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NO_x reduction based on N₂ dilution in a swirled-stabilized magnesium flame

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Abstract

New sources of clean energy with reduced greenhouse gas emissions must be considered because of global warming. Among these alternative sources, metal fuels such as aluminum or magnesium are promising. The present study focuses on the impacts of an air dilution by N₂ and of the equivalence ratio on the amount of nitrogen oxides (NO_x) produced by a swirled-stabilized magnesium flame. A polydispersed aerosol of pure magnesium in the size fraction 50-70 μm was injected in the combustion chamber. Different air-fuel ratios – from 0.14 to 1.0 – were obtained by changing the mass flowrate of the injected fuel. Thermocouples and a pyrometer were implemented to monitor the gas and flame temperatures. On-line paramagnetic analyzers were used to measure the oxygen (O₂) and NO_x concentrations. For each dilution condition (0, 30 and 50%), the NO_x mole fraction versus the equivalence ratio follows a parabolic shape. The amount of NO_x first increases when increasing the air-fuel ratio as the power dissipated by the flame increases, leading to the rise of the gas temperature. For the highest air-fuel ratios, the NO_x production is disfavored in the rich mixture. The maximal NO_x emissions are shifted to higher equivalent ratios when increasing the dilution level and it decreases by a factor 2 with a dilution of 50%, for a constant value of the air-fuel ratio. The gas temperature in the flame decreases due to the N₂ dilution. The NO_x emissions become negligible for an equivalence ratio of 1 at a 50% dilution. Nevertheless, under the tested experimental conditions, the level of NO_x never exceeded 7 gNO_x/kWh, which is lower than the maximal value of NO_x emissions from a gasoline engine.

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27

28 **Keywords.** Magnesium combustion; NO_x emissions; N₂ dilution; equivalence ratio

29

30 **1. Introduction**

31 The combustion of metal elements can be considered to produce a clean, renewable and carbon-free
32 energy. Metal powders such as magnesium or aluminum are indeed an energy source for the future, as
33 energetic as fossil resources and without CO₂ emissions [1,2]. In addition, the metal oxide particles
34 which are produced during the combustion are in solid form, whence they are fully recoverable at the
35 outlet of the system. They can be regenerated using, for instance, a solar oven [3,4]. Consequently, a
36 circular economy can be realized considering that the metal oxide produced can be collected and then
37 reduced using clean primary energy/electricity.

38 During the last decade, metal flames were obtained by Lomba et al. [5] and by Julien et al. [6]. Then, a
39 confined turbulent magnesium burner with swirl stabilization was designed by Garra et al. [7] and Laraqui
40 et al. [8]. Metal fuels consisting of micron-sized iron particles are also promising alternative fuels [9].

41 During metal combustion in air, NO_x are being produced. NO_x species include nitrogen monoxide NO,
42 nitrogen dioxide NO₂, nitrous oxide N₂O, nitrogen tetraoxide N₂O₄, nitrogen trioxide N₂O₃, nitrous acid
43 HNO₂, and nitric acid HNO₃ [10–12]. To our knowledge, the first studies concerning NO_x emissions
44 generated during metal combustion were [7], [8] and [13]. The objective of the present study is therefore
45 to reduce the knowledge gap concerning NO_x production from the magnesium combustion and the
46 impact of a dilution of the inlet air with nitrogen on these NO_x emissions. There are three possible paths
47 for nitrogen oxides formation: prompt NO, thermal NO_x and fuel NO_x [11,14]. For metal combustion,
48 only thermal NO_x are formed, through the combination of nitrogen from air with oxygen from air at high
49 temperatures [13]. The Zeldovich mechanism is usually used to describe the formation of thermal NO_x
50 [15]. This reaction highly depends on temperature, oxygen content and residence time of the fuel in the
51 flame. Nitrogen oxides cause acid rain which has a corrosive effect on materials as well as on the forest
52 ecosystem and fresh water [16,17]. Moreover, NO_x migrate into the troposphere and react with UV
53 radiation, contributing to the formation of ground-level ozone [12,18]. N₂O causes an increase in
54 greenhouse gases. In addition to these environmental impacts, nitrogen oxides have a negative effect
55 on human health by impairing respiratory function. Nitric oxide enters the alveoli of the lungs and
56 dissolves in the bloodstream, resulting in a decrease of the amount of oxygen carried by hemoglobin,

57 causing organs to suffer from a lack of oxygenation [10,11]. Numerous standards were introduced for
58 the various sources of NO_x emissions. For instance, Diesel vehicles must not emit more than 80
59 mgNO_x/km driven [19]. French regulations established an upper limit of 40 µg/m³ as an annual average
60 not to be exceeded, with a target of a 63% reduction in NO_x emissions at European level by 2030
61 compared to 2005 [20]. Two techniques can be implemented to contain the nitrogen oxides emissions
62 from combustion systems. Primary techniques limit the formation of NO_x in the combustion system.
63 Secondary techniques consist of reducing the NO_x formed with gas exhaust aftertreatment systems.
64 Primary techniques allow lowering the combustion temperature and/or reducing the amount of injected
65 oxygen to limit the rate of NO_x production [11,14,21]. The most common of these techniques is fuel-
66 staged combustion [14]. Its principle is to delay the injection of part of the fuel. Only a fraction of the fuel
67 is injected at the entrance of the flame zone. This creates a fuel-poor zone in the flame where the
68 temperature is consequently low. The remaining fuel is injected further down the flame. Thus, a second
69 combustion zone is generated, which is fuel rich, and this limits the NO_x formation. Combustion can
70 also be staged by air injection. Indeed, when insufficient air flow is injected into the burner zone,
71 combustion is therefore incomplete there. Additional air is then introduced downstream in the flame. The
72 temperature is lower in the area where there is oxygen excess, which limits the NO_x formation. Recycling
73 the flue gases and diluting them in the flame can also lower the flame temperature. This recycling can
74 be external or internal. External recycling or exhaust gas recirculation (EGR) consists to return the flue
75 gases from the combustion system upstream of the burner where they are mixed with the primary air.
76 This technique can reduce NO_x formation by up to 60%. Internal recycling consists to circulate the flue
77 gases near the burner [22,23]. Another technique consists to separate nitrogen from oxygen upstream
78 of the combustion system. This oxy-combustion technique is currently used for stationary combustion
79 sources, but it is not widely used for mobile sources as it is very costly. NO_x reduction by gas re-burning
80 is a secondary method which allows reducing nitrogen oxides with hydrocarbons in a slightly oxidizing
81 environment [24]. The NO_x formed in a first air-rich zone reacts in a second zone with hydrocarbons in
82 an oxygen-poor atmosphere. The unburnt fuel then reacts with the NO_x to reduce it. The unburned fuel
83 is fully oxidized in a third zone where excess air is added. The efficiency of this pollution control system
84 can be as high as 50%. Air staging and re-burning for NO_x reduction appear to be the most suitable for
85 metal combustion. These techniques enable the formation of NO_x to be limited by intervening in the
86 injection of air and fuel. They are to be favored in the first stage. Beside the primary techniques, the

87 secondary techniques allow reducing nitrogen oxides by using a reducing agent [25]. Secondary
88 techniques should only be used when primary techniques do not provide sufficiently conclusive results.
89 The only gaseous pollutants resulting from metal combustion are nitrogen oxides ($\text{NO}_x = \text{NO} + \text{NO}_2$) [8].
90 Additionally, N_2O could also be produced. During the combustion of magnesium, the 2000 °C hot flame
91 generates high levels of thermal NO_x emissions [11]. These NO_x levels must therefore be drastically
92 reduced. The rates of NO_x production depend on several factors, such as the temperature of the flame
93 but also the oxygen availability in the flame and consequently the equivalence ratio. In the previous work
94 [13], the NO_x emissions from metal combustion were investigated using a turbulent magnesium burner
95 where a swirl-stabilized flame was obtained in a combustion chamber. It was observed that the NO_x
96 emissions depend on the equivalence ratio and on the swirl. The NO_x yielding exhibited a bell-shape
97 curve versus the equivalence ratio, whatever the swirl. The NO_x emissions never exceeded 7 $\text{g}_{\text{NO}_x}/\text{kWh}$
98 and it was expected to reduce them.

99 The present study aims at investigating the reduction of NO_x emissions occurring during magnesium
100 combustion in a pilot burner. Various techniques may be used to reduce the NO_x emissions. They
101 include two types of systems: pre-treatment and post-treatment such as Selective Catalytic Reduction
102 (SCR) [26]. The objective of the present study is to reduce the NO_x levels using a pre-treatment strategy
103 which minimizes the NO_x formation in the combustion system. For such a strategy, staged combustion
104 is a very promising method which consist to delay the injection of a part of the fuel or of the oxidant in
105 the combustion chamber, to separate the zone where the temperature is very high from the zone where
106 the oxygen concentration is high [14,27]. Another method is the Exhaust Gas Recirculation (EGR), which
107 consists to reinject part of the gases and fumes at the engine outlet in the burner where they are mixed
108 with fresh air [22,23,28,29]. Considering the global mechanism proposed in [13] for NO_x production in
109 a magnesium flame, a relevant strategy for NO_x abatement could be to reduce the flame temperature
110 and the injected oxygen concentration. In the present study, the selected method simulates the EGR for
111 NO_x abatement. The confined combustion chamber with a stabilization by swirl is adapted for the
112 characterization of NO_x emissions and temperatures [8,13]. Nitrogen is mixed with air and injected
113 upstream of the combustion chamber to simulate a dilution with the exhaust gases. For the dilution of
114 hydrocarbons in a gasoline engine by recirculating gases or pure nitrogen, the dilution ratio cannot
115 exceed 30%, because the dilution causes a significant unburned hydrocarbon rate and it is therefore
116 necessary to find a compromise between the emissions of NO_x and particles [22]. In the case of metal

117 combustion, such an issue does not appear and it is then possible to reach dilution ratios up to 50%.
 118 This study is therefore focused on the effect of the N_2 dilution on NO_x emissions, as well as on the gas
 119 temperature in the combustion chamber, with dilution ratios of 0, 30 or 50%.

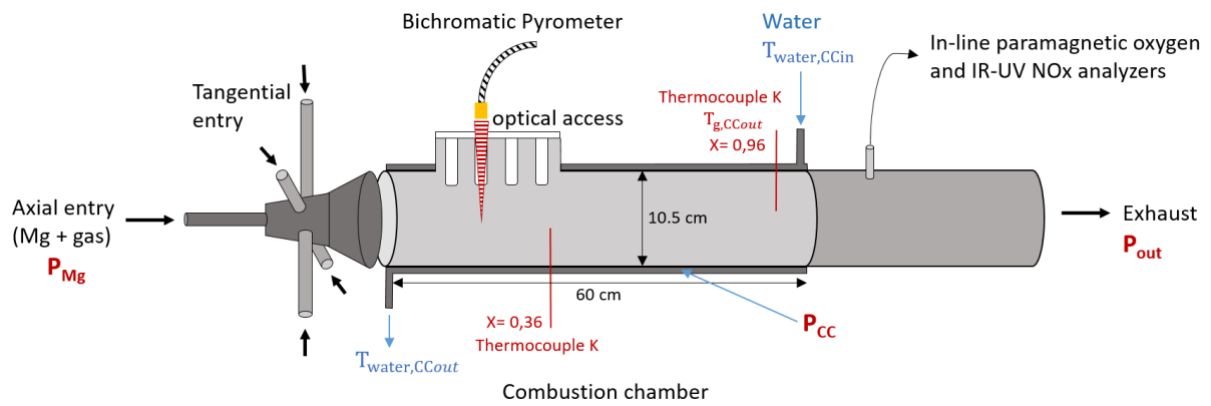
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121 2. Experimental approach

122 2.1. Metal-air burner

123 The metal-air burner used was schematically described in [30]. Figure 1 gives a schematic
 124 representation of the pilot system.

125



126
 127

Fig. 1. Scheme of the pilot system.

128 The pilot system is composed of three main parts: an air and magnesium injection, a combustion
 129 chamber with an optical access for flame observation and an outflow duct for gas sampling and analysis.
 130 The combustion chamber is 60 cm long for an internal diameter of 10.5 cm. It consists in a cooled
 131 double-walled stainless-steel pipe. Cooling-water flows between the two concentric walls of the
 132 combustion chamber in the opposite direction of the gas flow for heat recovery purposes. The chamber
 133 opens onto a long outlet pipe, called the free exhaust, to avoid any pressure drop in the combustion
 134 chamber.
 135

136 The fuel used is a magnesium powder with a purity greater than 99.8% and supplied by Carl Roth as
 137 CP20.2 reference. The particle size is characterized with a sifter RETSCH and it ranges from 50 to
 138 $70 \mu m$. The generated aerosol of magnesium powder is injected into the chamber. Solid fuel injection
 139 was conducted with a PALAS BEG 1000 type B. This system allows a continuous and homogeneous
 140 injection of particles.

141 The reconstituted air and nitrogen flows are controlled before entering the chamber by solenoid valves
142 and flow rates regulated by mass flowmeters. The injected airflow rate ranges between 2.3 and
143 8.6 Nm³.h⁻¹. Nitrogen is additionally injected (between 0 and 4.3 Nm³.h⁻¹), according to a given dilution
144 rate to mimic exhaust gas recirculation with a dilution of the air by nitrogen.

145 The flame is stabilized with a swirl system where a secondary air is injected by four tangential inlets in
146 the injection head. The swirl is characterized by its geometric swirl number S_g as already presented in
147 [13]. The actual swirl number is approximately $0,24 \times S_g$. Only the ratio between axial mass flow rate
148 and tangential mass flow rate affects the swirl number. In the present study, only one swirl condition is
149 considered with $S_g = 7.3$.

150 The equivalence ratio is modified by increasing the flow of the magnesium injected at constant air supply
151 injection. Equivalence ratio varies from 0.14 to 1.0. For such a range of equivalence ratio and depending
152 on the dilution, the power supplied by the combustion varies between 6 and 23 kW.

153

154 **2.2. Gas analyses and temperature measurements**

155 A sample of the outlet gas flow is collected in the combustion chamber using a tube with three bored
156 holes positioned perpendicularly to the flow. The holes are positioned back to the tube to avoid the holes
157 plugging by the particles of magnesium oxide. The NO and NO₂ emissions are monitored using infrared
158 and ultraviolet cells of X Stream Rosemount analysers. N₂O is measured using a Cascade analyser
159 (Emerson Rosemount ct5400 continuous gas analyser). NO_x are measured in ppm and are also
160 expressed in g.kWh⁻¹ relating to the power dissipated by the combustion for comparison with NO_x
161 emission from gasoline engines.

162 The oxygen concentration at the outlet of the system is monitored using a paramagnetic X Stream
163 Rosemount O₂ analyser, and expressed in %Vol.

164 The equivalence ratio can also be determined experimentally from:

$$165 \quad \varphi = \frac{\%O_{2,i} - \%O_{2,f} \times \frac{100 - \%O_{2,i}}{100 - \%O_{2,f}}}{\%O_{2,i}}$$

166 where $\%O_{2,i}$ is the inlet oxygen mole fraction and $\%O_{2,f}$ is the outlet oxygen mole fraction.

167 The temperature of the gas in the chamber is measured using a type K thermocouple located in the
168 flame at the normalized distance $x = L/L_{CC} = 0.36$, where L_{CC} is the total length of the combustion

169 chamber and L is the distance between the chamber entrance and the thermocouple. Additionally, a
 170 second type K thermocouple is positioned at the end of the chamber at $x = 0.93$. The thermocouples
 171 are gained Inconel 600 with an external diameter of 3 mm. The acquisition rate is 1 Hz. The gas
 172 temperatures reported are as measured by the thermocouples without any correction of the radiation.
 173 The position $x = 0.36$ corresponds to the heart of the flame, while the position $x = 0.93$ corresponds to
 174 the outlet of the combustion chamber. Moreover, the temperature of particles in the flame are measured
 175 using an IMPAC infrared ISR 12-LO bichromatic pyrometer ($\lambda_1=800$ nm, $\lambda_2=1050$ nm).

176

177 2.3. Dilution by N_2 of the injected air

178 An EGR (Exhaust Gas Recirculation) for reducing NO_x emissions is simulated by injecting nitrogen into
 179 the carrier gas. This dilution with nitrogen is done without any modification of the volume of the carrier
 180 gas. Air and nitrogen are mixed together before being injected in the axial and tangential gas inlet. The
 181 air/nitrogen mixtures allow obtaining dilution ratios of 0, 30 and 50% nitrogen, the dilution rates being
 182 expressed as a volume percentage. Table 1 summarizes the experimental conditions for the different
 183 dilution ratios which are defined with respect to the total air flow.

184

185 **Table 1:** Experimental conditions for each dilution ratio.

Dilution ratio	Axial entry (Nm ³ /h)	Tangential entry (Nm ³ /h)	Mg mass flow rate (g/h)	Equivalence ratio	Thermal power (kW)
0%	3.6 Air	5 Air	950 – 3150	0.14 – 0.59	6 – 22
30%	2.52 Air + 1.08 N ₂	2.1 Air + 1.5 N ₂	1050 – 3300	0.22 – 0.92	7 – 23
50%	1.8 Air + 1.8 N ₂	2.5 Air + 2.5 N ₂	900 – 3000	0.28 – 1	6 – 21

186

187 Then the mixture is injected into the system; the primary gas flow rate including the magnesium particles
 188 is 3.6 Nm³/h. The secondary/tangential gas flow rate is 5 Nm³/h for the swirl number $S_g = 7.3$.

189

190 3. Results and Discussion

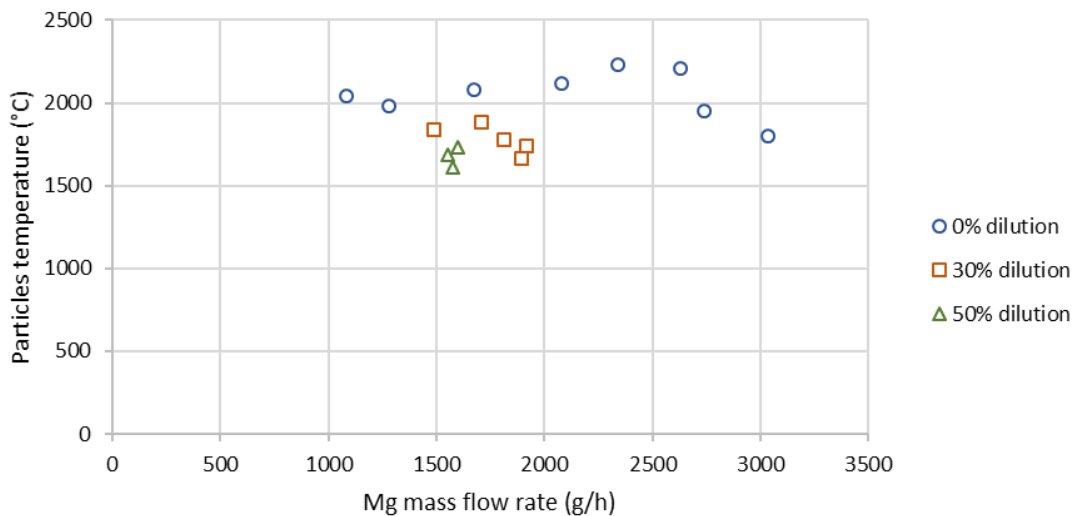
191 For metal combustion, only thermal NO_x have to be considered, as fuel NO_x and prompt NO are not
 192 relevant during the combustion of magnesium [13]. Zeldovich proposed a kinetic mechanism to simulate

193 the formation of thermal NO_x [15]. O and N radicals from the dissociation of O₂ and N₂ allow NO
194 formation. NO formation is also strongly influenced by the temperature. As a consequence, among all
195 the experimental parameters investigated, oxygen concentration and temperatures are those which
196 should significantly influence the thermal NO_x formation [15]. The particles and gas temperatures and
197 the oxygen mole fraction are first presented and discussed. The influences of the global equivalence
198 ratio and of the air dilution are analyzed for these experimental parameters. The NO_x emissions versus
199 the equivalence ratio and dilution by N₂ are then presented and discussed, considering their
200 dependence on temperatures and oxygen mole fraction. At last, the results are analyzed through the
201 global mechanism for NO_x formation presented in [13].

202

203 3.1. Particle temperature

204 The temperatures of the magnesia particles after recondensation are observed using a pyrometer.
205 Figure 2 gives the particles temperature as a function of the magnesium mass flow rate. The temperature
206 ranges between 1550 °C for a dilution of 50% to 2200 °C without any dilution.



207

208 **Fig. 2.** Particles temperature versus the magnesium mass flow rate for the three dilution rates.

209

210 The particles temperature is not significantly influenced by the magnesium mass flow rate, but it
211 decreases when the dilution ratio increases.

212 Because the magnesium combustion takes place in a homogeneous phase, MgO is formed in the gas
213 phase and recondenses in the solid phase. The magnesia particles formed are initially at a temperature
214 of 3040 K which is the maximal temperature that solid magnesia can reach [31]. The bichromatic

215 pyrometer returns the temperature of a solid in the flame area. This solid cannot be the inner wall of the
216 combustion chamber because it is cooled by water and therefore it remains at much lower temperatures
217 than those measured by the pyrometer. It cannot also be magnesium because it is in vapor form at these
218 temperatures. It is therefore surely the temperature of the magnesia.

219 The temperatures indicated in Fig. 2 are much lower than the theoretical value of 3000 K - which is close
220 to the adiabatic flame temperature – obtained by Shoshin et al. [32] and Lim et al. [33]. Such a difference
221 can be explained by the radiation and the convection heat losses in the combustion chamber that
222 decrease the average flame temperature. These heat losses are induced by both the cooled walls of
223 the combustion chamber and the optical access. They are enhanced by the turbulence in the swirled
224 flame.

225 It is important to notice that the temperature of the particles decreases when the dilution factor increases.

226 This could be explained by the depletion of oxygen in the flame when increasing the dilution ratio.

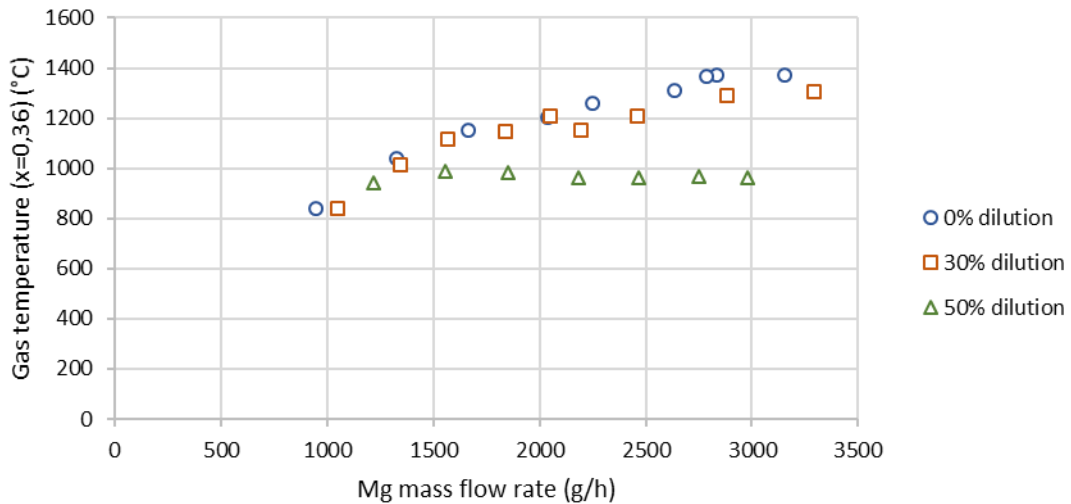
227 As the combustion rate of magnesium is a function of the O₂ concentration in the air-fuel mixture, when
228 the O₂ mole fraction decreases, the combustion rate decreases and consequently the energy released
229 by the combustion also decreases. Furthermore, nitrogen is a cold and non-reactive gas. A dilution with
230 N₂ of the injected air necessarily leads to a decrease of the gas temperature in which the magnesia
231 particles flow. Consequently, the magnesia particles cool faster in the gas. These two effects contribute
232 to the decrease of the temperature of the magnesia particles.

233

234 **3.2. Gas temperature**

235 The thermocouples positioned at the normalized positions $x = 0.36$ and $x = 0.93$ allow to investigate the
236 influence of the dilution on the gas temperature. The gas temperature does not significantly vary along
237 the axis of the combustion chamber in the first half of the chamber [34]. The temperature obtained for
238 $x = 0.36$ is a relevant indication of the average gas temperature in the flame zone. Moreover, the gas
239 temperature increases during a time length of 200 s before reaching a plateau where only small
240 fluctuations (lower than 20 °C) of the temperature versus time are observed.

241 Figure 3 presents the gas temperature at the position $x = 0.36$ as a function of the solid mass flow rate
242 of the magnesium.



243

244 **Fig. 3.** Gas temperature at $x=0.36$ versus the magnesium mass flow rate for the three dilutions.

245

246 Until a solid flow rate of 1500 g/h, the temperature proportionally increases with this solid flow rate.

247 Then, for higher solid flow rates, the temperature slowly increases for a nitrogen dilution of 0 and 30%

248 or even does not increase for a dilution of 50%. The position of the flame is potentially shifted back in

249 the chamber due to oxygen depletion when the dilution factor increases. Hence, the maximal heat

250 release is also shifted back and consequently the gas temperature slowly increases at the position

251 $x = 0.36$.

252 In addition, the gas temperature at $x = 0.36$ decreases with the nitrogen dilution. For a 50% dilution, the

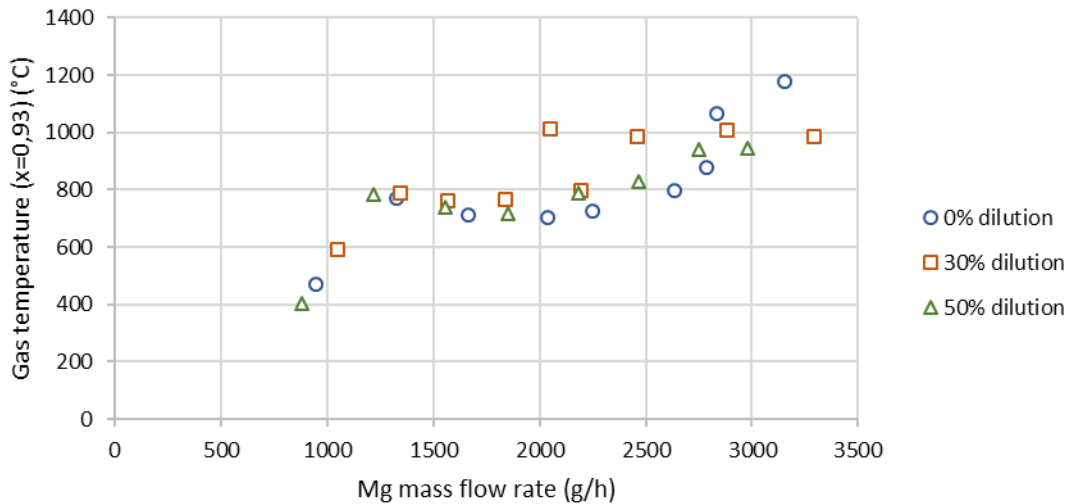
253 maximal gas temperature is 1000 °C to be compared to that 1400 °C obtained without any dilution. Such

254 a decrease in the gas temperature when increasing the dilution ratio is well correlated with the decrease

255 of the particle temperature in the same conditions as observed in Fig. 2.

256 Figure 4 depicts the gas temperatures as a function of the magnesium mass flow rate near the exit of

257 the combustion chamber at $x = 0.93$.



258

259 **Fig. 4.** Gas temperature at $x = 0.93$ versus the magnesium mass flow rate for the three dilutions.

260

261 The temperature ranges from 400 to 1200 °C, without any clear influence of the dilution on this
 262 temperature. The curves obtained for the three dilutions look quite similar. The temperature first
 263 increases when the magnesium mass flow rate increases from 800 to 1200 g/h. Then, a temperature
 264 plateau is observed for the magnesium flow rate ranging from 1200 to 2500 g/h. Finally, a slow increase
 265 of the temperature with the magnesium flow rate is observed. The two first stages are also observed for
 266 the temperature measured at $x = 0.36$, but with any clear influence of the dilution. Interestingly, the
 267 change of slope for the gas temperatures (at $x = 0.36$ and $x = 0.93$) appears for a magnesium mass flow
 268 rate of 1400 g/h. The position $x = 0.93$ is far from the flame zone which is located in the first half of the
 269 chamber, as previously shown by Laraqui et al. [30]. Consequently, the turbulence and the heat
 270 exchanges by thermal radiation and convection between the flame and the combustion chamber tend
 271 to homogenize the gas temperature from the middle to the exit of the chamber.

272

273 3.3. Heat Balance

274 A simplified heat balance is calculated, which accounts for the heat exchanges in the water-cooled
 275 combustion chamber and the heat exhausting the combustion chamber with the gas (N_2 , O_2 , NO and
 276 NO_2) and the solid (MgO). The heat exchange in the combustion chamber is calculated from both the
 277 inlet and outlet temperatures of the water circulating in the double wall exchanger and the water flowrate.
 278 The temperature of the gases and particles mixture leaving the combustion chamber is used to estimate
 279 the heat released at the chamber outflow.

280 The power dissipated by the combustion of the magnesium P_{Mg} , considering that it totally burns before
 281 leaving the chamber, the power recovered in the combustion chamber (P_{cc}) and that transported to the
 282 chamber outlet (P_{out}) are estimated using the following expressions:

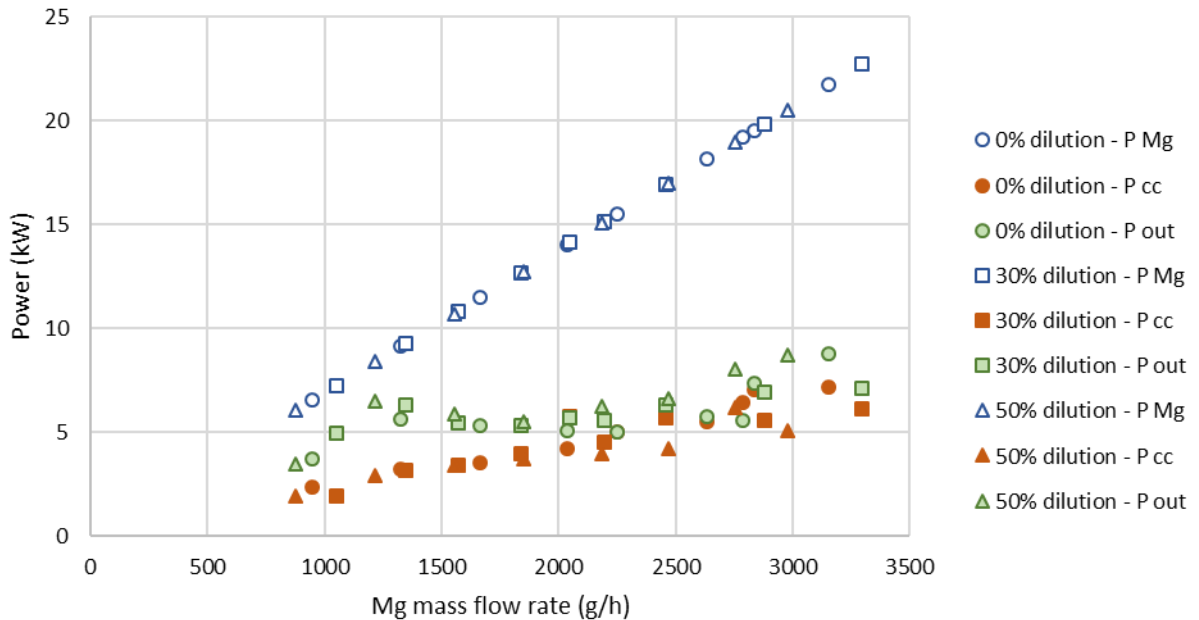
$$283 \quad P_{Mg} = \dot{m}_{Mg} \cdot p_{Mg}$$

$$284 \quad P_{CC} = \dot{m}_{water, chamber} \cdot C_{p, water} \cdot (T_{water, CCout} - T_{water, CCin})$$

$$285 \quad P_{out} = \dot{m}_{g, CCout} \cdot C_{p, air} \cdot (T_{g, CCout} - T_{g, ambient}) + \dot{m}_{MgO} \cdot C_{p, MgO} \cdot (T_{g, CCout} - T_{g, ambient})$$

286 The injected power P_{Mg} is determined using the mass flow rate of magnesium and the specific energy
 287 of magnesium combustion p_{Mg} (24.8 kJ.kg⁻¹). The specific thermal capacities are denoted $C_{p,x}$ for the
 288 fluid or element x under consideration (expressed in kJ kg⁻¹ K⁻¹). $T_{g, CCout}$ and $T_{g, ambient, l}$ are the gas
 289 temperatures measured with at the chamber outflow and the ambient temperature (°C). $T_{water, CCin}$ and
 290 $T_{water, CCout}$ are the temperatures of the water at the inlet and outlet of the cooling-water circuit (°C). \dot{m}_{Mg} ,
 291 $\dot{m}_{water, chamber}$, $\dot{m}_{g, CCout}$ and \dot{m}_{MgO} are the mass flow rate of Mg injected, cooling-water, gas, and MgO
 292 at the chamber (kg/s).

293 These balances are only indicative and comparative. They cannot be considered as absolute because
 294 after only 180 s thermal equilibrium is not reached in the system. P_{cc} and P_{out} are plotted versus P_{Mg} in
 295 Fig. 5, for the three dilution ratios.



296

297

Fig. 5. Simplified heat balance for the three dilutions (0, 30 and 50%).

298

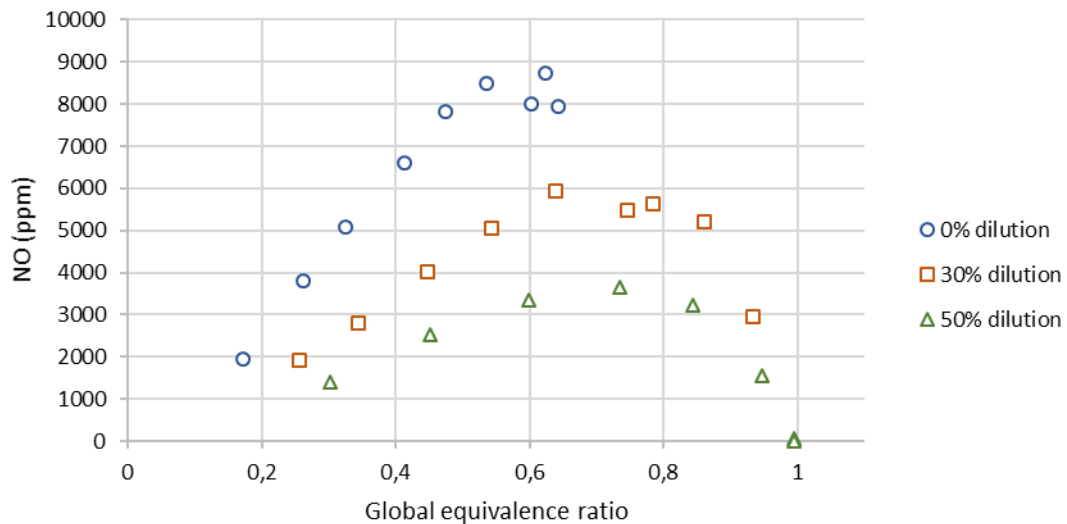
299 It appears that there is no significant effect of the dilution on the heat fluxes. This indicates that whatever
300 the dilution, it does not impact the power recovery from the combustion.

301 As shown in Fig. 5, the sum of P_{cc} and P_{out} is equal to P_{Mg} for magnesium flow rates lower than 1500 g/h.
302 This indicates that all the power delivered by the magnesium combustion is transmitted to the
303 combustion chamber and carried by the gas and solid leaving the combustion chamber. For a
304 magnesium flow rate greater than 1500 g/h, only a fraction of the power (approximately 66%) is
305 transferred to the chamber on the one hand and leaves the chamber on the other hand. To explain such
306 a behavior, it must be considered that the flame radiation is much less intense at low than at high
307 equivalence ratio. Therefore, the fraction of the power transferred by radiation to the different parts of
308 the burner, including the injection head, increases when the flow of injected magnesium increases. The
309 power emitted by the flame radiation at the combustion chamber outlet is also not accounted for. They
310 are not taken into account in the heat balance. This may explain the difference between the power
311 dissipated by magnesium combustion and the power collected.

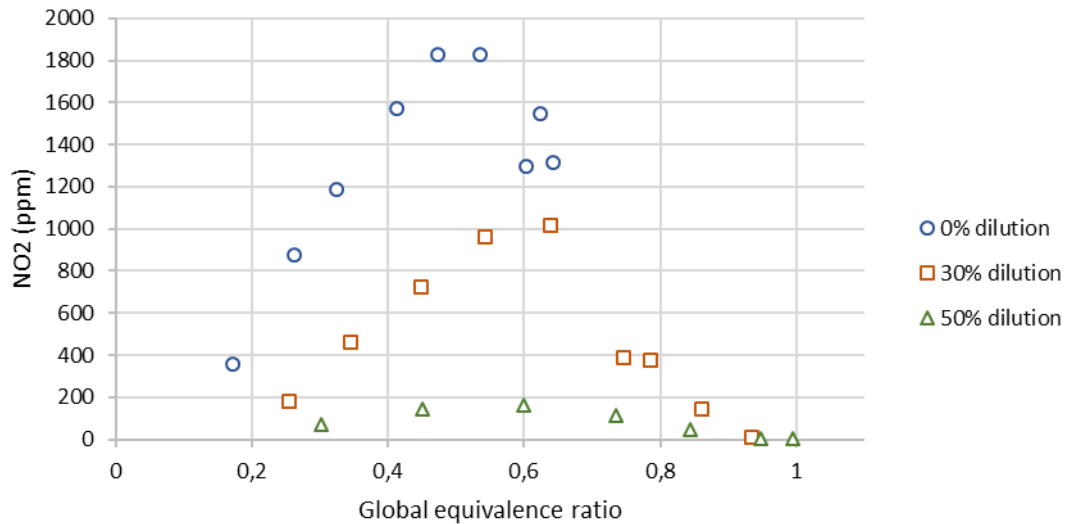
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313 3.4. NOx emissions

314 Figure 6 depicts the NO and NO₂ mole fractions expressed in ppm as a function of the global equivalence
315 ratio for the three tested dilutions.



316



317
 318 **Fig. 6.** NO and NO₂ emissions versus the global equivalence ratio for the three dilutions.
 319

320 The NO and NO₂ mole fractions exhibit a parabolic shape curve. They first increase with the global
 321 equivalence ratio until they reach a maximum. Then NO_x emissions decrease. The maximal values for
 322 NO mole fractions are 9000, 6000 and 3900 ppm for 0, 30 and 50% dilution, respectively. The effect of
 323 the dilution ratio on NO₂ mole fractions is more pronounced. NO₂ emissions drastically reduce from 1800
 324 to 100 ppm with a dilution ratio of 50%.

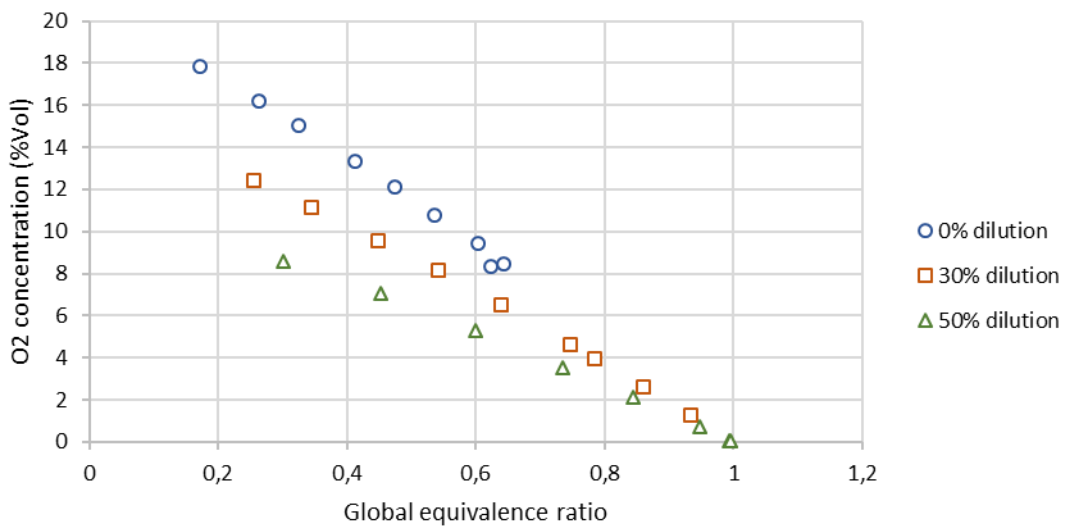
325 Considering the NO_x emissions as a function of the global equivalence ratio according to the nitrogen
 326 dilution rate, it first appears in Fig. 6 that the amount of NO formed is four times greater than the amount
 327 of NO₂ formed. NO is formed before NO₂ with much faster kinetics. With a 50% dilution, the amount of
 328 NO formed is divided by 2 and the amount of NO₂ formed is ten times less than without any dilution. In
 329 addition, for a global equivalence ratio greater than or equal to 1 for a dilution rate of 50%, there is no
 330 more oxygen to generate the formation of NO_x, the levels of NO and NO₂ are both reduced to 0 ppm.

331 N₂O was measured for experiments performed with 0% dilution and a range of equivalence ratios from
 332 0.57 to 0.97. No N₂O was observed whatever the experimental conditions tested.

333 The Mg particle burns in a micro-flame to produce MgO which recondenses. Increasing the Mg flow rate
 334 increases the heat released by combustion and also increases the subsequent heat transfers by
 335 convection and radiation between the MgO particles and the surrounding gas. This behavior can be
 336 explained by the rise of the concentration of the MgO particles produced by combustion.

337 The residence time of the gas in the combustion chamber is approximately equal to 0.5 s. For such a
 338 duration, a significant amount of thermal NO_x can be produced only if the gas temperature is higher

339 than 2000 K. Such an assumption is well supported by a model based on the Zeldovich mechanism
 340 [15], and the associated kinetic parameters [35]. The duration of Mg oxidation is approximately equal to
 341 few milliseconds [36,37]. Hence, NO_x formation surely occurs after Mg combustion is completed. It can
 342 occur in the gas phase in close contact with the surface of MgO particles where the gas temperature is
 343 higher than the mean gas temperature presented in Fig. 3.
 344 According to the global mechanism for NO_x formation proposed in [13], the NO_x yielding increases
 345 when increasing the Mg mass flow rate. Indeed, the total contact area between MgO surface and the
 346 gas increases as MgO concentration rises. As shown in Fig. 3, the mean gas temperature increases as
 347 a function of the global equivalence ratio which is a consequence on the one hand of the power
 348 dissipated by Mg combustion and on another hand of the rise in MgO particles concentration.
 349 For the highest equivalence ratios, the NO_x mole fraction decreases due to the depletion of the available
 350 oxygen. The amount of oxygen becomes a limiting factor as shown in Fig. 7, causing a decrease in NO_x
 351 emissions.



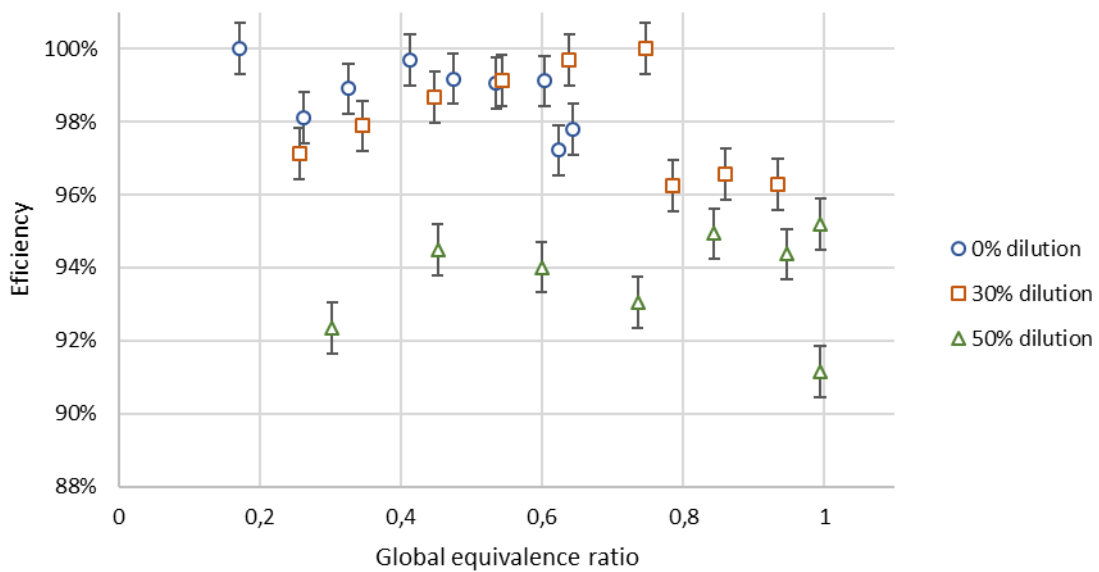
352
 353 **Fig. 7.** Oxygen concentration versus the global equivalence ratio for the three dilutions.

354
 355 As shown in Fig. 7, the oxygen mole fraction at the chamber outflow strongly decreases as the
 356 equivalence ratio increases. The oxygen defect for NO_x formation results in the Mg oxidation which
 357 occurs before NO_x formation. For a dilution of 50% and an equivalence ratio close to 1, oxygen
 358 concentration at the outflow is zero and this explain why no NO_x are produced under such experimental
 359 conditions. All the oxygen available is consumed to burn Mg.
 360

361 **3.5. Efficiency of magnesium combustion versus N₂ dilution**

362 All the injected metal powder Mg should be burnt and fully converted in MgO. A method to determine
363 the completion of Mg oxidation was already presented by Garra et al. in [7]. This method is here used
364 to determine the impact of the air dilution on the Mg oxidation. The combustion efficiencies of the solid
365 samples collected at the entrance of the combustion chamber ($0.0 < x < 0.36$) and at the end of the
366 combustion chamber ($x > 0.9$) are estimated.

367 For the particles collected at the chamber outflow, the combustion efficiency is close to 100%, whatever
368 the tested experimental conditions. Hence, the dilution efficiency does not affect the Mg oxidation
369 considering the overall chamber and there is no unburnt Mg at the chamber exit. Figure 8 depicts the
370 results obtained for the different equivalence ratios and dilution levels for the particles collected at the
371 entrance of the combustion chamber. The uncertainties are estimated from four experiments obtained
372 from the same collected particles and represented by the error bars in Fig. 8.



373

374 **Fig. 8.** Efficiency of the reaction at the entrance of the combustion chamber versus the global
375 equivalence ratio for the three dilutions.

376

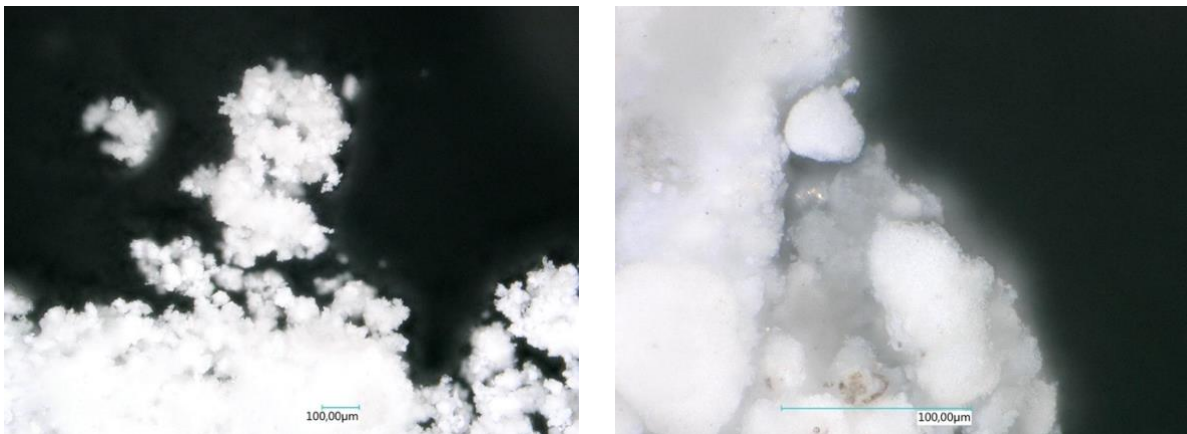
377 For the particles collected at the entrance of the combustion chamber, the combustion efficiency is very
378 high for the 0 and 30% dilutions and for equivalence ratios smaller than 0.7. It significantly decreases
379 (96%) for a dilution of 30% and for the highest equivalence ratio. For a dilution of 50%, the combustion
380 efficiency is smaller, lying between 91 to 95%. This means that the oxygen concentration is not high

381 enough to promote the full combustion of Mg with a 50% dilution. Some Mg particles are partially
382 oxidized and deposited on the chamber wall.

383 As the chamber is water-cooled, the wall temperature is too low for a slow oxidation [38] of the Mg
384 particles deposited here. If the level of oxygen is too low to promote the full oxidation of Mg, it is clear
385 that it is also too low for NO_x formation.

386 Figure 8 gives the yield of Mg to MgO conversion for the particles collected at the beginning of the
387 combustion chamber which approximately represent 20% of the mass of Mg/MgO collected. In the other
388 part of the combustion chamber, in the cyclones and in the secondary heat exchanger, the particles
389 collected are only MgO. They represent 80% of the mass of magnesia collected in the system. Thus,
390 taking into account the whole system, the overall conversion is always higher than 98%, whatever the
391 experimental conditions. The calculation of the equivalence ratio from the oxygen mole fractions at the
392 inlet and outlet of the system can therefore be considered as close to reality.

393 The collected particles were observed by microscopy with a Keyence optical microscope already used
394 by Moser et al. [38], see Fig. 9.



395
396 **Fig. 9.** Images of collected MgO (a) at the chamber outlet for 0% dilution and (b) at the chamber inlet
397 obtained for 50% dilution.

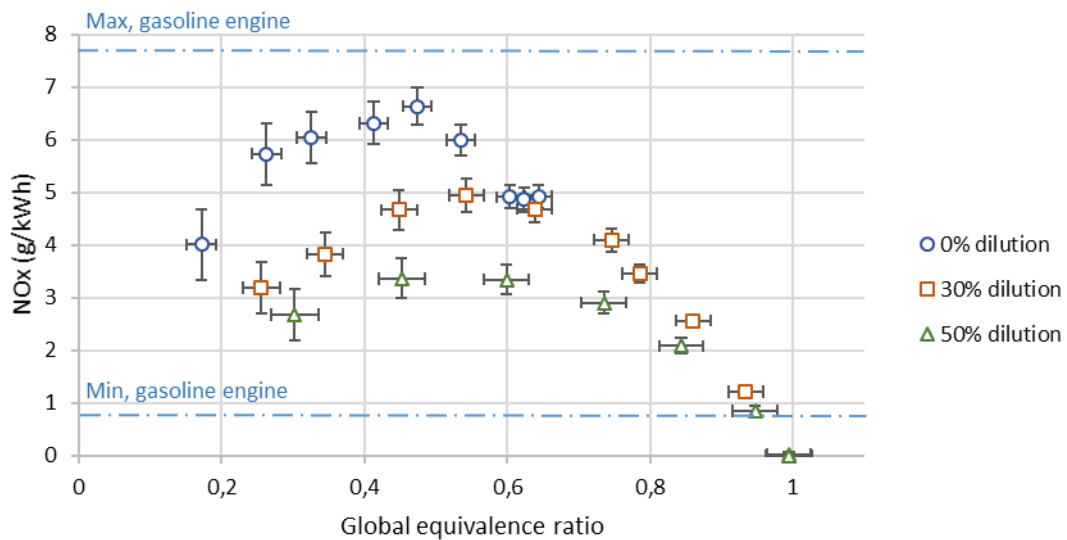
398
399 As shown in Fig. 9.a, the MgO particles collected at the end of the combustion chamber for the
400 experiments with 0% dilution are white aggregates of MgO. Such aggregates were already observed by
401 Laraqui et al. [8]. The particles collected at the entrance of the combustion chamber for a 50% dilution
402 (Fig. 9.b), exhibit some silvery metallic surface and appear to be not fully oxidised. The silvery metallic
403 surfaces are characteristic of magnesium inclusion which are not observed on the magnesia sample

404 collected at the chamber outlet for undiluted combustion. Moser et al. also observed Mg particles
405 partially oxidised in thermogravimetric experiments and Mg inclusions in MgO particles [38].

406

407 3.6. Comparison with NOx emissions of a gasoline engine

408 Figure 10 depicts the NOx emissions expressed in g/kWh as a function of the global equivalence ratio
409 or of the Mg mass flow rate according to the dilution rate. The vertical error bars are calculated from the
410 precision of the NOx analyser and the dissipated power. The horizontal error bars are calculated from
411 the precision of the oxygen analyser. The maximal and minimal NOx emissions for a gasoline engine
412 are also plotted for an easier comparison with a current gasoline engine as already presented for other
413 experimental conditions by Laraqui et al. [8].



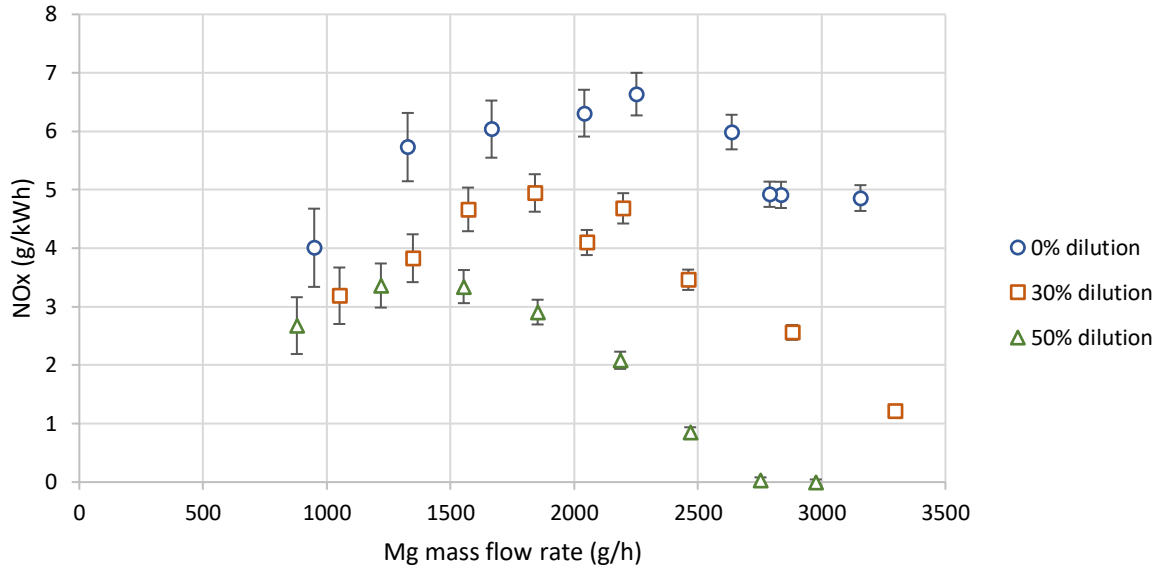
414

415 **Fig. 10.** NOx emission versus the global equivalence ratio for the three dilutions. Comparison with the
416 NOx emissions from a gasoline engine.

417

418 Interestingly, the NOx yielding is lower than the maximal value emitted by gasoline engines and falls
419 below the minimal value emitted by gasoline engines for a global equivalence ratio of 0.9 at a dilution of
420 50%. Moreover, as previously explained, there are no more NOx emitted at stoichiometry for a dilution
421 of 50%, while this is not the case for gasoline engine, as prompt and fuel NOx are additionally produced.
422 Such a result is very promising for the development of a technology using metal as energy carrier. The
423 NOx yielding is minimized by an air dilution with nitrogen and the level of NOx are negligible for the
424 highest values of equivalence ratio. 50% nitrogen dilution means that the injected gas is composed of a

425 mixture 50% air/50% nitrogen for a total gas flow rate similar to that injected for 0% dilution. Thus, half
 426 as much oxygen is brought into the burner. It is interesting to note that the NO_x emissions appear in
 427 different ways if they are plotted against the injected magnesium flow rate, as presented in Fig. 11.



428
 429 **Fig. 11.** NO_x emissions versus the Mg mass flow rate ratio for the three dilutions.

430
 431 Indeed, NO_x emissions are not significantly affected by nitrogen dilution for the lowest magnesium
 432 injection rates. In this case, even though the oxygen fraction is lower as the nitrogen dilution rate
 433 increases, oxygen remains in excess in the combustion chamber as shown in Fig. 7. Thermal NO_x can
 434 thus be formed in the gas phase in tight contact with magnesia. On the other hand, NO_x emissions are
 435 significantly reduced with N₂ dilution for magnesium injection rates higher than 1500 g/h. In this case,
 436 there is certainly a lack of oxygen in the flame which limits the NO_x production.

437 The maximal NO_x emissions from the magnesium combustion is 6.55 g_{NO_x}/kWh for 0% dilution and 3.25
 438 g_{NO_x}/kWh for 50% dilution. These maximal values are much higher than the amount currently emitted by
 439 coal power plant (from 0.2 to 0.4 g_{NO_x}/kWh) or biomass power plant (from 0.2 to 0.7 g_{NO_x}/kWh) [13].
 440 However, for a 50% dilution ratio and an equivalence ratio closed to 1, the NO_x emissions fall to 0 with
 441 the swirled-stabilized magnesium flame.

442

443 **4. Conclusion**

444 The study focused on the impact of N₂ dilution on the formation of thermal NO_x during the combustion
445 of magnesium in a swirled-stabilized turbulent flame. NO and NO₂ emissions versus the equivalence
446 ratio exhibit a parabolic shape. No N₂O is detected. The NO_x emissions (NO+NO₂) increase with the
447 equivalence ratio until they reach a maximum. Such an increase is explained by the rise of the
448 concentration of MgO particles in the combustion chamber. Indeed, NO_x are produced in the gas phase
449 in close contact with MgO particles where the temperature is high enough. After this maximum, NO_x
450 emissions decrease due to an oxygen depletion in the gas. Magnesium combustion reaction seems to
451 be faster than NO_x formation. The level of NO_x never exceeded 7 g_{NO_x}/kWh, which is lower than the
452 maximal value of NO_x emissions from a gasoline engine. A simulation of an exhaust gas recirculation
453 with a nitrogen dilution (0, 30 and 50%) allows significantly reducing the NO_x emissions, whatever the
454 equivalence ratio. The maximal NO_x emissions are shifted to higher equivalent ratios when increasing
455 the dilution level and it decreases by a factor 2 with a dilution of 50%, for a constant value of the air-fuel
456 ratio. The gas temperature in the flame decreases due to the N₂ dilution. Both such a temperature
457 decrease and the O₂ depletion lead to limit the NO_x formation. NO_x emission tends to zero for an
458 equivalent ratio of 1 and a nitrogen dilution of 50%.

459

460 *CRedit authorship contribution statement*

461 **Adeline Andrieu**: Methodology, Investigation, Writing – original draft. **Olivier Allgaier**: Investigation.
462 **Gontrand Leysens**: Conceptualization, Validation, Supervision. **Cornelius Schönnenbeck**:
463 Conceptualization, Validation, Supervision. **Jean-François Brillhac**: Conceptualization, Validation,
464 Supervision, Writing – original draft, Writing – review & editing, Project administration. **Alain Brillard**:
465 Validation, Writing – original draft, Writing – review & editing, Project administration. **Valérie**
466 **Tschamber**: Project supervision.

467 **Declaration of Competing Interest**

468 The authors declare that they have no known competing financial interests or personal relationships that
469 could have appeared to influence the work reported in this paper.

470

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474

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545 [magazine/r%C3%A9duction-des-nox-traitement-des-fum%C3%A9es-r%C3%A9duction-](http://atee.fr/energie-plus-magazine/r%C3%A9duction-des-nox-traitement-des-fum%C3%A9es-r%C3%A9duction-s%C3%A9lective-non-catalytique-ou)
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