Introduction of H2 in the Natural Gas Network

Tugay Anıl Turan

Thesis to obtain the Master of Science Degree in

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Supervisors: Prof. Francisco Manuel da Silva Lemos

Prof. António Manuel Ferreira Marques

Examination Committee

Chairperson: Prof. Jorge de Saldanha Gonçalves Matos

Supervisor: Prof. Francisco Manuel da Silva Lemos

Member of the Committee: Prof. Edgar Caetano Fernandes

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I declare that this document is an original work of my own authorship and that it fulfils
all the requirements of the Code of Conduct and Good Practices of the
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Abstract

This document is prepared as a master's degree thesis which addresses Instituto Superior Técnico and AGH University of Science and Technology. The study presented in the thesis is developed by the author, REN - Rede Eléctrica Nacional, IST Lisboa and AGH Krakow cooperatively. The research presented aims to give an overview and suggestions to REN in their hydrogen injection project. In the following years the company wants to give a transferring service to hydrogen producers. Therefore, there is a need for investigation of the current equipment and its adaptations to hydrogen injection. The focus of this study which is defined by REN is hydrogen injection into natural gas grid in Portugal. There are several hydrogen injection projects ongoing in the literature. The thesis points out the most recent developments, obstacles, and constraints to adapt hydrogen technologies into an existing natural gas grid. In the full scope of the project, assessment of the REN's natural gas transmission system, identification of the legislation, regulation and security standards of hydrogen injection, analysis of the adaptations of steel pipeline and other assets, new approaches to gas monitoring, potential derating of the infrastructure depending on hydrogen percentage injected are studied.

Keywords

Hydrogen injection, natural gas grid, hydrogen and natural gas mixture, energy management, renewable energy, green hydrogen.

Table of Contents

List of Figures

List of Tables

List of Abbreviations

ACER – European Union Agency for the Cooperation of Energy Regulations

ANL - Argonne National Laboratory

ANN - Artificial Neural Network

ASME – American Society of Mechanical Engineers

CARI - Combustion Air Requirement Index

CCUS – Carbon Capture Utilization and Storage

CEN - European Committee for Standardization

DOT – U.S. Department of Transportation

EASEE - European Association for the Streamlining of Energy Exchange

EC – European Commission

ETS - The emissions trading scheme

EU – European Union

GC – Gas Chromatograph

GHG – Green House Gases

GIE - Gas Infrastructure Europe

GW - Gigawatt

HE – Hydrogen Embrittlement

HHV – Higher Heating Value

HIC – Hydrogen Induced Cracking

HSC – Hydrogen Stress Cracking

IEA – International Energy Agency

ISO – International Organization for Standardization

kWh – Kilowatt hour

LEL – Lower Explosive Limit

LHV – Lower Heating Value

LNG – Liquid Natural Gas

NREL- The National Renewable Energy Laboratory

NTP – Normal Temperature and Pressure

PCI- Project of Common interest

PNNL - Pacific Northwest National Laboratory

RED II – Renewable Energy Directive

REN - Redes Energeticas Nacionais

RT – Roman Tile

SNL - Sandia National Laboratory

SSC – Sulfide Stress Cracking

TEN-E – Trans-European Networks for Energy

TSO – Transmission System Operator

UEL – Upper Explosive Limit

WI – Wobbe Index

Chapter 1: Introduction

In the introduction chapter, the scope of the work is introduced, and a brief overview is given. The possible effects and benefits of the hydrogen injection is considered. From the literature review, research on the specifications of hydrogen, applications of hydrogen injection are combined. REN’s hydrogen project is explained and discussed.

1.1 Overview

Clean energy production, energy security and climate change are the hottest topics which humanity tackles in today's world. Organizations and developed countries have changed their perception of energy production. Green energy production, decarbonization and a future with less fossil fuels is the new target. Although, there are many solutions available, the interest to hydrogen technologies keeps increasing. There are several reasons why hydrogen has a key role in the energy sector. Firstly, hydrogen has a unique carbon free nature and high calorific value when it is combusted. Every other fuel which is used today emits CO_2 or CO. It is a well-known chemical fact that when H_2 enters a reaction with O_2 the main products are heat and water. All around the world, organizations are trying to avoid emitting CO_2 due to its obvious hazards to human health and to the nature. If, hydrogen becomes the primary energy source in the world, the carbon emissions caused by energy production will be simply zero. Secondly, hydrogen may provide energy security everywhere in the world. Natural gas, oil, coal, and other fossil fuels are abundant in specific locations all around the globe. While some locations are rich and fortunate about their resources, some locations are without any energy sources. However, hydrogen can be produced from off-peak electricity and access electricity produced from renewable energy sources. This kind of hydrogen production gives a key alternative to energy storage methods which are still developing today. Thirdly, hydrogen has the potential to replace almost all the fossil fuels. It can be used in natural gas systems, powerplants and even in the mobility devices. Although, there are numerous benefits of hydrogen in energy sector, the infrastructure for hydrogen is still insufficient. That is why, one of the most logical usage of hydrogen is the injection into the natural gas networks. Within the scope of the thesis REN wants to investigate the hydrogen injection possibilities in the near future. According to the new strategy developed by Portuguese government 5% hydrogen injection into the natural gas system will be an obligation. The counterparts of these law will be seen in European Union in the near future as a part of decarbonization and green energy act. REN plans to transfer hydrogen provided by producers. The company will not produce hydrogen or develop a business plan focusing on hydrogen technologies. Instead, REN will be responsible for hydrogen and natural gas blending which will be relatively simple. The operation of hydrogen injection seems easier than the creating a circular hydrogen economy. However, there are still a lot of concerns about the system integration, operation costs, safety, and complexity. Under the scope of this master's thesis all the relative aspects are considered and investigated.

1.2 Motivation and Content

To date, the hydrogen injection is considered only up to a certain percent by volume. In the future, it is highly possible to witness the natural gas networks to become 100% hydrogen grids. It is fair to make this assumption because, with the current energy policy there is a trend towards using less and less fossil fuels. The main motivation of this thesis is to investigate hydrogen injection potential in RENs transmission network. While investigating this potential thesis points out crucial technical points, possible benefits and the disadvantages of this new business plan. Within this plan REN as the biggest transmission system operator (TSO) in Portugal should determine possible injection points and hydrogen injection quantities. After the determination, REN will inform H_2 producers about the process so that they can apply with their injection projects. While injecting hydrogen REN still has several responsibilities and contracts to maintain. Therefore, the thesis focuses on maximum possible hydrogen mixing with natural gas by volume while satisfying the previous requirements.

This thesis particularly motivated by a future which energy transition is provided by hydrogen technologies. Under current circumstances 100% hydrogen in the natural gas network looks like a dream that requires a lot of adaptations and investment. However, the purpose of the study is to justify hydrogen injection into today's natural gas infrastructure up to a certain percentage from technical and legal point of view. In order to determine whether the Portuguese transmission system is available for hydrogen injection, the thesis consists of several chapters. In these chapters, latest state of art literature, different applications, laws, constraints are presented. From REN's point of view, required adaptations, limitations, technical and economical constraints are considered and commented.

In this thesis there are 7 main chapters, and the general structure of the thesis is given below.

- Chapter 1- Introduction.
- Chapter 2- Assessment of hydrogen injection and its constraints on technical point of view.
- Chapter 3- Identification of legislation, regulation and security standards for H_2 injection in the natural gas network and also technical aspects
- Chapter 4- Analysis of the changes and adaptations in the system
- Chapter 5-Gas quality monitoring
- Chapter 6- Potential degrading of infrastructure due to hydrogen
- Chapter 7- Conclusion

1.3 Hydrogen in Energy Sector

Hydrogen is the simplest and most abundant element on earth. It consists of only one proton and one electron. Hydrogen can store and deliver usable energy, but it doesn't typically exist by itself in nature and must be produced from compounds that contain it. Hydrogen can be produced from diverse, domestic resources. Currently, most hydrogen is produced from fossil fuels, specifically natural gas.[1] However, the hydrogen is generally used to decrease the carbon emissions that is why producing hydrogen from off-peak electricity or from renewable sources such as solar and wind will be more common in the long term.

Hydrogen is an energy carrier, not an energy source and can deliver or store a tremendous amount of energy. Hydrogen can be used in fuel cells to generate electricity, or power and heat. Today, hydrogen is most used in petroleum refining and fertilizer production, while transportation and utilities are emerging markets.[1]

The nature of hydrogen separates it from other fossil fuels. When hydrogen consumed in a fuel cell, the reaction gives only water, electricity, and heat. That is why hydrogen plays a key role in a carbon neutral future. Clean hydrogen, being produced from renewables, nuclear or fossil fuels with carbon capture utilization and storage (CCUS) can help to decarbonize a range of sectors, including long-haul transport, chemicals, iron, and steel, where it is proven difficult to reduce emissions. [2] Considering the fact that hydrogen can be used commercial, industrial and transportation purposes, there is a high potential that hydrogen will replace most of our fossil fuel primary energy sources. It is known that many cities all around the world suffer from poor air quality. Besides all those industrial benefits, hydrogen can also help to improve air quality in cities and improve energy security. [2]

The physical properties of hydrogen are well known. It is the smallest of all atoms. Consequently, hydrogen is the lightest gas, about 8 times lighter than methane. In most of the applications, the volume available for fuel tanks is limited. This also applies to natural gas industry since, the diameter of pipelines cannot be changed at will. Therefore, for most meaningful assessment would refer the energy content of fuel gases to a reference volume. It is also proper to use the higher heating value for the energy analysis because it represents the true energy content of the fuel based on the energy conservation principle of the first law of thermodynamics. Since the production of hydrogen is governed by the heat of formation or the higher heating value, its use should also be related to its HHV energy content. The following volumetric higher heating values for hydrogen and methane at 1 bar and 25°C is presented in Table1. [3]

Table 1 Density and heating values of hydrogen and methane.[3]

	Units	Hydrogen (H_2)	Methane (CH_4)
Density at NTP	kg/m^3	0.09	0.72
Gravimetric HHV	MJ/kg	142.0	55.6
Volumetric HHV	MJ/m^3	12.7	40.0

1.4 Benefits of Hydrogen and Hydrogen Natural Gas Blend

Hydrogen projects and research activities related with it has been performed all around the globe because there are certain benefits. Firstly, adding hydrogen to natural gas can significantly reduce greenhouse gas emissions if the hydrogen is produced from low-carbon energy sources such as biomass, solar, wind, nuclear, or fossil resources with carbon capture and storage. Any social or environmental benefits associated with sustainable hydrogen pathways could arguably be attributed to natural gas with a hydrogen blend component in proportion to the hydrogen concentration.[4]. Secondly, because downstream extracted hydrogen can be used in automotive sector potential benefits arise from reducing petroleum consumption and improving air quality by reducing sulfur dioxide, oxides of nitrogen, and particulate emissions. [4] As the only carbon-free and possessing the highest energy content compared to any known fuel which is seen in Table 2, hydrogen is globally accepted as an environment friendly secondary form of renewable energy. Thirdly, hydrogen injection into natural gas does not require a massive change in the infrastructure. As it is investigated in this thesis when hydrogen is added to the natural gas in low volume percentages, there is no need to change the end user appliances and adaptations in the natural gas will be minor considering the side of the industry. According to Nikolaidis a further advantage is that, supported by appropriate storage technologies, hydrogen can be utilized for domestic consumption as it can be safely transported through conventional means, and to be fed to stationary fuel cells, it can be stored as compressed gas, cryogenic liquid or solid hydride. [5]

Table 2 Energy content of Hydrogen and other fuels.[5]

Fuel	State at ambient temperature and pressure	HHV(MJ/kg)	LHV(MJ/kg)
Hydrogen	Gas	141.9	119.9
Methane	Gas	55.5	50
Ethane	Gas	51.9	47.8
Gasoline	Liquid	47.5	44.5
Diesel	Liquid	44.8	42.5
Methanol	Liquid	20	18.1

One of the main problems in renewable energy sources is the energy storage. Hydrogen provides an alternative solution to this problem. Brandon mentions in his study which enlightens a great benefit of hydrogen technologies. Hydrogen can store larger amounts of energy per unit volume than other large-scale energy storage options being considered: it has over 200 times the volumetric energy storage density of pumped hydro storage and 50 times that of compressed air. Moreover, hydrogen can be used for both intra-day and inter-seasonal storage, enabling a greater degree of flexibility with day/night and seasonal variations. [6]

1.5 International Projects of Hydrogen and Natural Gas Blending

Hydrogen injection into natural gas has never done in Portugal before. The injection concept has come from the new legislations. Therefore, REN is not the first mover in the field of hydrogen. Globally projects have been developed with a broad range of injection percentages and investments. These projects can be listed as HyDeploy and H21 North of England from U.K. Snam from Italy, Dunkirk from France and HyBlend from U.S.A. Before understanding REN's hydrogen injection project, several projects have been chosen from different countries to give an overview on the current application, technological improvements, research, and development activities.

1.5.1 HyDeploy

HyDeploy is United Kingdom's first hydrogen blending deployment project. The project is developed by Cadent Gas, Northern gas networks and Klee University. The main goal of HyDeploy cooperation is to find scientific proof that hydrogen can be injected into the natural gas network up to 20% by volume. As it is claimed in the HyDeploy report if 20% of hydrogen is injected across the UK the carbon dioxide emissions would decrease 6 million tons annually. With this motivation HyDeploy tested a pilot network which includes 100 homes where 20% of hydrogen of hydrogen injected into their natural gas system under seven criteria. These criteria are listed below. [7]

1. Short term appliance behavior.
2. Long term appliance behavior.
3. Effect of hydrogen blend on material.
4. Risks of poor mixing.
5. Fire and explosion risk.
6. Hydrogen detection.
7. Customer perception.

The study claims that up to 20% hydrogen injection into UK's network is almost as safe as using 100% natural gas. Based on literature and the experiment conducted gas quality standards, material durability and end use safety/quality are satisfied.

1.5.2 H21 North of England

H21 North of England is the evolved version of H21 Leeds City Gate project. The main goal of the project is to investigate technical and economic feasibility of 100% Hydrogen conversion in the natural gas network in the north of England. Within the 2050 decarbonization vision, project was designed to deliver heat to the costumers with the same price and lowest carbon emissions possible. According to the studies performed up to now, the project claims the following results.

1)The gas network has the correct capacity for such a conversion.
2) It can be converted incrementally with minimal disruption to customers.
3) Minimal new energy infrastructure will be required compared to alternatives; and
4) The existing heat demand for Leeds can be met via steam methane reforming and salt cavern storage using technology in use around the world today. [8]

1.5.3 Hy Blend

The National Renewable Energy Laboratory (NREL) in the United States has started the HyBlend to accelerate the potential for blending hydrogen with natural gas. The project focuses on technical constraints of hydrogen injection into existing natural gas network in the U.S. According to their experience and expertise other important laboratory partners are put together in the collaborative research and development project. HyBlend states that there are several projects worldwide which demonstrates hydrogen blends up to 20% is economically and technically feasible. However, the effects on the materials and the equipment in the long run is not well-understood. That is why they have determined three research areas, and these areas are shared with other laboratories based on their capabilities and existing research profile. According to NREL’s document these areas and the responsibilities are given below.

1. **Hydrogen compatibility of piping and pipelines:** Sandia National Laboratory (SNL) and Pacific Northwest National Laboratory (PNNL) will conduct evaluations to estimate the life of metal and polymer piping and pipeline materials (e.g., steel and polyethylene) when blends are used. This information will be incorporated into a publicly available model that can be used to estimate pipeline life given key engineering assumptions.[9]
2. **Life-cycle analysis:** Argonne National Laboratory (ANL) will analyze the life-cycle emissions of technologies using hydrogen and natural gas blends, as well as alternative pathways such as synthetic natural gas.[9]
3. **Techno-economic analysis:** NREL will quantify the costs and opportunities for hydrogen production and blending within the natural gas network, as well as alternative pathways such as synthetic natural gas. [9]

Chapter 2: Assessment of Hydrogen Injection and Technical Constraints

In chapter 2 hydrogen injection into natural gas systems undergoes an assessment. The purpose of this assessment is to define and evaluate technical issues to be addressed when hydrogen injection starts. 4 significant assessment areas which may affect the current transmission system are found. These areas can be simply named as safety, impact to end user, durability and integrity.

2.1 Introduction

Every organization or company in the world would like to investigate the effects of their new business model to the existing model. In REN’s case, the company already has functioning business model and a transmission system. It is known that building a gas transmission system only for hydrogen will come with great capital costs. Therefore, introducing hydrogen into an existing pipeline network with small retrofitting and adaptations is more cost effective in the near future. In one hand, injecting hydrogen will provide numerous benefits like compliance with new law and decarbonization. On the other hand, there are possible disadvantages which may affect the current equipment and operations. REN wants to inject maximum possible hydrogen by volume into their natural gas network. While injecting hydrogen there are crucial topics to be examined. In this chapter, these topics are determined as safety, leakage, durability, integrity and impact to the end user.

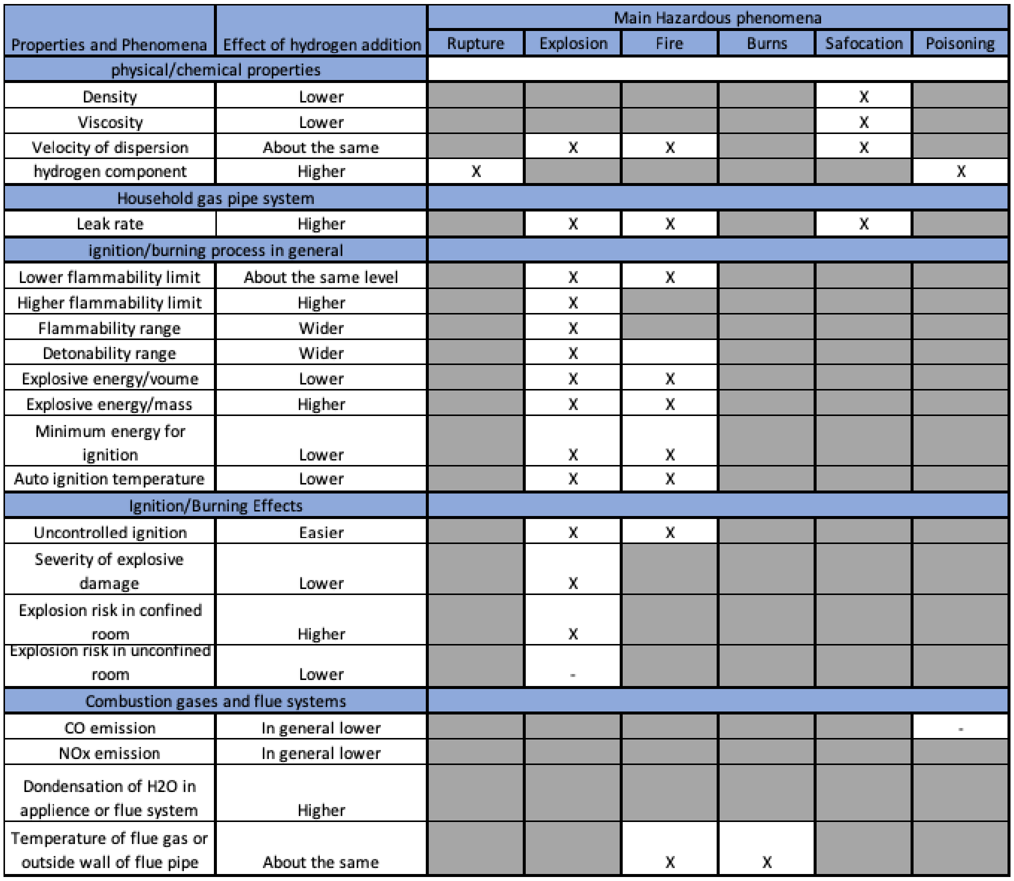
2.2 Safety

Safety is one the biggest concerns when hydrogen is the main topic. Whether it is industrial usage of hydrogen or cars fueled by hydrogen, scientific authorities and public raise questions about the safety. Hydrogen usage in several sectors is relatively a new technology and well-known flammable nature of it seems like the two important reasons why the technology faces a resistance. However, the natural gas industry has a developed approach to safety, because of the significant hazards that has been faced in the past. When the hazards from two industry is classified, there are two major areas which catch the attention. Both natural gas and hydrogen poses heat and explosion hazards due to their flammability and compression energy in large scale operations. Therefore, hydrogen injection into natural gas network will not alter the hazards significantly. Instead, the severity and the probability of the incidents will increase according to the hydrogen injection percentage by volume.

Pure hydrogen has several properties that would require a change in approach to safety. These include having a colourless flame, being incompatible with odorants used in natural gas, high ignition frequency, broad flammability limits, and a positive Joule-Thompson coefficient (that is, it heats up when expanding, whereas natural gas cools down). However, REN does not plan to build a 100% hydrogen network that is why if a lean-hydrogen mixture is used the impacts on safety will be more manageable. Surely, the parameters used for quantifying risk will change in order to have a complete risk assessment. There are multiple factors which must be considered to assess the safety concerns of hydrogen injection into existing Portugal’s natural gas network. That is why it is almost impossible to make general comments about the safety. Even from location to location the risk assessment results likely to vary since hydrogen has a broader range of conditions under which it will ignite. [4]

To fully understand the safety concerns caused by hydrogen injection the difference in specific phenomena compared to natural gas alone must be studied. IEA has identified properties, phenomena, hazards which are associated with natural gas. Under this study they also have investigated the effect of hydrogen addition into natural gas. The effects of hydrogen addition to natural gas properties and phenomena are shown in Table 3. [10]

Table 3 The effects of hydrogen addition to natural gas properties. (Adapted from IEA) [10]



Properties and Phenomena	Effect of hydrogen addition	Main Hazardous phenomena					
		Rupture	Explosion	Fire	Burns	Safocation	Poisoning
physical/chemical properties							
Density	Lower					X	
Viscosity	Lower					X	
Velocity of dispersion	About the same		X	X		X	
hydrogen component	Higher	X					X
Household gas pipe system							
Leak rate	Higher		X	X		X	
ignition/burning process in general							
Lower flammability limit	About the same level		X	X			
Higher flammability limit	Higher		X				
Flammability range	Wider		X				
Detonability range	Wider		X				
Explosive energy/voume	Lower		X	X			
Explosive energy/mass	Higher		X	X			
Minimum energy for ignition	Lower		X	X			
Auto ignition temperature	Lower		X	X			
Ignition/Burning Effects							
Uncontrolled ignition	Easier		X	X			
Severity of explosive damage	Lower		X				
Explosion risk in confined room	Higher		X				
Explosion risk in unconfined room	Lower		-				
Combustion gases and flue systems							
CO emission	In general lower						-
NOx emission	In general lower						
Dondensation of H2O in applience or flue system	Higher						
Temperature of flue gas or outside wall of flue pipe	About the same			X	X		

The overall risks posed by the existing natural gas pipeline system can be quantified, and these results are used as a baseline for comparing risks associated with hydrogen blends. However, in general, natural gas systems pose a lower risk of severe accidents than do other large-scale energy systems such as coal, petroleum, nuclear, and hydropower. [11]

Gas Technology Institute (GTI) has calculated risk factor of the natural gas network in United States. The severity of the hazards is quantified by a scale from zero (insignificant hazard) to fifty (severe hazard). The ranking of the hazards can be seen in Table 4. In this study they have also calculated the possible effects of hydrogen injection in to calculated risk factor. The research presents risk factor and overall risk of 100% natural gas, and various hydrogen injection levels by volume. [4]

Table 4 Significance of hazard classification by GTI.[4]

Significance of Hazard	Ranking Assigned
Severe	50
Moderate to Severe	40
Moderate	30
Minor to Moderate	20
Minor	10
None	0

Eight possible failure mode which are faced in the natural gas industry is taken from DOT 2007 Reports and evaluated under hydrogen injection conditions.[12] Although the natural gas network properties in the U.S. and Portugal are different, the risk assessment enlightens the future operations of REN. Especially, hydrogen addition under 20% gives clue on how risk factors will be affected in Portugal. According to GTI and NREL hydrogen injection less than 20% by volume would result in minor increase in the risk of ignition, explosion, and leakage. In the risk assessment the focus is the transmission lines. Naturally, in the service lines the overall risk differs because the service lines are often found in confined spaces where gas leakage would be more likely to accumulate. This study aligns with RENs vision because it was conducted on an existing pipeline and not in a newly designed pipeline system for hydrogen. The severity assessment of the failure modes in steel pipes are given in Table 5. The original study calculates the overall risk factor with the combination of different pipe material. Since, REN has only steel pipeline the overall risk factor becomes direct values of severity.

Table 5 Possible failure modes of steel pipes and their severity assessment.[12]

Failure Type	For Steel Pipe Material
Corrosion	50
Material Defect	30
Natural Force	30
Excavation	50
Other Outside Force	10
Equipment	30
Operation	30
Other	10

2.3 Impact to the End User

Natural gas from the distribution grid is most used in households for cooking or heating. As it is discussed in the safety part in this chapter, when hydrogen is injected into natural gas grid further safety concerns arise. In case of homes as end user, leakage and the risk of ignition are the most significant concerns. Therefore, depending on the hydrogen percentage it may be necessary to add an odorant to hydrogen to improve detectability. It may also be necessary to add a colourant as, unlike natural gas, a pure hydrogen flame is almost invisible. [13] However, several studies have discussed the issue of maximum hydrogen blend levels at which no or minor modifications would be needed for end-use systems, including appliances such as household boilers or stoves.[14]

Ranges noted as being acceptable generally for end-use systems fall within 5%–20% hydrogen, and most discussions note types of changes, precautions, or costs associated with higher blends. For example, Haines et al. (2003) estimate the cost of upgrades in the United Kingdom, Netherlands, and France with respect to modifications required for 3%, 12%, and 25% hydrogen blends[15] Whilst according to Hodges and Gerry most modern appliances should be capable of burning hydrogen blends of up to 20 vol%, above this level it is likely that appliances would need adjustments.[16] In REN's case it would not be a problem in the near future since their goal is to inject hydrogen less than 20% by volume initially.

As noted by Florisson (2009), end-use requirements are generally the most restrictive conditions on increasing hydrogen blend levels in natural gas. The conditions determining a maximum hydrogen blend level that does not adversely influence appliance operation or safety vary significantly and include the composition of the natural gas, the type of appliance, and the age of the appliance.[17] The impact of hydrogen blends on industrial facilities must be addressed on a case-by-case basis, and stationary gas engines likely will require changes to control systems.[18] These facilities are more likely to be connected to high pressure pipelines or have their own direct supply of natural gas. Introducing hydrogen blends into combustors for equipment such as gas turbines will alter the combustion characteristics. However, a considerable amount of work has been performed in recent years to design burners suited to these characteristics. [13]

In the scope of HyDeploy project in UK the home type of stoves are tested under different hydrogen injection conditions. The main conclusion of the project is 20% hydrogen addition by volume in natural gas would not affect household stoves in the short period of time. In Figure 1 stove flames with respect to hydrogen injection is given.

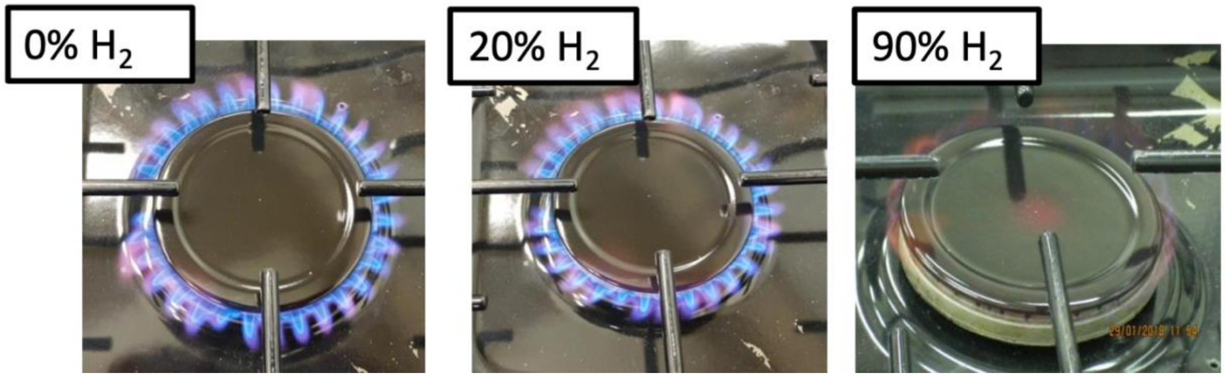


Figure 1 Stove flames with 0%,20% and 90% hydrogen injected in natural gas grid. (HyDeploy Webpage)

2.4 Integrity of the Pipeline

When injecting hydrogen into existing natural gas distribution system, the most important asset is the pipeline system. Therefore, its integrity plays a key role in all the operations of this project. The integrity management for natural gas pipelines is based on operation conditions for only transporting natural gas. If REN injects hydrogen into natural gas system, the operating environment will certainly change. This change may accelerate crack propagation or fatigue failures from the existing defects, and thus adversely impacts pipeline integrity. [4]. Depending on the hydrogen injection percentage by volume, there may be a need for a new integrity management criterion because it is possible that some certain defects which does not affect the integrity of the pipeline system under current criteria may become hazardous due to the material property change in hydrogen containing environments. Therefore, the maintenance costs for distribution systems under hydrogen service likely will increase because these systems will need to be inspected more frequently and likely will require additional leak detection systems. [18]

American Society of Mechanical Engineers (ASME) determined major threats to the integrity of a natural gas infrastructure. According to the ASME major threats are classified in 3 major areas namely time dependent threats, stable threats, and time independent threats. All these threats which are given in Table 6 required more caution and inspection when hydrogen gets injected into the system.[19]

Table 6 Major threats to distribution infrastructure according to ASME Standard B31.8S.[19]

Major Threats to Distribution Infrastructure According to ASME Standard B31.8S (Managing System Integrity of Gas Pipelines)		
Time Dependent Threat	Stable Threat	Time Independent
External Corrosion	Manufacturing Related	Third Party/Mechanical Damage
Internal Corrosion	Constuction Related	Incorrect Operation
Stress Corrosion Cracking	Equipment Related	Weather related/Outside Force

The above threats are defined primarily for natural gas transmission system which operate at high pressure and are constructed predominantly with high strength steels which are coated, wrapped or bare.

2.5 Durability

Hydrogen injection into the existing natural gas network is a pipeline extensive procedure. In other words, the pipeline system is the most important component which needs to be considered. European gas network is a diverse system which means there are many different types of pipes all around the Europe. However, the focus of this study will be on REN's pipeline assets which are API 5L Grade B, X42, X52, X60 and X70 steel pipelines. These type of steel pipelines have existed more than 50 years by now. Therefore, there are numerous research and experiment which are performed.

Hydrogen embrittlement (HE) is a metal's loss of ductility and reduction of load bearing capability due to the absorption of hydrogen atoms or molecules by the metal. The result of hydrogen embrittlement is that components crack and fracture at stresses less than the yield strength of the metal. [20] Moreover, hydrogen embrittlement is one of the most important phenomena explaining degrading of steel pipes like API 5L series under hydrogen involving conditions. It was found that HE has an important effect on the yield strength, tensile strength, elongation to failure, reduction of area, crack propagation resistance, fatigue life, hardening rate and nano-elastic and nano-hardness properties. [21]

API 5L X52 is the most used pipeline steel all around the Europe. That is why the current attention has been paid to HE of API 5L X52 steel under operating conditions.[22] There are different codes which indicate the failure pressure involving flow stress. The most used codes are ASME B31G, SHELL-92 and DNV RP F-101. Under the light of these codes several studies are performed to understand the effect of hydrogen embrittlement on mechanical properties of the pipeline steel material. Hadj Meliani at al. investigated the effect of hydrogen on the master failure curve of steel API 5L X52 and X70 and concluded his study with a decrease in the fracture toughness because of HE.[23]

Djukic et al. studied detailed analysis of deleterious hydrogen effects on the mechanical properties of steels at different scale from macro to nano and prescribe preventive measures to prevent failure in the pipeline systems.[24] There are also specific experiments and studies on the steel types that REN has in their pipeline system. For instance, Bellahcene et al. focused on the effect of hydrogen on mechanical properties of API 5L X70 pipeline steel. In this study significant decreases in the yield stress (8%), ultimate stress (12.7%) and elongation (31.9%) is determined. [25] Similarly Park et al. studied on API Grade 60 steal and observed decreases in yield stress (2%), ultimate stress (7%) and elongation (40%). [26] Finally, Elazzizi et al. performed experiments on API 5L X52 and concluded that there is a decrease in yield stress (2.5%) and elongation (38%) under hydrogen embrittlement phenomena.[27] The research mentioned above are summarized in Table7.

Table 7 Literature summary on different steel types and effect of hydrogen in their mechanical properties.

		Researcher and the Steel Type		
		Bellahecene et al. API 5L X70	Park et al. API Grade 60	Elazzi et al. API X52
Investigated Parameter	Yield Stress Decrease	8%	2%	2.5%
	Ultimate Stress Decrease	12.7%	7%	
	Elongation Decrease	31.9%	40%	38%

As it is shown in several research, hydrogen embrittlement and damage are one of the biggest concerns in high pressure transmission steel pipelines. However, the hydrogen effect on the steel strongly dependent on the hydrogen concentration in natural gas and operating pressure. That is why It is crucial to determine the acceptable hydrogen percentage which can be blended into natural gas without negatively impacting pipeline system.

The only impact of hydrogen in the steel pipelines is not hydrogen embrittlement. Carbon steels show accelerated fatigue crack growth and degradation in fatigue endurance limits when expose to hydrogen. Moreover, crack growth from existing defects may be enhanced by the addition of hydrogen due to the reduced ductility of steel, and fluctuation of the operating pressure in the pipeline may accelerate this effect. [28] When hydrogen embrittlement, crack growth from existing defects is considered, according to GTI, adding up to 50% hydrogen into the natural gas transmission pipelines may not cause catastrophic failure in NaturalHy project. [28]

Chapter 3: Identification of Legislation, Regulation, and Security Standards for H_2 Injection in the Natural Gas

Portugal has defined their hydrogen strategy under the light of European regulations. However, there is no solid law for hydrogen injection in Portugal. Therefore, different types of legislations and regulations are investigated to understand the possible limitations by law. Chapter 3 determines and combines laws, regulations, security standards for hydrogen injection into natural gas systems.

3.1 Introduction

European Union has announced its intention to be carbon neutral by 2050. This initiative on the path of a carbon neutral future has major impacts on the natural gas sector. Natural gas plays a significant role in the energy mixture for many countries that is why natural gas industry has well-established regulations, especially in Europe. However, these law and regulations need adaptations because of the developing nature of energy production and transportation. Hydrogen and other low carbon gases provide an opportunity to meet the decarbonization targets but even so, many of the law fails to mention hydrogen injection into natural gas systems. The purpose of this chapter is making hydrogen injection into natural gas network safe, reliable, consumer-friendly, efficient, and environmentally friendly by law. Therefore, European law and legislations are examined under several categories.

3.2 A Review of European Union’s Decarbonization Policy

3.2.1 Clean Planet for All

The European Commission came up with the vision of “Clean Planet for All” as a part of its 2050 long term strategic plan on climate change. [29] It was published in November 2018 and pointed out a vision for a sustainable future.[30] The purpose of this long-term strategy is not to set targets, but to create a vision and sense of direction, plan for it, and inspire as well as enable stakeholders, researchers, entrepreneurs and citizens alike to develop new and innovative industries, businesses and associated jobs. In alignment with the purpose of the long-term strategic plan, The Clean Planet for All looked at the combinations of available options for Member states which will lead to a climate neutral European economy.[31] Finally, the vision identified seven strategic areas which requires a joint action to achieve carbon neutrality. These strategic areas can be named as:

1) Maximizing of the benefits from Energy Efficiency including zero emission buildings. [31]

2) Maximizing the deployment of renewables and the use of electricity to fully decarbonize Europe’s energy supply. [31]

3) Clean, safe, and connected mobility. [31]

4) A competitive EU industry and the circular economy as a key enabler to reduce greenhouse gas emissions. [31]

5) Develop an adequate smart network infrastructure and inter-connections. [31]

6) Reap the full benefits of bioeconomy and create essential carbon sinks. [31]

7) Tackle remaining CO2 emissions with carbon capture and storage. [31]

3.2.2 European Green Deal

On 11th December 2019 the EU Commission presented its vision for a European Green Deal for the EU and its citizens with the aim of becoming climate neutral by 2050. [32] The Deal provides more detail of how the zero carbon targets can be met. It resets the Commission's commitment to tackling climate and environmental-related challenges that is this generation's defining task. The European Green Deal is a response to these challenges. It is a new growth strategy that aims to transform the EU into a fair and prosperous society, with a modern, resource-efficient, and competitive economy where there are no net emissions of greenhouse gases in 2050 and where economic growth is decoupled from resource use. [33] Since REN's activities and Hydrogen economy mainly depends on gas industry, the key aspects related to the gas sector is examined in this chapter. The important aspects related to the gas industry include in the European Green Deal is listed below:

- Power generation to be based on renewables complemented by phasing out of coal and decarbonizing gas. [34]
- Decarbonization of gas sector to be facilitated by support for development of decarbonized gases, forward looking design for a competitive decarbonized gas market, and addressing energy related methane emissions. [34]
- Foster development of hydrogen networks or CCUS, energy storage and sector integration.
- Support for “climate and resource frontrunners” to develop first commercial applications of breakthrough technologies. Priority areas include clean hydrogen, fuel cells, energy storage, CCUS. One example is EU support for clean steel breakthrough technologies. [34]
- Regulatory framework for infrastructure (incl. TEN-E Regulation and Projects of Common Interest (PCIs)) reviewed to ensure consistency with climate neutrality. Foster development of hydrogen networks or CCUS, energy storage and sector integration. [34]
- Renovation of buildings and housing, especially social housing, to reduce emissions. [34]
- Ramp up production and deployment of sustainable alternative transport fuels. [34]

The EU has developed and published several policies and strategies to decrease greenhouse emissions all around the Europe. In the alignment of this thesis' focus, the items related with natural gas sector and hydrogen are pointed out one by one.

3.2.3 Promotion of Low Carbon Technologies

The EU Innovation Fund has closed the first call for proposals for large-scale projects on 29 October 2020 and for small-scale projects on 10 March 2021. The focus of the fund is the innovation of the low carbon technologies and processes in energy intensive sectors.[35**]** The budged of the fund is determined as 10 billion euros in between 2020-2030. In this period the fund will support projects related with carbon capture and storage, renewable energy generation and energy storage. 10 billion euros is one of the biggest funds all around the world dedicated only for low carbon technologies. It aims to create financial incentives for investment in next generation technology and helping them to reach the market. [35]

3.2.4 Promoting Renewable Energy

The revised Renewable Energy Directive (RED II) entered into force in December 2018. It establishes a legally binding target of 32% renewables by 2030 including gases renewable gases such as biomethane and green hydrogen. Member States must assess the need to expand gas infrastructure to integrate and accommodate renewable gases. [36] Nevertheless, RED II directive does not provide clear definitions for renewable gases which causes a legal gap in hydrogen.

3.2.5 The Emissions Trading Scheme (ETS)

The emissions trading scheme is one of the biggest developments in EU's policy to fight against the climate change. The scheme is the world’s the first and the biggest carbon market. It has been seen a key tool to reduce greenhouse gas emissions cost effectively.[37] In the scope of the ETS around 10000 installations in the energy and manufacturing industry is limited for their emissions. Moreover, airlines operating in the European Union is also subjected to this policy.[37] These 10000 installations and the aviation sector in Europe covers more than 40% of the GHG emissions in European Union.[37] The ETS is known as a cap-and-trade system which allows trading emission allowances freely. The scheme resulted in creating the concept of carbon price in the EU.

3.2.6 The 2030 Climate and Energy Framework

The 2030 climate and energy framework focus on EU-wide targets and policy objectives for the period from 2021 to 2030. The framework has 3 key targets which is listed below. [38]

1)At least 40% cuts in GHG emissions compared to 1990 levels by 2030.[38]

2)At least a 32% share for renewable energy.[38]

3)At least a 32.5% improvement in efficiency.[38]

3.2.7 Energy Union

Overall energy policy is summarized by the Energy Union. The key principles of both the regulatory framework and the Energy Union are often cited in discussions concerning the development of future regulation for the gas industry as it transitions to a zero-carbon future.[39] The energy union strategy was published on 25 February 2015. The main goal of the strategy was to create an energy union that provides EU consumers, households, and businesses secure, sustainable, competitive, and affordable energy. [39] According to the European Commission The energy union builds five closely related and mutually reinforcing dimensions:

- Security, solidarity, and trust - diversifying Europe's sources of energy and ensuring energy security through solidarity and cooperation between EU countries. [40]
- A fully integrated internal energy market - enabling the free flow of energy through the EU through adequate infrastructure and without technical or regulatory barriers. [41]
- Energy efficiency- improved energy efficiency will reduce dependence on energy imports, lower emissions, and drive jobs and growth. [42]
- Climate action, decarbonizing the economy - the EU is committed to a quick ratification of the Paris Agreement and to retaining its leadership in renewable energy. [43]
- Research, innovation and competitiveness- supporting breakthroughs in low-carbon and clean energy technologies by prioritizing research and innovation to drive the energy transition and improve competitiveness." [39]

3.3 EU Directives and Regulations Relevant to Hydrogen and Natural Gas Infrastructure

Natural gas is one of the primer energies sources in European Union. Therefore, the EU has a well-established regulative framework for natural gas. The current framework was designed to create a competitive European market for natural gas, building on the existing mature industry and market. [44] At the center of these regulations there is the Third Gas Directive and there are other regulations which is associated with the gas directive. Although many of the regulations fail to hydrogen in natural gas sector, some of the legal acts include hydrogen technologies and its infrastructure. The current regulatory framework does not hinder hydrogen injection into natural gas network. However, they create barriers related with deployment, safety, environment, and transportation of hydrogen. Since, there is not any direct legal act or law on hydrogen injection, the directives and regulations require an adaptation to hydrogen. The key regulative framework which is related to hydrogen technologies are listed below with their summaries.

3.3.1 The Third Gas Directive

The directive is also known as 2009/73/EC, and it is the updated version of 2003/55/EC. Like the previous gas directives 2009/73/EC sets regulations starting from supply, storage, transmission, and distribution of the natural gas in European Union. Moreover, the directive mentions laying down the rules relating to the organization and functioning of the natural gas sector, access to the market, the criteria, and procedures applicable to the granting of authorizations for operation of systems. [45] The Directive 2009/73/EC does not mention anything related with hydrogen injection into natural gas system nor hydrogen technologies. Instead, hydrogen and biogas can be classified under the category of other gases. These gases are also granted non-discriminatory access to the gas system by sub article 41.[46] Sub article 41 states that "Member States should ensure that, considering the necessary quality requirements, biogas and gas from bio- mass or other types of gas are granted non-discriminatory access to the gas system, provided such access is permanently compatible with the relevant technical rules and safety standards. Those rules and standards should ensure that those gases can technically and safely be injected into and transported through the natural gas system and should also address their chemical characteristics." [46] Article 1.2 of the Directive supports the sub article 41 and points out "The rules established by this Directive for natural gas, including LNG, shall also apply in a non-discriminatory way to biogas and gas from biomass or other types of gas in so far as such gases can technically and safely be injected into, and transported through, the natural gas system." [46]

3.3.2 Directive 2019/692/EC

This directive amends Directive 2009/692/EC concerning common rules for the internal market in natural gas. This amendment, however, refers to a new EC approach on the regulation of the gas transmission lines connecting Member States with third countries. Although this Directive is important for addressing barriers to the realization of the internal natural gas market, it does not address the need of fundamentally revising the Gas Directive to incorporate provisions for hydrogen. [47]

3.3.3 Regulation (EC) 715/2009

(EC) 715/2009 is also known as the Gas Regulation mentions a variety of topics such as definitions, certifications of TSOs, monitoring, regional cooperation, and tariffs in the natural gas sector. The main reason why this regulation is to set non-discriminatory rules for access conditions to natural gas transmission systems considering the special characteristics of national and regional markets with a view to ensuring the proper functioning of the internal market in gas. [48]

3.3.4 Regulation (EC) 2015/703

This Regulation establishes a network code which sets out rules regarding interoperability and data exchange as well as harmonized rules for the operation of gas transmission systems. In addition to interconnection points, Article 17 applies to other points on transmission network where the gas quality is measured. [49] Regulation EC 2015/703 also aligns the technical procedures used by transmission system operators within the EU.

3.3.5 Regulation (EU) 2017/460

The regulation of EU 2017/460 establishes a network code setting out the rules on harmonized transmission tariff structure for gas, including rules on the application of a reference price methodology, the associated consultation and publication requirements as well as the calculation of reserve prices for standard capacity products. [50]

3.3.6 Regulation (EU) 2019/942

The regulation (EU) 2019/942 establishes a European Union Agency for the Cooperation of Energy Regulators to assist the regulatory authorities in various regulatory tasks. [51] The organizational structure of ACER and its tasks in various fields are determined. Article 8 sets the Agency's tasks as regards terms and conditions for access to and operational security of cross border infrastructure. [51]

3.3.7 Regulation (EU) 559/2014

Regulation EU 559/2014 establishes the Fuel Cell Hydrogen 2 Joint Undertaking with the aim of increasing the effectiveness of hydrogen production from renewable energy sources and demonstrating use of hydrogen at large scale, to integrate renewable energy sources with the energy system.[52]

3.4 National Hydrogen Regulations and Strategies

With the recent development in the decarbonization policies hydrogen technologies gained significant momentum. It is observed from the literature the number of countries which have published their national strategies is increasing every day. However, national strategies will not be enough to utilize hydrogen in full scale. Laws and legislative framework for every county is needed because hydrogen has the potential to be used in tens of different sectors. Under this section specific strategies and law of pioneer countries are examined and compared. Since the focus of the thesis is on hydrogen injection in natural gas systems, allowable hydrogen content in the natural gas network in different national frameworks are investigated.

By early September 2020, France, Germany, The Netherlands, Portugal, Spain, and Norway had released national hydrogen strategies. After this development more European Union countries like Austria, Estonia, Luxembourg, Poland, Slovakia is expected to publish their strategies. Figure 2 demonstrates the finalized national hydrogen strategies and developing hydrogen strategies in Europe.[53]

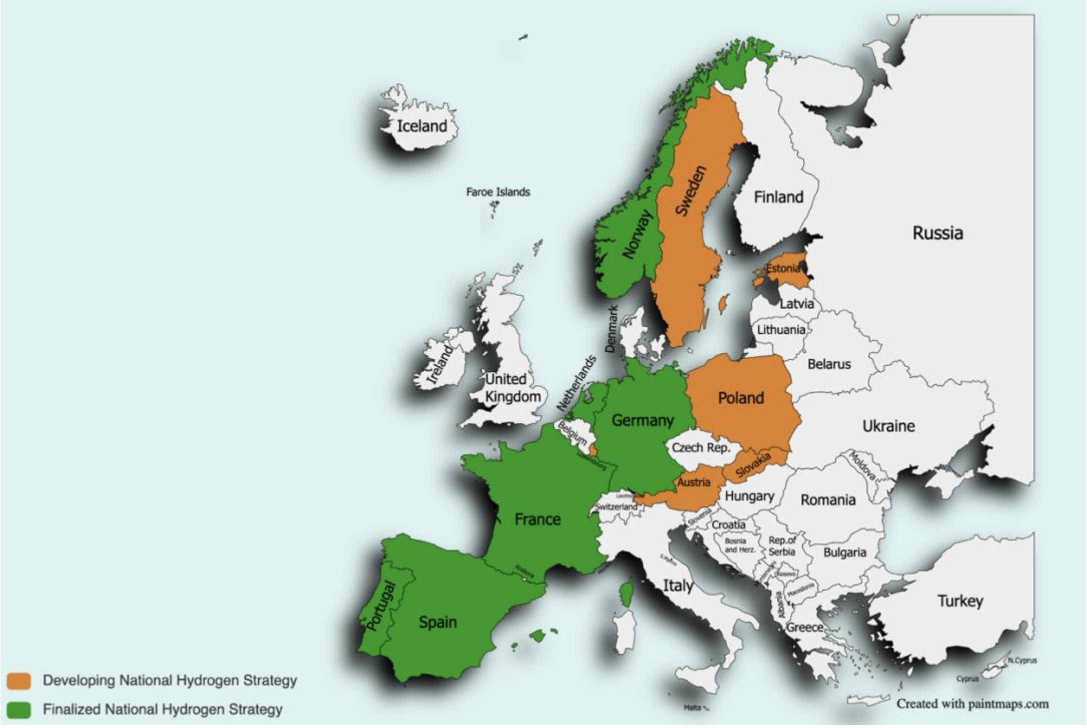


Figure 2 Map of Europe showing finalized and developing national hydrogen strategies. Adapted from [53]

Since focus of the thesis is the hydrogen injection into existing natural gas network, current available hydrogen injection percentages from pioneer countries are investigated. The allowed hydrogen levels in the network and their governing law are presented in Table 8.

Table 8 Allowable hydrogen injection levels in different countries and its governing law.

Country	Allowed Percentage of Hydrogen Injection	Law/ Legislation	Pure Hydrogen Injection
Austria	4% By Volume	OVGW 31	Yes
Belgium	There is no limit. However 2% is accepted by TSO.	Not Applicable	No
Bulgaria	Hydrogen injection is not allowed.	Not Applicable	No
France	6% By Volume	Law No 2015-992	No
Finland	1% By Volume is decided by TSO.	Decree 551/2009	No
Germany	10% By Volume	DVGW Worksheets G 260 and G 262	Yes

Netherlands	0.02%(mol)	Ministerial Decree ‘Gas Quality’	No
Ireland	0.1% By Volume	Not Applicable	No
Italy	1% By Volume.	UNI/TS 11537:2019	No
Latvia	0.1% By Volume	Not Applicable	No
Lithuania	2% By Volume	Not Public	No
Portugal	0.1% By Volume (For2021)	Decree-Law 62	Not specified
Spain	5% By Volume	34/1998	Not specified
Sweden	2% for safety of steel sector determined by TSO.	Natural gas regulations MSBFS 2009:7	Clarification is needed
United Kingdom	0.1% By Volume	EN437:2003 T10	No

3.4.1 France’s Law and Strategy

A specific law has not been established in France yet. However, hydrogen is mentioned in general laws on energy and mobility. On 17 August 2015 Law No 2015-992 of 17 relating to the Energy Transition for a Green Growth is published. Article 121 of Law No 2015-992 states that the French government shall establish a *"*development plan for the storage of renewable energies using decarbonated hydrogen*".* With this article hydrogen distribution infrastructure and power-to-gas business is encouraged. [54]

On June 2018, France became the first EU country to develop a national strategy for hydrogen. The strategy was a milestone in France’s energy transition plan. [53] The first hydrogen strategy had three main objectives which can be named as “greening” hydrogen for industrial use, using hydrogen for mobility to complement the battery sector and stabilizing energy networks. In September 2020, the second version of the strategy is released with significant updates.[53] From 2020 to 2030 7 billion euros of public funding is allocated for hydrogen technologies and its development. One of the main goals of the strategy is to reach an electrolyzer capacity of 6.5 GW by 2030.[53]

3.4.2 Germany’s Law and Strategy

In June 2020 Germany has released their National Hydrogen strategy. Until 2030, the strategy aims to build 5 GW electroyzer capacity.[53] To increase the phase of hydrogen technologies development 9 million euros of public funding became available under the scope of the strategy. In Germany's National Hydrogen Strategy utilization of the hydrogen is as important as the production. That is why they plan to adapt their current gas network by building pipelines dedicated for hydrogen. In The German Energy Industry Act (Energiewirtschaftsgesetz) hydrogen and other new gases which are accepted as renewable source defined as "biogas" In this context, the Gas Network Access Ordinance sets the regulatory framework enabling the "biogas" injection into the natural gas networks.[45] On 10 February 2021 the German government passed the draft of an amendment to the Energy Act which focuses on the regulation of hydrogen networks.[55] The purpose of the amendment to the Energy Act is to gradually build up a hydrogen infrastructure in Germany. The provisions are intended as a transitional solution until corresponding European guidelines are available. [55]

3.4.3 The Netherlands's Law and Strategy

In April 2020 the Dutch Government has released their National Hydrogen Strategy. According to the strategy 4 GW of electrolizer capacity is planned to build by 2030.[53] The Dutch authorities know this amount of hydrogen is difficult to utilize without injecting it to the national gas network. Therefore, under the scope of the strategy a four-year program is developed to identify and investigate the safety issues of blending hydrogen with natural gas. Currently, Gas Quality Decree, allows a level of 0.5% hydrogen in the regional networks and a level of 0.2% in the national networks.[56] The Gas Act will need adaptations to allow for a higher percentage of hydrogen blending. Physical blending of up to 2% is already achievable in The Netherlands with minor adjustments and the government expects that this amount may be increased.[56]

3.4.4 Portugal's Law and Strategy

On 14 August 2020 the Portuguese National Hydrogen strategy was formally adopted by the Council of Ministers (Ministry Council Resolution 63/2020). In alignment of the EU hydrogen strategy's timeline the strategy has three stages. According to Portugal's plan these three stages is decided as follows:[53]

Stage 1: Establishment of the regulatory framework and first projects from 2020 till 2023.[53]

Stage 2: Consolidation and roll-out of projects at national level from 2024 till 2030. The target of 2030 has a crucial role in the strategy because by 2030, hydrogen should have a share of 5% in the country's final energy consumption. This final target of 5% is planned to be achieved by up to 15% hydrogen injection into natural gas grid and 2 to 2.5 GW electrolizer capacity including a 1GW project in Sines.[53]

Stage 3: The full development of the national hydrogen market from 2030 till 2050.[53]

Since REN's hydrogen injection projects highly dependent on the Portuguese Law and Portuguese National Hydrogen Strategy, Decree-Law 62 becomes a driving force in the project. Decree-Law 62 sets the legal framework for gas in Portugal and includes renewable gases and low carbon gases. In the decree, the definition of renewable gases is given as "Renewable gases are those produced from renewable sources; low carbon gases are those produced from non-renewable sources but with emissions bellow 36,4gCO_2 eq/MJ". In the scope of the law, the business model for injecting renewable gases or hydrogen has a framework. According to this framework, the producer of the renewable gas or hydrogen must pay for the connection to the grid. The grid connection costs are shared by all producers using that connection. Even if a company decides to provide hydrogen to the grid after the connection is build, the costs will be projected and shared by those who connect later. Most importantly to maintain the 5% share of hydrogen in the energy mix, there will be an obligation to incorporate renewable and low carbon gases in the national grid and the minimum share will be defined by a Ministerial Order. Therefore, the suppliers will have to assure a minimum share of low carbon gases in their gas purchases.

3.4.5 Spain's Law and Strategy

When Spain released its hydrogen strategy, 9 billion euros worth of investment in the hydrogen sector is announced.[53] The main goal of the strategy is to build 4 GW of electrolizer capacity and to achieve 25% of renewable hydrogen within the industrial hydrogen mix by 2030.[53] Like many other countries there is currently no specific legislation for hydrogen in Spain. However, it is destined to change after the publication of national hydrogen strategy. Currently there is no specific limit for hydrogen injection in the grid and every project is assessed on case by case. PD-01 protocol provides basis on hydrogen injection into natural gas network. [57] In the document, there are technical specifications and requirements of the gas circulating in the grid. Mostly this document makes references to European standard of ENE-EN 16726. [57]

Chapter 4: Analysis of the Changes and Adaptations in the System

Introducing hydrogen in the natural gas network will change several conditions in the property of the gas and the operation conditions. Changes in the fluid dynamics, gas quality, energy content, pressure are calculated and analyzed in Chapter 4. Possible adaptations for REN’s infrastructure are discussed and suggested.

4.1 Changes in the Fluid Dynamics and Gas Quality Parameters

Hydrogen injection into natural gas network may result in several changes in the system and the operation conditions depending on the hydrogen concentration. One of the most important changes that is faced would be the properties of natural gas mixture. In the gas transmission business fluid mechanics and gas quality parameters are crucial for sustainability and profitability purposes. Under this sub-chapter the changes in the gas quality are primary calculated with assumptions. Furthermore, the possible alteration effects and adaptations are analyzed. The hydrogen ranges are determined under the light of Portuguese governments and REN’s hydrogen strategy. Therefore, many of the calculations are performed up to 20%-30% of hydrogen concentration in natural gas.

4.1.1 Pseudo Compressibility Factor

One of the base parameters while transportation of natural gas is pseudo compressibility factor. Therefore, the analysis of the changes in the natural gas properties will start from it. Pseudo compressibility factor (Z-factor) represents the proportion of volume a given amount of gas to the ideal volume of it at the common conditions and defined pressure and temperature.[58] Gas compressibility makes the difference between ideal gas and real gas. [58] In order to calculate the Z-factor Peng-Robinson Equation is used and the cubic equation created from Peng-Robinson parameters is solved by Microsoft Excel.

$$a(T) = 0.45724\left(\frac{R^2T_c^2}{Pc}\right)\left(1 + k\left[1 - \left(\frac{T}{Tc}\right)^{0.5}\right]\right)^2$$

Equation 1[59]

$$k = 0.37464 + 1.5422\omega - 0.26922\omega^2$$

Equation 2 [59]

$$b = 0.07780\,\frac{RTc}{Pc}$$

Equation 3 [59]

where ω is the acentric factor, Tc and Pc are the critical temperature and pressure, respectively, and R is the universal gas constant. Mixing rules can be derived from statistical mechanics, or by using a phenomenological point of view from classical thermodynamics [59] The widely used van der Waals Mixing Rule are:[59]

$$a = \sum_{i}^{c}\sum_{j}^{c} x_i * x_j * a_{ij}$$

Equation 4 [59]

$$b = \sum_{i}^{c}\sum_{j}^{c} x_i * x_j * b_{ij}$$

Equation 5 [59]

where xi is the molar fraction of species i, c is the number of different species in the mixture, a_{ii} and b_{ii} are the pure component parameters. a_{ij} is cross parameters and represented as follows:

$$aij = (1 - k_{ij})\sqrt{(a_i * a_j)}$$

Equation 6 [59]

For the simplicity purposes the calculation is performed only for the dominant gases in the mixture which are methane, ethane, and hydrogen. The important parameters for these calculations are given in Table 9. and Table 10

Table 9 Critical temperature, critical pressure, covolume, acentric factor, binary interactions and attraction parameters required for the calculations.

Required Parameters for Peng-Robinson Equation			
Component Name	Methane	Ethane	Hydrogen
Critical Temperature, Tci /K	190,56	305,32	33,1
Critical Pressure, Pci /MPa	16,04	30,07	2,016
bi	7,68495205	6,56803726	10,6206537
ωi	0,0115	0,105	-0,215
κi	0,39234029	0,53360143	0,03057705
ai	59244,1522	100997,536	15190,2845

Table 10 Binary interactions of methane, ethane and hydrogen

k_{ij}			
i↓ / *j*→	Methane	Ethane	Hydrogen
Methane	0,00000	0,00340	0,11260
Ethane	0,00340	0,00000	0,09120
Hydrogen	0,11260	0,09120	0,00000

When pseudo-compressibility factor is calculated in iteration steps for hydrogen concentrations of 5%,10%,15%,25% and 30%, an increase in the Z-factor is observed which is presented in Figure 3.

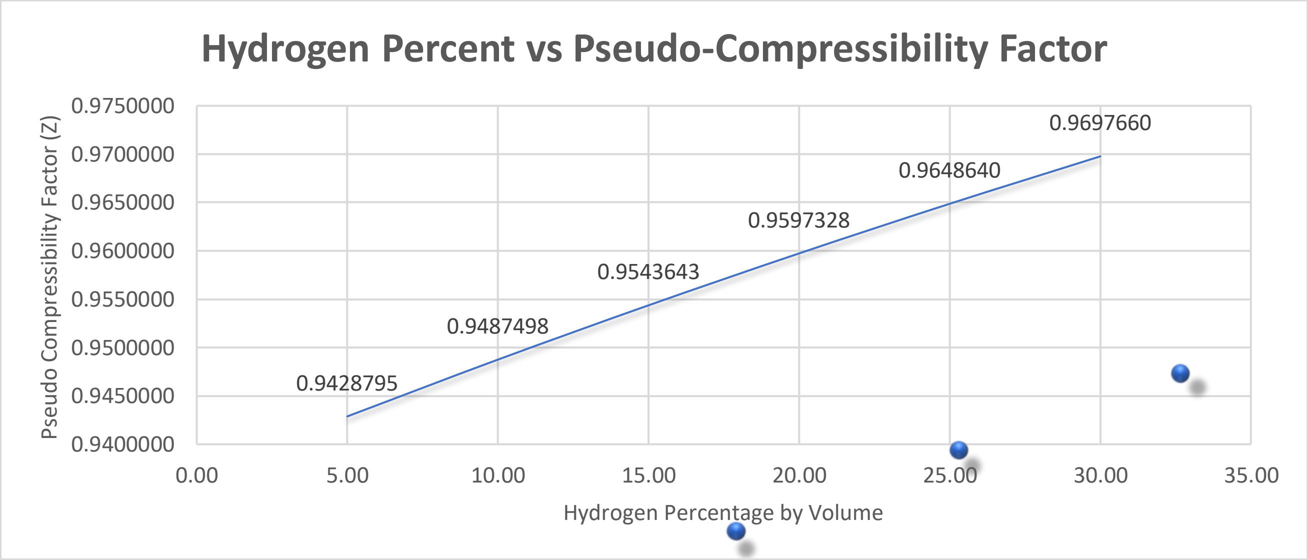


Figure 3 Change in pseudo compressibility factor depending on the hydrogen percent injected into natural gas mixture.

4.1.2 Natural Gas Mixture Density and Viscosity

After calculating the change in Z-factor the change in gas density is calculated. Gas density is useful and important for many of the calculations in natural gas industry. Moreover, there are gas density standards that needs to be satisfied. Therefore, understanding the possible change in density will provide meaningful insight. Since, molecular mass of hydrogen is very low compared to other components in the natural gas, density of the mixture is expected to decrease with the increasing hydrogen concentration. The Density of the mixture is calculated under certain temperature (288.2K) and pressure(8.4Mpa) conditions. These conditions are chosen because they are the design parameters for REN's pipeline infrastructure. Calculated change for the density of the mixture is presented in Table 4.

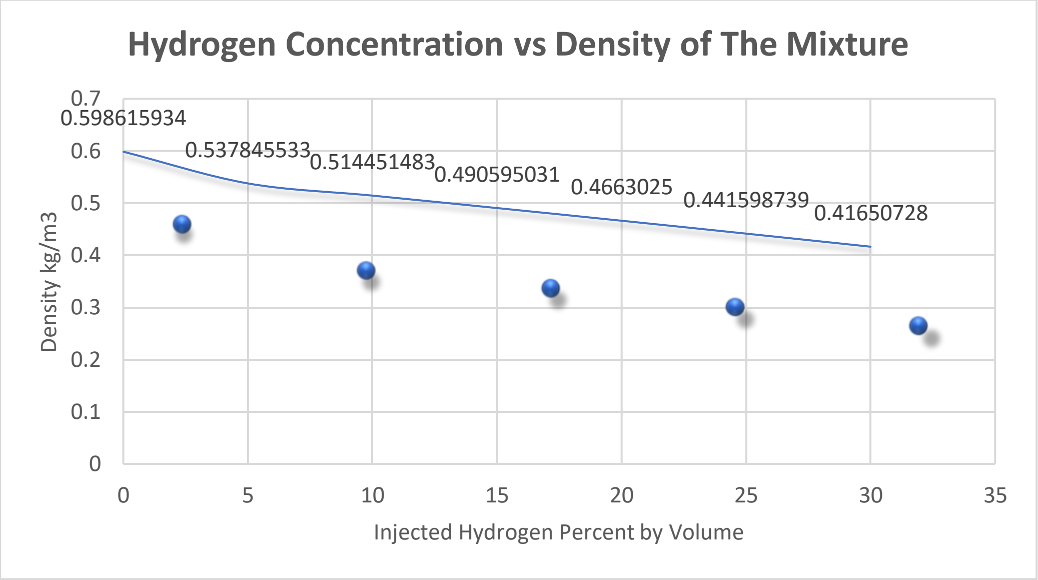


Figure 4 The change in gas density with respect to injected hydrogen volume in natural gas.

The dynamic viscosity change is also calculated at 288.2 Kelvin for the further calculations of pressure. With the increased hydrogen percentage by volume decreased viscosity is observed. Although there is an obvious decrease in the viscosity. This decrease does not look significant. Therefore, it would not affect any operation. The change in dynamic viscosity is shown in Figure 5.

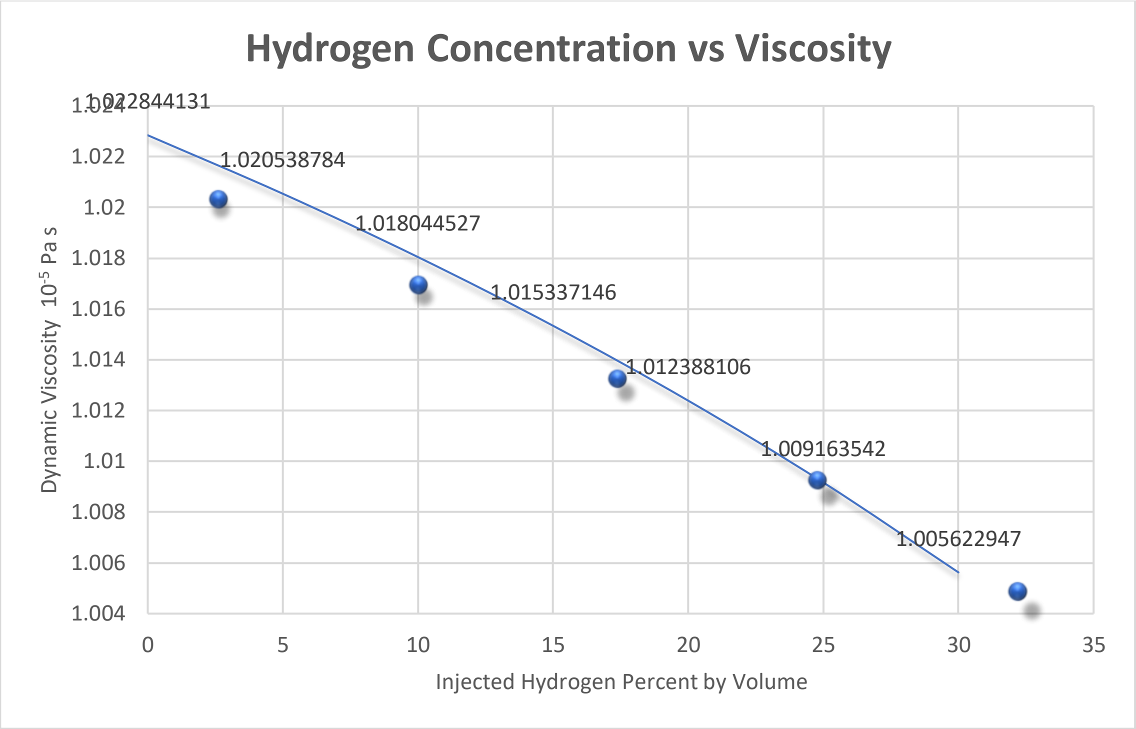


Figure 5 The change in viscosity at 288.2 Kelvin with respect to injected hydrogen volume in natural gas.

4.1.3 Upper Explosive Limit and Lower Explosive Limit of the Natural Gas Mixture

One of the biggest concerns about utilization of hydrogen is its flammability as discussed in Chapter 2 previously. In this subsection explosive limits of the natural gas and hydrogen mixture is investigated. Explosive limits express the concentration range of a gas in air which explode in the presence of an ignition source. The explosive limits are usually measured or calculated as the percentage by volume. [60] Two types of explosive limits are calculated which are upper explosive limit (UEL) and lower explosive limit (LEL). As the name implies UEL is the highest concentration of a gas in air capable of producing a flash of fire and LEL is the lowest concentration of a gas in air capable of producing a flash of fire. [61] Hydrogen is a gas characterized by high explosive range. Its lower explosive limit is 4%, and its upper explosive limit is as high as 76%.[62]

The decrease in the lower explosive limits and the increase in the upper explosive limits clearly increases the explosiveness of the mixture. Therefore, it raises safety concerns about transmission network. However, this mixture also decreases explosiveness of the hydrogen. Therefore, hydrogen transportation with natural gas is safer than transporting hydrogen alone. The calculated change in lower explosive limits and upper explosive limits are shown in Figure 6 and Figure 7 respectively.

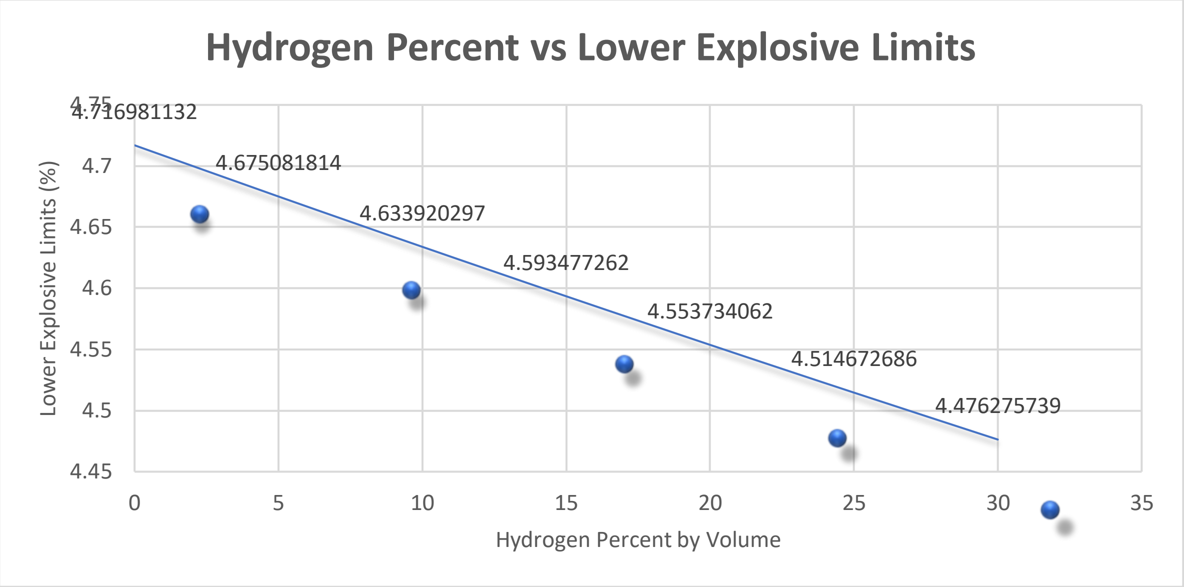


Figure 6 Change in the lower explosive limit of the natural gas according to the added hydrogen into the system

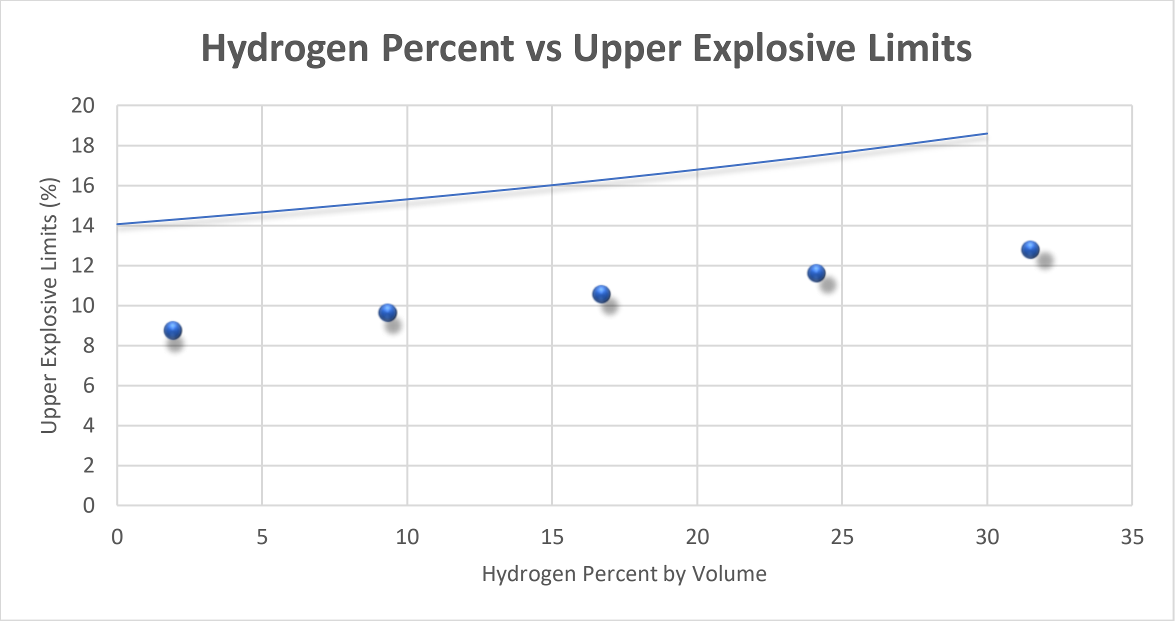


Figure 7 Change in higher explosive limits due to added hydrogen influence.

4.1.4 Change in Heating Values and Wobbe Index

The importance of the heating values and wobbe index will be examined in detail later in the Chapter 5 (Gas Quality Monitoring). These parameters are the most important indicators to explain the quality of the natural gas mixture. Most of the natural gas purchasers decide which natural gas to buy according to their needs of heating values and wobbe index. Moreover, these parameters are also standardized by ISO and European union. Therefore, while transporting hydrogen companies must stay in the standardized ranges for these parameters. Lower heating values of pure methane, ethane and hydrogen is given in the Table 11. Furthermore, the wobbe index is calculated by equation 7.

Table 11 Lower heating value and higher heating value of pure Hydrogen, Methane and Ethane.

	Hydrogen	Methane	Ethane
Lower Heating Value (MJ/m3)	10,80	35,80	27,30
Higher Heating Value (MJ/m3)	12,70	39,80	29,70

$$Wobbe\ Index = \frac{Higher\ Heating\ Value\ of\ the\ Mixture}{\sqrt{Specific\ Gravity\ of\ the\ Mixture}}$$

Equation 7 [63]

Figure 8, Figure 9 and Figure 10 shows the increasing hydrogen concentration effect on lower heating value, higher heating value and wobbe index respectively.

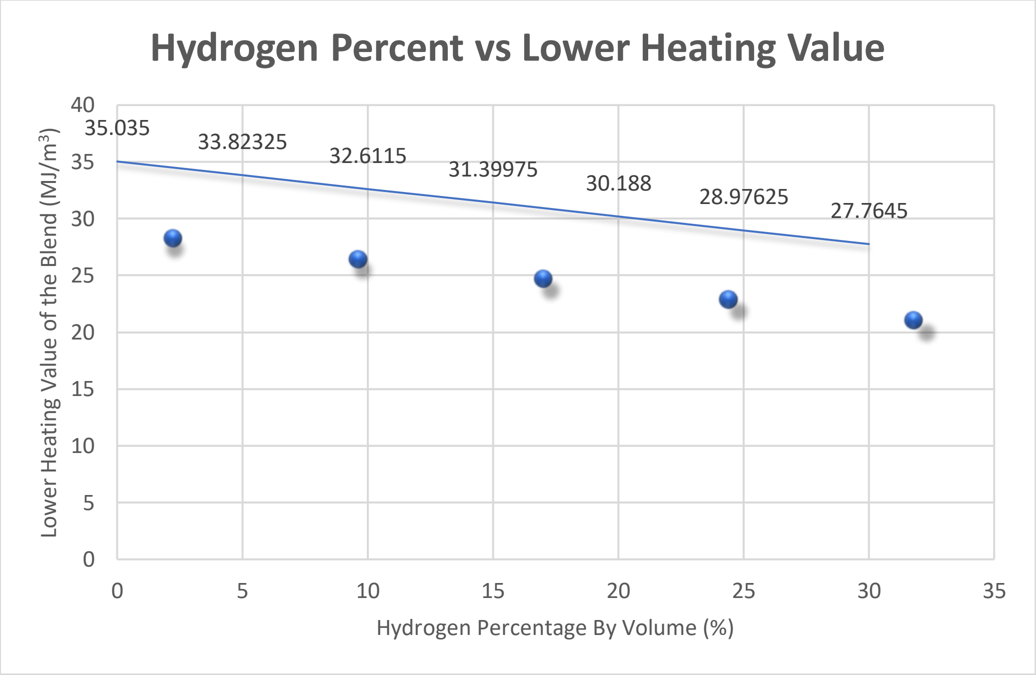


Figure 8 Change in lower heating value depending on hydrogen percentage

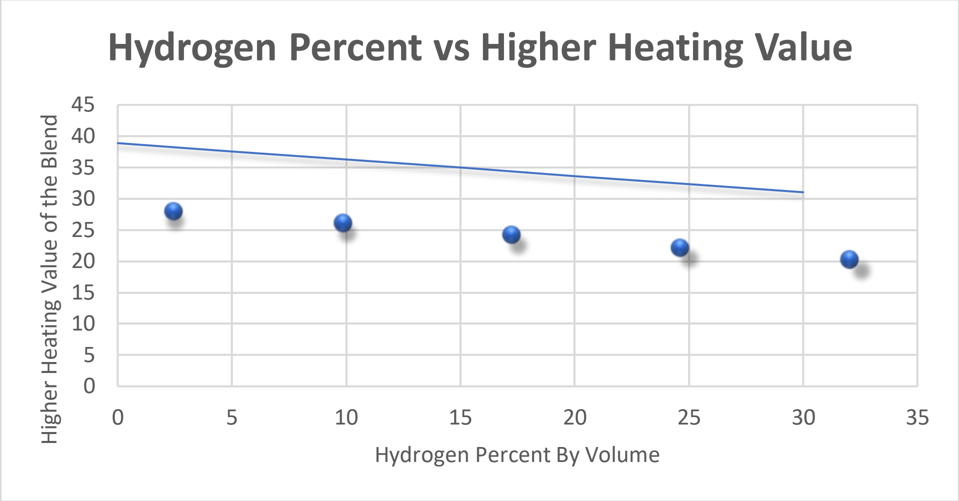


Figure 9 The change in higher heating value in different hydrogen injection levels by volume.

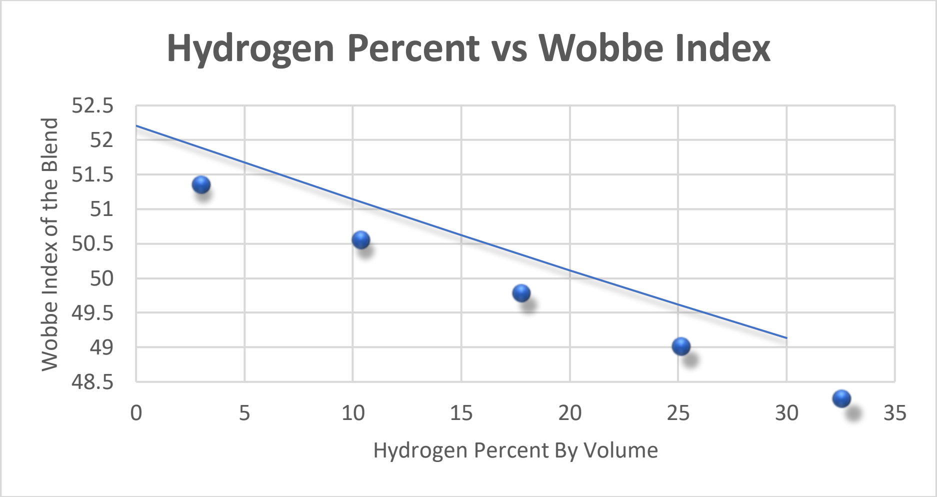


Figure 10 Calculated wobbe index change according to different hydrogen injection percentages by volume.

Table 12 Decrease in lower heating value, higher heating value and wobbe index with increasing hydrogen percent by volume

Hydrogen percent	LHV Decrease(%)	HHV Decrease(%)	Wobbe Index Decrease (%)
0	0	0	0
5	3,458684173	3,367231493	1,020415038
10	6,917368346	6,734462986	2,030913409
15	10,37605252	10,10169448	3,02778987
20	13,83473669	13,46892597	4,00639935
25	17,29342086	16,83615747	4,960870921
30	20,75210504	20,20338896	5,883713878

On one hand a significant decrease is observed in the lower hating value and higher heating value of the natural gas mixture, especially when hydrogen concentration exceeds 30%. On the other hand, the decrease in the wobbe index is relatively smaller. Initially REN plans to blend the natural gas by 5%. According to REN's plan approximately 3.46%, 3.37% and 1.02% decrease is expected in lower heating value, higher heating value and wobbe index respectively. These changes are shown in percentage in Table 12. Initially this drawback does not require an adaptation because it can be fixed by changing some of the service conditions. However, in the future if hydrogen concentration exceeds 20% in the gas mixture there will be a need for further research and adaptations.

4.2 Changes in the Delivered Energy Content

4.2.1 Amount of Energy Delivered Per Cubic Meter of Delivered Gas

Meeting the energy demand of a region is one of the key responsibilities of a natural gas operator. That is why the energy amount delivered per cubic meter is a critical parameter. To keep the energy flow constant, the operator needs to change the operating conditions even if the quality of the natural gas decreases. This is exactly what many operators will be doing if they start injecting hydrogen into their natural gas systems. Since, energy content of the hydrogen per cubic meter is significantly lower than the regular natural gas mixture. The hydrogen affects the natural gas by simply decreasing the amount of energy in 1 m3 of gas mixture. This effect can be observed in Figure 11. The simple solution of this decrease in the energy content will be the increase of pressure and gas velocity in the natural gas network if the system parameters allow.

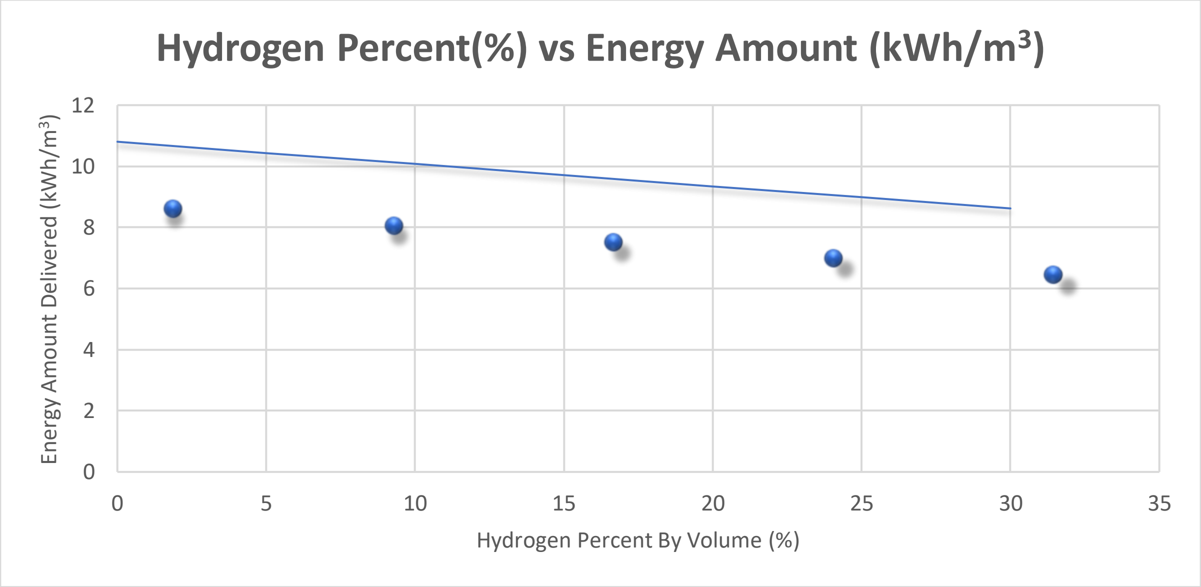


Figure 11 Energy amount delivered per cubic meters with increasing amount of hydrogen injection into the system.

4.2.2 Changes in the Line pack in REN’s Pipeline Network

REN’s natural gas network connects many locations, and these locations are connected with pipelines which has different diameters according to the needs. The specifications of the pipelines which connects major points are given in Table 13. Therefore, every single connecting pipeline contains different amount of energy. This energy is represented by the term called Line Pack and it is the inventory of gas in a pipeline.[64] In this sub-section four pipelines which has different diameters are chosen to calculate hydrogen injection effect on line-pack. The chosen pipelines are highlighted in orange color in Table 13.

Table 13 REN’s major pipeline connections are given with their distance and pipe diameter.

The Transmission Line	Distance (km)	Pipe Size (mm)
Setubal-Leiria	173	700
Leiria-Gondomar	164	700
Gondomar-Braga	50	500
Campo Major- leira	220	700
Braga- Valença	74	500
Monforte-Guarda	184	300
Mealhada-Viseu	68	500
Sines Setubal	87	800

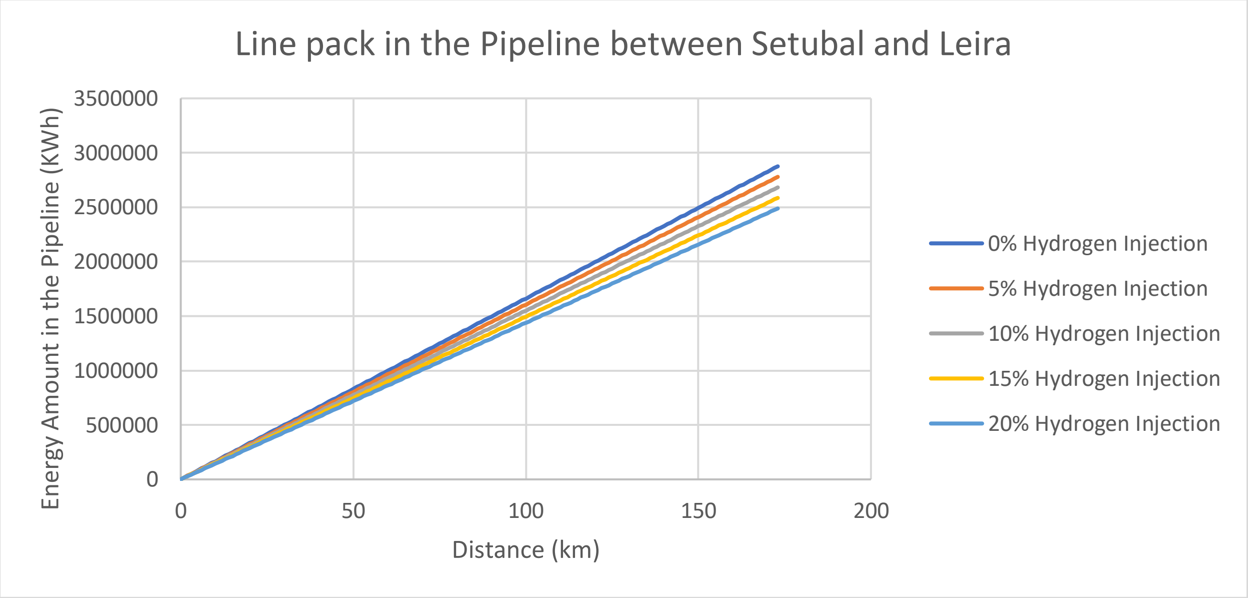


Figure 12 The effect of increasing hydrogen concentration on the line pack between Setubal and Leira.

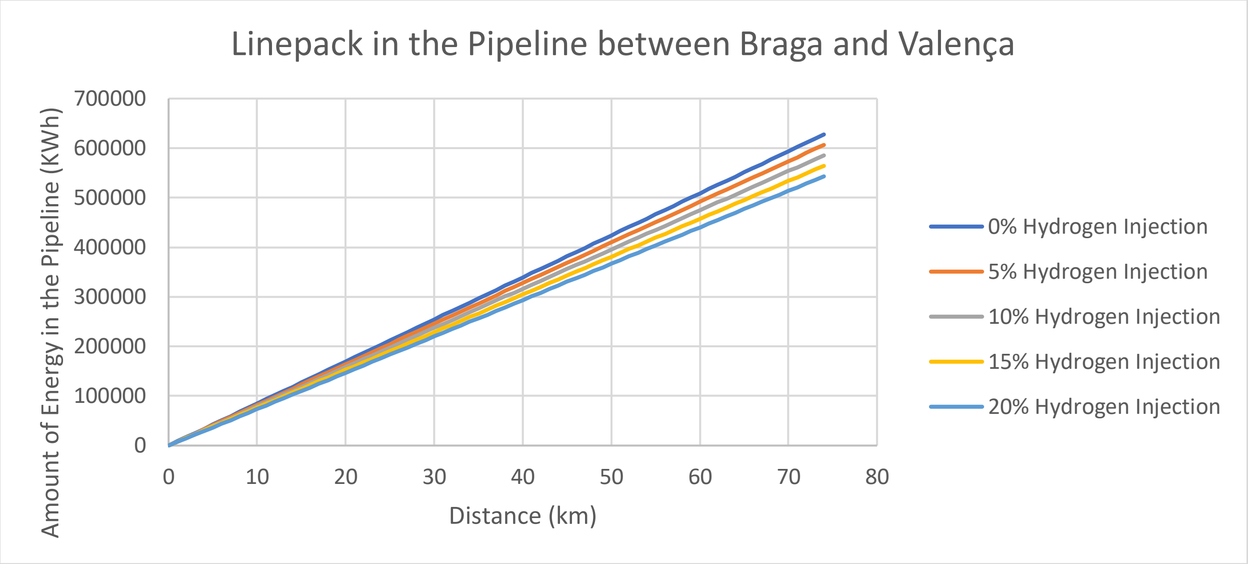


Figure 13 The effect of increasing hydrogen concentration on the line pack between Braga and Valença.

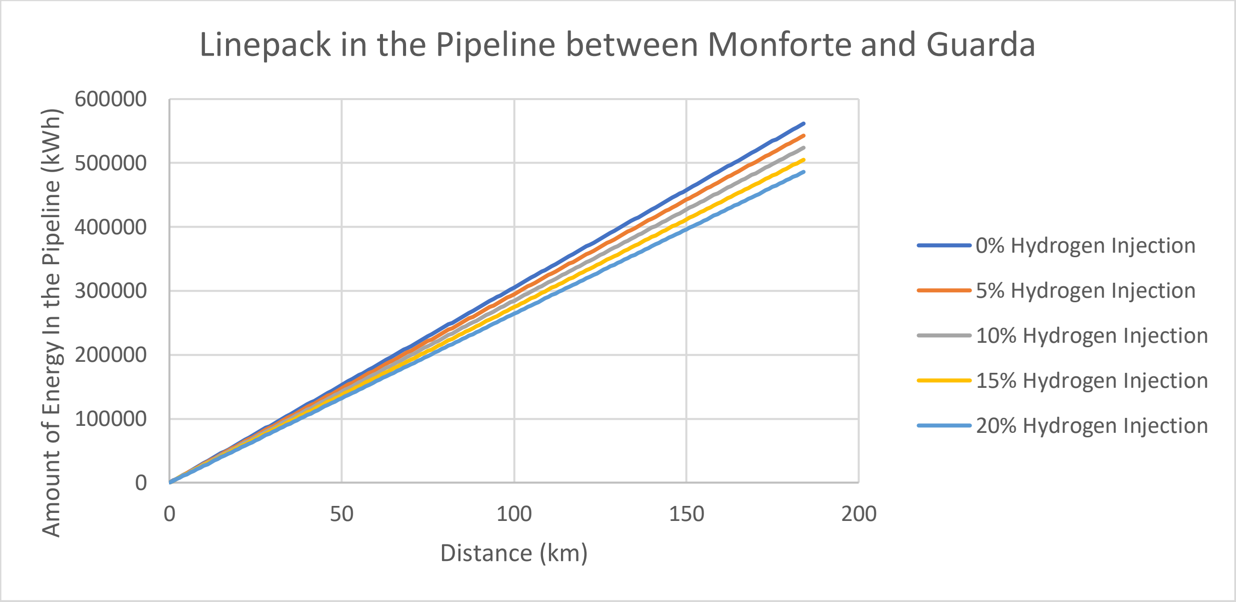


Figure 14 The effect of increasing hydrogen concentration on the line pack between Monforte and Guarda.

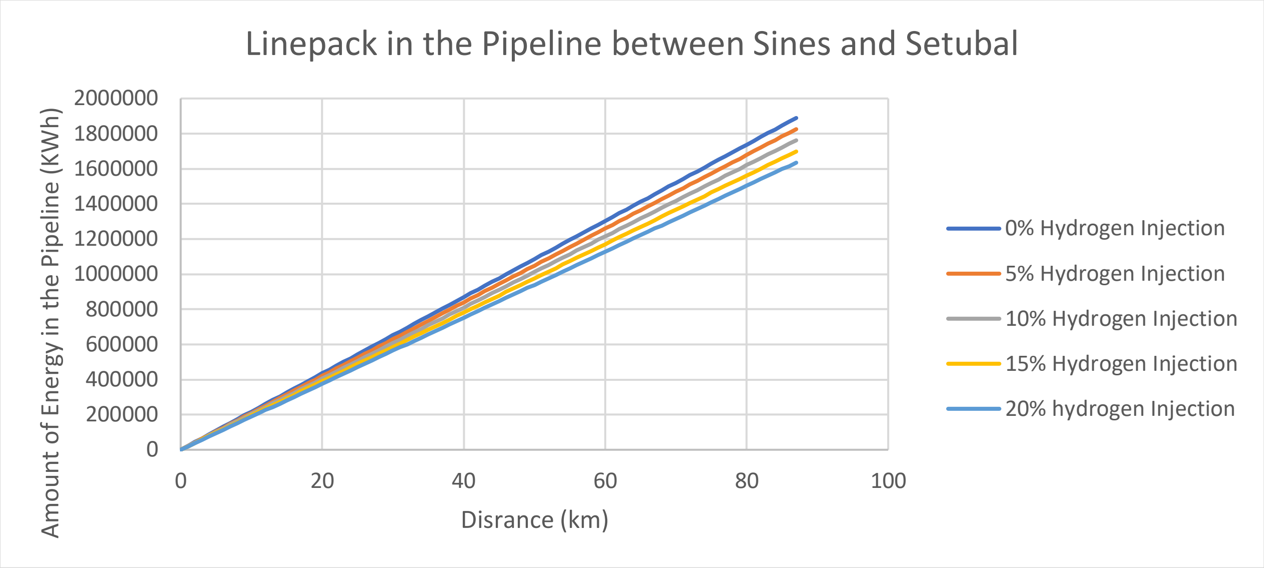


Figure 15 The effect of increasing hydrogen concentration on the line pack between Sines and Setubal.

Table 14 Changes in the line-pack depending on hydrogen percentage through pipeline.

Hydrogen Percentage	Line Pack Between Given Locations in KWh			
	Setubal Leira	Braga Valença	Monforte Guarda	Sines Setubal
0% Hydrogen	2875763,604	627599,7012	561786,5433	1888905,479
5% Hydrogen	2778929,986	606466,9664	542869,8899	1825301,659
10% Hydrogen	2682096,368	585334,2316	523953,2365	1761697,839
15% Hydrogen	2585262,751	564201,4968	505036,5831	1698094,019
20% Hydrogen	2488429,133	543068,762	486119,9297	1634490,198

Line pack is directly proportional to the volume of the pipeline. That is why pipelines with higher diameter and longer distances suffer from the line pack lose more than others.

4.3 Changes in the Pressure

Pressure is one of the most important parameters in the natural gas industry because it has the potential to affect every single equipment and component. As it was mentioned previously in this chapter pressure change also has the potential to balance delivered energy amount loss due to hydrogen injection. Moreover, the planning and design of the stations and pipeline is done according to the pressure modification in the transmission network. Clearly without the increased levels of pressure, it would not be possible to transport hydrogen or natural gas for long distances. Therefore, terms like pressure drop and operating pressure plays a key role in the industry. That is why it is fundamentally important to understand how hydrogen injection would affect the pressure. For pressure drop calculations in the pipeline Panhandle B equation is used because of its suitability for high pressure and turbulent flow characteristics. The pipeline is assumed to be completely horizontal. Therefore, change in potential energy is neglected.

$$Qn = 108.080\ \frac{1}{\mu^{0.02}} * \frac{Tn}{Pn}\left[\frac{(P_1^2 - P_2^2)}{L * d^{0.9608} * T_{avg} * Z_{avg}}\right]^{0.51} * D^{2.53}$$

Equation 8 [62]

Where:

Qn= Flow rate (m^3/s) ➔ 5760000 m^3/day is assumed.

μ= Dynamic viscosity of the mixture (Pa.s) ➔ Changes with changing hydrogen concentration (For 5% hydrogen injection 1,02054E-05 Pa.s is the calculated value.

Tn= Normal temperature (K) ➔ 273.15K.

Tavg= Average temperature of the gas mixture ➔ 288.15K is assumed.

Pn= Normal pressure (Pa) ➔ 101325 Pa

P_1= Upstream gas pressure (Pa) ➔ 7000000 Pa is Assumed

P_2= Downstream gas pressure (Pa) ➔ Is calculated by the Panhandle B equation

L= Length of the pipe (m) ➔ In every 10km the calculations are performed iteratively. 170km is the limit.

d= Relative gas density ➔ Changes according to hydrogen concentration 0.463 is calculated for 5% of hydrogen injection.

Z= Pseudo compressibility factor ➔ Changes depending on hydrogen concentration 0.943 is calculated for 5% hydrogen injection.

D= Diameter of the pipe (m) ➔ 0.7 meters of diameter is chosen.

The calculated pressure drops with in 17km distance is shown in Figure 16 with changing hydrogen percentages. The obvious conclusion from that figure is when hydrogen concentration increases in the natural gas mixture, the pressure drop will be less. The main reason behind this conclusion is the increase in pseudo compressibility factor, decrease in the viscosity and the decrease in the relative density with increased level of hydrogen injection.

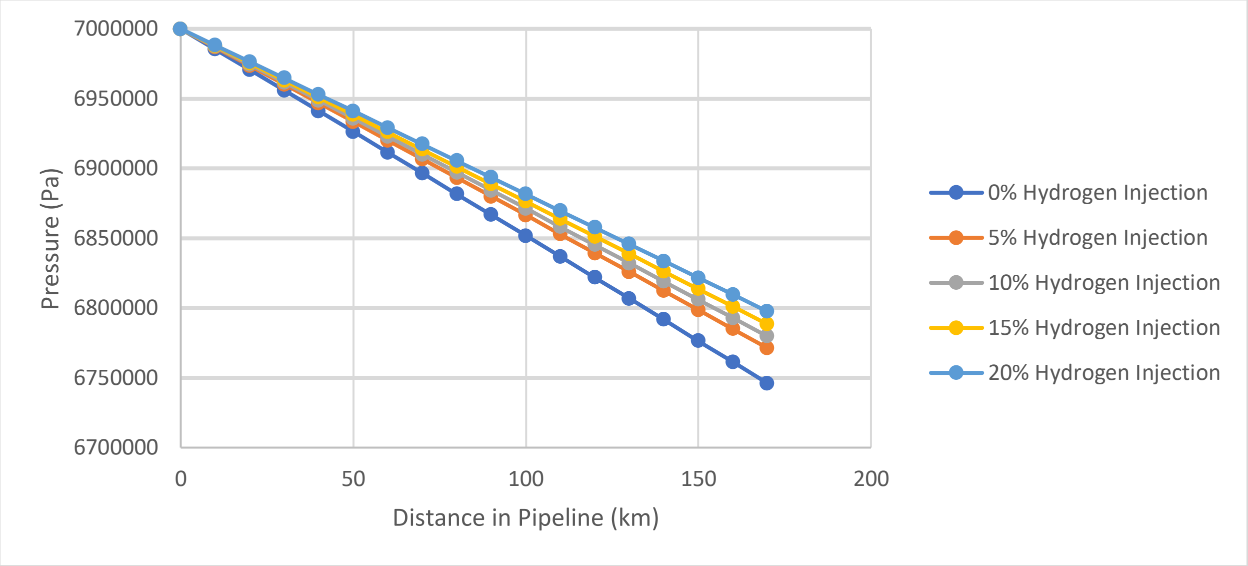


Figure 16 The effect of hydrogen injection in pressure drop.

4.4 Adaptations Required for Blending

In REN's hydrogen strategy there is a major adaptation, and it is building a blending station. This adaptation is based on the business case of the project. According to the case, hydrogen producers are responsible for connecting their hydrogen to the natural gas operator. Therefore, REN will not produce hydrogen instead they will be responsible only for the transportation of it. Due to possible mixing problems and possible agglomeration of hydrogen, blending of natural gas and hydrogen before injection is desired. The location, the size and the capacity of the blending station has not been decided. However, one thing is certain, the blending equipment must be big enough to blend REN's transmission volume. With this type of blending operation REN aims to homogenize and control hydrogen concentration through the natural gas network.

Chapter 5: Gas Quality Monitoring

Gas quality and its monitoring are critical terms in natural gas industry. With the introduction of new and renewable gases into the sector, the quality of the gas varies significantly from region to region. Therefore, gas quality and monitoring are more important than ever. In Chapter 5 gas quality parameters, quality standards in European Union and gas quality measurement methods and equipment are examined and explained in detail. Since the main goal of the thesis is to investigate hydrogen injection into natural gas system in REN, new approaches of gas monitoring from the literature are gathered and discussed. Overall, chapter 5 aims to explain the current and future practices of gas monitoring.

5.1 Introduction

The concept of gas quality determines general properties of gas in terms of chemical composition and thermodynamics. Natural gas usually has a complex chemical composition which includes different types of hydrocarbons and trace elements. The main components of a natural gas mixture can be listed as CH_4, C_2H_6, C_3H_8, C_4H_{10}, N_2 and CO_2.The chemical composition of the natural gas is often required by chemical and process industries. However, instead of defining the gas by its composition some key parameters are used. Some of the most important parameters are used to explain the quality of the gas. These parameters are known as superior calorific value, inferior calorific value, density, wobbe index, hydrocarbon dew point, Joule-Thompson coefficient, and methane number.[65] Each parameter is used for a specific purpose, and they are explained in detail later in this chapter. The composition and quality parameters of natural gas vary depending on the origin, upgrading and transport routes. [66] The thermodynamic state plays an important role with respect to reference condition of storage and transportation. Moreover, it is shaped by temperature and pressure of the gas.

The measurement of the quality of the gas is the basis of commerce among producers, royalty owners, transporters, process plants, and federal government authorities. Because of the size of the natural gas sector, measurement of hydrocarbons has a significant impact on the Gross National Product of exporting and importing countries. [67] Any error in the measurement of the parameters may affect the profit both in short and long term. In addition to the effects on profit, the inaccurate measurements can cause legal liabilities, loss of costumers and adverse publicity. [67] In many gas markets, gas quality must be known prior to supply and transportation, and therefore it is often a prerequisite for custody transfer. [68] That is why in the European market standards are strictly audited and companies are expected to base their business on accurate measurement of the gas quality.

In general, gas quality measurement even of natural gas improves process quality with regard to safety, efficiency, and emissions. This indicates an emerging need for gas quality metering instruments with changed specifications addressing especially the renewable gases. The share of renewable gases like hydrogen and biogas in the market is increasing because of the national and international climate change policies and laws. For this reason, the quality of the natural gas is expected to chance more often than usual. [69]

In this chapter the definitions and the importance of gas quality parameters are mentioned. Different approaches of determining gas quality are presented and examined based on the physicochemical properties. Moreover, essential regulatory framework and standards of gas quality is introduced. In addition, recent developments and new approaches to gas quality monitoring is investigated.

5.2 Gas Quality Parameters

REN follows a uniform standard for natural gas quality which puts specific limits to the most important physical and chemical characteristics of natural gas. This approach provides a range for gas composition which allows REN to have a sustainable business. As mentioned before the key parameters for REN include superior calorific value, inferior calorific value, wobbe Index, and gas density. Following sub-chapters give an overview to the crucial parameters of natural gas and the impact of hydrogen injection into natural gas system.

5.2.1 Gas Composition

Natural gas consists of hydrocarbons, inert gases and trace amounts of other compounds. Although, methane is the predominant component of natural gas, gases like ethane, propane, butane, pentane are often present in the mixture. There are also natural gas impurities which include traces of compounds such as oxygen, hydrogen sulphide, and water. The composition of the natural gas cannot be constant due to the nature of production. In other words, the percentage ranges of compounds in the natural gas fluctuate due to several factors.

The gas composition is used for the calculation of some physical parameters. These parameters often define the technical performance of the gas. Moreover, the composition is one of the biggest reasons why certain type of material (steel, polyetelyne) is selected in the natural gas network. When certain amount of hydrogen is injected to the natural gas, the gas composition completely changes. For example, addition of 20% hydrogen into natural gas will result in decrease of the other hydrocarbons and trace components by 20% in total.

5.2.2 Higher Heating Value

The volumetric higher heating value represents the energy content in a volume of gas when completely burnt in air at standard conditions. [70] volumetric HHV for a gas composition is the sum of the individual components weighted percentage of the components heating values. Methane has a volumetric HHV of 37.7MJ/Sm^3 while hydrogen is 12.1 MJ/Sm^3 at standard conditions. [71]

The likely technical consequence of natural gas with hydrogen used as a fuel in reciprocating engines or gas turbines, without tuning, is a loss of efficiency. [72] Higher heating value is a key to commercial action of the cost of natural gas as it refers to the energy value of the gas. The higher heating value is used as one of the parameters to calculate the cost of gas. [8]

To achieve the same energy throughput for a lower HHV gas composition an increase in flow quantity would be required. Higher volumes of gas may require addition or modification to existing infrastructure such as supply regulators, metering, and increased pipe diameter.

5.2.3 Wobbe Index

The Wobbe Index, is a physical parameter and it represents the measure of the heat output to gas appliances. It is expressed in MJ/Sm^3 and is calculated when the higher heating value of the gas is divided by the square root of the relative density of that same gas.[73]

Gases with different compositions but equal wobbe indexes and pressures before a nozzle have the same thermal power; thereby this parameter reflects information about a thermal load for many systems. [74] The high and low gas quality limits define the values beyond which the WI is not permitted to vary. Within these limits appliances have been designed and tested to operate safely. For gases that are outside the defined limits there may be technical, safety, regulatory and commercial impacts. [8] From the previous chapters it is known that increasing hydrogen percent in the natural gas decreases the wobbe index.

5.2.4 Superior Calorific Value

The amount of heat which would be released by the complete combustion in air of a specified quantity of gas, in such a way that the pressure at which the reaction takes place remains constant, and all the products of combustion are returned to the same specified temperature t_1 as that of the reactants, all of these products being in the gaseous state except for water formed by combustion, which is condensed to the liquid state at t_1. [75]

5.2.5 Inferior Calorific Value

The amount of heat which would be released by the complete combustion in air of a specified quantity of gas, in such a way that the pressure p_1 at which the reaction takes place remains constant, and all the products of combustion are returned to the same specified temperature t_1 as that of the reactants, all of these products being in the gaseous state. [75]

5.2.6 Relative Density

The density of a gas divided by the density of dry air of standard composition at the same specified conditions of pressure and temperature. The term ideal relative density applies when both gas and air are considered as fluids which obey the ideal gas law, the term real relative density applies when both gas and air are considered as real fluids.

5.3 European Gas Quality Standards

Challenges of a nonunified gas quality standards have been discussed in European level for so many years. That is why The European Commission has started working on harmonizing the gas quality standards. The project has been executed by the cooperation of European Association for the Streamlining of Energy Exchange (EASEE), Gas Infrastructure Europe (GIE) and European Committee for Standardization (CEN). This cooperation has resulted in gas quality CBP in 2005.[76] Mainly CBP 2005-001/02 defines important combustion and non-combustion gas quality parameters and these parameters ranges.[76] All of these parameters and ranges are accepted by the European Union countries. [76] To be specific 2005-001/0210 set limits on wobbe index, relative density, sulfur content, oxygen content, carbon dioxide content and the hydrocarbon content within natural gas. [77]

After the first projects unifying gas market, CEN developed the CEN/TC 234. These standards simply address to operations, maintenance, design and construction of the gas infrastructure to ensure technical operability between countries. In comparison to CBP 2005-001/02, CEN/TC 24 proposed wider range of wobbe index and lower sulfur content in the natural gas. In addition to European standards, there are international standards which has a significant impact on the CEN/TC 234. ISO EN 16726 defined gas quality parameter ranges and their measurement conditions.

Currently REN follows International Standard ISO 13686 and quality specification published by Portuguese legislation regarding natural gas quality (Despacho no: 19 624-A/2006) as well as many other standards.

5.4 Gas Quality Measurement Methods and Devices

There are 3 major categories of methodologies of determining gas quality. These categories are known as direct measurement approach, indirect measurement approach and inferential measurement approach. All of these approaches are used to measure quality parameters like wobbe Index, calorific value, density viscosity. The direct approach measures the heat released from a defined specific combustion with a calorimeter. [78] In the indirect measurement approach stoichiometric combustion is involved. Furthermore, inferential approach uses the composition-based calculations or correlation with other measured parameters. [79] Currently REN prefers to use inferential approach of Gas Chromatographs to measure gas quality. There are several criteria to consider while deciding a measurement method for any system. These criteria can be listed as accuracy of the measurement, the desired gas quality parameter, the cost of the measurement, volume of natural gas to be measured and the time response characteristics. [74] All of these criteria represent a constraint to the gas quality measurement and monitoring. According to the needs of business an optimized method of measurement must be selected.

5.4.1 The direct measurement approach

The direct measurement of the calorific values or wobbe index is based on direct measurement of the heat release to a heat exchange fluid, typically an air flow or water heat sink, while completely combusting a defined quantity of gas. Although, the gas outdated calorimeters were replaced with gas chromatographs, there is a growing interest in calorimeters because of its ability to measure the energy content directly. [80] There are numerous calorimeters in the market and some of them are listed below. Most of them offers high accuracy, relatively low capital cost and fast response rate for continuous monitoring of the natural gas sector.

Union Instruments produces a calorimeter called CWD 2005 which aims to determine the gas quality of process gases. The calorimeter's main component is an acoustic specific density measurement cell. Within this cell wobbe index and the specific density are measured directly, calorific value and CARI are calculated from these values. It is suitable for all combustible gases - even low calorific gases can be measured with a support gas supply. [81]

Yokogawa Electric Corporation produces the CM6G Gas Calorimeter is used to measure and control the calorific value or wobbe Index of the sample gas. In this calorimeter, the sample gas is burnt at the burner with air and the temperature difference between the combustion exhaust gas and the feed air at the burner inlet is detected by using a thermocouple. [82]

GasCVD Model CVM400 is developed by Azbil Corporation. The calorimeter conforms to international measurement standards. The main advantage of the device is the low capital cost. The company states that Model CVM400 costs 20-30% less than a traditional gas chromatographs in the market. Moreover, it provides a response rate of 5 seconds at the fastest which is usually 30 times faster than the ordinary gas chromatographs. [83]

5.4.2 The indirect measurement approach

Indirect measurement method is based on correlative relationship between different gas quality parameters. To be specific, the linear relationship between the residual oxygen content and the inferior calorific value is used. The stoichiometric mixture of the fuel and air is determined by either thermocouple prob measuring the maximum flame temperature or the lambda prob measures the residual oxygen. With this method the combustion air requirement index (CARI) or the air demand can be determined. [78] In the market several leading companies offer a variety of indirect measurement devices some of which are listed below.

AMS RHADOX 7300 determines wobbe Index and air demand by measuring residual oxygen after oxidation. Moreover, the device measures gas density and the heat values. The determination of the Gas density is performed according to the Bernaulli method. To achieve high accuracy in the measurement, the parameters pressure and temperature must be constant within low limits. A reference measurement of ambient pressure and temperature at the location of the device compensates the ambient influence. [84]

The COSA 9610™ Wobbe Index Meter's measuring principle is based on the analysis of the oxygen content in the flue gas after combustion of the sample. A continuous gas sample is mixed with dry air at a precisely maintained constant ration. The fuel air mixture is oxidized in a combustion furnace in the presence of a catalyst at 800’C, and the oxygen concentration of the combusted sample is measured by a zirconia oxide cell. The residual oxygen provides an accurate measurement for the combustion air requirement of the sample gas, which can be correlated accurately to the wobbe Index of the gas. [85]

5.4.3 The inferential measurement approach

The inferential measurement methods are the most used approach in the modern natural gas industry. This category of measurement includes gas chromatographs, process gas chromatographs, mass spectrometers and optical gas analyzers.

5.4.3.1 Gas chromatography as inferential approach

Gas chromatography (GC) is a separation technique used to isolate volatile components of a mixture depending on differences in the mode of partitioning between a flowing mobile phase and a stationary phase. In the natural gas industry gas chromatographs are widely used because of the complex chemical composition of the gas mixture. The operator collects the gas from the gas field, saves it in the gas storage tank, and then determines the gas composition and the concentration via the column chromatographic separation technique in the laboratory. [86] The main working principle of the gas chromatographs is to separate all the gas components into columns and process every gas individually with detectors. With the measured fractions of each component, gas chromatographs calculate the desired gas quality parameters such as relative density, wobbe index and calorific value.

Hydrogen injection into natural gas networks is a recent development for the sector. Therefore, some of the effects of hydrogen on the grid equipment is still uncertain. However, there is a certain effect of hydrogen into the typical gas chromatographs. Most of the chromatographs in the sector utilizes helium as a mobile phase carrier. Since, the thermal conductivity values of helium and hydrogen is exceptionally high compared to other gases and close to each other, higher concentrations of hydrogen in the natural gas results in errors during measurements. [74] Therefore, hydrogen concentration in the natural gas must not exceed 8% if traditional gas chromatographs are in use in the network. [74] Some of the producers have adapted their products by adding a second mobile phase carrier (Argon or Nitrogen) for enabling accurate measurement when hydrogen concentration is high in the natural gas mixture. Some of the producers offer gas chromatographs which allows hydrogen in the system. These devices and their producers are given in Table 15.

Table 15 Gas chromatography devices, their producers, the mobile phase carrier and hydrogen acceptance.

Name of the Device	Producer	Mobile Phase Carrier	Hydrogen Acceptance in Natural Gas
EnCal 3000 [87]	Elster	He	Up to 5%
PGC 9303 [88]	RMG	He	Up to 5%
PGC 9304[88]	RMG	He and Ar	Up to 20%
SAM-Bio [89]	Siemens/Marquis	He	Up to 3%
SAM- Complete [89]	Siemens/Marquis	He	Up to 5%
SAM- Complete-Advance [89]	Siemens/Marquis	He and N2	Up to 25%

5.4.3.2 Mass spectrometry as an inferential approach

Mass spectrometers are another alternative for the gas quality measurement. This technology can measure major and minor components within the natural gas mixture. The accuracy strongly depends on the design of the spectrometer and the complexity of the natural gas mixture. to measure principal components and many minor components with varying accuracy, depending on the mass spectrometer design and the complexity of the gas mixture. [90] Mass spectrometers which are used in the natural gas sector typically have a software that measures gas concentrations. This software allows users to calculate wobbe index, mass balances real-time. [90] There are many mass spectrometers which can operate in the natural gas industry. Some of these devices are mentioned below.

Thermo Scientific Prima Pro process mass spectrometers have been used to provide fast, online, accurate analysis of the properties of a wide range of fuel gases for many years. The MS measured component concentrations and calculated standard energy parameters as derived Values, including Calorific Value (Upper & Lower), Specific Gravity, wobbe Index and Density according to ISO 6976. Moreover, Prima PRO provides significantly faster and more accurate hydrogen analysis than a thermal conductivity detector, enabling rapid detection of water leaks into the furnace. It can also be used to determine the water content in flux additions. [91]

The Extrel's MAX100-BTU uses quadrupole mass spectrometer is designed to deliver a continuous online analysis of natural gas. The device analyzes hydrocarbons, H_2, CO, CO_2, H_2O, H_2S and other gases. At the end of the analysis wobbe index, CARI, specific gravity and density is measured with high accuracy. It has the speed necessary to analyze the total composition of the sample and report the Heating Value in seconds. Furthermore, the maintenance and calibration requirements are relatively low compared to other devices. [92]

5.4.3.3 Optical gas analyzers as inferential approach

In contrast, gas sensors based on optical absorption offer fast responses (time constants below 1 s are possible), minimal drift and high gas specificity, with zero cross- response to other gases as long as their design is carefully considered. Measurements can be made in real time and *in situ* without disturbing the gas sample, which can be important in process control. [93] Optical gas analyzers normally employ physical displacement of an optical element to perform the sensor channel (wavelength) scan. In order to achieve high resolution scanning over a large bandwidth, a long optical path and a bright source are required. However, unlike the mass spectroscopy there are some species which are transparent in the IR and so produce no optical signal. Hence, all optical spectroscopy techniques are blind to some materials and the optical sensor channels must be supplemented with some complimentary sensor technology. [90] There are a couple of products available on the market as optical gas analyzers. However, they are not successful on measuring hydrogen content since hydrogen in infrared inactive.

The Precisive 5-282 Gas Analyzer is designed specifically for online gas quality monitoring of natural gas products. wobbe index, gas density, specific gravity, the gas composition, and the calorific value are measured accurately. The analyzer is based on MKS' Tunable Filter Spectroscopy platform which directly measures the component concentrations in the gas. This device is used for pipeline gas fiscal metering and natural gas quality measurement in LNG terminals. [94]

5.5 Alternative Approaches to Gas Quality Monitoring

Most of the gas quality instruments presented in the proceeding sections require relatively high investment and operational costs. In the natural gas sector, most of the measurement devices are based on gas chromatography. [95] There are two main disadvantages of the GC measurement technology. Firstly, it usually takes hours to get a meaningful result. Secondly, the method itself requires technically skilled operators. That is why relatively cheaper electrical sensors increased their share in the gas monitoring market. [86] Nevertheless, these sensors on its own are not suitable for gas composition analysis because of the cross sensitivity of different gases in the natural gas. Recent research and development in the optimization area have solved this problem by using artificial neural networks (ANN). It is claimed that the data can be rectified by ANN to eliminate the effect of cross-sensitivity. [96] Similarly the following researchers used different types of optimizations to measure a specific property of the gas mixture.

Table 16 Alternative approaches of gas monitoring, their research, optimization model and used sensors.

Research	Optimization Method	Sensors	Measured Property
Areej Shahid et al.	Artificial Neural Network	SnO_2 sensor	CH_4 and CO concentration
Maria Gabriella Xibilia et al.	Deep Neural Network	Soft Sensor	Gas Concentrations
Tanghao Jia et al	Multi-layer perception neural network	Infrared Gas Sensor	Gas Concentrations

Regardless of the optimization methods there are several corelative gas quality measurement devices available in the market. Generally, these devices conduct measurements based on thermal conductivity, speed of sound, temperature, pressure, and infrared spectrometry. The combination of these measurements provides important gas quality parameters with the help of microelectromechanical systems. For instance, GasPT2 is a corelative gas quality device produced by Advantica. It uses speed of sound sensor and thermal conductivity sensor to analyze gas compositions, wobbe index and calorific value.

Chapter 6: Potential Degrading of the Infrastructure

One of the biggest concerns about the hydrogen transportation in the existing natural gas network is the degradation of steel infrastructure due to hydrogen effect. Chapter 6 focuses on the effect of hydrogen on steel pipelines. These effects are defined and classified. The term of hydrogen embrittlement is analyzed in detail and a few factors affecting it are discussed. Moreover, previous research which confirm hydrogen caused degrading in the steel pipelines are included and investigated.

6.1 Introduction

Pipelines are the safest method of transporting huge volumes of oil and gas. [22] Hydrogen transport in the existing natural gas pipeline system is proposed to increase the share of hydrogen in the energy mixture. The durability of some metal pipes can be degraded when exposed to hydrogen over a long period of time, particularly with injection at high hydrogen concentration and pressure. [21] The effect of hydrogen in the pipeline systems depends on the operating conditions and the material of the pipeline. Since REN's infrastructure of pipeline is based on various carbon steel types of API 5L B, X42, X52, X60, X70 this chapter focuses on the hydrogen effect on mentioned pipeline steel.

Hydrogen induced failures and degradations are considered as a major integrity concern for the steel pipelines under the pressure levels of natural gas transmission and distribution. [21] Because of the nature of the operations in natural gas sector, the pipelines are continuously exposed to severe conditions. Moreover, the combination of these conditions and diffused hydrogen in the pipeline may result in failures caused by hydrogen induced damage. [97]

It is rather easy to claim that hydrogen has a degrading effect on steel pipeline, but the limits and the condition of this degrading is an ongoing investigation for the natural gas and hydrogen sector. Therefore, the driving force behind this chapter is to introduce hydrogen degradation mechanisms and the effect of increased hydrogen levels in the system.

6.2 The Degradation in Carbon Steel Pipelines Caused by Hydrogen

There are several mechanisms involving hydrogen which have caused embrittlement or damage to steel pipelines. Some of these mechanisms are investigated in detail under this section. Hydrogen Embrittlement (HE) is the general term which explains all the detrimental effects which can be observed on pure metals or alloys in hydrogen including environments. [98] Some of the effects are more serious than others. The operating conditions, mechanical properties of the pipeline steel and the chemical composition will have a significant influence on the occurrence of the different hydrogen embrittlement modes.[98] In the scope of this thesis most relevant hydrogen caused degradation modes are selected and presented.

6.2.1 Hydrogen Induced Cracking (HIC)

Hydrogen-induced cracking is a degradation mode which occurs in three steps. Firstly, there is formation of hydrogen atoms at the steel surface and adsorption on the surface. Secondly, adsorbed hydrogen atoms diffuse into the steel substrate. Lastly, the accumulation of hydrogen at hydrogen traps like voids around inclusions in the steel matrix leads to increase in internal pressure. [99] Simply, the increase in the internal pressure becomes the main reason of crack initiation and propagation. [99] Cracking requires the production of nascent hydrogen atoms (H_0) at the steel surface, usually by a corrosion reaction in an H2S-containing gas. Under the service conditions, hydrogen atoms that permeate through the steel may cause embrittlement or failure.[99]

6.2.2 Hydrogen Stress Cracking (HSC)

If there is hydrogen gas in a metallic environment, the material can become brittle. Because of increased brittle behavior the material may fail when the stress level is high. [99] Hydrogen stress cracking is observed when the sustained load in hydrogen environment causes a brittle fracture on a typically ductile alloy. Usually, the brittle fracture occurs at sustained loads below the yield strength of the material. [99] This cracking mechanism depends on, strength of the material, microstructure, applied stress, and temperature. Most of the steels have a threshold stress. If this threshold stress is exceeded hydrogen stress cracking takes place. The threshold stress is characterized by the yield and tensile strength of the steel. When the strength of the material increase, the threshold stress decreases, and the material becomes vulnerable for hydrogen stress cracking.

6.2.3 Reduction of Tensile Ductility and Notched Tensile Strength

Hydrogen gas will reduce the tensile strength/ductility and notched tensile strength of susceptible materials in high pressure environments. The effect can be determined by conducting tensile tests of candidate materials in dry air or inert gas comparing the results of similar tests conducted in a hydrogen environment. Premature failures of burst discs have been attributed to this phenomenon. [98]

6.2.4 Sulfide stress cracking (SSC)

This is a subset of hydrogen embrittlement in which the cathodic reaction controls the cracking reaction. [98] It is usually reported in localized hard zones in weldment of susceptible materials. [98] In the heat-affected zones adjacent to welds, there are often very narrow hard zones combined with regions of high residual tensile stress. When these zones become embrittled to such an extent by dissolved atomic hydrogen that they crack [99]

6.3 The Effect of Microstructure and Micro Alloying Composition on Hydrogen Embrittlement

As it is discussed previously in this chapter, mobile dislocations can accumulate hydrogen on defects which constitute obstacles to their movement. This hydrogen accumulation meets with existing residual stress, and initiation or propagation of cracks are promoted. [100] The local accumulation of hydrogen is favored by a coplanar movement of dislocations and depends on the nature and distribution of defects in the microstructure. [21] When hydrogen enters the material, it tends to accumulate in a wide variety of locations within the microstructures such as grain boundaries, inclusions, voids, and dislocation entanglements. [101] Therefore, it is important to understand the microstructures and their possible effects on hydrogen embrittlement.

During the production, thermomechanical processing leads to a wide range of different microstructures depending on the properties of the steel. Ferritic pearlite, polygonal ferrite, acicular ferrite, bainitic ferrite, bainite and martensitic microstructural phases are the most observed ones.[102][103]. The steel processing parameters like cooling rate have a significant influence on the formation of the mixture of these microstructure phases. [104] For example, fast cooling rates and low transformation temperatures produces bainitic ferrite, followed by acicular ferrites in decreasing order.[97] Additionally, higher transformation temperatures and slower cooling rates often generate polygonal ferrite microstructure. The relationship between microstructures and hydrogen damage has been investigated by many authors. Bhandeshia claimed retained austenite structure is the most suitable to avoid hydrogen induced cracking. [105 Moreover, Mori G et al. concluded his study that bainitic and acicular ferrite structures have resistance to hydrogen damage. Since, retained austenite are commonly found dispersed within acicular ferrite and bainitic ferrite microstructures these two studies support and justify each other. Furthermore, Ramirez et al. observed the presence of martensitic phase in pipeline steel increased susceptibility to hydrogen damage. [106]

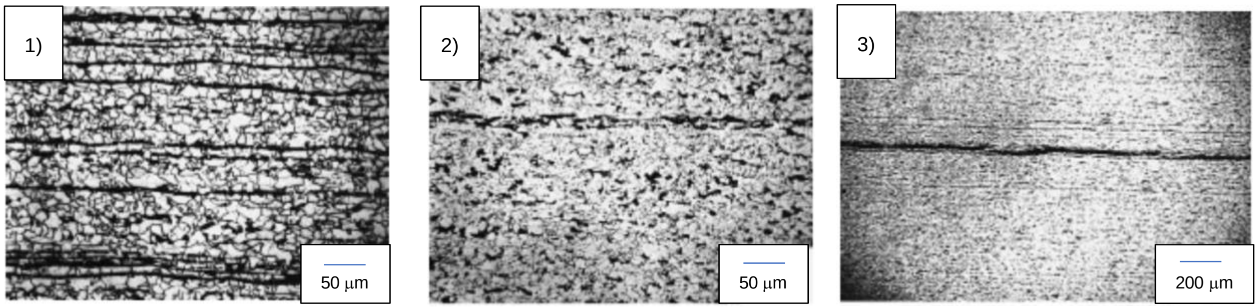


Figure 17 Optical micrographs of 1) Pearlitic bands across X52 steel. 2) Segregation of pearlitic bands in X60 steel. 3) Crack at the ferrite-pearlite banding in X60 steel.[107]

Figure 17 shows banded microstructural phases which are common features in lower grade pipeline steels like X52, X60. While Figure 17.1 presents pearlite bands uniformly distributed across the microstructure of X52 pipeline steel, Figure 17.2 shows pearlitic bands in X60 steel which has a higher strength. Hydrogen induced cracking is usually occurs in these banded regions. For instance, Figure 17.3 highlights the crack which is propagated along the ferrite-pearlite banded zone of X60 pipeline steel.

Micro alloying was developed as one of the biggest breakthroughs in the steel industry. In steels, small amounts of niobium or vanadium or titanium were added, and significant grain refinement was obtained due to retarding the recrystallization by precipitates of carbon-nitrides of these microelements. [108]

The purpose of adding Nb, V and T was to decrease the amount of microstructural grain boundaries and limit the grain growth in pipeline steel. [109] Clearly every micro alloying element will have a different effect on the pipeline steel. Under this sub-section micro alloying elements which has the biggest impact on hydrogen degradation is chosen from literature and listed below:

1) According to Shi et. al. Copper (Cu) addition during production increases the strength in X120 pipeline steel. Because copper rich precipitates in the microstructure acts as hydrogen trap sites. Since, hydrogen is trapped in the Cu rich traps irreversibly, the hydrogen diffusion decreases significantly. Therefore, the risk of hydrogen degradation is minimized.[110]

2) Similarly, Baba et al. states in this research, alloying with Cupper (Cu) decreases hydrogen diffusion and corrosion rate in the pipeline steels.[111]

3) Chen et. all investigated the effect of adding Niobium (Nb) in steels and experimentally proved that micro alloying with Niobium improved the mechanical properties significantly.[112]

4) Capdevilla et. al. claimed that without Titanium (Ti) the risk of hydrogen degradation in the high strength steels is increased.[113]

5) Haddadi et. al. Stated that adding small amounts Manganese (Mn) and Phosphorus (P) increase the strength of the steel. [114]

6) Haq et al. investigated micro alloying with different amounts of Manganese (Mn) in X70 pipeline steel. When 0.5%wt of Mn is alloyed, the decrease in hydrogen diffusivity is observed. However, when 1.2%wt of Mn is alloyed the increase in hydrogen diffusivity is found. [115]

This research clearly state that micro alloying is a sensitive process and finding the right amount of micro alloying element might be tricky. However, a general list of micro alloying elements and their effects are given by Ohaeri and Shirband and the modified version is presented in Table 17.

Table 17 Micro alloying element and its influence on hydrogen trapping, hydrogen diffusion and precipitation strengthening. [116][97]

Micro Alloying Element	Hydrogen Trapping	Hydrogen Diffusion	Precipitation Strengthening
Aluminum (Al)	▨	Decrease	▨
Carbon (C)	▨	Increase	▨
Chromium (Cr)	Increase	Decrease	Increase
Copper (Cu)	▨	Decrease	▨
Manganese (Mn)	▨	Increase	▨
Molybdenum (Mo)	Increase	▨	Increase
Nickel (Ni)	▨	Decrease	
Niobium (Nb)	Increase	Increase	Increase
Phosphorous (P)	▨	Increase	▨
Silicon (Si)	▨	Decrease	▨
Sulphur (S)	▨	Increase	▨
Titanium (Ti)	Increase	▨	Increase
Vanadium (V)	Increase	▨	Increase

6.4 The Effects of Hydrogen on Mechanical Properties of the Pipeline Steels

Boukart measured the fracture toughness of several API steel by using notched Roman tile specimens. In the experiment procedure same measurement are repeated with electrolytic hydrogen charging. The fracture is compared before and after hydrogen charging. According to this comparison hydrogen-air fracture toughness ratio as a function of yield strength decreased approximately by 33%. This decrease is observed in API X52 and X70 pipeline steel. Figure 18. shows the difference in the elongation at the rapture caused by hydrogen embrittlement effect. [21]

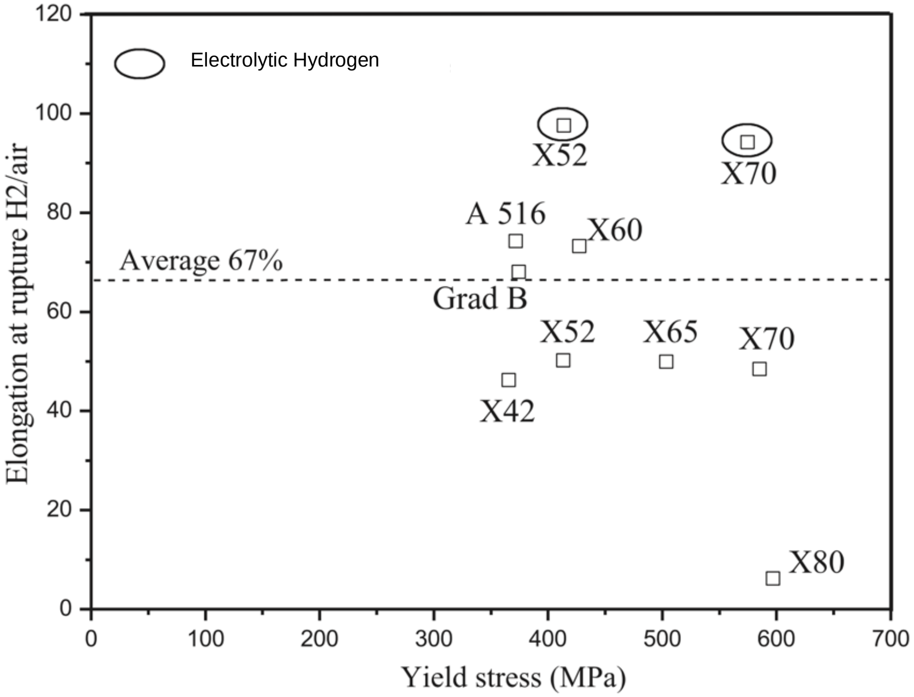


Figure 18 Effect of hydrogen embrittlement in the fracture toughness as a function of yield strength.[21]

Similarly, many researchers have experimented on the topic. For example, Capelle et all investigated the hydrogen effect on API X52, X70 and X100 with Roman tile specimens. According to a set of experiments fracture toughness decreased for X52 and X70 steel pipeline by 10.46% and 4.74% respectively.[22] The change in the fracture toughness can be observed from Capelle er al.'s graph which is shown in Figure 19.

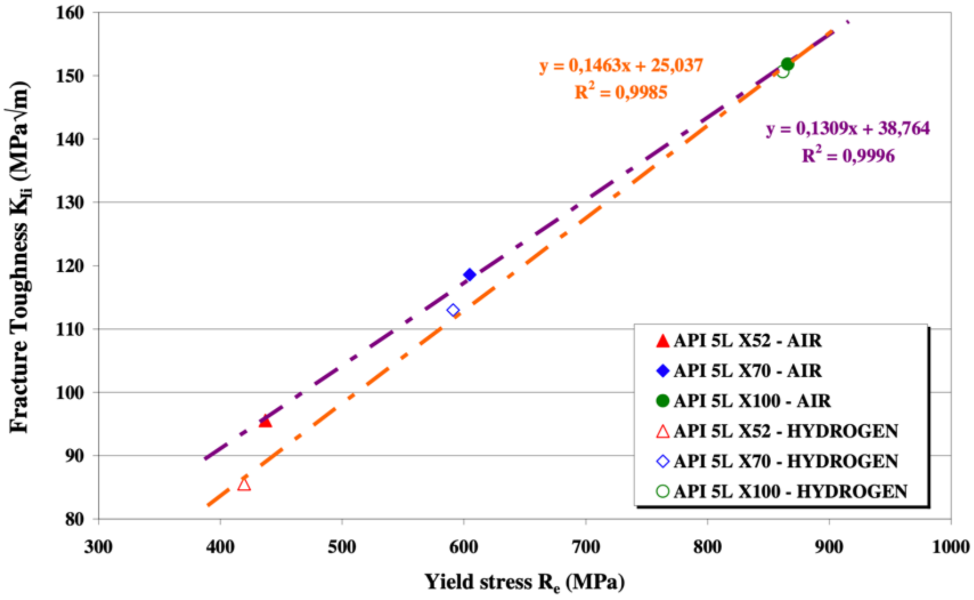


Figure 19 The change in fracture toughness of API 5 L X52, X70 and X100 pipeline steels in hydrogen charging.[22]

Meliani et al. experimented on API 5L steels with different types of specimens. (SENT, CT, DCB, and Roman tile). As a result of this study a material failure curve based on the critical notch intensity factor is plotted.[23] As an experimental approach, all the specimens are exposed to hydrogen charging for 30 days. Typically, the results for Roman tile specimen are the focus of this thesis because of its pipeline like shape. According to the research, because of the presence of hydrogen charging the notch stress intensity factor decreases 9.8% compared to the control specimen of API 5L X52 steel.[23] The difference in the notch stress intensity factor due to hydrogen charging is shown in Figure20.

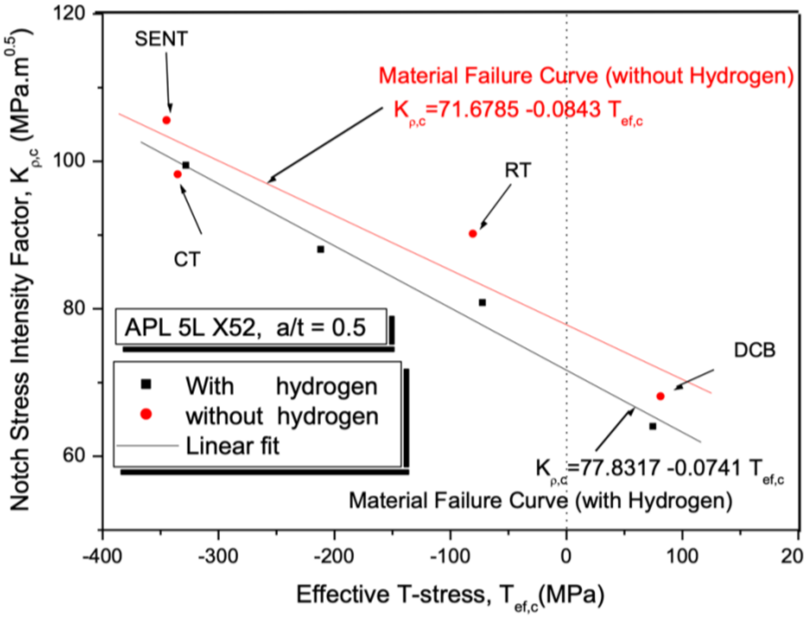


Figure 20 The change in notch stress intensity factor because of hydrogen charging.[23]

Chapter 7

Conclusions

In chapter 7, the general overview of the study is given with results. A summary of the important points from previous 6 chapters are given. The business model of hydrogen injection is presented with constraints and technical results. Proof of technical possibility of hydrogen injection is given with examples. Moreover, the future research necessities and opportunities are mentioned.

Conclusions

The development of laws and policies against the greenhouse gases has increased the importance of carbon free energy solutions in the world. Hydrogen has a high potential of being the primer force in this energy transition because of its low carbon nature and its wide range of applications. It is known that hydrogen can be utilized in energy production, energy storage, transportation as a fuel and heat source for many industries. However, one of the most interesting utilization methods of hydrogen is the injection to natural gas networks. Transportation cost of hydrogen in pressurized vessels are genuinely high, that is why hydrogen transportation and usage in natural gas industry is proposed as a solution.

Many of the member states in European Union started investing on hydrogen production and utilization methods because current policies of European Commission promote the green hydrogen economy. In case of hydrogen injection into natural gas network many countries still investigate the possible outcomes of such application while some countries like France, Germany and Spain allow significant percentages of hydrogen in their system. With development of the new hydrogen strategy Portugal aims to be one of the countries which injects hydrogen in their transmission system up to 5% by 2025 10% by 2028 and by 15% by 2030. REN as the main TSO in Portugal will play a key role in this energy transition process. The principal responsibility of REN will be receiving hydrogen from producers and blend it to the natural gas mixture. While producers are building the connection to natural gas REN will supervise and control the sustainability and the durability of the hydrogen injection into the network. There are several projects all around the world which proves that regular natural gas networks can handle 10% hydrogen in the system with minor operations. These projects are HyDeploy by U.K., Snam by Italy, Dunkirk by France, and HyBlend by U.S.A.

Although, it is proven by several research and development projects hydrogen injection is possible up to 20% with minor adaptations, the changes in the system cannot be ignored. In the chapter 4 these changes are analyzed and calculated. The expected changes in the system from 0% hydrogen injection to 15% hydrogen injection is given in Table 18.

Table 18 Summary of the changes in the system

Change between 0% and 15% Hydrogen Injection			
The Change in the System	Unit	Change in %	Decrease/Increase
Gas Density	kg/m^3	14,0574127	Decreases
Viscosity	*10^{-5} Pa s*	0,733932435	Decreases
Lower Heating Value	MJ/Nm^3	10,02731905	Decreases
Higher Heating Value	MJ/Nm^3	9,740184439	Decreases
Wobbe Index	MJ/Nm^3	2,637833381	Decreases
Lower Explosive Limit	%	2,61828204	Decreases
Upper Explosive Limit	%	12,1884984	Increases
Pressure Drop	Pa	0,622899086	Decreases
Linepack	KWh	10,10169447	Decreases

As it can be seen in Table 18. most of the changes are related with natural gas quality. Therefore, gas quality monitoring has a critical importance. Because of this reason, gas quality monitoring methods, equipment and new approaches are investigated under Chapter 5. Gas chromatographs which has hydrogen tolerance are presented and suggested. Additionally, because of the high capital cost of conventional gas chromatographs the usage of relatively cheaper gas sensors and application of artificial neural network method is investigated and suggested for increasing hydrogen volumes in the gas mixture.

Finally, one of the biggest concerns in the natural gas industry which is possible degradations due to hydrogen content is studied under Chapter 6. The hydrogen embrittlement and its modes are given and their degrading effect on REN's pipeline infrastructure is discussed. From literature, experiments including roman tile specimens are selected and hydrogen's negative effect on the mechanical properties of carbon steel pipeline is presented.

To conclude, hydrogen blending with natural gas will technically be feasible with minor adjustments in the current infrastructure for blends which has less than 10% hydrogen. If the hydrogen percentage exceeds this limit additional research and adaptations will be required especially for hydrogen degrading effect and gas quality monitoring. For the first years of the project the main investment for REN will be building the blending station and designing the blending equipment for large quantities of natural gas and hydrogen blend.

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