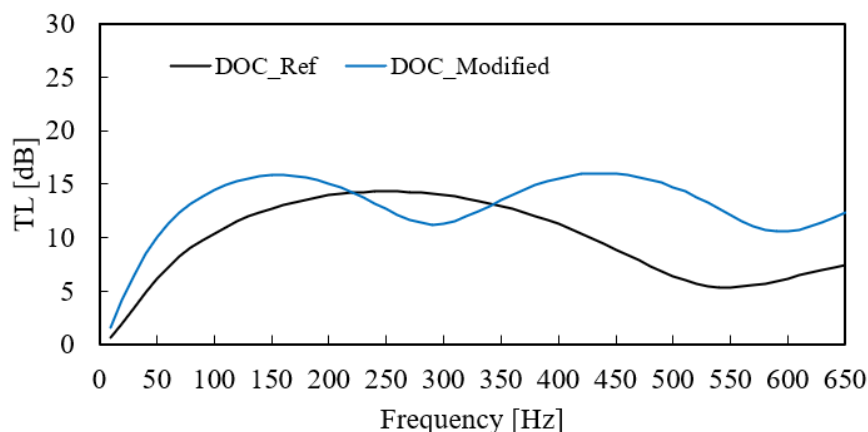


Figure 8. TL comparison, reference DOC vs modified one.

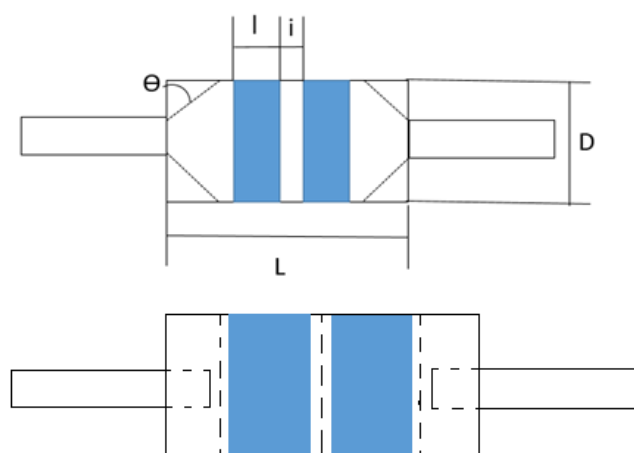


Figure 9. Comparison between geometry of reference DOC and modified one.

Table 1. Reference DOC geometry parameters vs modified DOC.

DOC	D	$\Theta$	L	l	i	$n_p$	R	OAR	$L_f$
Ref	297	40	510	90	60	0	678	0.92	0
Mod	297	0	765	180	90	3	678	0.92	127.5

## Conclusions

In the present work, an interdisciplinary approach to the development of an advanced multifunctional EGCS is presented. The use of a DOC for NO<sub>x</sub> abatement is proposed, replacing traditional systems on board ships such as SCR and EGR. The DOC is designed to also have noise reduction properties, aiming to also eliminate the silencer along the exhaust line of marine diesel engines, thanks to the systems integration. The wet scrubber present downstream of the DOC, can be used also for the reduction of SO<sub>x</sub>, always with a view of system integration to obtain a compact exhaust line that allow to save space on board while be compliant with the international regulations. The preliminary study presented in this paper focused on the possible doping to increase the resistance of the catalyst to sulfure poisoning and to the oxidation of SO<sub>2</sub>. It has been seen that tungsten is the most promising doping in this sense. A good correlation between laboratory-scale measurements and DOC on a diesel Genset mockup has been highlighted and good results are obtained in increasing the acoustic properties of the DOC. Other studies are ongoing to further increase the efficiency of NO<sub>x</sub> oxidation within the DOC.

## Scientific Ethics Declaration



The authors declare that the scientific ethical and legal responsibility of this article published in EPSTEM journal belongs to the authors.

## Acknowledgements or Notes

\* This article was presented as oral presentation at the International Conference on Basic Sciences, Engineering and Technology ([www.icbaset.net](http://www.icbaset.net)) held in Marmaris/Turkey on April 27-30, 2023.

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