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#### Abstract

To prevent the public from harmful emissions a further reduction of the air pollution is worldwide in the main focus of the authorities. For amendment of the environmental performance in the cokemaking industry ThyssenKrupp Uhde has developed and improved several processes to fulfill the ever increasing stringent environmental demands. New technologies were implemented at several new installations e.g. for low emission quench towers and for the application of the single chamber pressure control system PROven. These projects are now under realization at several sites worldwide. The paper will give an overview of the basic fundamentals and technological features of the applied processes, the actual status of the projects and the emissions reduction to be expected.

Key words: Cokemaking; Emissions; Air pollution.

<sup>&</sup>lt;sup>1</sup> Technical contribution to the 6<sup>th</sup> International Congress on the Science and Technology of Ironmaking – ICSTI, 42<sup>nd</sup> International Meeting on Ironmaking and 13<sup>th</sup> International Symposium on Iron Ore, October 14<sup>th</sup> to 18<sup>th</sup>, 2012, Rio de Janeiro, RJ, Brazil.

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#### **1 INTRODUCTION**

Coke plant operation entailing the lowest possible emission levels has become ever more important in view of environmental protection and industrial hygiene. Worldwide new environmental rules were defined giving also new impetus toward the development of eco-friendly technologies.

Worldwide non-attainment concerning ambient air quality can be stated in many industrial areas<sup>[1]</sup>. For a reduction of pollution the emission limit values were tightened by the authorities significantly, e.g. in Germany by the revised regulation "TA-Luft"<sup>[2]</sup> since 2006. In the US the implementation of the 2006 Clean Air Act (CAA) according to NESHAP (40CFR Part 63 –National Emission Standards for Coke Oven Batteries) is under progress<sup>[3]</sup>. Programs to realize technical measures for a reduction of these emissions by industrial sources have been initiated:

- The aspired reduction of particulate matter (PM) emissions has promoted new LEQT (Low Emission Quench Tower) projects in the US.
- In many countries the application of an individual oven pressure control system is now considered as best available technology to reduce effectively fugitive emissions at the oven closures and therefore is now mandatory for by-product coke plants in many countries.

ThyssenKrupp Uhde as a technology-driven plant builder tackled these issues more intensively and pushed constructive as well as process technology developments for an improved environmental protection forward.

The main focus in this paper will be given on actual projects in North America.

# 2 PART I: NEW LEQT PROJECTS IN THE US FOR PARTICULATE MATTER REDUCTION

As the main target of a quenching system the pushed red-hot coke has to be cooled down from approx. 1050°C to ambient temperature to enable further processing and transportation. In western states the most applied technology is wet quenching. Of major technical concern for a coke plant operator is the coke moisture content that has to be consistently lower than 5% for coke usage in a blast furnace.

Despite these only few technical targets there are objectionable consequences of the quenching process – *emissions*. Of major importance in this context are the particulate matter (PM) emissions, in particular the respirable dust as PM10 (<10µm) resp. PM2.5 (<2.5µm) because of its health effects - beneath the additional developing gaseous emissions resulting mainly from carbon gasification and burn-off. In most countries worldwide only the filterable PM emissions are considered. In some countries additionally the so-called condensables are considered, which comprise the primary condensable PM (material directly condensable under the conditions at the quench tower exhaust) and the secondary condensable PM (forming in the atmosphere via complex photochemical reactions e.g. from the emitted SO2, NH3, NOx as condensable PM to sulfates, nitrates etc.)<sup>[4]</sup>.

In the US the (filterable plus condensable) PM emissions limitations are defined in the permit for each planned new quenchtower and are based on an emitted dust load per year. After conversion of these values specific emission factors can be derived. Whereas in former times values of more than 300 g/t<sub>coke</sub> could be found, nowadays values of only approx. 130 g/t<sub>coke</sub> are allowed for the new LEQT (low emission quench tower)-projects in the US<sup>[5]</sup>.

Such low PM emission levels are possible only by application of advanced technological developments.

# 2.1 Sources for PM Emissions at Quench Towers and Discussion of the Technical Measures for Reduction

The following sources principally contribute to PM emissions:

- i. the spalling of the coke pieces by the "thermo-shock" from the contact of water with red-hot coke,
- ii. PM deposits at the quench tower internals and baffles, if they were not cleaned in a regular manner after each quench and constitute a buffer for PM deposits
- iii. the freshwater which is used for preparation of the quenchwater which contains a certain amount of TDS (total dissolved solids) i. e. particles of a size <2  $\mu$ m, molecular, ionic, colloidal or fine particles mainly the salty constituents.
- iv. the quenchwater itself that is enriching with the TDS by each quench until a balance value is achieved in the US certain limit values exist. Furthermore the constituents TSS (total suspended solids, particle size >2µm and the settleable solids from material of any size nor remaining suspended or dissolved constitute PM sources.

From the desription of the sources the possible measures for the reduction of PM emissions in LEQT's are to be derived:

- trapping of the spalled PM by a "highspeed" and intense cooling procedure, which guarantees a fast water penetration throughout the whole charge,
- a fine distributed plume spray for pre-condensation/-separation of the ascending PM;
- baffles for separation of the PM (Figure 1),
- flushing water sprays for a regular baffle cleaning after approx. each quenching procedure to avoid PM deposits; to this end the utilized water should be quite clean,
- an improved quench water settling plant incl. an automatic scraper for optimum water cleaning respectively removing of PM introduced by the quench water,
- a sealing between the quench car and -tower to avoid side-emissions at the quench tower bottom as far as possible.

In general, an appropriate design of the whole quenching system including the way and chronological sequence of the quench water feed can be used to affect the vapor flow conditions, the access of outside air etc. in such a way that the dust separation takes place as far as possible already within the tower. For this design practical experience, growing from project to project will continue to play the most important role in the next years; up to now for a full theoretical description of the dust separation in a quench tower the detailed - highly unsteady - processes in a quench tower are far too complex. However, for further future improvements of the quenching behaviour more research and development is imperative.

#### 2.2 Methods of PM Monitoring

Worldwide different PM monitoring procedures are common practice – as demanded by the respective authorities. Frequently applied and practically proven is the so-

called "Mohrhauer method" (former standard VDI 2066). A deficiency of this method is the underproportional capture of fine dust. So-called isokinetic methods as e.g. US EPA method 5 (and others) try to avoid this drawback; this method is well established for stationary waste gas stacks sources. Because in a quench tower the conditions are quite different (e.g. high vapor content, highly unsteady vapor flow velocity varying between up to 15 m/s and near 0 m/s within one quench procedure) this test method does not allow strict conformance for this application and can be used only with certain drawbacks – also from a practical standpoint.

ISSN 2176-3135

It has also to be stated that a comparison of emissions levels determined according to different standards can lead to wrong conclusions and has to be strictly avoided.

Obviously, there is a high demand on improving PM monitoring methods for quench towers - concerning as well the reliability as also the practicability of application in the difficult technical surrounding.



Figure 1. LEQT baffle system and flushing liquor cleaning system.

## 2.3 CSQ Quenching System for PM Emission Reduction

The development of the most advanced wet quenching system CSQ<sup>[6-9]</sup> was guided by the sump quenching technology<sup>[7]</sup> which was developed for one spot quench cars and their typical high charging levels at modern high-capacity coke ovens. CSQ includes the features mentioned before plus additional improvements (see below) all giving rise to a low environmental impact. It was derived as an environmental friendly alternative to the coke dry quenching (CDQ) technology. The PM emissions come close to a CDQ-system, the gaseous emissions resulting from post-gasification reactions are even less<sup>[6-7]</sup> - the fast water flooding stops the gas generation almost instantaneously. On the other side investment, operation and maintenance costs are substantially lower. The process itself is a combination of bottom (major part) and top quenching (minor part) providing extreme short cooling time. During this "highspeed" quenching procedure the coke is popped up; by this high mechanical load on the coke particles in the bottom part of the shaft a high dust load is generated which has to be cleaned before being emitted into the atmosphere. An additional hood inside the quenchtower prevents the down falling coke from being deposed outside the quench car and additionally reduces the level of side steam emissions at the bottom. The coke is fairly stabilized by the CSQ quenching procedure; herefrom the name CSQ (Coke Stabilization Quenching) is derived.

To reduce the dust load of the emerging quenching vapors considerably the emission control baffels for CSQ were arranged in two levels one above the other.

Meanwhile the CSQ-technology was applied at many new coke plants in the world as the Schwelgern, POSCO Gwangyang and Hyundai plants. During the commissioning of these towers the high performance of the CSQ technology concerning PM emissions was confirmed: The PM emissions could be reduced safely to below 15 g/t of coke (determined acc. to VDI 2066). This is a low value compared to the current limit values between 25g/t and 100 g/t which are demanded by the authorities in most parts of the world for conventional quench towers.

Figure 2 shows the CSQ installations at the Hyundai and Schwelgern coke plants.





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Figure 2. CSQ quenchtowers at Hyundai and Schwelgern coke plant.

#### 2.4 Features of the New LEQT's in the US

Many of the CSQ-features are also implemented at the new LEQT's under erection in the US - 3 at USS Clairton MonValley works (Figure 3), 1 at USS Granite city, i. e.:

- Optimized water settling plant to clean the quenchwater
- the existing USS-LOMO quenching system<sup>[10]</sup> was not changed; compared to conventional top quenching processes the LOMO-type of water addition provides for an enhanced water penetration into the depth of the quench car bed and hereby allows also an intense and short time single spot quenching (Mon Valley)

- Double baffle system
- Upper plume spray for pre-condensation/additional scrubbing of the plume

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Upper baffle cleaning system

One of the most important items concerning the PM emissions reduction is the double baffle system. The efficiency of this system was estimated for the Schwelgern CSQ-system of ThyssenKrupp Steel by Zeimes:

- The upper level baffles aim mainly for fine dust separation. They are made of plastics, have smaller inner distances between the louvers and have an separation efficiency of 74% for the separation of the relevant PM size.
- The lower level baffles are made of stainless steel (to prevent any burning risk) and are designed by greater inner distances for the separation of coarser dust. Their separation efficiency was estimated 92%.
- From these both separation systems a combined separation efficiency of 98% results.

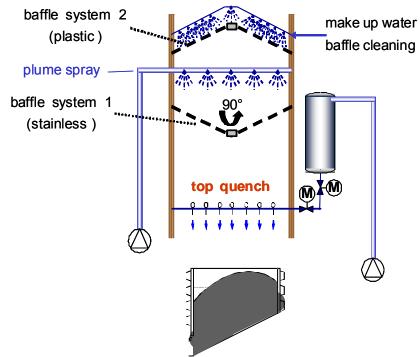


Figure 3. Scheme of the new LEQT's for USS Mon Valley plant.

The new LEQT's will come into operation within 2012/2013. Results concerning their performance will be reported in future after commissioning.

## 3 PART II: INDIVIDUAL OVEN PRESSURE CONTROL SYSTEM PROVEN<sup>™</sup>

Even though battery emissions have been reduced permanently in manifold ways, the respective developments reached process-technical limits: One factor deciding a fundamental limit is the inseparable relationship between the pressure in the collecting main and the pressure in the chamber. From this relation it follows that for conventional operated plants the choice of the necessary collecting main pressure has to be considered only as a foul compromise. The resulting variable pressure conditions in each single oven may give rise to fugitive emissions if the metallic

sealings of oven closures are not tight enough. To avoid such emissions an individual oven pressure control system was developed in the 90's<sup>[11-13]</sup> by DMT in cooperation with ThyssenKrupp Steel. By means of a decoupling of the pressure conditions in the oven and in the g.c. main an individual regulation of the pressure in oven chambers was made possible. This system has opened up absolutely new possibilities for an operation of coke oven batteries entailing substantially less emissions compared to conventional coking technology.

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ThyssenKrupp Uhde had already secured itself the base of further development at an early stage. Since the first ever industrial application at a 6m battery of ThyssenKrupp Steel in 1999 and following in the year 2003 at the two coke oven batteries of the modern high-capacity coke plant Schwelgern, this so-called PROven<sup>™</sup> system was enhanced (Pressure Regulated Oven) by own patented features. Several developments as e.g. an internal overflow regulation system, an advanced process control, tools for a reduction of the ammonia liquor consumption etc. <sup>[14-17]</sup> provided the basis for a system reliable operating in the harsh industrial environment of a coke plant.

From then on it has been rendering a noticeable contribution to improving environmental protection worldwide at more than 2000 coke ovens with a coke production capacity of 30 Mio. t (Figure 4).

The PROven<sup>™</sup> system has not only been implemented on numerous new coke plants all over the world. It has also been retrofitted on many coke plants that have already been in operation.

New batteries       rmany     8.4     2     140     2.6       7.8     1     70     1.1       ina     7.6     12     840     12.0       Korea     7.6     10     580     10.0       6.7     1     75     0.8       A     6.1     1     84     0.9       Retrofitted batteries       azil     6.6     3     147     1.8       6.0     3     105     1.1		Contracts for	the PROve	n <sup>™</sup> -Systen	n
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Figure 4. Worldwide PROven<sup>™</sup> installations.

PROven<sup>™</sup> has set the environmental protection standard for new installations not only in Germany, but meanwhile also in the US and in Canada: in all these countries the supervisory authorities prescribe a chamber pressure regulation system as a mandatory unit for new coke oven plants.

## 3.1 Functional Principle of the PROven<sup>™</sup> System<sup>[15]</sup>

The essential functional elements of the PROven<sup>TM</sup> system are schematically shown in Figure 5. A central component of this system is a water cup (so-called FixCup) by which a pressure-wise separation is accomplished between the actual coke oven chamber and the gas collecting main. In the FixCup a variable flow resistance for the

raw gas is built-up via the internal water level. Hereby the pressure in the oven chamber is regulated individually by the aid of the afore mentioned overflow regulation device in conformity with the coking progress, i.e. ensuring

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- a low pressure at the start of carbonization eases the strain on the oven sealing systems and hereby reduces the fugitive emissions at the oven closures; furthermore for older coke ovens any stack emissions from raw gas leakages can be reduced hereby in a causal manner,
- a gradually rising back-pressure as carbonization time advances so that the pressure in the coke oven chamber despite a greatly different gas development during the coking time is constantly slightly positive to prevent ingress of air into the oven.

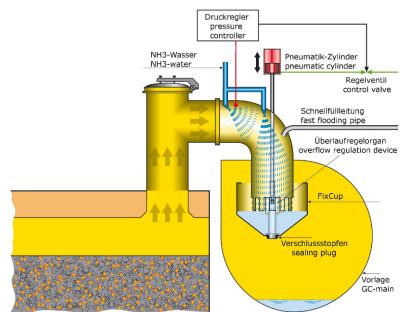


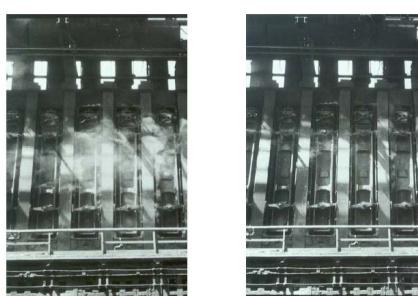
Figure 5. Schematic View of the Single Chamber Pressure Regulation using PROven<sup>™</sup>.

In contrast with conventionally operated coke ovens in which the pressure in the gas collecting main must lie in the positive range (at approx. +1 mbar) on account of the non-existing disconnection towards the chamber, the PROven system advantageously permits even an operation at suction (approx. -3 mbar). This allows for a substantially more efficient suction of gases on coke oven charging than achievable with hitherto applied conventional systems.

#### **3.2 Reduction of Diffuse Emissions**

The positive effect of the PROven system on the emission pattern became evident already during the first installations at the old ThyssenKrupp Steel coke plants in the mid 90's: Figure 6 shows clearly that emissions from the - severely emitting - oven door area using a conventional suction system (Figure 6 at left), can be reduced substantially by applying the single chamber pressure control (Figure 6 at right).

\* International Congress on the Science and Technology of Ironmaking - ICSTI \* International Congress on the Science and Technology of Ironmaking - ICSTI \* Ironmaking and Raw Materials Seminari 42° Seminário de Redução de Minério de Ferro e Matérias-primas \* Brazillan Symposium on Iron Ore / 13° Seminário Brasileiro de Minério de Ferro



**Figure 6.** Door emissions on the former coke plant August Thyssen: at left – coking process without PROven; at right: coking process with PROven application.

Comprehensive series of studies by ThyssenKrupp Steel <sup>[14]</sup> served to review and to quantify the efficiency of the PROven system. These investigations determined the influence exerted by various chamber pressure rates on the pressure at the sealing system and as consequence on the evolution of emissions.

Figure 7 shows the results of these measurements: The implementation of the PROven system leads to a significant reduction of PAH emissions both on pusher side and on coke side during the most critical 5h after charging. The reduction of emissions evolving there accounts for approx. 70% compared to the status without application of PROven.

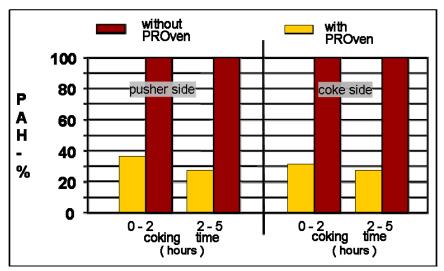


Figure 7. PAH emissions at oven doors with and without application of the PROven-System.

This marked emission reduction, measured in an old plant, was the reason for installing the system including its several improvements at Schwelgern and most following modern coke plant erections by ThyssenKrupp Uhde.



The almost complete absence of fugitive emissions at all oven closures is the typical appearance of all the new coke oven batteries equipped with the PROven<sup>™</sup>-system as is underlined by figure 8.



Figure 8. The Schwelgern battery in operation - emission free thanks to PROven.™

#### 3.3 Newest Development - MINI-PROven

The "Normal" PROven system has constructively been developed for the dimensions prevailing in the gas collecting main area of high-capacity coke ovens (i.e. having chamber heights over 6m). Figure 6 shows the structural conditions exemplary for the PROven equipment at a high-capacity coke oven battery. But the relatively tight space conditions become already evident even with these large ovens having an oven height of 7.6 m.



**Figure 9.** Space Conditions in Standpipe/Gas Collecting Main Area with a "Normal" PROven-Equipment at a High-Capacity Coke Oven Battery. For an installation at small-sized coke oven batteries with oven heights of only 4 to 6 m, space conditions are still much tighter; the standardized structural PROven elements, in particular the regulation device, are significantly oversized so that it has hitherto been impossible to offer their retrofit with PROven. However, such mostly older batteries exist in great number on the world market.

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To be able to give also these coke plants access to the environmental friendly PROven – technology the so-called Mini-PROven system<sup>[17]</sup> was developed; the adaptations required for this purpose have been elaborated within the scope of a recent internal development project.

In this project the following tasks and objectives posing great challenges in design and process engineering have been solved without having to abandon the proven basic functional and regulating features of the PROven system:

- General downsizing of structural elements dimensions.
- Adaptation of adjacent equipment components and maintenance ports to create the largest possible accessibility for service work by operating staff despite tight space conditions.
- Complete revision of the important regulation device developed by ThyssenKrupp Uhde so as to allow for an unhindered gas passage despite the smaller construction size on the one hand while preventing tar accumulation on the other hand.
- Adaptation of the regulation characteristics to the reduced crude gas volume by accordingly redesigned built-in elements to allow for appropriate regulating characteristics.

The function of the newly devised system was tested in a 1:1 scale on a test rack (Figure 10), investigating and studying especially the control behavior with the new overflow regulation valve, the wetting behavior of all components which is significant for a faultless operation as well as the mechanical adaptation to movements within the system.



Figure 10. MINI-PROven Test Rack.

# 3.4 First Industrial Application of the Downsized MINI-PROven at Essar Algoma/Canada

Until the end of 2011 the first successful large-scale implementation of the new Mini-PROven concept was realized. Within the scope of the refurbishment of the old 5m battery no. 9 of Essar Algoma in Canada the new MINI-PROven system (in combination with a new gas collecting main) was installed (Figure 8). This system has led to a significant reduction of the diffuse emissions at the oven closures (by  $70\%^{[18]}$ ); additionally the stack emissions were reduced considerably because of the decreasing oven to flue leakage.

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Figure 11. New Mini-PROven Installation at the Pre-Existing Coke Oven Battery No. 9 at Essar-Algoma in Canada.

## 4 CONCLUSIONS

Technological improvements of the environmental performance of conventional coke plants in North America are in good progress:

New LEQT installations (3 for US Steel MonValley works under erection, 1 for Granite city under commissioning) with advanced features for PM separation will give in future a significant contribution to the improvement of the ambient air quality

The worldwide applied individual oven pressure control system PROven<sup>™</sup> is meanwhile also under erection at US Steel MonValley works (battery C) and will contribute to a reduction of the fugitive emission levels. Results will be reported after commissioning.

The first industrial MINI-PROven installation was retrofitted at the Essar Algoma coke plant in Canada. Hereby, even small and medium-sized coke ovens can now be refurbished with an individual oven pressure control system. The application of the PROven<sup>TM</sup> principle is thus made accessible also for the complete spectrum of small-sized up to high-capacity coke ovens.

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