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Development of refining industry and reduction of greenhouse gas emission

Introduction

Crude oil refineries are industrial plants which are characterized by high consumption of energy and at the same time high emission of greenhouse gases (GHG). Owing to this fact, they are in the centre of dispute on the possibilities of reducing the GHG emission, especially in view of accepted obligations in this respect which result from the law stipulations in the European Community and in the world.

Reduction of greenhouse gas emission in the oil sector is a difficult task due to continuously rising demand for engine fuels and light combustion oils, which makes it necessary to increase crude oil processing and thus involves larger consumption of energy. The second element of higher GHG emission in the refining industry is continuous growth of requirements concerning the quality of fuels. Production of larger and larger volume of light, highly refined fuels requires higher energy consumption and results in increased GHG emission.

Legal situation

The results of decisions of the Framework Convention on Climate Change [21] and Kyoto Protocol [5] are obligations undertaken by particular countries-signatories to reduce the emission of greenhouse gases according to the accepted program which makes it possible to apply supporting mechanisms including trading schemes of permitted emission values.

The principal legal act on the emission trading scheme in the European Union (EU-ETS) is the Directive 2003/87/EC dated 13 October 2003 [4] setting the trading scheme with EC permitted greenhouse gas emission and changing the directive of the Council 96/61/EC. Meanwhile, a similar legal act in Poland is the Act dated 22 December 2004 on trading with permitted emission of greenhouse gases and other substances into the air [14].

The above regulations establish the periods of balancing, concerning verification and reports of greenhouse gas emission, i.e. CO_2, CH_4 and N_2O in particular member states. The first trial period lasted three years (2005–2007), the current one will last five years (2008–2012). The European Commission allocates to the member countries limits of permitted emission for a particular balancing period on the basis of elaborated State Plans of Distribution of GHG Emission. The governments of particular countries distribute obtained permissions among owned installations included in the EU ETS system. In Poland, a register of enterprises covering the emission trade was created: 'The list of installations registered in the KRU [national register of emission permits]'. According to the above regulations, by the year 2020, planned reduction of joint greenhouse gas emission in the Community will be at least 20% lower than the one recorded in 1990.

In order to improve and expand the Community greenhouse gas trading scheme, the Directive 2003/87/EC was substituted with the Directive of the European Parliament and Council 2009/29/EC dated 23 April 2009 (the EU ETS Directive) [19]. Due to the fact that the Community greenhouse gas trading scheme covers currently about 10 thousand installations in the energy generation sector and other industry branches which are responsible for almost half of the emission of CO_2 and 40% emission of all greenhouse gases, the EU ETS system was considered to be the basic tool for lowering the emission in the Community.

According to the Directive 2009/29/EC in the third stage of implementation of the EU ETS scheme in years 2013–2020, the greenhouse gas emission should be decreased by 21% in comparison to the emission levels in 2005. This fact determines the average number of permissions allowed for a business entity covered by the scheme, regardless of the method of distribution.

Directive 2009/29/EC introduced changes in respect of the possibilities of purchasing permissions for emission in the EU ETS scheme. Industrial sectors may obtain 90% of free of charge permissions in different ways. Three groups of industrial sectors were determined:

- exposed to CO_2 leakage (obtain 100% free permissions upon fulfilling certain conditions),
- electric energy producers,
- remaining industrial sectors, including heat producers.

In the two latter groups of sectors the share of free of charge permissions for emission will diminish successively: from 80% in 2013 to 30% in 2020 and 0% in 2027.

Thus, effective reduction of greenhouse gas emission by the business entities belonging to the EU-ETS scheme, including refineries, will become a necessity.

Sources of generating greenhouse gas emission in refineries

In European refineries over 90% of GHG emissions originate from burning fuels both in the process installation and heat, steam and electric energy generation. The remaining 10% of GHG emission comprise: carbon dioxide emitted in production of hydrogen used in desulfurization and saturation of different streams of hydrocarbons, methane whose emission may occur in exhaust gas combustion (torches, waste incinerators), in safety valves which release gases to atmosphere and due to leakages [7].

The volume of greenhouse gas emission in oil refineries depends on many elements which include first of all:

- type of refined mineral resources i.e. crude oil,
- level of complexity of crude oil processing (process diagram),
- type of refined fuels,
- technical solutions concerning production and use of energy,
- level of optimization of energy usage,
- application of biocomponent additives in production of ready fuels.

Quality of raw materials

The European Community is dependent on the import of crude oil; in 2000 even 75% of supplies of processed oil came from import which included about 37% from the OPEC, 27% from the OECD, about 29% from Russia and the former Soviet Republic states and 7% from other sources [11].

The oils coming from different regions of the world differ substantially in their properties. Over a long period of time, in respect of changing oil quality, a switch tendency has been observed from sweet crude to more and more acid crude and from light to heavy one.

In the year 2000, in the Community countries together with Norway and Switzerland (EC 27 + 2) about 695 MT of crude oil was processed and current use amounts to about 715 MT. By 2020, further oil consumption increase is planned to about 765 MT. Table 1 presents current and future volumes of processing various kinds of oil in the EC countries 27 + 2 [11].

It is common knowledge that the processing of lighter oil (crude oil) renders the possibility of reduced CO_2 emission. Heavier oil requires larger amounts of energy to obtain the same output of light products due to the necessity of converting larger volumes of remaining material and the necessity to remove more amount of sulphur. Processing of light oil will be more difficult to execute due to increased share of heavier oils with larger contents of sulphur in the market.

Table 1. Volume of processing various kinds of oil in the EC countries 27 + 2 in years 2000–2020

MT/year	2000	2005	2010	2015	2020
Brent*	2281	238.2	254.7	265.4	265.7
Nigerian	58.7	58.7	58.7	58.7	58.7
Algerian condensed	1.7	1.7	1.7	1.7	1.7
Iranian light	143.0	143.0	143.0	143.0	143.0
Ural	139.0	128.9	112.4	101.7	101.4
Kuwait	71.3	94.7	As needed		

*Apart from crude oil, about 20 MT/year of Brent oil vacuum remnants is processed in addition

So far, this trend has almost passed unnoticed in Europe on account of favorable logistics of light oils purchase, e.g. from the North Sea, even though increasing consumption of heavy oil from the Middle East has also been recorded [11].

It should be noted that in the recent years possible increase of Ural oil consumption has been compensated by light oil from the North Sea, i.e. Brent oil which constituted about 10% of the total raw material feed in Europe. This fact resulted in decreased energy consumption in refineries by 4.2% and decreased CO_2 emission by 6.8%.

However, larger changes in respect of used raw materials are not possible due to the fact that many Community member states have no deposits and no possibilities to change their system of crude oil import [6].

Diagram of crude oil processing in refineries

Crude oil processing may follow different diagrams depending on the level of raw material conversion, i.e. share of mass production of combustion engine fuels in relation to the mass of processed oil. The most frequently occurring processing diagrams are presented in Table 2 [1, 11].

About 70% of capacities in European refineries work according to diagram no. 2: FCC + VB (DC) and diagram no. 6: FCC + HC + VB, while according to diagram no. 7 (complete conversion), only 4% of total capacity of refining installations operate in Europe. In the USA this diagram of processing is commonly used.

Emissions of GHG from particular processes of crude oil processing in refineries demonstrate substantial variety and their share in total emission of GHG largely depends on the diagram of crude oil processing.

Table 3 presents diversity of percentage shares for particular processes of crude oil processing in total emission of CO_2 in relation to different diagrams of crude oil processing that function in the refineries of North-Western Europe [1].

In case of simple crude oil processing (diagram no. 1) the largest emission of CO_2 is generated by the atmospheric distillation unit which processes the whole raw material feed. Together with increased conversion level of processed crude oil, the share of atmospheric distillation unit in the total CO_2 emission decreases. In semi-complex process diagrams the unit with substantial share in CO_2 emission is fluidal catalytic cracking (FCC). The hydro cracking unit generally causes smaller CO_2 discharge than the FCC. However, when we add to the emission of CO_2 generated from the HC units the emission from production of additional hydrogen in steam methane reforming, this situation becomes reversed [1].

According to forecasts by specialists, in the years 2010–2020 there will be further expansion of crude oil processing in the European oil sector caused both by stricter requirements in respect of the quality of combustion engine fuels and combustion oils and by increased demand for medium distillates used for production of diesel oil and avgas and by planned, substantially stricter requirements for ship propellant oils [2].

Table 2. Diagrams of crude oil processing in refineries

Processing category	Processing diagram	Description of processing
1 Simple	Topping	Refinery includes only atmospheric distillation unit.
	Hydroskimming (HSK)	Refinery produces chiefly heavy fuel oil (HFO). Light fuels are gasoline and diesel oil.
2 and 3 Semi- complex	HSK + Fluidal catalytic cracking (FCC) + Vis breaking (VB) (2) or delayed coking (DC) (3)	Fluidal catalytic cracking increases production of gasoline by processing heavy fractions and remnants. Coke is removed in catalyst regeneration process. Cracking gasoline requires hydro desulfurization.
4 and 5 Semi-complex	HSK + Hydro cracking (HC) + VB (4) or DC (5)	Installations for hydro cracking increase production of gasoline and medium distillates (it gives flexibility of processing); good quality diesel oil is obtained.
6 Complex	HSK + FCC + HC	Smaller production of gasoline than in the version FCC + DC but larger than in the system HCU + DC. In case of an additional installation IGCC all the remnants are processed. The only heavy product is asphalt.
7 Complete conversion	HSK +HC + FCC + DC	Delayed coking is used for reducing the production of heavy combustion oil. Presence of DC increases production of fuel, additionally coke is produced.

Table 3. CO_2 emission in particular crude oil processing units in refineries of North-Western Europe (NWE)

Processing unit	Crude oil processing diagram				
	1. HSK	2. HSK + FCC + VB	4. HSK + HCU + VB	5. HSK + HCU + DC	6. HSK + FCC + HCU + VB
Crude oil in thousand ton	26 762	150 809	31 027	14 232	42 298
Atmospheric distillation	47%	28%	30%	26%	27%
Vacuum distillation	13%	8%	8%	8%	9%
FCC	-	22%	-	-	11%
Isomerization	2%	2%	0%	1%	1%
Hydro refining of paraffin oil	-	1%	-	-	-
Hydro refining of diesel oil	8%	5%	5%	4%	4%
Hydro cracking of HCU	-	-	12%	13%	9%
Vis breaking of VB	4%	2%	2%	-	3%
Delayed coking DC	-	-	-	5%	-
Hydro treating	5%	5%	5%	4%	3%
Reforming 1/2R	11%	8%	11%	1%	1%
Reforming CCR	3%	7%	10%	16%	13%
Reforming SPLIT	8%	10%	12%	10%	8%
Other	1%	2%	0%	-1%	-1%
Total units	100%	100%	95%	86%	91%
Steam methane reforming	-	-	5%	14%	9%
Total	100%	100%	100%	100%	100%

Refinery fuel

The main fuel burned at European refineries are light process gases (C_1–C_2) and coke from the FCC installation. The remaining fuel, used on average in 25% as compared to the total consumed fuel, is liquid one – usually crude oil residues of low value.

The most beneficial fuel for European refineries is natural gas which does not require any processing and produces low emission. The European refineries demonstrate large diversity in proportion of consumed liquid and gas fuels from the sole use of gas e.g. in Norway, New Zealand and Austria through majority of gas (Slovakia, the USA, Finland, Holland) and majority of combustion oil (Poland, Turkey) to sole use of combustion oil e.g. in Sweden and Portugal [1, 11].

Substitution of liquid fuel with imported natural gas is an effective method of limiting the CO_2 emission. In general, 5 to 10% energy is produced from natural gas in the European refineries [6].

However, in order to assess the benefits of switching from liquid refinery fuel to natural gas in the area of the Community, the result in the global scale has to be taken into account. Increasing demand for natural gas in Europe will bring about higher price of this fuel, and thus, consequently, lack of possibility of buying it by entities with poorer financial position which will be forced to use larger volumes of liquid fuels. In this way, CO_2 reduction in Europe will result in its increased amount in the remaining regions of the world [6].

Technical modernization in energy production and usage

On account of high energy consumption of crude oil processing, for many years there have been activities in refineries aiming at increasing energy ‘efficiency’ i.e. consuming lower amounts of energy to power the same devices and equipment units. Only between 1995 and 2005 the efficiency of energy use increased in the refineries of the Community by 13%. It was connected with optimization of heat exchange of particular processes and operations and using new pumps and compressors or modernization of the existing ones. Central management of energy allowed to implement further savings, especially in new refineries with modern technical equipment [1, 6].

Electric energy as well as heating steam in a refinery can be produced with the use of cogenerated heat and power system (CHP), gas/steam turbines or IGCC (*Integrated Gasification Combined Cycle)* installations – a gasification complex for refinery residues. The latter method makes it possible to obtain synthesis gas and then electric energy with efficiency higher by 45÷55% than conventional coal-fueled power plants with very low emissions of NO_x, SO_2 and CO_2 [8]. Some refineries use external sources for production of electric energy, among which the ones with the highest emission are the sources of obtaining energy from combustion of hard or brown coal.

Carbon capture and storage – CCS process

A prospective method of limiting CO_2 emission is its capture and storage (CCS – *Carbon Capture and Storage*) i.e. capture and storage of carbon dioxide. Up till now, the agent used for capturing (absorption) of CO_2 from exhaust gases was solution of alkyl amine. As it is the principal source of costs of the CCS process, currently intensive research is being done concerning the possibilities of substituting alkyl amine with another agent or method of capturing carbon dioxide. The next stages are: compression of CO_2 and its transport to the storage site. Carbon dioxide is stored underground in natural geological formations e.g. exploited oil and gas deposits on land or off-shore under the bottom of the sea [10].

The CCS process was created for 'ecological' application of mined coal as fuel. Industrial implementation will concern chiefly large emitters of carbon dioxide such as heat and power plants, power plants and cement plants, where it is easier for economical justification of necessary investments. Projects of starting up industrial CCS installations are forecast for 2020. All the three stages of the process (CO_2 capture, transport and storage) have to be considered from the technical point of view and financial profitability. Some legislative actions are necessary too, which will legalize accumulation of large volumes of CO_2 underground.

The first industrial solutions of the CCS process may be implemented in refineries due to the fact that gases from hydrogenation processes are relatively little diluted and the CO_2 emitters produce smaller output [10].

Application of biocomponents

Less effective than the previous variant of CO_2 emission reduction at refineries is the application of the first-generation biofuel for production of engine fuels. Currently this variant is executed by production of a particular batch of petrol with addition of bioethanol and specific batch of diesel oil with addition of fatty acid esters originating from rape or other vegetable oils. The characteristics of first-generation biofuel is presented in Table 4 [13].

Table 4. The first generation biofuels and raw material for their production [12]

Biofuel type	Raw material	Production process
Bioethanol	Sugar beet, grains, sugar cane, corn	Hydrolysis + fermentation
Pure vegetable oil	Oil plants e.g. rape seeds	Cold pressing Extraction
Biodiesel RME, FAME/FAEE	Oil plants e.g. rape, soya, sunflower, palm oil	Cold pressing + trans- estrification
	Used cooking oils	Transestrification
Bio ETBE	Bioethanol	Chemical synthesis

According to the Act dated 25 August 2006 concerning biocomponents and liquid biofuels [15] which is the implementation of the Directive 2003/30/EC of the European Parliament and Council dated 8 May 2003 concerning supporting the application of biofuels or other renewable source fuels in transport [18], the introduction of biocomponents and liquid biofuels to commerce is performed according to the indications of the National Indicative Programme. On this basis 'the entity which executes the National Indicative Program is obliged to provide in a particular year at least the minimal, determined by the Council of Ministers share of biocomponents and other renewable source fuels in the total volume of liquid fuels and liquid biofuels sold or otherwise transferred or consumed for its own needs'.

National Indicative Programme for years 2005–2007 was published in the Report for 2005 elaborated for the European Commission [12] and it was implemented by the Council of Ministers' Order dated 15 June, 2007 for the period 2008–2013 [20].

In general, at the domestic refineries petrol and diesel oils are produced including about 5% (v/v) of biocomponents and small volumes of biofuel B-20 (20% of biocomponent in diesel oil) and B-100 (pure ester type FAME).

On the basis of the announcement of the Minister of Economy for the European Commission concerning supporting the application of biofuels or other renewable source fuels in transport in 2008 [16], the real national indices of biocomponent and biofuel share in the volume of liquid fuels consumed in transport in years 2005–2007 were much lower than the indicated values. However, in 2008 there was an increase in the share of biofuels in the total sale up to 3.66%, owing to which the Indicative Plan was exceeded [16].

Table 5 presents the real consumption of fuels and biofuels in transport and the real index of the share of biofuels in the years 2000–2008 and also planned index of biofuels share in the years 2005–2008.

The presented data demonstrate that there are still enormous reserves in respect of increasing the share of liquid renewable fuels to the level of National Indicative Programme planned for the following years. Complete utilization of these reserves at refineries by producing engine fuels containing maximal permissible share of biocomponents can be the way to reduce CO_2 emission and simultaneously execute accepted indicative plans.

Table 5. Consumption of fuels and biofuels in transport and planned and real indices of the share of biocomponents and biofuels in the volume of liquid fuels

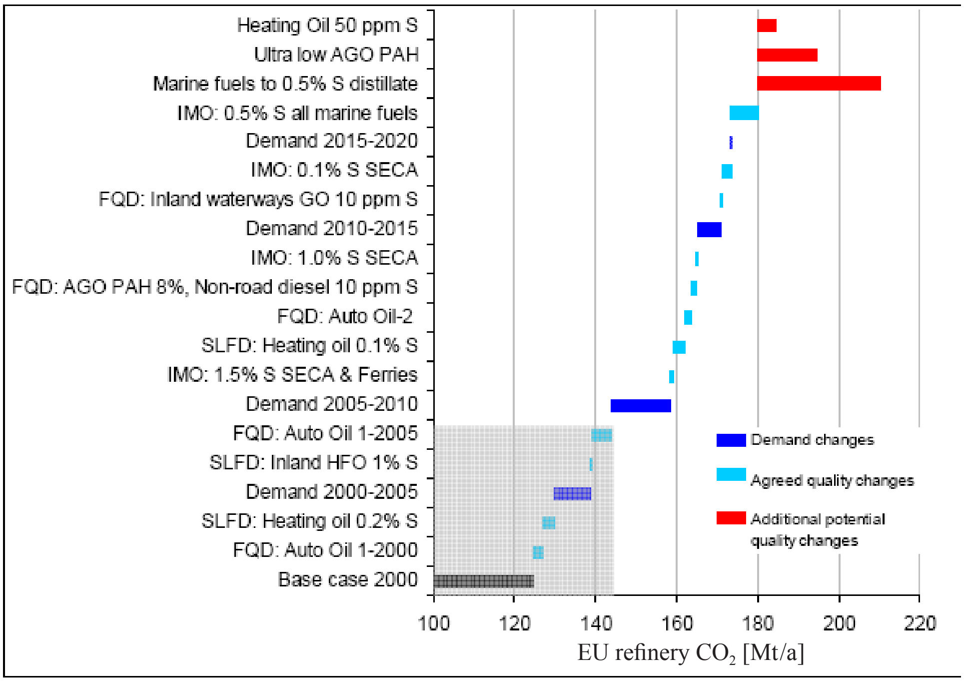
Year	Consumption in transport (thousand ton)				Index according to calorific value in %	
	petrol	Diesel oil	bioethanol	esters	real	planned
2000	4.841	2.343	40.6	0	0.35	
2001	4.484	2.562	52.4	0	0.46	
2002	4.109	2.940	65.3	0	0.57	
2003	3.941	3.606	60.1	0	0.49	
2004	4.011	4.303	38.3	0	0.29	
2005	3.915	5.075	42.8	17.1	0.47	0.5
2006	4.048	6.042	84.3	44.9	0.92	1.5
2007	3.997	7.212	70.8	37.3	0.68	2.3
2008	**4.109**	**10.069**	**185.6**	**479.9**	**3.66**	**3.45**
2009						**4.60**
2010						**5.75**
2011						**6.20**
2012						**6.65**
2013						**7.10**

Forecast greenhouse gas emission volume from the refinery sector of the European Union by 2020

According to the analysis of the CONCAWE [1, 2], the emission of carbon dioxide in the oil sector of the Community was about 122 MT/year in 2000, while in 2005 – about 144 MT/year. By 2020, further successive increase of CO_2 emission is forecast due to increasing demand for middle distillates (diesel oils and avgas) and also successively implemented or planned stricter quality requirements for engine fuels, combustion oils and ship propellant oils. Emission of CO_2 forecast on the basis of implemented and already determined changes in respect of fuel quality will amount to the level of 180 MT/year in 2020, while taking into consideration the successive planned changes of fuel quality will bring about further increase of carbon dioxide emission to about 210 MT/year (Drawing 1) [1].

According to the results of performed analysis, the execution of listed qualitative changes for fuels which will have been implemented by 2020 will require investments in the refinery sector in the amount of about 50 billion USD. Successive propositions of the IMO and SECA in respect of deep desulphurization of marine fuels to sulphur content below 0.1% m/m will increase investment costs by further 10 billion USD. The change in the diagram of oil processing towards intensification of fuel production, chiefly due to a deficit of middle distillates in Europe, will require investments of about 65 billion USD. Apart from the investment related to desulphurization of residue refinery streams, it will be necessary to abandon production of heavy marine fuels and to invest in processes of their conversion to products of higher value such as diesel oil and petrol [2].

According to the execution of the Kyoto Protocol obligations and regulations which implement trading with greenhouse gas emission permissions in the European



Legend

	Changes related to increasing demand for fuels
	Introduced or already determined quality changes
	Planned additional quality changes

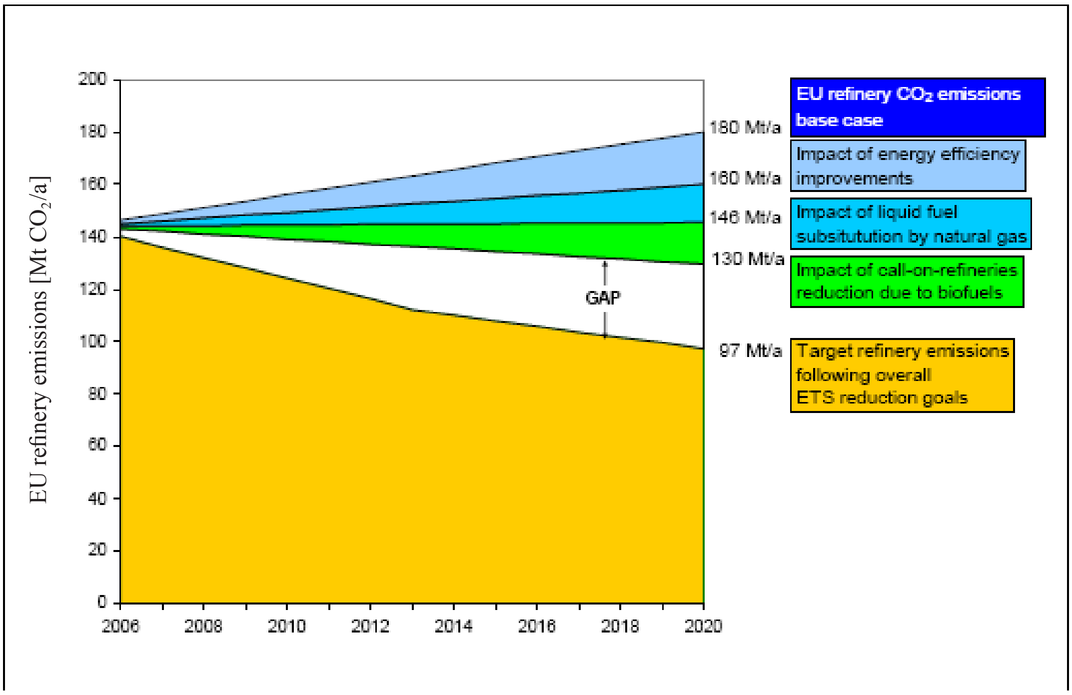
Drawing description

'FQD' (*fuels quality directive*) concerns changes in respect of obligatory requirements for engine fuels introduced by the Directive 98/70/EC, then amended in 2005 and 2009 and other quality changes planned for consecutive years.

'SLFD' (*sulphur in liquid fuels directive*) concerns changes resulting from stricter requirements in respect of sulphur contents in combustion oils: already implemented (Directive 1999/32/EC and its amendments in 2000, 2003 and 2008) and planned in consecutive years,

'IMO' (*International Marine Organization*) and 'SECA' (*Sulfur Emission Control Area*) concern potential changes of sulphur in ship propellant fuels suggested by the above organizations.

Drawing 1. Forecast CO_2 emission increase at European refineries in years 2000–2020



Legend:

	Reduction of emission due to improvements and modernization
	Reduction of emission due to change of fuel
	Reduction of emission due to application of biofuels
	Target CO_2 emission in accord with Directive EU-ETS

Drawing 2. Forecast volume of CO_2 emission at EC refineries (27 + 2)

Union (Directive 2003/87/EC replaced with Directive of the European Parliament and Council 2009/29/EC dated 23 April, 2009 – the EU ETS directive), due to emission reduction index accepted by the Act, calculated greenhouse emission volume should be in 2020 at the level of 97 MT eq CO_2/year. The existing 'excess' of the GHG emission, which in 2020 will amount to 83 MT CO_2 eq/year should be reduced by actions recommended for the refining industry such as:

- change of processed raw material i.e. crude oil, to lighter one with smaller content of sulphur,
- change of refining fuel, which is oil residues and heavy combustion oils and refinery gas, towards increasing the share of low emission natural gas originating from purchase,
- implementation of energy management system and modernization of energy production and consumption system which consists in the application of modern equipment and technical solutions and optimization of heat consumption,
- use of biocomponents for production of oil fuels,
- application of other methods, including carbon capture and storage by extracting it from emission gases, transport and underground storage (CCS technology).

According to estimations by the CONCAWE [1], the execution of above described actions will enable overall reduction of CO_2 emission in the EC refinery sector (27) + Norway and Switzerland to the volume of about 133 megaton/year in 2020 (Drawing 2).

In relation to presented estimations, a surplus is created between the forecast amount of carbon dioxide emission by the Community refinery sector, considering the application of all possibilities of its reduction, and the volume of emission resulting from accepted reduction obligations, which in 2020 will amount to 33 MT eq CO_2/year. Currently, it is not possible to eradicate or reduce it efficiently.

Summary

Conclusions resulting from numerous specialists' elaborations and factual forecasts concerning the possibility of reducing greenhouse gas emission in the European crude oil sector have demonstrated that ecology supporting objectives accepted by the Community, which result from the international regulations and stipulations of the Directive 2009/29/EC aiming at achieving high level of reduction of GHG emissions of particular countries and especially the emission generated by entities operating in the EU-ETS system are not possible to achieve due to particular ecology oriented objectives focused on successive stricter quality requirements for engine fuels, combustion fuels and marine fuels.

The 'double track' of ecology oriented activities pointed out in the forecasts should be subjected to detailed analysis and broad discussion within the European Commission and conclusions accepted as a result, should be taken into consideration in the process of determining and then accepting successive, more and more ambitious levels of reduction of GHG emission by the Community member states, including entities operating within the EU-ETS system.

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Mgr inż. Halina SYREK – starszy specjalista badawczo-techniczny INiG, kierownik Laboratorium Produktów Bloku Olejowego w Zakładzie Olejów, Środków Smarowych i Asfaltów. Absolwentka PK – Wydział Chemii, Technologia Ropy i Gazu; studia podyplomowe na PW – Inżynieria Procesowa i Aparatura Chemiczna. Specjalizacja zawodowa – woski naftowe.

Mgr inż. Delfina ROGOWSKA – starszy specjalista badawczo-techniczny, zastępca Kierownika Zakładu Paliw i Procesów Katalitycznych Instytutu Nafty i Gazu w Krakowie. Od 10 lat zajmuje się tematyką technologii benzyn silnikowych zawierających związki tlenowe, w tym biokomponenty.