Hydrogen-methane mixtures: vehicle tests performed by ENEA

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Abstract

Waiting for commercial ICE vehicles fuelled with 100% of hydrogen, a first approach towards a clean urban mobility can be sustained by the use of alternative fuel based on a blend of hydrogen and methane gases. In the last years many experiments were carried out to verify the opportunity to use Hydrogen-natural gas (HCNG) blend in natural gas (CNG) engines. This paper reports the results of experimental tests performed in ENEA Casaccia Research Center aimed to evaluate the environmental performances of a CNG vehicle when fuelled with a HCNG blend

Introduction

Among the strategies put in place to contain the environment deterioration, of the utmost interest are those processes aimed at improving the mobility of the resident population by addressing both the optimization of traffic flows and transport modes and the reduction of vehicles specific emissions.

In Italy the transport sector suffers from several criticalities, among them being the high rate of motorization, the highest in Europe, amounting to 0.59 vehicles per capita (source ACI 2009), indicating a clear polarization of mobility to the use of private vehicles (82% of terrestrial movements). Therefore the public transport certainly suffers the consequences of widespread car ownership, offering a service not in line with users demands, so not promoting measures such as modal shift and others.

In this operational framework, apparently blocked, vehicles emissions are subjectet to increasingly stringent legislation to limit the quantities of pollutants.

This continuous effort to reduce emissions harmful to human health is combined with the need to reduce greenhouse gas emissions, to achieve the objectives of the Kyoto Protocol. Also in this respect the Italian situation appear disappointing, with an emission of CO2 in 2008 (source ISPRA 2010), of 471 million tons. Withstanding the reduction target of 6.5% by 2012 (year of reference 1990) gross emissions have seen a growth of 7.6% and transportation has contributed to the increase in CO2 emissions for a 28%, witnessing the increased use of vehicles.

The difficulties experienced by the transport sector should also consider another equally important aspect, the dependency on fossil fuels. In fact, this subordination exposes the sector to market fluctuations in relation to political tensions in the areas of extraction. Moreover, it is common opinion, that the availability of fossil resources is limited in time with forecasts indicating a time horizon of 50 years for crude oil.

Facing these constraints requires the development of all available technological resources to make a change towards a net reduction in the consumption of fossil fuels in transportation.

In this scenario, electric vehicles are already a chance for the automotive industry, pursuing actively the policy of placing them on the market, but the psychological barrier is also stronger than the technological one, slowing the spreading of this technology. A truly epochal change seems to have been recognized by the "hydrogen revolution" with the use of hydrogen as clean energy and democratic, as defined by the economist J. Rifkin. Obviously this definition is function of the energy source used for its production, because hydrogen does not exist in nature and must be obtained from other compounds where is alloyed with other elements. The use of renewable energy along with the use of thermal and chemical processes is the best way to produce hydrogen without using fossil fuels and with almost no impact on emissions of CO_2 . The alternative is to use electrolysis to separate hydrogen from water (through last generation technologies able to assure efficiencies over 70%) using electricity from renewable sources. Availability of hydrogen for road transport has the main obstacle in the fact that at the moment there are no vehicles, currently available on the market, that can use hydrogen as fuel, and, last but not least, there is not an hydrogen distribution and refuelling network. Several experiments were conducted to demonstrate the feasibility of hydrogen in automotive applications using "fuel cells", but despite the good performance shown in the energy field, there are still problems related to performance degradation and system costs. Time will be required before vehicles could be run using fuel cell in a manner quite similar to conventional applications.

Hydrogen and methane mixtures for ICE: state of the art

Waiting for mature technologies for efficient use of hydrogen, a new possibility is offered by the mixture of hydrogen and methane. These mixtures are often referred to as "the bridge", which will make possible the transition to a hydrogen economy. In fact, the idea to combine a fossil fuel such as methane (itself already environmentally beneficial with respect to gasoline or diesel) and hydrogen can achieve several benefits. On the U.S. market hydrogen mixed with natural gas is already a reality with a registered trademark ® by hythane Co. LLC, which sells a mixture of 20% hydrogen (by volume) methane, corresponding to an energy content of 7 % the whole mixture. In other states research and demonstration projects are focusing on the possibility of using mixtures of hydrogen and methane with hydrogen content between 10% and 30%.

But what are the advantages of mixing of hydrogen and methane? Firstly, it should be noted that the addition of hydrogen to natural gas reduces the total carbon content of fuel and therefore it produces a drop in CO_2 emissions by a percentage proportional to the fraction of hydrogen introduced. The already effective action to reduce emissions of greenhouse gases produced by the use of methane in place of diesel / gasoline undergoes an additional positive effect with the hydrogenated mixtures. Another characteristic of hydrogen is to improve the combustion process as the flame front spreads more quickly, allowing a more complete carbon oxidation. The result is improved efficiency of the engine, so reducing the fuel consumption for the same distance.

Basically it is a "leverage effect" that cuts CO_2 emissions beyond the theoretical limit expected from the simple replacement of methane molecules with those of hydrogen. Moreover, due to the higher burning rate we are able to operate the engine with air to fuel ratios higher than those used with pure methane. Mixtures leaning lower nitrogen oxides emissions and is normally pushed up to tolerable threshold value, beyond which it is not possible to further reduce the fuel content due to incipient misfiring. The introduction of hydrogen, thanks to its superior combustion flame speed, can overcome this limitation and further reduce NO_x emissions. Indeed experimental studies have confirmed the lower emissions of NO_x with reductions of 50% compared to natural gas.

Among these, the experiments conducted in the laboratories of ENEA Casaccia, since 2006, two European projects, Hy-BONG and MHyBus, and a national projects, the “Hydromethane project”.

Infrastructures and normative aspects

Mixtures used in internal combustion engines does not require a high degree of purity, as in the case of fuel cells, and it is possible to use hydrogen produced from fossil fuels or as a by-product of other industrial processes. In the latter case, hydrogen production is mostly reused within the same plant, e.g. for energy production, leaving little quantities available for the use of motor vehicles.

Currently, most processes used to produce hydrogen generators are based on steam reforming from a fossil matrix (usually natural gas), hydrogen is obtained with CO_2 and it is necessary to rethink the processes of production hydrogen process to ensure a "CO_2 free".

An interesting process for the production of methane from the mixture is the so called "Carbon-saver" process in which the mix is directly obtained from a flow of natural gas through a plasma

process. It is not required to add hydrogen to the methane produced, and CO_2 emissions are with a lower if compared to conventional steam reforming.
Even if you use methane to produce hydrogen to be added to natural gas for the formation of mixture it remains a profit, due to the already mentioned leverage effects, with reduced CO_2 emissions. There is the possibility of capturing carbon dioxide and confine it in natural reservoirs to remove it from the atmosphere, but it is still in process of identifying the optimal technology choices still have to be defined.
The final prospects of the mixtures are then linked to the real capacity to produce hydrogen from renewable sources in sufficient quantities to meet the demand of transport.
In terms of distribution, in Italy the complete adaptability of the structures of final distribution to the needs of the mixture has been demonstrated by ENI (Colle Salvetti, near Livorno). Other studies have evaluated the susceptibility to the distribution of hydrogen in mixtures with methane, taking advantage of the pipeline network which already exists. The results were encouraging for both security and for compatibility, indicating a percentage of hydrogen from 15 to 20% as the maximum transportable current network.
From the safety point of view, current CNG engines are compatible with the use of mixtures, provided changes to the combustion control. It needs also check the components of the storage system and power supply, to ensure their compatibility with the percentages of hydrogen in the mixture to limit the effects of embrittlement of unsuitable materials.
With a view to a penetration of hydrogen into the world of transportation, it should be noted that the development of technology suffered until now a lack of legislation that would allow to put hydrogen powered vehicles on the road
The process to put on the road such vehicles have always been addressed through appropriate tests, agreed with the authorities responsible for issuing certifications. The need to expand this area to make a way for the commercialization of hydrogen vehicles has finally been implemented recently by the EU which has issued Regulation 79/2009, concerning the approval of hydrogen vehicles. The Regulation 406/2010 indicates needed requirements, for the vehicle and its components, in order to achieve approval. In particular for the components it is required to verify the compatibility between the materials and hydrogen in order to ensure the capacity of mechanical strength over time. The EU Directive stipulates that, for mixtures of hydrogen-methane will be issued a special regulation with regard to the mixing ratio and its use, but it points out clearly that the vehicles are comparable to hydro, for approval and to vehicles pure hydrogen: *"Hydrogen mixtures could be used as a transition fuel towards the use of pure hydrogen, to facilitate the introduction of hydrogen-powered vehicles in Member States where the natural gas infrastructure is good. The Commission should therefore develop requirements for the use of mixtures of hydrogen and natural gas/biomethane, especially a mixing ratio of hydrogen and gas which takes account of technical feasibility and environmental benefits. Regulation 2009/79 / EC)".*

The Hydromethane project

The region Emilia Romagna has shown interest in the use of hydrogenated mixtures of methane as fuel for transport applications. In particular the subject of interest is the possibility of introducing the Hydrogen-enhanced fuel within public transport sector. As a result of this attention the Region has launched a study program that developed into a demonstration project for the evaluation of the performance of Hydromethane mixtures for public transport vehicles. The "Hydromethane project" has involved two public transport companies, ATM and ATR, respectively of the Provinces of Ravenna and Forlì-Cesena, which participated actively in the project through the provision of two vehicles to perform the tests foreseen by the project.
The experiment aims to verify the performance in terms of emissions and energy consumptions of two natural gas vehicles fuelled with Hydromethane blends and in particular:
- to verify the operability of the means of transport with Hydromethane mixtures:
- to define could be the optimal mix to reduce energy consumption;
- to assess the reduction of CO_2 emissions;
- to characterize the emissions of pollutants subject to pollution control (HC, CO and NOx).

This involves testing of various hydrogen-methane mixtures in order to identify the mixture able to provide the best performance without producing collateral effects on the stability of combustion. Advance degree was changed in order to optimize combustion and reduce emissions of any unburned hydrocarbons and nitrogen oxides. No changes were done on air to fuel ratio with respect to pure methane usage, as the engine optimization by acting simultaneously on two parameters exclusively on road tests resulted too complex.

Experiments methodology

The experiments were carried out in two phases: the first one employing a long vehicle (12 mts.) provided by ATR Forli and the second a short vehicle (8 meters) from ATM Ravenna, a company that has a long experience (the first in Italy) with the use of natural gas in public transport.
In the first phase only a mixture of H2-CH4 (by volume in hydrogen content of 5%) was used and the results were compared with pure methane usage. In the second phase various formulations of the methane-hydrogen mixture were tested (5%, 10%, 5%, 20% and 25% by volume of hydrogen) to develop a full spectrum of benefits obtainable by the adoption of hydrogen mixtures for road transport. Hydrogen/methane mixtures were provided by Air Liquid, vehicles refuelling was possible thanks to a compression mobile station developed by ENEA in a previous programme, Fig.1.
During the first part of the activity sub-urban test cycles were used, urban cycle in the second phase. Further tests were additionally performed:
- Consumption measurements according to CUNA cycle;
- Test of PM emission in steady state conditions;
- Test for emissions of VOC in steady state conditions.

The tests were conducted on a street circuit which reproduces a typical operative path of the bus.
Emissions were measured directly on board by a proper instrumentation.

Tested vehicles

The first phase of tests was carried out using a 12 meters long vehicle produced by Bredamenarinibus, an Avancity Model 240 CNG owned by ATR Forli. The vehicle is a classic 12 meters one whose laden weight reaches 17 tonnes and can carry 93 people at a maximum speed of 60 km / h.
The vehicle is homologated as an EEV (Enhanced Environmentally friendly Vehicle, a term that indicates in a European standard emissive low-emission vehicles over 3.5 tonnes in category M2 or M3). Traction is provided by a four-stroke engine powered by natural gas with a maximum power of 205 kW @ 2200 rpm and maximum torque of 1000 Nm at 1400 rpm. The engine is a Mercedes-Benz M906 LAG turbocharged with intercooler features. The total displacement is 6880 cm3 six-cylinder in line. The storage facility for natural gas is composed of four cylinders on the roof of the vehicle for a total capacity of 1284 lt.
The second vehicle used for road tests is a short vehicle of Bredamenarini - 8 meters long - model Vivacity CNG, Fig.2, owned by ATM Ravenna; its laden weight reaches 13 tonnes, can carry 65 people at a maximum speed of 60 km / h. Equipped with the same engine Mercedes of Avancity model with the same engine capacity but weakened to 170 kW. The tanks for natural gas are always four and are located on the roof of the vehicles.
The engine is lean-burn type and this can reduce NOx emissions and obtain high efficiencies. Lean burn engines work in excess air reducing fuel consumption, being able to fully exploit the fuel used in the mixture. The more difficult ignition of the mixture is overcome by an appropriate mixing between air and fuel to make the air-fuel mixture more homogeneous in the cylinder.
In this way you avoid the nasty knock or misfire of the mixture (no burst). However, for a given output power (BMEP Brake Mean Effective Pressure on the Chart 1) engine knocks or engine misfire may occur when varying excess air and therefore it is necessary to monitor carefully the operating point of the engine in order to avoid critical areas.

Both vehicles were ballast using sandbags to simulate during the tests a payload equal to half the maximum load.

The mission

The road tests were conducted on a circuit located within the ENEA Research Center “Casaccia”. The choice to run the tests at this location was dictated by the need to be able to move safely on a road comparable to a typical bus path. The path chosen is representative of the needs of operability of the vehicle on the roadway for both the size of the roadway and the altitude. The size of the roadway is an important factor especially for the tests with the long vehicle. In fact, some sections are characterized by a 90 ° bend, (wich can usually be found at town road intersections), and the size of the roadway is such that the steering of the vehicle in relation to its overall dimensions is allowed. The circuit is 3.8 km long with an altitude varying between 132 and 152 m above sea level. The route alternates flat part with uphill and downhill that show no noticeable slope except for a single stretch of the journey.
The tests carried out on the road represented two types of transport services in urban and sub-urban areas. Therefore along the route obligatory stops have been placed every 250 m, equal to the typical distance between two stops for the urban public transport service. In total, the stops are 16 (the last segment has a reduced distance). During the simulation of the sub-urban service the bus has stopped every two stops in order to drive for stretches of 500 m. In the simulation of the urban service stops have been performed systematically in order to drive for stretches of 250 m.


Fig. 1: Refuelling station

Fig. 2: Tested bus


Fig. 3: Testing circuit

	Urban	Sub-urban
Length	3800 m	
Mean speed	20 km/h	22 km/h
Max speed	40 km/h	47 km/h
Time	730 sec	700 sec

Tab.1. Characteristics of the cycles

Results and Comments

Hereafter are the results of tests performed on the vehicles described above in accordance with procedures identified and agreed.

Tests on the circuit: energy consumption

The consumption evaluation is done by using two indirect methods: the carbon balance and the measure of the air-fuel ratio. The first method uses emissions (CO_2, CO and HC) from which the weight of total carbon emitted is derived. The carbon in the exhaust may only come from the fuel and therefore, for a known mass composition of the fuel, the amount of fuel consumed can be worked out. The air-fuel ratio (AFR) tells how much fuel is burned in relation to the input air. Knowing the load of the exhaust gases, the quantity of fuel burned in relation to the air can be traced. The measures of consumption shows similar results for both methods (see Table 2).

On the basis of the experimental results, it can be stated that using hydromethane mixtures produces savings in terms of mass of fuel used. The consumption reduction, in mass, varies between 5% and 20% for the short vehicle while it is about 7% for the long vehicle.
It is more representative to express the consumptions as natural gas equivalent quantitative (Table 2). In fact, since the mixtures have different levels of hydrogen content, they have different energy content and a proper comparison should be performed on the same metrics, such as bringing them back to kWh / km of fuel or energy content of methane, or indicating the G20 consumption that would have been necessary in order to provide the same energy content of the tested mixture.
By comparing the results of the CH4 consumption equivalent it can be noted that the mixtures at 15% and 20% show the same results, while the 25% mixture brings to a further decrease in energy consumption. The reduction in consumption can be ascribed to the improved performance of the engine due to the positive effect of the higher burning rate of hydrogen added to methane.
The efficiency appears to increase in a range between 4% and 15%, depending on the mixture.

Fuel consumption - g/km (Difference in consumption- methane- equivalent - %)				
	8 m		12 m	
	AFR method	Carbon method	AFR method	Carbon method
CH4	309.18	307.31	385.26	380.94
Hy 5%	293.56 (-4.2%)	292.98 (-3.8%)	358.96 (-6.0%)	352.20 (-6.7%)
Hy 10%	274.43 (-9.5%)	277.15 (-8.1%)		
Hy 15%	259.73 (-13.5%)	261.26 (-12.4%)		
Hy 20%	256.82 (-13.4%)	257.23 (-12.7)		
Hy 25%	248.76 (-15.0)	246.39 (-15.3)		

Table 2: Consumption measured on the cycle

CO2

Increasing the hydrogen content, CO2 emission decreases because carbon atoms are replaced with hydrogen atoms. CO2 emission, in moles, is equal to the percentage by volume of methane.
Table 3 shows the theoretical decrease in CO2 emissions obtainable by substitution of the carbon atoms with those of the hydrogen atom:

CV_{CH4}%	moles CO2/mole mixture	gCO2/mole mixture	$Kg_{Co2}/kg_{mixture}$
95	0.95	41.8	2.73
90	0.90	39.6	2.71
85	0.85	37.4	2.69
80	0.80	35.2	2.67
75	0.75	33	2.64

Table 3: Theoretical CO2 produced per mole of mixture

Table 4 shows the actual, measured data and the percentage reductions related to methane: the use of hydrogen-methane mixtures produces a reduction that goes beyond the mere substitution of carbon atoms.

CO2 emissions - g/km								
	8 m				12 m			
	CO2 g/km	ΔCO2 g/km	ΔCO2%	kgCO2/kg mixture	CO2 g/km	ΔCO2 g/km	ΔCO2 %	kgCO2/kg mixture
Hy 5%	782.06	-51.26	-6.2	2.67	950.75	-81.71	-7.9	2.70
Hy 10%	734.44	-98.88	-11.9	2.65				
Hy 15%	691.75	-141.56	-17.0	2.65				
Hy 20%	671.00	-162.32	-19.5	2.62				
Hy 25%	640.86	-192.46	-23.1	2.60				

Table 4: CO2 emissions

As a matter of fact, the reduction found is due to two factors: the reduction of the carbon atoms in the mixture and the decrease in consumption due to an increased engine performance.

If we define leverage effect the ratio between the actual and theoretical reduction, a reduction factor of 3 to 5 points (i.e. reduced emissions from 3 to 5 times the theoretical value) can be calculated, as shown in Table 5.

Leverage factor - reduction of CO2 emissions		
	8 m	**12 m**
CH4		
Hy 5%	3.97	5.11
Hy 5% 1g	4.93	
Hy 10%	3.68	
Hy 15%	3.38	
Hy 20%	2.79	
Hy 25%	3.97	

Table 5: Leverage factor

CO emissions

CO emissions were very low, down to the limit of instrument sensitivity, and did not show significant variation respect to the values already at the limit. Table 6 shows the emissions for CO observed with the different mixtures used:
HC emissions are substantially unchanged. The measure is inclusive of all HC compounds, both methane and non-methane. Most of the residues of organic compounds detected is still attributable to the unburnt methane emitted while the non-methane component should be very low.

NOx emissions

In literature it is reported that the use of hydrogen mixed with natural gas produces substantial reductions in emissions of nitrogen oxides (up to 50%) compared with emissions of mere natural gas. Even if the hydrogen would produce an increase in temperature in the combustion chamber and consequently increased emissions of NOx, the reduction of ignition timing advance and the leaning of the fuel / air ratio can offset this tendency and produce a reduction in the emissions of nitrogen oxides .
Table 6 shows the NOx emissions for both vehicles in urban and sub-urban cycles.
For the long vehicle, whose map of ignition was not changed, emission of NOx was reduced by 6%.
For the short vehicle, using the same mixture without altering the map of ignition (Hy 5%) the emission reduction is close to null.

	CO emissions - g / km		**NOx emissions - g / km**	
	8 m	**12 m**	**8 m**	**12 m**
CH4	0.07	0.64	3.76	3.56
Hy 5%	0.09	0.18	3.75	3.35
Hy 5% 1g	0.10		2.02	
Hy 10%	0.18		2.34	
Hy 15%	0.20		2.15	
Hy 20%	0.11		3.91	
Hy 25%	0.12		5.47	

Table 8: CO and NOx emissions (g / km)

Moving the adjustment of ignition of 1 degree in delay, the emission of NOx in the short vehicle is reduced by 46%. For values of 20 and 25% of hydrogen there is an increase of NOx for the reasons already mentioned.
It has to be remarked that on the short vehicle the interventions on the engine has been limited to the ignition advance and did not regard the degree of enrichment of the mixture (leaner mixture).
Increasing the levels of H2, the lower emissions of NOx are kept moving further forward the advance of ignition. With mixtures with high hydrogen content the adjustment of the advance is no

longer sufficient to guarantee a reduction of emissions. With 25% of hydrogen content with 4 degrees of advance you get an increased levels of NOx emitted at both low and high loads (see Fig. 4).

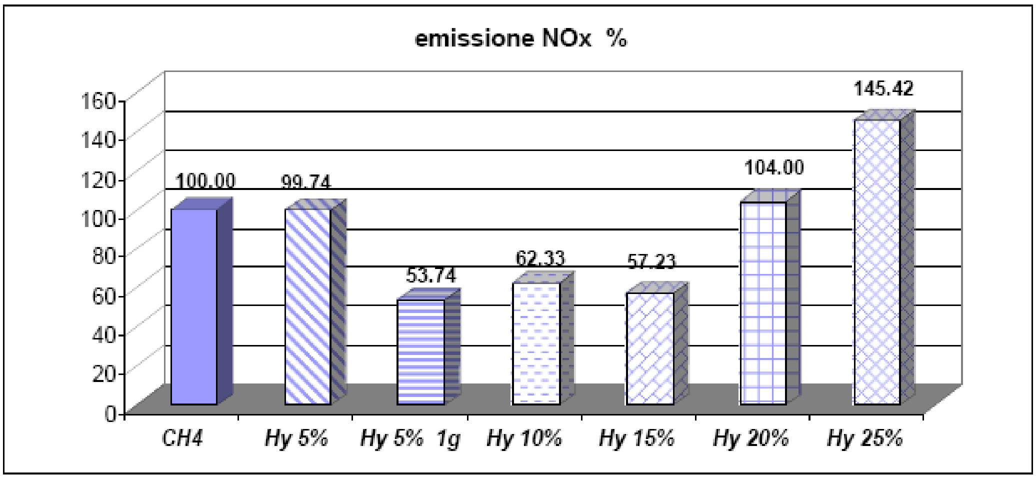

Fig. 4: NOx emissions in urban cycle

Conclusions

The experiment brought to positive conclusions on the use of hydrogen mixed with methane but also highlighted some aspects of technology for the best use of methane-hydrogen mixtures in normal commercial applications.

- The perspective of reducing CO2 emissions is one of the strengths for the use of hydrogen and methane mixtures. From the surveys performed, the actual reduction is beyond the theoretical one expected for the simple reduction of the carbon atoms with hydrogen atoms. This essentially occurs as a leverage effect happens, able to amplify the reduction in CO2 emissions beyond the theoretical limit. The increase of the reduction is attributed to the improved fuel efficiency that reduces energy consumption, and therefore the emission of combustion products. The leverage effect observed is between 3 to 5 times the expected theoretical value, calculated with the assumption of invariance of engine performance. The mixture of 25% produces a 25% reduction in CO2 emissions compared with theoretical 9% expected and is the largest absolute reduction detected. In relative terms, compared to the theoretical reduction, the 5% blend coupled with the delay of 1° of the ignition advance, is the one that has the strongest leverage effect.
- CO emissions are very low and show no appreciable changes at the top or bottom. Detections are at the limit of instrument sensitivity but the measures have shown that the CO are below the limits imposed by European standards, particularly for class EEV. Emissive CO levels do not seem to be strongly influenced by the driving cycle used for the tests, being very small in both the urban cycle Casaccia and the cycle CUNA.
- NOx have a more varying behavior compared to CO. In urban cycle Casaccia NOx emissions occur up to 47% less, using the 5% blend with 1° of delay in the anticipation of ignition. Similar values also occur with mixtures of 10 and 15% (with delays in advance of ignition of 1° and 2° respectively). By increasing the H2 content in the mixture, the NOx emissions are increasing and can not be recovered with the advance of ignition adopted (up to 4° at 25%). The strategy for the reduction of NOx, beside increasing the advance of ignition, is based on greater leaning of the fuel / air mixture. Indeed the addition of hydrogen allows better and faster combustion, which allows using a mixture of fuel poor even further reducing the emission of NOx.
- The emission measurements in urban cycle Casaccia showed that the vehicle is fully within Euro III for all the mixtures used. But only for 5% modified, 10% and 15% values are within the EEV standards. In cycle CUNA NOx emissions are within EEV classes only for the mixture of 5 to 15% and with sole methane. The other two formulations of the hydrogenated mixtures exceed these limits. The increase in emissions for blends of 20 and 25% is indicative of higher

temperatures in the combustion chamber due to the presence of hydrogen in large quantities. The reduction in burning time (reduced ignition advance of 4°) or the leaning of the mixture may serve to reduce NOx emissions. This is true even at levels of H2 over 15%.

New trials

In line with the just completed trial and in order to contribute to a sustainable mobility ATM is partner together with ENEA and ASTER of a project financed by the EC under the LIFE+ program, under the coordination of the Emilia-Romagna.
This new project, called MHyBus will last three years and intends to obtain ministerial authorization to use a mixture of hydrogen and methane as fuel for public transport.
More details on the project objectives and updates on its results can be found in the project website: www.mhybus.eu.

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