Analysis of Petroleum Coke Consumption in Some Industrial Sectors

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Abstract

The world production of petroleum coke has been growing in the last years, due to a growing supply of heavy oils. The market is divided among the green coke consumers and the calcinate coke consumers. When taking pollutants into consideration, the quality of petcoke depends on the type of processed petroleum and the operational conditions of the production unit. Three different processes can produce petcoke. The cost of petcoke is inversely proportional to its sulfur content. Due to the consumer process type, there are those who can only use coke with low sulfur content. However, there are consumers that can burn coke with larger sulfur content without needing exhausted gas desulfurization treatments, such as power plants with fluidized bed boilers, and in the cement industry where the sulfurous gases incorporate to the clinker in the form of calcium sulfate.

Keywords

Petroleum Processing; Petcoke; Desulfurization; Power Plants; Cement Industry

Introduction

Petroleum coke is a byproduct of the oil refining industry. It has a high calorific value and a low cost. Due to higher amounts of processed heavy oils, petroleum coke production has been increasing. Its low cost and abundance has made it an attractive residue for the industrial sector, especially for the electric power and cement production sectors (Wang et al, 2004). As oils become heavier, their densities in API degrees decrease and their contaminant content, especially sulfur compounds, may increase. This tendency becomes evident as presented in the data in Figure 1, part of a survey done on about 15.5 million barrels of oil processed per day in the U.S., of which two-thirds are currently imported from various parts of the world (EIA, 2012).

Due to the demand for petroleum with lighter production fractions such as naphtha, gasoline, kerosene and diesel, oil refineries have begun processing heavier oils to meet the market demand for light and medium products. Thus, along with the fluid catalytic cracking, which converts diesel into gases and gasoline, delayed coking has been increasing and gaining more and more importance in refineries (ANP, 2003). Since petroleum coke is a product derived from the bottom of the barrel oil processing system, this derivative has more or less sulfur content based on the type of oil from which it originated. Therefore cokes can have high and low sulfur content (Salvador et al, 2003; Barros et al, 2003). The high sulfur is a great problem because the sulfur emission control becomes the challenging. In literature there are many technologies for SO_2 emission control , as example using CaO sorbent or thermal desulfurization process (Wang et al, 2004).

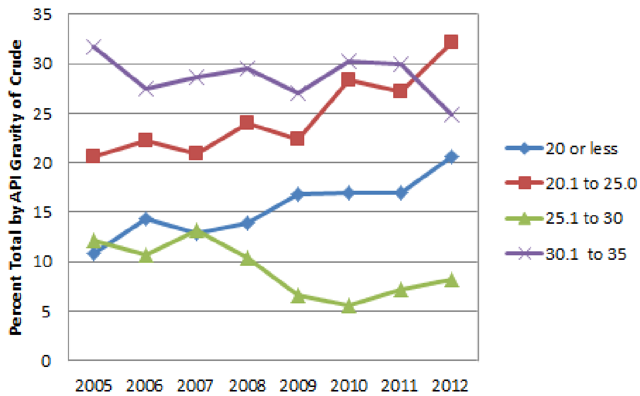

FIGURE 1. PERCENTAGE EVOLUTION OF IMPORTED CRUDE OIL, BY API GRAVITY BY USA, OF 2005 TO 2012 (EIA, 2012).

Types of Production Processes of Petroleum Coke

According to Speight (2004), coke production can be achieved by three different processes: delayed coking, coking in a fluidized bed and coking in a fluidized bed with gasification. Since coke originates from heavier petroleum fractions, it is natural that its denser impurities become concentrated, like metals and sulfur compounds; the content of these components depends

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directly on the quality of the processed oil. Therefore, there can be cokes in the international market with sulfur contents ranging from about 4.0 to 7.5% by mass (Salvador et al, 2003). The coke produced in Brazil has sulfur contents averaging between 0.7 and 0.8 % by mass (Barros et al, 2003).

Comparative Composition of Coke Produced

All three of the production processes of petroleum coke have different process and operational configurations, which have a direct effect on the composition of the coke produced by each one of the three processes mentioned above (Speight, 2004). Table 1 shows the composition of the coke produced by the three processes, using traditional loads of vacuum residue, but originated from different oils. The table only shows the H/C relation to the condition of the process, since the loads do not have the same origin.

TABLE 1. COMPARISON BETWEEN THE PETROLEUM COKE PROCESS.

Composition (% in mass)	PetroleumCoke–ProductionProcess		
	Delayed	Fluidized Bed	Fluidized Bed with Gasification
C	87.9[(1)]	86.3	94.9
H	3.51[(1)]	2.2	0.3
H / C	0.47[(1)]	0.31	0.04
N	1.61[(1)]	2.4	1.1
S	7.5[(1)]	6.9	2.8
O	-	0.9	0
Ashes	0.33[(1)]	1.3	1.0
d (g / cm^3)	2.00[(2)]	0.80	0.96

Source: Adapted of Furimsky (2000), Salvador et al.(2003)[(1)] and Garcia (2002)[(2)].

World Production of Petroleum Coke

The world production of petroleum coke reached 81 Mt (million tons) in 2001, 83 Mt in 2002 and was expected to surpass 88 Mt in 2005 (Dynamis, 2004). The United States of America is the largest producer of petroleum coke, accounting for about 66% of the world production. About 57% of the American production comes from the Gulf of Mexico (Texas and Louisiana). In the United States, about 35 refineries produce petroleum coke in considerable quantities (over 1000 t/day). Table 2 shows world production of coke, with the percentage share by region (Dynamis, 2004). Countries that export petcoke, according to Petcoke Report - April - 2007 (Energy Publishing, 2007) are presented in Table 3 and shows the absolute supremacy of the U.S. as a producer and exporter of petroleum coke

The total market value of petroleum coke was around 100 million tons in 2011 (Roskill, 2012). The United States accounted for 40% of the supply, while China produced 24 million tons. The market is bound by the demands of five major industries: petroleum refining, electric power generation, cement, steel and aluminum. The aluminum and steel industries require coke with higher purity, low sulfur and low levels of metals, especially in the aluminum industry for the production of high quality electrodes.

TABLE 2. WORLD PRODUCTION OF PETROLEUM COKE.

Regions Producers	Participation (%)
North America	69.5
South America	9.1
Europe	8.5
Asia	6.9
Ex-URSS	5.0
Africa	0.5
Oceania	0.5

Source: Dynamics (2004).

Price of the Petroleum Coke

Several factors influence the petroleum coke market price. When the international price of coal increases, the demand for petroleum coke also increases. According to analysts, the rise of the international coal price is generally what most influences the high price of the petroleum coke, usually with a delay of about 3 months. Figure 2 presents the evolution, in US$/million of BTU, of the petroleum coke price in relation to the cost of natural and synthetic gases produced by petroleum coke gasification (GCPA, 2005).

TABLE 3. EXPORTING COUNTRIES OF PETROLEUM COKE IN JANUARY AND FEBRUARY 2007.

Country	January - 2007				February - 2007			
	PetroleumCoke(t)				Petroleum Coke (t)			
	Green	Calcined	Total	%	Green	Calcined	Total	%
Canada	12052	6577	18629	0.66	8155	5347	13507	0.65
China	77815	55006	132821	4.71	80519	51078	131597	6.28
United Kingdom	20035	17899	37934	1.35	4932	24564	29496	1.41
United States	2383861	245021	2628882	93.28	1619898	299736	1919634	91.7
Total (jan/2007)			2818266		Total (fev/2007)		2094234	

Source: Energy Publishing (2007).

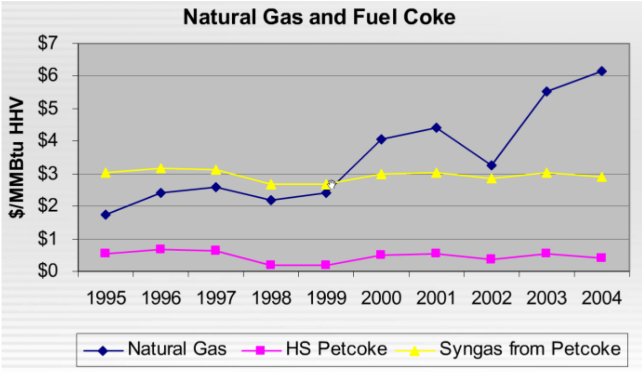

FIGURE 2. EVOLUTION OF THE PRICE OF PETROLEUM COKE, GAS NATURAL AND SYNTHETIC GAS DERIVED FROM PETROLEUM COKE (GCPA, 2005).

Table 4 shows a price comparison (relative to December 2012) between the cokes produced in the gulf region of the U.S. and in Venezuela. It shows that sulfur content interferes more with the prices than the HGI - Hardgrove Index. With the exception of the coke produced in the West Coast of the U.S., there was a price uptrend (Energy Publishing, 2012). The price stability of petroleum coke in recent years has also been an incentive for its use. Figure 3 shows a price comparison (March of 2007) between the petcoke produced in the U.S. gulf region and in Venezuela (Energy Publishing, 2007).

TABLE 4. PRICES OF PETROLEUM COKE IN FUNCTION AND SULPHUR CONTENT OF HARDNESS HGI.

Origin	Shulphur (%)	HGI	Price (US$/t)
United States (Gulf region) and Venezuela	4 – 5	<50	62.00 a 68.00
	6	35 a 45	50.00 a 56.00
	6	50 a 70	51.00 a 57.00
United States (West Coast)	3	45 a 50	80.00 a 87.00
	4+	45 a 50	73.00 a 79.00

Source: Energy Publishing (2012).

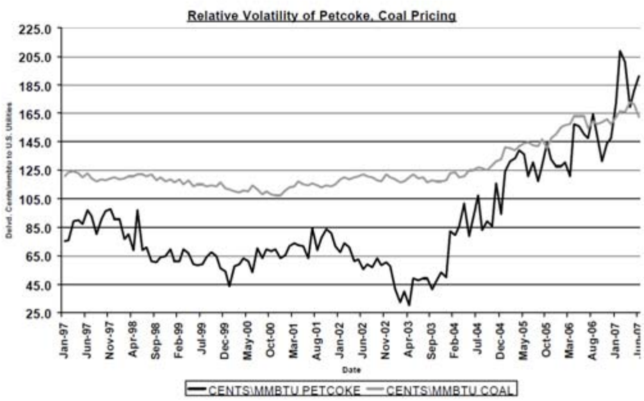

FIGURE 3. VOLATILITY ON BETWEEN THE PRICES OF COAL AND PETROLEUM COKE.
Source: Energy Publishing (2007).

Figure 3 shows that petroleum coke price was always below coal. However, from July of 2004, the increase of petroleum coke prices led them increasingly close to that of coal. In July of 2006 and January of 2007, petroleum coke prices surpassed the price of coal. On the other hand, the amount of petroleum coke which a refinery produces has little or nothing to do with the market value. In fact, refineries do not decide to produce more petroleum coke when the market demand increases and prices rise.

The sole reason for the production of petroleum coke growth is when greater amounts of heavy petroleum fractions are converted into lighter fractions such as in aviation fuel and gasoline, which create more residues. However, if a refinery is producing more heavy fractions, such as fuel oil, the process produces less petroleum coke.

Green Petroleum Coke (GPC) and Calcined Petroleum Coke (CPC)

The coke obtained directly from the delayed coking process is called green coke due to the greater volatile content in its composition. In another unit of operation, green coke is subjected to a calcination process, forming the so-called calcined coke, whose volatile content is greatly reduced. In the green coke calcination process, the sensible and latent heat of the volatile matter are recovered in a heat recovery boiler, which generates steam in a suitable pressure level for use in other units in the plant or sold to nearby green coke calcinations plants. Therefore, the production of steam can be considered an indirect consumable product obtained in the petroleum coke production. Table 5 shows the comparative composition between the green coke and the calcined coke by anode grade. This Table shows the great difference between the volatile matter content contained in green coke and calcined coke. Table 5 also shows that calcined coke HGI is smaller than that of the green coke HGI. As the HGI is inversely proportional to the coke's grindability, it shows that green coke is more suitable for used as fuel when compared to the calcined coke, because less energy is used in the grinding process.

The global market of calcined coke is a highly competitive one with participants from various countries. Petroleum coke is used as fuel in India, China, Japan, Russia, Germany, the U.K., France, the U.S., Canada, Venezuela, Mexico, Argentina and Brazil. (Martinez and Bartholomew, 1998 apud Ellis and Paul, 2000; Globaldata, 2012).

In 2009, Brazil imported about 3.75 million tons of petroleum coke and produced 3.5 million tons. The steel industry consumed about 825,000 tons. However, the largest consumer of petroleum coke was the cement industry. Currently in Brazil, the only green petroleum coke calcinator is Petrocoque S.A., located in the city Cubatão, in the São Paulo State, in an area close to the PresidenteBernardes Refinery. Petrocoque receives the green coke from PresidenteBernardes Refinery of Cubatão, a Petrobras unit, and via a calcination process, in which the rotary kiln operates at 1.8 rpm and 1300 °C, produces the calcined coke, whose main application is the manufacture of electrodes for aluminum metallurgy (Lind, 2003).

TABLE 5. COMPOSITION CHARACTERISTIC OF GREEN AND CALCINED COKE (ANODE).

Composition	Green Coke	Calcined Coke (anode)
Volatile Matter (%)	9.0 ~ 10.5	0.08 ~ 0.15
Moisture (%)[(1)]	8.0 ~ 14	0.2 ~ 0.4
Ashes (%)	0.09 ~ 0.14	0.10 ~ 0.18
Sulphur (%)	0.70 ~ 0.85	0.70 ~ 0.78
Vanadium (ppm)	180 ~ 230	200 ~ 250
Nickel (ppm)	180 ~ 200	200 ~ 220
Silicon (ppm)	40 ~ 80	20 ~ 60
Iron (ppm)	80 ~ 120	60 ~ 100
Sodium (ppm)	70 ~ 90	50 ~ 100
Calcium (ppm)	20 ~ 40	20 ~ 40
Hardgrove Index (HGI) [(1)]	70 ~ 80	20 ~ 40

Source: PETROCOQUE (2003) and Hammond et al.(2003)[(1)].

Processes that Uses Petroleum Coke

Depending on the supply to the consumer market, there are cases where the calcined coke is used as fuel in thermoelectric power plants and cement plants with high volatile content (12%t) and the high value of HGI (100). Other advantage is that the calcined petroleum coke has a high energy content compared to mineral coal and low ash content (typically 0.1 wt%).

The thermoelectric power plant usually use coal with low sulfur content mixed with petroleum coke with high sulfur content, with aim to emit SO_2 in acceptable values for the environmental legislation.

The work of Olmeda et al. (2012) introduces a new way to use petroleum coke as lightweight aggregate in cement mortars to make sound barriers. The acoustic behaviour herein was assessed by constructing a large dimension mortar slab (made of cement and coke as aggregate) used as floor covering and measuring. Results showed that coke addition leads to a decrease in mechanical properties of resultant mortars, this is principally due to an increase of the porosity (60%). A gradual increase of impact noise insulation was observed in light weight floor covering from middle to higher frequencies tested, reaching, within this range, a remarkable improvement of sound insulation compared to control slab (14 dB).

The green coke may have the following uses (Ellis and Paul, 1998): fuel coke used in cement production and in fluidized bed boilers to generate steam and electrical energy, using limestone to remove sulfur compounds; if it presents low-sulfur content, the petroleum coke may have metallurgical use in the form of mixtures composed with mineral coke, for blast furnace feed; undergoing partial oxidation, the petroleum coke can be used in gasification processes for applications in water steam production in electric power generation; and in the production of gas for various industrial applications. The green coke can also be used directly in the blast furnace, in a process known as injection of coal fines. In this process the green coke, also used as fuel, is blended with other coals, pulverized and injected directly into the blast furnaces.

Other applications,suchassupplementing metallurgical coalusedin coke batteriesforsteel productionorthe direct use inthe production of siliconcarbide, alsoprovide highvaluemarketsfor green coke (Garbarino, 2007). Table 6 presents a summary of the uses of green coke.

TABLE 6. APPLICATTIONS TO GREEN PETROLEUM COKE.

Applications of the Green Coke	Markets	Required Quality
Raw material for calcination	Aluminum TiO_2	Low volatile (Max. 12%) Low levels of metals Lowsulfur (<2%)
Carbon-basedreducer	Pig Iron Iron Alloys Carbides	Low sulfur (< 1.0%) High fixed carbon (>90%) Low levels of metals Granulometry (iron alloys and pig iron)
Raw material for coke kiln	Foundry	Low sulfur (<1.0%) High fix carbon (>90%)
Raw material for coke kiln	Greatsteel industries	Low sulfur (<1.0%) High fix carbon (>90%)
Fuel	Lime Great steel industries Ceramic red Pelletizing/Sintering	Low sulfur (<10%) High calorific value
Fuel	Cement Kiln Power Plants	High sulfur (>4%) High calorif value

Source: Carvalho and Assis(2007).

In Brazil, the larger demand for calcined petroleum coke occurs in the aluminum industry that uses it to produce carbon anodes. Its purpose is to conduct electric current in orderto obtain an electrolytic reaction of alumina dissociation for the production of primary aluminum (Silva et al, 2009).

Beside this application, other industrial application is the titanium production from titanium oxide and calcine petroleum coke. The main process steps are chlorination, reduction and electrolysis igneous, this technology is called Kroll process (Zahner, 1995). Table 7 shows the worldwide consumption of calcined coke.

TABLE 7. WORLDWIDE CONSUMPTION OF CALCINED COKE.

Industrial Segment	Consumption of petroleum coke (tons)
Aluminum Industry	9,880,000
Steel Industry	1,040,000
Re-carburizing market	910,000
Titanium dioxide	650,000
Industry others	520,000

Source: Dickie, 2006.

These applications are being used successfully in Brazil, in COSIPA – CompanhiaSiderúrgicaPaulista in Cubatão, SP, making the steel sector yet another consumer of petroleum coke (BR-N° 20, 2005).

Figure 4 shows the electrical energy production in the U.S. in 2009, based on the type of fuel used, showing that the thermoelectric plants contributed with 50.1% of the total production, presenting itself as a potential consumer of petroleum coke (FCEL, 2005; EIA, 2012). The World Energy Council recommends that the mixture of mineral coal and petroleum coke contain at most 20% of coke, due to the low content of volatile material in the petroleum coke (WEC, 2005).

New Technologies for Consumption of Petroleum Coke

According to Furimsky (1999), the supply of heavy oils has increased in the world market. Processing these oils has generated less light products and more heavy residues. Thus, there will be a tendency for refineries to use residue from gasification processes in refineries of origin, producing gas streams, which may serve as raw materials for other processes.

Edison International and British Petroleum had planned on developing a 1 billion dollar power plant in an oil refinery located in California. The plant would produce 500 MW using the byproduct of the petroleum coke gasification, with operation expected in 2011 (Reuters, 2006).

Wang et al (2004) proposed a process having zero SO_2 and CO_2 emissions with an expected application for petroleum gasification. The coke is burned in a fluidized bed, using CaO for absorbing SO_2 and CO_2. A calciner, burning coke, may be used to regenerate the CaO and obtain CO_2 for other industrial processes. The loss of CaO will be due to its conversion into $CaSO_4$, forcing the replacement of $CaCO_3$ through the calciner.

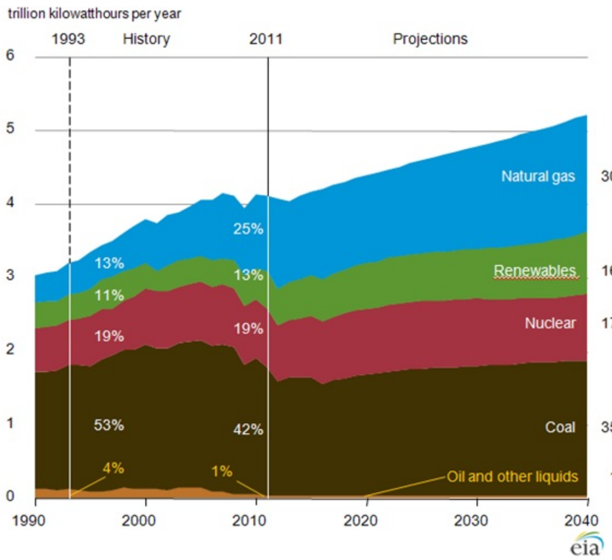

FIGURE 4. ELECTRIC POWER GENERATION IN THE UNITED STATES, IN 2010 (EIA, 2012).

Environmental Legislation

Due to sulfur contents, normally present in much of the petroleum coke available in the international market, among the parameters controlled by the Environmental Legislation, this study is primarily concerned with the emission limits of SO_2, due to the fact that this pollutant is produced by burning petroleum coke.

In the U.S., the emissions standard for SO_2 established by EPA for kilns of cement clinker production depends on the location, atmospheric dispersion conditions and proximity to population centers. In the European Union, the established standards vary from country to country, but they are measured under the conditions of 25 °C, 101.3 kPa and 11% O_2 corrected, in the exhausted gases, in dry basis. Under these conditions, it is considered that, for emissions below to 1000 mg SO_2/Nm^3, it is recommended gases be treated by using limestone and, for emissions slightly over 1000 mg SO_2/Nm^3, systems of dry or moist scrubbers are used. For the kilns of clinker production it is recommended to keep emissions between 170 and 340 mg SO_2/Nm^3 (PA, 2005).

In Brazil, CONAMA Resolution Nº 5 of 06/15/89, according to the Revision on September 19th, 2005 limited emissions nationally by typology of sources of priority pollutants. The CONAMA Resolution No 8 of 12/06/90, which is in a revision process, establishes that for fixed sources with power less than 70 MW the emissions limit is 5,000 g SO_2/Mkcal (coal or fuel oil). For fixed sources with power greater than 70 MW, the emissions limit is 2,000 g SO_2/Mkcal (coal or fuel oil). The CONAMA Resolution Nº 382 of 12/26/2006 establishes the maximum emission limits for air pollutants from fixed sources. In Article 7, paragraph 1, it cites that the Environmental Agency may establish values less restrictive than the maximum emission limits established in this Resolution, considering the technological limitations and the impact in the local conditions according to the provisions of CONAMA Resolution Nº 5 of 06/15/89. Article 8 established that limits and criteria established by CONAMA Resolution Nº 8 of 12/06/90 for the processes of heat generation not covered by this Resolution should remain applicable. CONAMA Resolution N° 382 in ANNEX XI - Emission limits for air pollutants from Portland cement industry does not establish emission limit for sulfur oxides (SO_X), advocating on its item 7 that "according to the local characteristics of the area of influence of the polluting source to the air quality, the Environmental Agency may establish more restrictive emission limits, inclusively considering the alternative use of fuels with lower pollution potential". In the State of São Paulo, the emission standard for the cement industry is from 350 mg SO_2/Nm^3 to 7% O_2 corrected in exhausted gases, in dry base (Busato, 2004). In Paraná State, Brazil, Resolution N° 041/02 establishes emission limits of SO_2 for source generators of thermal potency, according to the Table 8 (SEMA-PR, 2002). For clinker production kilns, the same resolution establishes the limit of 400 mg SO_2/Nm^3, with 7% O_2 corrected in the exhausted gases in dry basis.

TABLE 8. EMISSION LIMITS FOR SO_2, TO 7% O_2 CORRECTED IN THE EXHAUST GASES IN DRY BASIS, AS RESOLUTION 041/02, STATE OF PARANÁ-BRAZIL.

Thermal Power (MW)	mg SO_2/Nm^3
< 10	-
10 – 50	3,000
50 – 100	1,400
> 100	400

Source: SEMA – PR (2002).

Conclusions

The increase of petroleum coke production is a natural consequence of the increase of oil API degree, currently available in the international market. This trend is reflected in the discovery of large non-conventional oil reservoirs, extremely heavy and with high sulfur content. Thus, the current consumer market of petroleum coke must be amplified, whether by encouraging current customers to increase their quotas, or including new technologies for the use of petroleum coke. It must also be taken into account the possibility of coal blends with low sulfur content, aiming to reach the important consumer market of thermoelectric power plants. Therefore, there is a balance between the production and the consumption of this residue of oil processing. However, whichever route is to be taken for the use of the petroleum coke, the consumers must comply with the emission standards for pollutants, especially in relation to the SO_2.

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