



# Magnesium/air combustion at pilot scale and subsequent PM and NO<sub>x</sub> emissions



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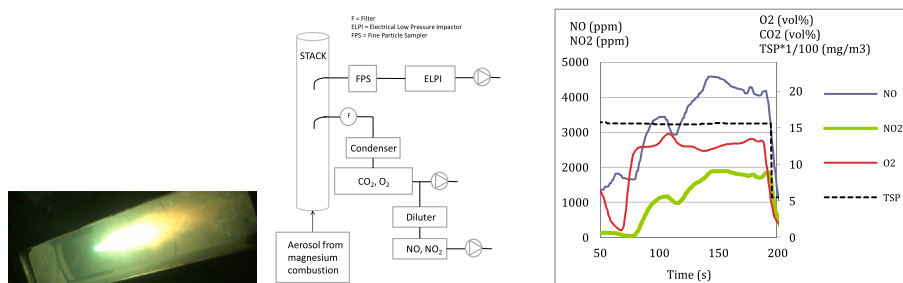
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## HIGHLIGHTS

- The feasibility of magnesium energy release at a pilot scale (from 3.0 to 5.0 kW) is demonstrated.
- Magnesium/air stable flames are obtained from raw magnesium powder and combustion completion ratios ranges from 91 to 100%.
- Total suspended particles (TSP) emissions, estimated from ELPI measurements, ranges from 1 to 35 g/(N)m<sup>3</sup>.
- NO<sub>x</sub> emissions are higher for the 20–50 μm fraction than for the 50–70 μm fraction (NO average of 1100 ± 140 mg/(N)m<sup>3</sup>).

## GRAPHICAL ABSTRACT



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## ABSTRACT

Fossil fuel scarcity, global warming and non-constant energy production through renewable energies (wind turbines and photovoltaic cells for example), lead to investigate innovative energy sources and new ways for energy storage. In the present study, magnesium powder has been considered as a new possible energy carrier. In order to analyze more deeply the magnesium combustion and the generated by-products, short time stable magnesium/air flames have been realized in a combustion chamber using an oxy-acetylene flame for ignition. Sieved magnesium samples with two fractions were combusted: 20–50 μm and 50–70 μm. The power delivered by the Mg/air flame was estimated in the range 3–5 kW. The gaseous emissions (O<sub>2</sub>, CO<sub>2</sub> from oxy-acetylene combustion, NO and NO<sub>2</sub>) were analyzed with on-line analyzers and the particulate emissions were analyzed with an Electrical Low Pressure Impactor (ELPI). The mass concentration of emitted particles whose size is smaller than 10 μm was proved to be very high (up to 35 g/(N)m<sup>3</sup>) and the emitted particles were mainly bigger than 1 μm (84–97 wt%). NO<sub>x</sub> emissions were higher for the 20–50 μm Mg fraction (NO average of 4300 ± 200 mg/(N)m<sup>3</sup>) than for the 50–70 μm Mg fraction (NO average of 1100 ± 140 mg/(N)m<sup>3</sup>).

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## 1. Introduction

Fossil fuel scarcity and global warming lead to diversification of the energy sources. The main goals for new energy production and storage consist in limiting greenhouse gases emissions such as CO<sub>2</sub>,

in promoting renewable energies and limiting their environmental impacts. Renewable energy sources have known downsides. For instance, wind turbines [1–3] or photovoltaic cells [4,5] present non-constant energy production and release. Therefore innovative ways to perform energy storage and production have to be investigated. Among such innovative ways are production of sustainable biofuels using a great variety of resources that do not compete with edible crops and new conversion technologies. An example is hydrogen ( $H_2$ ) energy storage using electrolyzers [6] and release in fuel cells [7,8] for vehicle applications or production of a totally renewable biodiesel through supercritical triglycerides ethanolyse [9].

Most of the renewable energy sources depends on environmental conditions such as solar and wind and are not portable. Energy storage is then a key issue concerning a transition towards zero-carbon systems. Energy storage in a chemically reduced form for subsequent combustion and energy release is a valuable option. In this context metal particles could be considered as a renewable energy carrier [10].

The investigations of energy storage using metal powder are scarce [11] and mainly aluminum and magnesium are considered as a metal candidate for such an application. Magnesium is currently the 8th most abundant element in the earth's crust with 2.1 wt% [12]. Its electrolysis is already widely documented in order to obtain metal Mg from Mg oxides [13]. Many studies consider magnesium combustion under different combustion atmospheres, among which are carbon dioxide at low pressure [14], water vapour [15] and  $O_2$ -inert atmosphere [16]. Fundamental researches on burning processes and mechanisms are also widely referenced [16–19]. The ignition and burning of metal particles were investigated by many authors from the nineteen-sixties [20,21] until today [11,22,23]. The investigations considered micron-sized metal particles or nano-sized particles [24] exhibiting shorter burning times. Magnesium combustion is mainly used in punctual energy release applications, for example in nanothermites pyrotechnics [24], mixed with aluminum building alloys for welding applications [25–27] or for space rockets [28–31]. But these applications are currently not considered as potentially interesting for magnesium-based energy storage, as they do not involve a steady continuous release of energy through magnesium combustion and the products, i.e. magnesium oxide, are released into the atmosphere.

Recently, the direct combustion of recyclable metal fuels has been thoroughly considered [32]. Metal particles exhibit energy density similar to liquid hydrocarbons making them relevant for transportation systems. Metal particles seem compliant with an external combustion engine. This vector is expected to be safer for the on-board storage compared to hydrogen commonly considered as the new energetic vector for mobility [33,34]. Another advantage of metal as an energy vector is its ability to be reduced; the oxide produced from combustion can be recycled using primary energy in a separate process. Consequently an essential requirement for the combustion of metal fuels is the complete capture of all the metal oxide formed [32]; solid combustion-product morphology and size have to be investigated on metal-fuelled burners. Since particulate matter emissions (PM) is an important issue for nowadays societies [35], fine ( $<1\ \mu m$ ) and ultrafine ( $<100\ nm$ ) particulate matters are especially pointed out as they can penetrate the deepest part of the lungs [36]. Therefore PM emissions generated by metal combustion have to be considered. Emissions of Nitrogen Oxides ( $NO_x$ ) cause significant damage like smog formation, acid rain or ozone formation at low altitudes [37]. The high flame temperature leads to high  $NO_x$  pollutant emissions [32] and this could be an issue for the development of the use of metal powders as a new energetic vector.

The future of energy generation for terrestrial applications could be the use of metal as fuel. However the pollutants emitted by the combustion of metal particles is not well documented in comparison to those emitted by the combustion of hydrocarbon fuels. The present study focuses on the magnesium combustion and the subsequent pollutants emitted. The metal recycling step – reduction of MgO to Mg using primary energy [32] as solar energy for instance [38] is out of the scope of this work. Two questions are then addressed: (i) what are the solid products of the combustion process occurring in a burning chamber with a continuous injection of Mg delivering a power suitable for use in automobile sectors? (ii) what are the pollutants emitted during the combustion process?

In order to investigate the efficiency of magnesium combustion and to analyze the by-products generated – including emissions of particulate matter and  $NO_x$  – stable magnesium/air flames have been produced in this study in a totally new laboratory scale combustion chamber. The power delivered by the Mg/air flame in this boiler has been estimated to be in the range 3–5 kW. This boiler is implemented in a combustion chamber. A cyclone allows trapping of particles in the chamber outflow. This enables the analysis of the solid and gaseous products emitted during the Mg/air reaction occurring in realistic situations. The gaseous emissions ( $O_2$ , NO and  $NO_2$ ) are monitored using an online analyzer. The particulate emissions are analyzed with an Electrical Low Pressure Impactor (ELPI).

The experimental set-up is described in the first section of the paper. The second part of the paper presents mass and energy balances within such a combustion system. The evaluation of the combustion efficiency has been performed using innovative ways based on the thermogravimetric analysis of the residues produced by the combustion process, but also on the dynamic assessment of magnesium and  $O_2$  consumption.  $NO_x$  and PM emissions are presented and discussed in the third part of the paper.

## 2. Materials and methods

### 2.1. The combustion system

The magnesium powders used in this work are pure ( $>99.8\%$ ), supplied by Carl Roth® (France) as CP20.2 reference. The raw powder was sieved using a RETSCH GmbH (Germany) AS200 sifting machine. Two size fractions were studied: 20–50  $\mu m$  and 50–70  $\mu m$ . A scheme of the combustion system is depicted in Fig. 1. The system is composed of four main parts: magnesium aerosol generator, boiler, combustion chamber and cyclone. The nozzle inner diameter of the boiler is 22 mm; the combustion chamber diameter is 106 mm. The magnesium/air aerosol is generated using a PALAS GmbH (Germany) BEG 1000B device, allowing the injection of solid powder in the range 0.1–200  $\mu m$ . Aerosol generation complies with norm VDI 3491 for reliable aerosol mass flow rate output. The combustion process may be decomposed in the following stages:

- setting air flow rates of the burner without magnesium particles injection,
- ignition of the oxy-acetylene flame in the combustion chamber,
- starting Mg powder injection at a small mass flow rate,
- increasing potentiometer of the PALAS to the nominal mass flow rate value for magnesium injection,
- stopping the oxy-acetylene flame once a stable Mg/Air flame is obtained,
- stopping the magnesium particles injection and immediate nitrogen inerting, from the solenoid valve releasing pressurized  $N_2$ , and stopping air injection in the combustion chamber.

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