

THE NEXT GENERATION OF MAGNESIUM BASED MATERIAL TO SUSTAIN THE INTERGOVERNMENTAL PANEL ON CLIMATE CHANGE POLICY

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Abstract

Current Mg alloys have several drawbacks that limit wide and profitable utilization in the industrial sector. From an environmental point of view, lighter metals like magnesium are currently considered unclean products as they require energy-intensive. But they have been proven to be "clean" in the transport sector, as they can reduce fuel consumption. Here the potential of magnesium based materials is addressed through double-tasking: a) establish innovative lean-manufacturing processes, avoid the classic melting step to substantially reduce carbon footprint of the magnesium products; b) encourage the using of no-melt processes, realizing high-resistant ultra-fined microstructures. The "Green Metallurgy 2020", a project funded by European Community in the LIFE+ 2009 Program, started in September 2010, coordinated by Politecnico di Milano (ITA) aims to scale to industrial route such impressive results experienced by CENIM (SPA) for some ultrafine bi-phase Mg-Zr (-Y) produced by no-melting route that achieved up to 400 MPa UTS and elongation capability of about 13%.

Introduction

While there is considerable debate about when global oil and natural gas production is likely to peak, there is no debate that fossil fuels like oil, coal, and natural gas constitute a non-renewable, finite resource. A resource that has powered cars, trucks, power plants and factories, and caused the recent relative and dramatic build up of greenhouse gases. As a basic requirement for the strategy to cut CO₂ levels in the transport sector, the IPCC report claims more fuel-efficient vehicles are needed in the future [1]. These would include advanced electric and hybrid vehicles with more powerful and reliable batteries, cleaner diesel vehicles, bio-fuels, modal shifts from road transport to rail and public transport systems, non-motorized transport (cycling, walking), and higher efficiency aircraft. In Europe, 12% of man-made CO₂ comes from passenger cars, with overall transport producing 26%. Between 1995 and 2003, motorists in the EU-25 increased their annual mileage by 16.4%. While a major challenge, it is clear that vehicle emissions are part of a much larger jigsaw [1]. The new industrial modern era is facing a difficult challenge: on the one hand there is the problem of increasing power energy, coupled with the dramatic increase of world energy consumption related to the growing economies in ASIA and AFRICA. On the other hand, there is an urgent and strong need to decrease the GHGs produced as a byproduct of this growth. These opposing forces bring an environmental imperative to the economic accountability of the actual cost of power production and need to include all of the environmental perimeters of our energy use. While fossil fuels have been considered to be relatively inexpensive in the past, a more modern approach starts with the realization that the atmosphere's assimilation of the byproducts produced by fossil fuel combustion

is not without consequences. Today their economic impact on environmental and health must also be considered. For this reason a more modern approach focuses on supply chains and total product life cycles which involve minerals, energy resource processing, manufacturing steps, logistics, re-use, and recycling routes. Among these, Life Cycle Assessment (LCA) concerns over the limitations of raw materials and energy resources sparked interest in finding ways to cumulatively account for energy use and to project future resource supplies and use. LCA analysis adopts a schematic method of assessing the entire product life cycle, which is usually divided into the following stages: a) cradle-to-entry gate (raw material extraction and refining); b) entry-gate-to exit gate (product manufacture) and c) exit gate-to-grave (product use, recycling, and disposal). The risk, however, may be that reduced fuel consumption, i.e. the cut CO₂ in an internal combustion engine (ICE) vehicle, cannot balance the equivalent carbon dioxide¹ produced during the cradle-to-exit gate.

Achieving a Net Positive Balance of CO₂ Emission in Transport

The ICE is expected to continue to be the dominant technology for the short-medium term, as the ICE can maintain similar performance standards by utilizing more lightweight designs that directly can reduce CO₂ emissions, and promote further engine downsizing. For the long-term, more lightweight hybrid, fully electric or hydrogen cars that can travel long distances will surely be a key-factor to saving energy. According to the European Automobile Manufacturers Associations (ACEA), automakers will have to reduce CO₂ emissions² in new cars to 130 grams per kilometer by 2012/15, with an additional 10 gram reduction coming from 'complementary measures' including a greater use of bio-fuels. And a new objective of 95 grams per kilometer was established for 2020 [2]. From 2012 onwards, manufacturers who do not meet their targets must pay an "excess emissions penalty"³, which may become law as early as 2012, may become an incentive for automakers to reduce CO₂ emissions by levying fines on automakers who don't respect emission thresholds (stated on 130gCO₂/Km).

¹ The kgCO₂ equivalent emissions is a parameter used for directly accounting of Global Warming; in fact it is also refereed as Global Warming Potential (GWP).

² In 2009, the market share of cars emitting 120 gCO₂/km had risen to 25%. Cars with emission above 160 gCO₂/km accounted for 23% of the market, compared to 39% in 2006 and to 80% in 1995 (source: ACEA).

³ Excess emission penalty shall be calculated as the excess emissions (over EU targets), multiplied by the number of new passenger cars (yearly basis) multiplied by the excess emissions premium that shall vary accordingly to the calendar year (for the year 2012, 20 Euros, for the year 2013, 35 Euros; for the year 2014, 60 Euros; 2015 and subsequent calendar years, 95 Euros).

If forced to pay a fine on cars that do not meet EU emission standards⁴, automakers, especially those in the medium-high quality area, would be encouraged to look for new short-term emission reducing strategies, and reducing the weight of each car may be the most practical, and least invasive way of reaching that goal. In fact, alternative solutions like cutting-performance or eliminating accessories would affect client's perceptions and expectations of the product.

The Magnesium Perspective for Vehicles: Superlight Structural Components Target Environmental Problems

Thanks to their low density, magnesium alloys offer one of the best opportunities. With a density of 1.81 kg/dm³, they are one lowest structural metals and are excellent alternative to heavier materials like steels and even aluminum. Current environmental points of view state that while lighter metals like magnesium are currently unclean products to make, they could be "very clean to use". This can be attributed to the substantial weight reduction they offer and the resulting reduction of fuel consumption and CO₂ on ICE vehicles. With that in mind, consider the following key points regarding internal combustion engine transportation taken from [3]:

- 2.85 kg CO₂eq is the average emissions per 1 liter of gasoline consumption;
- 10.83 kg CO₂ per 1000 km is the calculated amount of reduced emissions for every 100 kg mass saved on passenger vehicles;
- if 200,000 km is the total life distance of a common passenger vehicle, a total amount of 2,166 kg CO₂eq can be saved through 100 kg weight reduction.

Although magnesium density is less than 25% of that of steel, its lower mechanical resistance is an impediment to achieving a 1 to 1 substitution ratio. Conservatively speaking, 1 to 2.65 is the (volumetric) substitution ratio to guarantee similar resistance of components (Table I).

Table I. Total Average Pollution Emissions Over Medium-size Vehicle Life.

	Total amount	Metrics
Car mass	1,300	Kg
Emissions	2.85	kg CO ₂ eq/liter
Average consumption	8.5	liter / 100 km
Total CO ₂ emission over 200,000 km	48,450	kg CO ₂ eq per car

Such a statement comes from the mechanical resistance of conventional Mg alloys, that is about 35% of common medium-

⁴ The European Commission calculates it will cost on average EUR 1500 per car to comply with new EU CO₂ targets. In addition, there will be the costs for the 10gCO₂/km 'complementary measures' (gear-shift indicators, tire-pressure monitoring, low-rolling resistance tires, air-conditioning and bio-fuels) (source: Proposal for a REGULATION OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL, "Setting emission performance standards for new passenger cars as part of the Community's integrated approach to reduce CO₂ emissions from light-duty vehicles", presented by Commission, Brussels, 19 December 2007).

carbon steels (50% for very low carbon steels). Substituting medium carbon steel with conventional magnesium alloys on structural components will result in an increase of the sections and volume of the components. On the other hand, thanks to magnesium's lower density, an overall weight reduction of approximately 40% can be expected using a magnesium component rather than a medium-carbon steel component (Table II).

Table II. Mg-alloy Design and Weight Saving by Substituting Medium Carbon Steel with Conventional (wrought) Mg-based Alloys.

Features	Mg	Steel
Density (kg/dm ³)	1.81	7.87
Resistance limit (MPa)	170	450
Usage ratio	2.65	1.0
Volume (mm ³)	265	100
Mass total (kg)	4.8	7.9
Total weight saving if 100 kg steel is substituted with magnesium conventional alloy (kg)	39	

To evaluate the impact of magnesium usage on medium-size passenger cars in terms of final net reduction of kgCO₂eq over the vehicle's travel-life distance, it is necessary to calculate the negative impact on the environment. The impact can be expressed in terms of kgCO₂eq, as measure of the Global Warming Potential (GWP) of magnesium-based products manufactured⁵. In the LCA of magnesium product to use onboard, the net GWP shall be calculated starting from the GWP of the cradle-to-exit gate path, but subtracting the reduction of CO₂ emissions obtained by reduced fuel consumption for the reduced total weight onboard. The importance of properly calculate the net GWP of magnesium product is due to the fact all magnesium processes are energy intensive and expensive, in comparison with steel-making⁶. The availability of primary minerals, the energy sources used for basic extracting process route, the power generation method employed for electricity in case of electrolytic process route⁷, drive the entire GWP process, specifically the carbon-footprint of the production of raw materials.

⁵ Great impact is ascribable to the use of sulfur hexafluoride (SF₆) as a cover gas, yet it is used in various low-cost plants. SF₆ is used to protect molten magnesium against oxidation both in primary magnesium smelters and in secondary magnesium production (alloy making). However, in Europe SF₆ is banned while in the United States the use is optional for industry. The SF₆ (used often in the Chinese magnesium industry) GWP impact has been calculated to be 22,800 kg CO₂eq per kg of SF₆ used (in other words, 22,800 times greater than 1 kg of CO₂ emitted). Usually 1 kg of SF₆ is required as cover gas per ton of melting magnesium, resulting in a 22,800 kg CO₂ equivalent per kilogram of melt magnesium.

⁶ The GWP average value for steel production by the integrated route is around 2.3 kg CO₂eq/kg steel.

⁷ Actually, the emissions from the electrolytic route strongly depend on energy sources for electricity generation (or energy-mix adopted) in the producer Country.

As raw materials are supplied, manufacturing and assembly steps include various conventional steps that also contribute to the carbon footprint of the final magnesium-based product⁸: a) transportation of ingots of the purchased alloy to the casting plant; b) ingot preheating, melting, and holding; c) casting into precision dies; d) finishing (i.e., fettling and grinding); e) quality control, with an average product reject rate of 3%; f) and transportation and assembly [4].

Table III shows data comparing different raw magnesium extraction processes for a simplified calculation of GWP per kg of magnesium less costly cast product.

Table III. Environmental Impact of Usual Magnesium Component (“Cradle-to-Gate” path) Expressed in GWP per kg of Magnesium Manufactured.

Plants	Primary Mg Production Manufacturing and Total Alloy Making (*) [kg CO ₂ eq /kg]	Manufacturing and assembly (**) [kg CO ₂ eq /kg]	Total cradle-to-gate [kg CO ₂ eq /kg]	References
Hydro Magnesium (I)	16.1	19.4	35.5	Tharumarajah et al.
Pigeon (China) (II)	43.3	19.4	62.7	Tharumarajah et al.
AM (Australia) (III)	27.9	19.4	47.3	Ramakrishnan et al.
Bolzano (Brazil) (IV)	13.8	19.4	33.2	Cherubini et al.
Magnetherm (France) (V)	17.6	19.4	37.0	Cherubini et al.

Footnotes:

(I) Recycling ratio: 70% primary: 30% secondary. All upstream stages included in alloy making. Use of SF₆ as cover gas.

(II) Mg ingots produced in China. For manufacturing and assembly stages: Mg ingots transported to North America or Europe for AZ91D alloy making.

(III) Electrolytic process employed in Australia (now ceased, but an interesting case for comparison). HFC-134a used as cover gas instead of SF₆. Alloy making by AM-Cast.

(IV) Current mix for electricity and natural gas for plant operations in Brazil. Alloy making as for the AM-Cast process.

(V) Thermal process formerly used in France up until 2001. Alloy making as for AM-Cast process.

(*) For comparison: GWP values for steel and aluminum production (not comprehensive of alloy making) are 2.3 kg CO₂ eq / kg steel and 22.4 kg CO₂ eq / kg aluminum respectively.

(**) GWP of common manufacturing steps derived from magnesium product LCA [4]; manufacturing steps include preheating of magnesium alloy ingots, melting, precision sand casting, fettling in a common open-loop recycling cycle.

⁸ In material flow, 80% of the recycling material has been assumed to be from scraps, rejects, and fettling waste.

Table IV shows the calculation of the CO₂ cut over the exit-gate-to-grave path, based on the general assumptions reported in Table II.

Table IV. Percentage Reduction of CO₂ Estimated with the Increasing Usage of Conventional Magnesium-alloys for the Manufacturing of Components Substituting Heavy Materials on Medium-size Passenger Vehicles.

Features	Scenarios			
	I	II	III	IV
Magnesium usage (kg)	93	125	219	256
Weight reduction (kg)	60	80	140	165
Total GWP of Bolzano route (kgCO ₂ eq)	-926	-176	2,004	3,405
Total percentage of CO ₂ cut ^(*)	-1.9%	-0.4%	4%	7%

(*) The percentage is calculated over the total amount of 48,450 KgCO₂ estimated for 200,000 km average travel-life distance.

As Friedrich and Schumann [5,6] stated in their early study on the potential uses of magnesium components for Volkswagen vehicles, a total average usage of around 150 kg could potentially be achieved in upcoming years. This value was actually close to the 154 kg targeted by a report issued in 2004 by the United States Automotive Materials Partnership (USAMP), the Automotive Metals Division (AMD) for the United States Council for Automotive Research (USCAR) entitled “Magnesium Vision 2020: A North American Automotive Strategic Vision for Magnesium” [7].

Taking into consideration the “unclean side” of the product cycle, around 150 kg of magnesium usage would be not sufficient for substantial benefits in terms of CO₂ percentage reduction, as shown in Table IV.

The challenge suggests the implementation of a double correlated target:

- from the alloy-design side, it is mandatory to reduce the substitution ratio (namely, usage ratio, refer to Table 2) from 1 to 2.64, switching to a 1 to 1.8, in order to reduce mass part of magnesium component substituting heavy materials;
- and from the process-design side, it is mandatory to drastically reduced the carbon-footprint of the unclean route steps needed in the manufacturing process of magnesium components.

While we think these two statements should be considered fundamental to the realization of novel, eco-sustainable magnesium-based alloy development [8,9], in the following we mainly focus on the alloy-design side issue.

The Alloy-Design Approach: The Environmental Problem Targeted

As previously explained and illustrated in Table IV, it is possible to refine the carbon-footprint of the entire life product cycle and decrease the amount of CO₂ by reducing the use of heavy materials. This assumption is tied to the strategy of alloy-design that should lead to a substantial reduction of the substitution ratio for low-carbon steel (namely, usage ratio) from 1: 2.65, as shown in Table 2, to a more effective 1: 1.8 value, calculated in Table V.

Table V. Mg-alloy Design and Weight Saving by Substitution of Medium Carbon Steel with High-Performance Novel Mg-Based Alloys.

Features	Mg	Steel
Density (kg/dm ³)	1.81	7.87
Resistance limit (MPa)	250	450
Usage ratio	1.8	1.0
Volume (mm ³)	180	100
Mass total (kg)	3.3	7.9
Total weight saving if 100 kg steel is substituted with magnesium High-Performance Novel Mg-Based Alloys (kg)	39	

(*) Mg-alloy Design and Weight Saving by Substitution of Medium Carbon Steel with High-Performance Novel Mg-Based Alloys.

Based on the LCA of the entire magnesium product manufacturing cycle, a positive balance of emissions over cradle-to-grave is achievable using a highly environmentally friendly and energy-saving program. There can be a drastic cut of kgCO₂eq emitted during the manufacturing steps by simply switching from 19.4 kgCO₂eq in Table 3 to a value close to 1-2 kgCO₂eq per kg of processed magnesium, primarily attributed to the use of a SF₆-free route and net-finishing shaping that eliminates additional machining and finishing. The total GWP could be decreased even further by using fully-electrical machines powered by renewable sources which is produced on-site or centralized, and by increasing the market share of the recycled magnesium. As a cleaner manufacturing cycle could be adopted, the GWP of cradle-to-exit gate cycle is lowered considerably. If approximately 175 kg of high performance materials are used onboard, around 11% of CO₂ can be saved on ICE vehicles, as shown in Table VI.

Table VI. – Percentage Reduction of CO₂ Estimated with the Increasing Usage of High-Performance Magnesium-Alloys Produced with Lower Environmental Impact Manufacturing Cycle.

Features	Scenarios			
	I	II	III	IV
Magnesium usage (kg)	63	85	149	175
Weight reduction (kg)	59	121	211	247
Total GWP of Bolzano route (kgCO ₂ eq)	1,260	3,556	6,891	5,592
Total percentage of CO ₂ cut(*)	2.6%	7.3%	14.2%	11.5%

(*) The percentage is calculated over the total amount of 48,450 KgCO₂ estimated for 200,000 km average travel-life distance.

Using this premise, the way to reduce CO₂ is to evaluate the performance of magnesium-based alloys and target its most effective usage, and do it in the most environmentally friendly process possible. Electric or hybrid motors can also benefit from the use of magnesium structural components. The potential

quantity of magnesium that can be used in electric cars is high because they do not reach the high temperatures that you find on ICE motors. This is a very interesting perspective as a relevant reduction in weight would greatly enhance the distance that vehicles powered with electric batteries can travel, which is currently the main objection to their wide spread use.

Integration of Process-Design and Alloy-Design Strategies for Lowering Carbon Footprint in Transport

Reducing the grain-size of metals is the key to improve mechanical properties of metal alloys in general. Nanostructured materials in particular (i.e. less than 1 micron grain size) achieve the highest performances allowed for a metallic material, with increased strength, toughness, corrosion resistance, and creep resistance. Various laboratory-scale methods exist to produce ultrafine structures through top-down routes, namely, solidification grain refining, severe plastic deformation of billet, etc., but nanostructured materials cannot be produced for large parts and no examples exist of industrial process lines for the fabrication of cost-effective components made of nanostructured metallic materials.

Currently, there are two approaches to produce bulk nanostructured materials:

- TOP-DOWN, consists of breaking down the bulk material into particles of nanometer size grain (in metals, e.g.: mechanical alloying, equal channel angular pressing, high-energy ball milling, accumulative roll bonding, high-pressure torsion).
- BOTTOM-UP, in which individual atoms or clusters are put together to form required nanoparticles (e.i: gas-phase synthesis, inert gas condensation, physical/chemical vapor condensation, plasma synthesis, etc.).

Extensive research has been conducted at CENIM [10-15] on ultrafine and nano-grain size magnesium-base alloys directly produced from a solid state (from Rapidly Solidified Powders, RSP), proving the effectiveness of Mg-based direct-powder extrusion. The effect of extrusion temperatures on the final microstructure and mechanical behavior for various Mg-based bi-phase systems has been extensively investigated. It was found that warm (below 0.5 T_m) extrusion of Mg-based alloys produced using the RSP route showed that the lower the compacting extrusion temperature, the higher the yield stress of monolithic product.

Several Mg-based bi- and ternary-phases systems were studied and optimized at CENIM. Generally speaking, microstructures provide high strength as RSPs are compacted by the extrusion process at around 250 °C. This is the temperature at which the inner shearing realized during extrusion is sufficient to finely distribute second phase particles. As very fine particles homogeneously dispersed in the Mg-matrix, a further strengthening mechanism begins, reinforcing the grain-refinement strengthening mechanism. For example, the MgZn_{3.3}Y_{0.43} alloy showed a yield stress of 410 MPa with 12% elongation [10]; this effect agrees with the general Hall-Petch equation valid for metals associated with the metallurgical phenomenon that produces mechanical properties according to grain-size refinement. Such high grain-refining leads to further benefits in terms of increased physical properties related to corrosion and creep resistance, and formability. At medium-low temperatures RSPs compacted by warm-extrusion exhibit super-

plastic behavior. These temperatures can vary from 200°C to 400°C, depending on the metallurgical Mg-system and the targeted results, as well as at high strain rates, attaining elongations to failure around 500% [15]. Generally speaking, impressive mechanical behavior was obtained at CENIM for this new process of magnesium alloy, as reported in Table VII.

Table VII. – Main key-features relative to ultra-fine powder warm extrusion of dual-phase magnesium alloys [10] versus average values for conventional steels.

Mg-based High Performance Novel Alloys from PWD metallurgy	Low-carbon steels	Medium carbon steels
up to 350 MPa YS	from 300 – 400 YS MPa	from 550 – 700 YS MPa
about 400 MPa UTS	from 500 – 600 UTS MPa	from 600 – 850 UTS MPa
elongation about 13%	elongation about 20%	elongation about 15%

Basing on such previous experience, the authors are currently involved in the European Project titled “Green Metallurgy 2020”⁹, a 2 ML€ project financed by European Community as one of the successful proposals submitted to the EU LIFE+ 2009 Program. This 3-years project officially started on the 1th of September 2010 and it aims to overcome main historic barriers that currently prevent magnesium from widely being applied as substitute ultra-lightweight material for the transport sector. The main scope of this project is to industrially scale to the production plant such impressive results obtained in laboratories, in order to commercially introduce a no-melting route that will be characterized by very low carbon footprint. This outcome shall be realized through application of specific technological steps tested for several years in CENIM’s laboratories and engineered with the support of the Politecnico di Milano’s Advanced Material Research Group in order to figure out how the basic process steps can be integrated in a single or in a double-step plant machinery. This fact is actually fundamental to greatly reduce the high costs related to magnesium part manufacturing using conventional melting processes. According to preliminary experiments, grain coarsening can be avoided. This prevents decaying of final mechanical properties of finished components thanks to maintain the ultrafine grain-size of the precursor material. As the historic barriers for high-performance usage of magnesium-base materials are removed, the magnesium industry will be able to increase their market share by capturing segments of the market now dominated by heavier materials like aluminum alloys and steels.

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⁹ This project is in accordance bases on the project proposal finally approved on 31 May 2010 for the co-financing by the EU Commission, Directorate of the LIFE+ Program.