STUDY OF THERMAL, THERMO-MECHANICAL AND MINERALOGICAL CHANGES OF USED SILICA BRICKS AFTER MORE THAN 40YEARS OF LIFE IN COKE OVEN BATTERY 3 OF TATA STEEL

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ABSTRACT

Present study has been carried out for the full investigation of used Silica Bricks of coke ovens Battery No#3 of TATA STEEL Jamshedpur, India after more than 40 years of operation. Coke Oven Battery no#3 first came into operation on June 11, 1940. It was pulled down in 1958 and re-built, increasing its capacity and size from 55 to 57 ovens. The second phase of the Battery extended till 1975, when it was re-built again for Phase#3 , Coke Oven Battery No #3 has the unique distinction of being the first Battery in India to last for 40 years . Tata Steel has truly shown the way that it is possible in India to enhance the life of coke ovens .The Coke Oven Battery #3 , that was re-built by M/s Otto India Pvt Ltd and was light up on May 19th , 1975, consisting of two sub-batteries of 27 ovens each , The Top charge Battery no #3 could carbonize 1,450 tonnes of coal daily , with a rated production capacity of 1000 tonnes of blast furnace coke per day .The long life of Coke Oven Battery #3 can be attributed to the state of the art maintenance techniques deployed and focus on environment .It has been found that, though the outer appearance remain same as before use , but there is change in the appearance inside the brick making it denser in the area exposed to flame /flue side . These bricks are tested for chemical , physical [AP, BD, TSG] , thermal [RTE] , thermo-mechanical [Creep in compression , RUL] and mineralogical [XRD , Microscopy] . The results has been found , these used bricks have less amount of cristoballite , more amount of tridymite phase and glass phase . These changes are more predominant in the area , which is exposed towards the flue /flame side .

Coke Oven Battery-3 details		
Number of Ovens		54
OVEN DIMENSIONS	Length of sole	13760 mm
	Width of Ram side	390 mm
	Width of Coke side	450 mm
Mean		420 mm
Height		4570 mm
Oven Centres to centres distance		1100 mm
Oven Stretcher Thickness		100 mm
Waste Gas chimney height		90 m
Mean Battery Temp		1244 Deg C Max

Key-words

Residual quartz (RQ), Cristoballite, Tridymite, Glassy phase, Apparent porosity (A.P),Bulk density (B.D),True specific gravity (TSG),Reversible thermal expansion (RTE), Refractoriness under load (RUL) , X-Ray diffractometer, Creep-in compression and DIN 1089 Part 1.

INTRODUCTION

The life of coke ovens are greatly depends upon the design, quality of silica bricks used, operational parameters of a coke oven and coal quality .During operation of a coke oven, silica bricks used in chamber walls are subjected to thermo-mechanical stresses and chemical infiltration of coal used. Due to the above, the physical, chemical, mineralogical and thermal properties of silica bricks undergo significant changes from the original properties. The extent of such changes depends on the age of silica brick in operation, temperature prevailed in side coking chamber and the flue temperature. It is also depends upon amount of solid particle infiltration from coke side as well as gaseous infiltration in flue chamber. Previously people were using 4 – 4.5 mtr tall battery and the temperatures of coking were 1000°C – 1100°C and cooking time was much higher compared to the present coking time As 30 to 50% of space above the coal was kept vacant, the stresses were on the lower portion of chamber walls. The main problem of coke oven operation is the pollution from coke oven, higher the frequency of opening of oven doors, more gases escape from ovens and create pollution in the environment. In order to have less pollution following measures were taken.

a) Proper Sealing of the doors.
b) Wider Coke Oven chamber. However there is a limitation to the width of the coke oven chamber.
c) Tall Coke ovens: more throughput is obtained with less number of door openings. The silica brick used in the tall coke oven has to with stand the pressure of pusher on the wall. Therefore many deliberations have been made before finalizing the specification of coke oven refractories in Europe and in India .The first such specification is DIN 1089, Part1, was formulated and the latest revision was made in 1995. The changes particularly A.P. (Apparent Porosity), Sp.Gr. (Specific Gravity) and RQ (Residual Quartz) in the specification of silica bricks for coke ovens over the years (Tab. 1)

Tab. 1: Specification of silica bricks

Specification	% A.P.	Sp.Gr	RQ
DIN 1089, part 1, 1956	≤28	≤2.40	--
GOST 8023, 1956	≤23	≤2.37	--
MECON S/1210/A/10/ 01/0/001 (A), 1985	≤19	≤2.35	≤5.0
DIN 1089 part 1, 1995 Creep at 1450°C/5-25hr	22.5 0.12 max	--	≤6/1.5

Many pioneers have carried out studies of used silica bricks in coke ovens in the past. In 1926, S.S.Cole, [1] studied properties of silica bricks from coke oven walls. In 1938 W.C.Rueckel, [2] studied the failure of coke oven walls by reaction with coal ash. In 1940, F.H. Clews et. al, [3] studied the silica brick work in a vertical retort. In 1956 AO, T et al, [4] studied the corrosion of retractions in a coke oven. In 1968 K.Konopicky et al, [5] studied the silica bricks from chamber wall of coke oven.

After a gap of about 25 years, in 1995, Dr.J.D.Panda et al, [6] have studied used silica bricks from 4.5 Mtr coke ovens after use over 22 years and 25 years in 2 different steel plants in India and found that, most significant change in silica bricks are those of conversion of SiO_2 Polymorphs. Similarly in 1996, Dalmia R.H. et al, [7] studied mineralogical and physical changes in used Indian coke oven silica bricks used in 4.5 mtr. Coke oven battery.In 2003, Dr. B.K. Panda et al, [8]studied used Silica in a Tall Coke Oven battery in India after 10 years of operation.

In continuation of the study of used coke oven silica bricks, the present investigation is about used silica coke oven bricks from Battery-3 of Tata Steel in India after more than 40 years of operation. In a coke oven, silica bricks are subjected to severe thermo-mechanical stresses.. To enhance the life of tall coke ovens, these silica bricks should have lower R.Q. (Residual Quartz) content, higher creep resistance and should have homogeneous thermal behavior during operation. As The coke oven battery has got highest campaign life till date in Tata Steel, detailed investigations were carried out to know the reasons of getting such high life.The present investigation includes mineralogical, physical, chemical and thermo-mechanical properties of used silica brick samples which were collected after dismantling of the coke oven.

SCOPE OF STUDY

Materials

Used silica bricks were collected after more than 40 years of use in coke oven battery-3 of Tata Steel from the following areas:-

1. Wall bricks of Ram side - Middle position ,18th layer
2. Wall bricks of Coke side - Bottom position, 11th layer
3. Gas riser bricks in the regenerator zone
4 . Wall bricks (header bricks)

Some unused silica bricks (wall shapes) from the coke oven yard were also collected for comparative study.

Methods of study

A.P. (Apparent Porosity), B.D. (Bulk Density), R.Q. (Residual Quartz), Creep-in-Compression & Chemistry, were studied according to DIN 1089, part(1) 1995 (Refractories for use in Coke Ovens, silica bricks requirement and testing Feb.1995). QXRD (Quantitative X-Ray Diffractometer) was used for estimating residual quartz, cristoballite and tridymite. As the tridymite XRD intensity is found to be dependent on the crystal size,[6]only relative tridymite pulse counts were used for comparison. Reversible thermal expansion (RTE) were studied in a Dilatometer as per DIN 50145.

Sample Preparation

The used silica bricks as mentioned above are cut at equal intervals to obtain 3 pieces of 20 mm thickness and marked as section (F) for the portion in contact with gas face and section (C) for the portion in contact with coke face and as section M for the middle portion of the brick. These 3 samples were taken for study of physical properties such as (AP/BD/ASG). After the study, from these cut samples, samples specimens were prepared for thermal study(RTE),chemical property, mineralogical properties such as residual quartz, cristoballite, tridymite by XRD. From the used bricks, 50mmdia sample were drilled from the gas face as well as coke face and taken for creep and RUL study .Similar study will be carried out on the available bricks for comparison.

RESULTS

Physical Properties

The physical properties of silica brick samples collected from different location of Bat-3 after dismantling (Tab. 2). It is found that, the apparent porosity of all 4 samples varies between 14.1 to 21.6%, bulk density varies from 1.76 to 1.95 and apparent specific gravity varies from 2.224 to 2.298.Though all bricks got densified (lower apparent porosity and higher bulk density) from the original condition, the header brick (sample 4) got highest densification. Further, from the values and range of apparent specific gravity, all the bricks moves towards tridymite and glass phases as specific gravities of quartz, cristoballite, tridymite and quartz glass are 2.65, 2.33, 2.27 and 2.21 respectively. The reason behind more amount of glass are due to infiltration of solid particles from coke side and gaseous particles from flue gas side.

Tab. 2: Physical properties of used Silica Bricks

Sl. No.	Brick Description	Location of brick in Coke Oven	Cut brick sample facing side	AP (%)	BD Gm/cc	ASG
1	Wall brick	Ram Side Middle position 18^{th}.Layer	Flue Chamber side	16.9	1.85	2.22
			Middle side	18.9	1.85	2.28
			Coke Chamber side	16.9	1.88	2.27
2	Wall brick	Coke side	Coke Chamber side	20.9	1.81	2.29
		Bottom position	Middle side	20.4	1.83	2.3
		11^{th}. Layer	Flue Chamber side	19.5	1.85	2.3
3	Gas riser nozzle brick	Regenerator Zone	Flue Chamber side	20.5	1.78	2.23
			Middle side(Flue gas hole)	19.9	1.8	2.25
			Coke Chamber side	21.6	1.76	2.25
4	Header brick	Header zone	Coke Chamber side	16.3	1.91	2.28
			Middle side	16.3	1.91	2.29
			Flue Chamber side	14.1	1.95	2.27
5	Original brick			**24.5**	**1.74**	**2.35**

Mineral phase identification by XRD

The mineral phases present in 4 silica brick samples are determined by X-Ray Diffractometer method (Tab.3)

Tab. 3: Mineralogical phases in silica bricks

Brick No.	% Tridymite + Glass	% Cristobalite	% Residual Quartz
1	92	8	0
2	90	8	2
3	95	5	0
4	93	7	0
Original silica brick	44	51	5.0

From the above test results, it is found that, the tridymite with glass varies from 90-95 %, cristoballite phase varies from 5-8% and residual quartz varies from nil to 2 %.It is clear that, all silica brick samples has under gone a drastic change in their phase composition from the original brick composition and all samples were having more amount of tridymite and glass confirming the apparent specific gravities data found in all samples (Tab. 2). This has happened due to longer period of operation and exposure to the temperature zone (870 °C -1470 °C) where more tridymite will be formed.

Chemical Property

The chemical analysis of unused and used silica bricks are given in Tab. 4.

Tab. 4: Chemical Analysis of unused and used silica bricks

Section	C	M	F	
Chemical (wt%)	Coke side	Middle portion	Flue side	Original Silica Brick
CaO	2.2	1.8	2.2	2.2
MgO	Tr.	Tr.	Tr.	Tr
Al_2O_3	1.35	1.40	1.50	1.1
Fe_2O_3	0.89	0.95	1.2	0.7
Na_2O	0.21	0.28	0.35	0.14
K_2O	0.23	0.3	0.5	0.12
SiO_2	94.7	94.3	94.0	94.9
SO_3	**0.11**	**0.15**	**0.18**	NIL

From the above chemical analysis it reveals that, highest amount of infiltration found in section F which is facing flue side. At the same time there is a gradual increase of alumina, alkali and iron oxide fromcoke side to flue side. It is also observed that there is a decrease of calcium oxide in flue face (section F). From these data, it can be concluded that, infiltration of impurities takes place in silica bricks during use. These impurities forms low eutectic glass with silica and increases the glass amount as well as tridymite content.

Thermal Property (RTE)

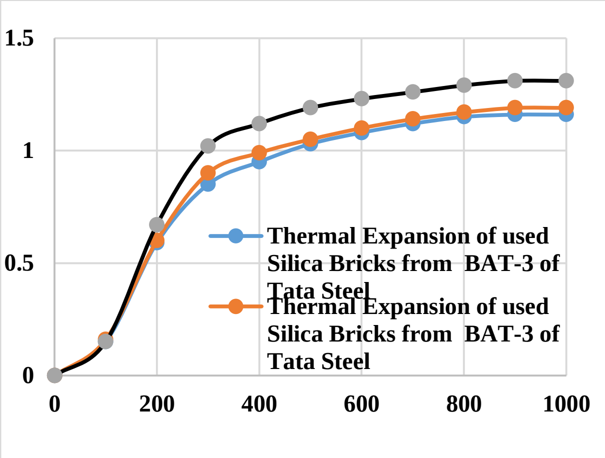

Fig. 1: RTE of used and unused silica bricks

From the Fig. 1, it is found that, RTE value has decrease in both samples and it is lowest in sample from flue side. As the thermal expansion of tridymite phase is the lowest in order of 1.0-1.1% at 1000 °C, used sample contains more amount of tridymite from the original used bricks.

Thermo-mechanical property (Creep- in- compression).

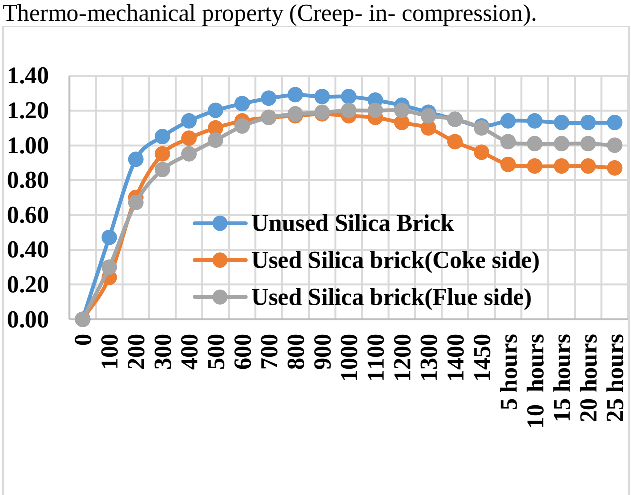

Fig. 2: Creep-in-compression of used and unused silica bricks.

From Fig. 2, it is found that, the creep deformation at 1450 °C after-0.09, 25 hours are +0.02,-0.09 and-0.10 took place for unused silica brick, used silica brick(coke side) and used silica brick(flue side) respectively .This indicates the unused silica brick have higher amount of residual quartz and cristobalite when compared to used silica brick.

Physical and Structural change in silica brick after use

The physical and structural change of silica brick after use is shown in Fig- 3, 4 and 5.

Fig. 3: Physical appearance of unused silica brick

Fig. 4: Physical appearance of used silica brick as recieved

Fig. 5: Physical appearance of used silica brick (cut faces)

Fig.3 shows the colour and structure of original silica brick while Fig. 4 shows the colour and structure of used silica bricks. Fig. 5 shows the cut surface of used silica bricks. It is found that, there is drastically change in colour and structure occurred inside used silica bricks. These changes are attributed to the mineral phase changes that took place along with more amount of glass formation.

DISCUSSION

The most significant change in silica polymorph has been observed in used silica brick samples (Tab. 3). It is found that the amount of cristoballite, which was originally 51% in original brick, has decreased to 5 to 8% overall in samples under investigation.. At the same time, tridymite+glass has increased from 44% in the original brick to 90-95 % in used bricks. This is due to prevailing temperature of 1150°C to 1450°C in different portions of coke ovens for longer period which is the polymorphic transformation temperature of cristoballite in to tridymite.

From Fig. 2, it is found that the difference between the creep values of the coke face and gas face differs slightly. This is due to the lesser amount of infiltration from coal due to lower porosity which is in conformity to the observation of J.D.Pandaet. al, [6&7]

K.Konopicky et al,[5]have taken a number of used samples from different coke oven battery A to H having a life of 4.5 years to 15 years whereas used sample one each were taken from coke oven battery I and K of Republic steel company, Cleveland, USA which was used for 16 years and 42 years respectively. In their study the apparent porosity of coke face of used brick was 27% max where as in 1995 J.D.Pandaet. al, [6&7]studied the silica brick in which the apparent porosity was 23% max. In our present study the apparent porosity of original brick is 19% max. The infiltration in coke face of silica bricks from coke were very high in the silica bricks investigated by K.Konopicky,,[5]. The amount of infiltration in coke face of used silica bricks investigated by J.D.Pandaet. al,[6&7] were lower than that silica bricks investigated by K.Konopicky,[5]. In our present investigations the amount of infiltration is highest. This is due to Higher porosity (25% max) of the silica bricks under investigation when compared to that silica bricks investigation by K.Konopicky et al,,[5]and J.D.Panda et al,[6&7].

K.Konopicky et al,,[5]and J.D.Panda et al,[6&7] found that the tridymite content is highest in gas face of used silica brick. The same phenomena were also observed in silica bricks under investigation. This confirms the cristobalite transformation in to tridymite at the operating temperature of coke oven.

K.Konopicky et al,[5] have found that there is increasing trend of cristobalite from coke side towards fire side where as J.D.Panda et. al,[6&7] found a drastic reduction of cristobalite in gas face of used silica brick when compared to coke face of used silica brick. The same phenomena were also observed in used silica brick under investigation. This is due to the fact that the residual quartz content which was very high (15 – 20%) in coke face of used silica brick investigated by K.Konopicky et al,[5] transformed into cristobalite during use. As the residual quartz in coke face of used silica bricks investigated by J.D.Pandaet. al,[6&7] is 1.8% the amount of transformation of quartz into cristobalite was little. The similartype phenomena were also observed in used silica brick of present investigation.

K.Konopicky et al,[5] have found that the conversion of residual quartz in to cristoballite is dependent on temperature as well as period of use. The bricks which are in use (11 years) converts into cristobalite at temperature in 1030 – 1080°C where as in bricks which are 4 ½ years of service converts slowly and converts around 1100°C. These types of phenomena are not pronounced very much in silica bricks under investigation due to presence of very low amount (<1%) of residual quartz.

CONCLUSION

Silica brick used in coke ovens for more than 40 years undergo significant mineralogical changes. Degree of conversion of the SiO2-Polymorphs depends on the location of the bricks in the walls of coke ovens as well as time and temperature.

The amount of infiltration and change in creep behavior of used silica brick is dependent on porosity and year of use. Due to higher porosity in the original silica bricks and due to prolong period (> 40 years) of use the silica bricks under investigation, the infiltration from coke into brick structure is very high resulting reduction of apparent porosity and increased amount of glass.

Due to prolonged use of silica bricks between 900 °C and 1450 °C , which is the stability temperature range of tridymite, appreciable amount of tridymite has been formed all bricks which is evident from the mineralogical investigation carried out in XRD.

The lower thermal expansion at 1000 °C found in used silica bricks when compared to original brick expansion, further confirms the formation of high amount of tridymite in the used silica bricks as tridymite has the lowest thermal expansion amongst other silica polymorphs namely quartz and cristoballite.

The higher positive creep value and high D_{max} value found in original unused silica bricks also further confirms conforms the presence of higher amount of residual quartz. The lower creep value and lower D_{max} value of used silica bricks confirms the formation of high amount of tridymite in used silica bricks after use.

For longevity and thermal stability of coke ovens, the silica bricks should be dense and should have low residual quartz and lower creep in compression .This helps in efficient heat conduction in chamber walls and brings thermal stability and will have homogeneous thermal expansion.

As the properties of used silica bricks under present investigation are much superior and found structurally stable and used bricks are found intact at their positions, coupled with adherence of good operational practice, the coke battery life has gone to the highest in the history of Tata Steel.

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