MaxH2DR

Maximise H2 Enrichment in Direct Reduction Shaft Furnaces

Procedures for the determination of physical properties of DR raw materials, intermediate and products - Report on bed-scale experiments on two different pellet types

Deliverable D1.2 by WP1. Partners: TS, UNISA, M22

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1. Introduction

Most of the work is carried out by TATA STEEL NEDERLAND TECHNOLOGY B.V. (TS) and UNIVERSITA DEGLI STUDI DI SALERNO (UNISA) within the frame of the task 1.2 “Experimental investigations of physical properties of raw materials, intermediates and products” for the first 22 months of the MaxH2DR project. The main objective of deliverable D1.2 (by WP1. Partners TS, UNISA, M22) was to characterize the main relevant physical properties of the raw materials, reduced products, and final product behavior under dominant hydrogen atmosphere.

In this report, two major investigations are highlighted.

Tata Steel Nederland Technology B.V (TS) focused on physical, chemical and metallurgical aspects investigation of different grade of pellets. In this report, different grade of pellets were characterized under Blast Furnace (BF) conditions. In the next phase, these pellets will undergo reduction and degradation investigation under high hydrogen conditions mimicking Direct Reduced Iron (DRI) operating line. Metallurgical Lab Ironmaking (MLI) located at IJmuiden, state of the art metallurgical testing facility was used to characterize different grade pellets. Fundamentally, pellet tests are developed for two different purposes. Firstly, quality control, to compare different batches of pellets using international standardized tests. Secondly, to simulate the behavior of pellets under process conditions of BF or DRI. International standardized tests are developed to enable consistent comparison between different pellets for quality control. International standardized tests are used by pellet producers (usually iron ore mines) and customers (steel factories) to determine pellet quality between themselves. The most commonly used standardized tests are ISO (International Organization for Standardization) tests. Conversely, process simulation tests are developed for research purposes, to simulate process conditions and to investigate the behavior of pellets to be able to optimize the properties.

Universita Degli Studi Di Salerno (UNISA) has presented status of new hot shear cell which aims at to study the rolling friction and the sticking behavior of iron ore pellets under high hydrogen conditions.

2. Standard tests for Blast Furnace and DRI process

Globally, extensive knowhow on fired pellet quality exists for both BF and DRI process. Quality aspects of the fired pellets depend primarily on physical and metallurgical properties. Physical characteristics of the fired pellets are assessed by Tumbler Index TI & Abrasion Index AI, Cold Compression Strength (CCS), density, porosity, and size distribution. The metallurgical characteristics of the fired pellets are reduction degradation index, reducibility index, softening - melting behavior, and swelling properties. These tests are commonly used for the BF grade pellets. They are reported based on standard ISO tests as listed in Table 1. DRI operators are also widely using these tests for assessing physical properties of Direct Reduced (DR) grade pellets. However, for metallurgical characteristics, DRI operators mainly rely on standard tests such as clustering and sticking index, reduction degradation index or Linder test, and reducibility index. ISO tests commonly performed to assess DR grade pellets are highlighted in (see Table 2). Most of these tests are compatible for DR process like Midrex and HYL /Energiron, where hydrogen content in tests were limited to 45-61%. However, in the framework of the MaxH2DR project, new tests are being investigated under operating conditions of DR shaft up to 100% hydrogen application.

Table 1. List of ISO test for BF and DRI process.

S. N	Test	Purpose	ISO Reference	Test conditions	Suitable
1	TI and AI	Fines generation due to mechanical degradation during transportation.	ISO 3271	Sample: 15 kg Total no of revolutions: 200 rpm @ 25 rpm	BF DRI
2.	Decrepitation Index DI	Measure for evaluating the degree of size degradation caused by rapid heating of iron ore lump.	ISO 8371	Temperature: 700°C Time: 30 min	BF DRI
3.	Swelling	Swelling behavior of pellets	ISO 4698	Sample 18 pellets Temperature: 900°C Gas composition: 70% N2, 30% CO Gas flow rate: 15 Nl/min Time: 60 min	BF
4.	CCS	Cold Compression Strength	ISO 4700	Sample: > 60 pellets Compression speed: 10-20mm/min Capacity of load: 1000 daN/pellet End of test: When load or contraction is <50%	BF DRI
5.	Reduction Degradation Index RDI	Degradation behavior/ fines generation after reduction	ISO 4696-1	Temperature: 500 °C Gas composition: 20% CO, 20% CO2, 2%H2O, 58% N2 Gas flow rate: 20 L/min Time: 60 min	BF
5.	Reduction Degradation Index RDI	Degradation behavior/ fines generation after reduction	ISO 4696-2	Temperature: 550 °C Gas Composition: 30% CO, 70% N2 Gas flow rate: 15 L/min Time: 30 min	BF
6.	LTD (static)	Low Temperature Degradation	ISO 13930	Temperature: 500 °C Gas composition: 30% CO, 70% N2 Gas flow rate: 20 L/min Time: 60 min	BF
7.	Reducibility Index RI	Reducibility Index	ISO 4695	Temperature: 950 °C Gas: 40% CO, 60% N2 Gas flow: 50 Nl/min Time: 240 min max	BF
8	Reduction Under Load RUL	Reduction behavior under load condition	ISO 7992	Temp 1050 °C Gas composition: 40% CO, 2% H2, 58% N2 Gas flow: 83 NL/min Load: 20 mbar Time: 240 min	BF

Table 2. List of ISO test for DRI process.

S.N	Test	Purpose	ISO Reference	Test conditions	Suitable
1	Clustering and Sticking Index %	Quantification of clustering and sticking tendency of pellets	ISO 11256	Temperature: 850 °C Gas composition: 30% CO, 45% H2, 15% CO2, 10% N2 Gas flow = 40 L/min atmospheric pressure Load: 147 kPa Time = 60 min	DRI
2.	Linder Test RDI -DR Reduction - Disintegration (Dynamic reducibility test)	The Linder test was designed primarily for low temperature degradation and reducibility for DRI conditions (Midrex)	ISO 11257	Temperature: 760 °C Gas composition: 55 H2, 36% CO, 5% CO2, and 4% CH4 while the sample is tumbling in a rotating drum Gas flow: 13 L/min Time: 300 min	DRI
3.	Reducibility DR90	Reducibility Index for direct reduction process	ISO 11258	Temperature: 800 °C Gas composition: 45% H2, 30% CO, 15% CO2, and 10% N2 Gas flow: 50 L/min Time: 90 min	DRI
4.	COREM R180	Reducibility for direct reduction	COREM R180	Temperature: 400 to 850°C Gas composition: Mix 1: 61 H2, 5.5% CO, 28.5% CO2, and 5% N2 Mix 2: 61% H2, 14.0% CO, 20% CO2, and 5% N2 Mix 3: 61% H2, 26.0% CO, 8% CO2, and 5% N2 Gas flow: 30 L/min Time: 180 min	DRI

3. Test Procedure and Results

3.1 Chemical Composition

In the framework of the MaxH2DR project, two variants of pellets, i.e., BF grade and DR grade pellet were collected, screened and distributed to all partners for experimental work. They were screened in two different sizes; standard size of 10-13 mm and slightly bigger size of 13-15 mm since DR process prefers mainly large pellets. Two types of BF grade pellets were differentiated based on their basicity CaO/SiO_2 (low and high), whereas DR grade pellets were distinguished based on their coating types (lime coated and bauxite coated). Chemical analysis of these pellets is shown in Table 3. Total iron content in BF grade pellets were relatively low at 64.6-64.9% whereas the total iron contents in DR grade pellets were high at 66.4-67%. Accordingly, the gangue content in BF and DR grade pellets were about 6.8 % and 3.6 %, respectively. Two extreme basicity levels were considered for BF grade pellets with low to high range of 0.22 to 0.76. Magnesium oxide (MgO) content in the BF grade pellets low value of 0.38% and high value at 1.34%. On the contrary, both basicity and magnesium content in DR grade pellets were varied in narrow range of 0.44 - 0.55 and 0.12 - 0.28% respectively (see Table 3). All types of pellets were completely assessed w.r.t physical and metallurgical properties highlighted in Table 4.

Table 3. Chemistry of BF and DR grade pellets.

Pellet	Fe %	SiO_2 %	CaO %	MgO %	Al_2O_3 %	FeO %	MnO %	P_2O_5 %	TiO_2 %	B2 %	Gangue %
A1	64.9	5.0	1.0	0.39	0.34	0.15	0.15	0.02	0.05	0.21	6.7
A2	64.6	5.1	1.1	0.38	0.35	0.16	0.16	0.02	0.04	0.21	6.9
B1	64.6	2.8	2.1	1.34	0.64	<0.1	0.27	0.06	0.21	0.75	6.8
B2	64.9	2.8	2.1	1.33	0.64	0.15	0.27	0.06	0.21	0.77	6.8
C1	66.9	1.7	1.0	0.28	0.43	0.69	0.05	0.03	0.21	0.57	3.4
C2	67.0	1.7	0.9	0.29	0.40	0.73	0.04	0.02	0.22	0.53	3.3
D1	66.4	2.3	1.0	0.12	0.73	0.71	0.15	0.06	0.09	0.41	4.1
D2	67.1	2.0	0.9	0.12	0.56	<0.1	0.14	0.06	0.08	0.47	3.6

A1: BF grade pellet A 10-13 mm; A2: BF grade pellet A 13-15 mm; B1: BF grade pellet B 10-13 mm; B2: BF grade pellet B 13-15 mm; C1: DR grade pellet C 10-13 mm; C2: DR grade pellet C 13-15 mm; D1: DR grade pellet D 10-13 mm; D2: DR grade pellet C 13-15 mm; gangue = $CaO+SiO_2+Al_2O_3+MgO$

Table 4. List of chemical, physical and metallurgical tests and their applicability to DRI.

Test type	Applicable to BF/DR
Chemistry	BF + DR
Porosity	BF + DR
Density	BF + DR
Mini ASTM	BF + DR
Cold Compression Strength	BF + DR
Swelling Index	Applicable to BF only
Q-Hosim	Applicable to BF only.

3.2 Porosity measurement

Porosity is an important physical property of the pellets because it directly influences reducibility. The higher the porosity, the better is the reducibility of the pellets. The GeoPyc 1360 instruments is commonly used for measuring bulk density or envelop density (see Figure 1). Porosity is measured, if true density of the pellets is known, using the AccuPyc II 1340 (explained above). In the GeoPyc instrument (envelop density or bulk density analyzer), the sample volume was analyzed by packing it in silica sand, dry flow. Such device is compatible to measure the bulk/envelop density of porous pellet [1] [2].

First, the sample chamber was filled with sand only and packed under rotating movement to a desired pressure. From the position of the piston, the volume of the sand was calculated (blank measurement). Thereafter, the sample chamber was opened, and the weighed sample was transferred inside (~10 pellets weighed and inserted in the chamber). The packing to desired pressure was repeated and the total volume of sand and sample was measured (sample measurement). From the difference in volume between the sample and blank measurements, the sample envelope volume was calculated. The porosity was calculated by subtracting the volume of particles from the envelope volume.

$$\varepsilon = \left(\left(Ve - m/(\rho a)\right)\right)/Ve \quad (1)$$

ε = fractional porosity
Ve = envelope volume, cm3
m = sample mass, g
ρa = absolute density (particle or skeletal density measured by gas pycnometer) g/cm3

Figure 1. GeoPyc 1360 device for pellets porosity measurement.

All four pellets types (BF and DR grades, A, B, C, & D) in their two different sizes were studied in the GeoPyc 1360 device. Initially, the envelop density (i.e., bulk density) of all pellets were measured in the GeoPyc (see Figure 2). Envelop or bulk density of both BF and DR grade pellets were varied between 3.50-3.73 g/cm^3. Porosity was measured using fractional porosity equation. Porosity of all pellets were observed in the range of 27.4% to 30.3% (see Figure 3). The average porosity of both BF and DR grade pellets were 28.9% and 29.6% respectively. Comparatively, bigger pellets (13-15 mm) were more porous than smaller pellets (10-13 mm). The difference between bigger and smaller pellets is 0.7%.

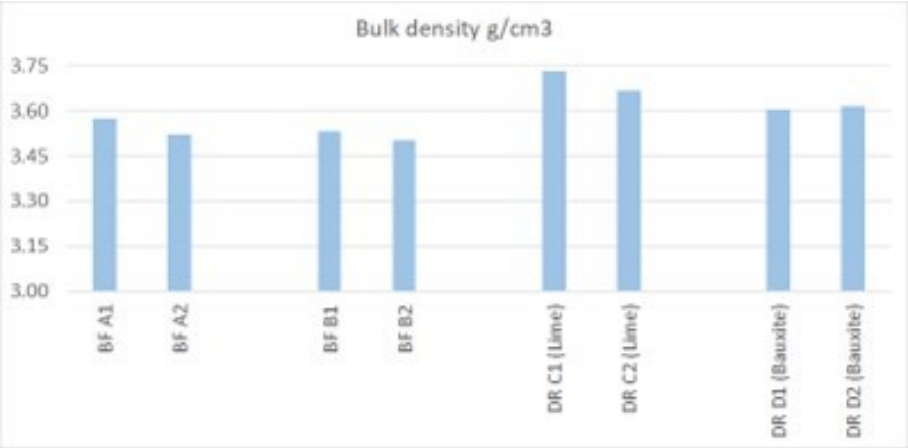


Figure 2. Bulk density measurement of BF and DR grade pellets.

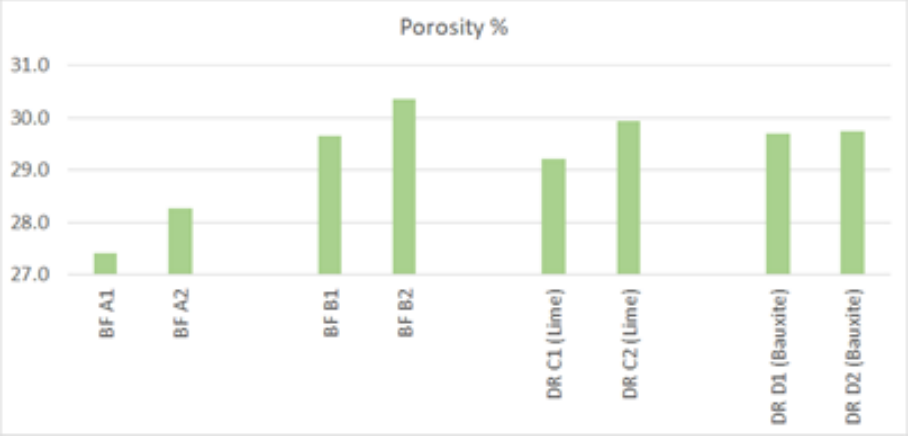


Figure 2. Porosity measurement of BF and DR grade pellets.

3.3 Density measurement

True or Absolute Density measurement

The AccuPyc II 1340 Pycnometer is a gas displacement pycnometer (Figure 4). It determines density and volume by measuring the pressure change of helium within calibrated volumes. The sample volume can be measured, from which density can be derived automatically if the sample mass is known [1] [2]. Three pellets are weighed and inserted in the chamber. True density of pellet is computed using the helium gas displacement method.

Figure 4. AccuPyc II 1340 device for measuring density.

True or absolute density of all pellets were measured in the AccuPyc II 1340 (see Figure 5). True density of both BF and DR grade pellets were varied between 4.92-5.19 g/cm^3. Average true density of DR grade pellets (~5.16 g/cm^3) is higher than BF grade pellets (~ 4.97 g/cm^3). It can be easily explained by higher iron content in DR grade pellets.

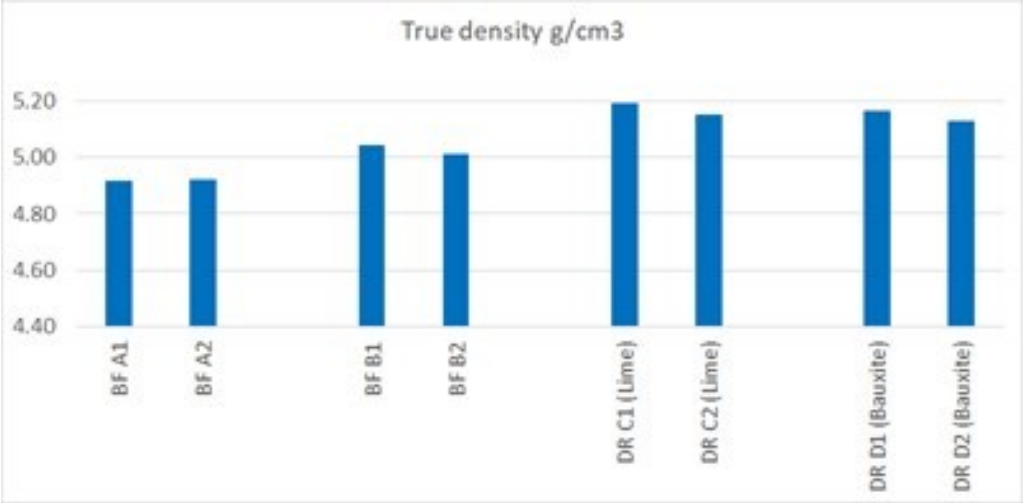


Figure 5. True or absolute density measurement of BF and DR grade pellets.

3.4 Tumbler and Abrasion Index measurement/Mini ASTM

The most commonly used method to quantify the formation of fines by mechanical degradation is by determining the TI and AI. The pellet sample is tumbled in a tumbling drum and then sieved to determine the percentage intact pellets (tumble index), broken pellets and fines (AI). The test is meant to simulate conditions during transportation of pellets.

Relatively, mini-ASTM test can be used as an indicator for fines generation (AI) as broken pellets and fines inside a DR shaft furnace. Mini ASTM tumble tests can be used as relevant standards for differentiating quality of pellets (both BF as well as DR grade) w.r.t to its mechanical degradation (see Figure 6)

In the ISO standard (ISO3271:2007) [3], the AI is determined by tumbling a batch of fired pellets in a rotating tumble drum and by determining the fraction smaller than 0.5 mm (%< 0.5 mm, AI)

and the fraction larger than 6.3 mm (%> 6.3 mm, TI). First, the total weight before tumbling is recorded (m_0), then the sample is tumbled and sieved using a 6.3 mm sieve and a 0.5 mm sieve, to collect a fraction >6.3 mm (m_1) and a fraction of 0.5– 6.3 mm (m_2). The difference in weight of the starting weight before tumbling and the two collected sieving fractions are the fines <0.5 mm. The tumble index (eq. 2) and abrasion Index (eq. 3) are calculated as follows:

Tumble Index: TI% = $m_1/m_0 \times 100\%$ (2)

Abrasion Index: AI% = $\big(m_0 - (m_1 + m_2)\big)/m_0 \times 100\ \%$ (3)

where m_0 is the total weight before tumbling, m_1 is the fraction > 6.3 mm and m_2 is the fraction of 0.5 – 6.3 mm. At TS, a smaller version of the tumble drum that requires only 500 g of material is used, called the mini-ASTM (see Figure 6). Table 5 shows comparison between the ISO test and mini-ASTM. ISO test requires relatively larger batch of sample up to 15 kg which undergoes tumbling in bigger drum of ~1000 mm length and 500 mm diameter with two lifters.

Figure 6. Small drum mini- ASTM device for measuring tumbler and abrasion index.

Table 5. Comparison of ISO 3271 (2007) and Mini ASTM conditions for TI and AI.

Test method	ISO 3271 (2007) International Standard	Mini-ASTM
Apparatus	1000 mm OD 500 mm wide 2 lifters 50 x 5 mm	130 mm ID (OD = 140) 195 mm wide 2 lifters 20 x 5 mm
Number of revolutions	200 at 25 rpm	600 at 30 rpm
Weight of materials (dry)	15 kg	0.5 kg
Material tested	Pellets, sinter or lump ore	Pellets, sinter or lump ore
Size of pellets	Pellets 6.3 – 40 mm	Pellets 10 – 13 mm
Expression of results	Tumbler index: wt% > 6.3 mm, Abrasion index wt% < 0.5 mm	Mini-ASTM value wt% > 6.35 mm wt% < 1 mm

OD = out diameter, ID = inner diameter

All four pellets types (2 BF, 2 DR) in their two different sizes were studied in themini-ASTM. TI (+6.35 mm, %) of both BF and DR grade pellets were varied between 96.6-98.1% (see Figure 7). AI (<1mm, %) of all pellets were varied between 1.8-3.2% (see Figure 8). There is a marginal difference observed between BF and DR grade pellets w.r.t to their TI and AI. Among all pellets, BF grade pellet

A1 has high TI up to 98%. Comparatively, DR grade pellet D1 (bauxite coated) generated slightly higher fines than BF grade pellet A1 ~3.2 and 1.8%, respectively. Also, bigger size pellets generated 0.5% higher fines compared to their smaller size pellets of 10-13 mm.

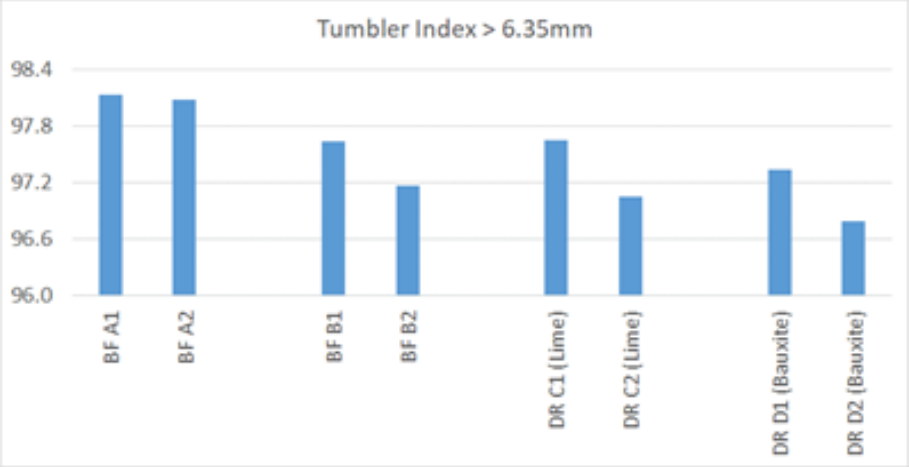


Figure 7. Tumbler index of BF and DR grade pellets.

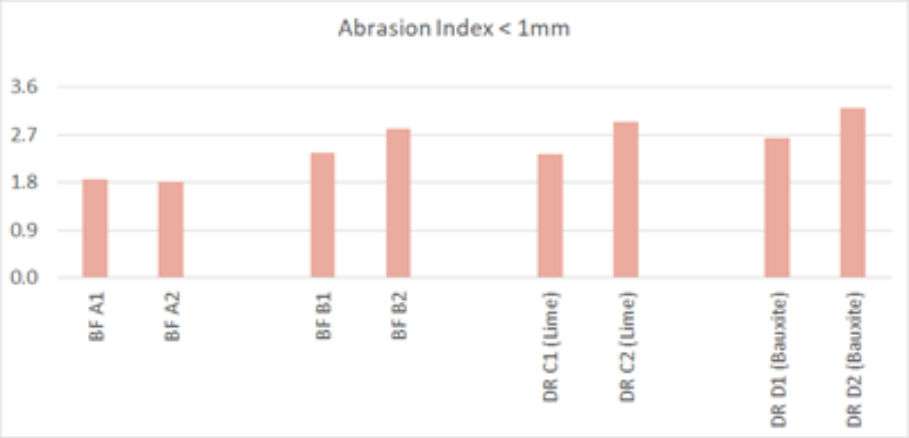


Figure 8. Abrasion index of BF and DR grade pellets.

3.5 Cold Compression Strength (CCS)

The most common method to determine pellet strength, resistance against body breakage, is the CCS. The international standard for this test is ISO 4700:2015. Fired pellets are sieved to a size of 10 – 12.5 mm. At least 60 pellets are tested to determine the average cold compression strength as per ISO whereas ~118 pellets were crushed as per TS standard. The test is complete when the load falls to a value of 50% or more of the maximum load recorded, or when the plate gap has reduced to 50% of the initial test-piece diameter. Comparison of ISO test and ISO CSS at TS are highlighted in Table 6. Cold compression test apparatus at TS IJmuiden is shown in Figure 9.

Table 6. Different test methods to measure the cold crushing strength (CCS) of fired pellets [4] [5]

Test condition	ISO CCS 4700:2015	ISO CCS TS Standard
Pellets screened to size	10 – 12.5 mm	12 – 13 mm
Capacity of load cell	1000 daN/pellet*	1000 daN/pellet
End of test	When load or contraction is <50%	When load or contraction is <50%
Average taken of number of pellets	>60	>118

*1 daN = 1.02 kgf

Figure 9. Determination of the cold crushing strength.

The CCS (ISO) of both BF and DR grade pellets were varied between 240-280 daN (see Table 7). Among all pellets, BF grade pellets (A1, B1) and DR grade pellet (D1) were comparable with CCS within 275-282 daN. However, DR grade pellet C1 (lime coated) has observed lowest CCS up to 241 daN with weak pellet fraction (<120 daN) as high as 10.5% (see Figure 10).

Table 7. Cold crushing strength (CCS) of BF and DR grade pellets.

Pellet	CCS ISO	CCS STDEV	<60 kg	<120
BF A1	275	83.1	2.6	4.4
BF B1	282	95.5	0.0	4.6
DR C1	241	79.0	0.9	10.5
DR D1	277	97.9	0.0	4.5

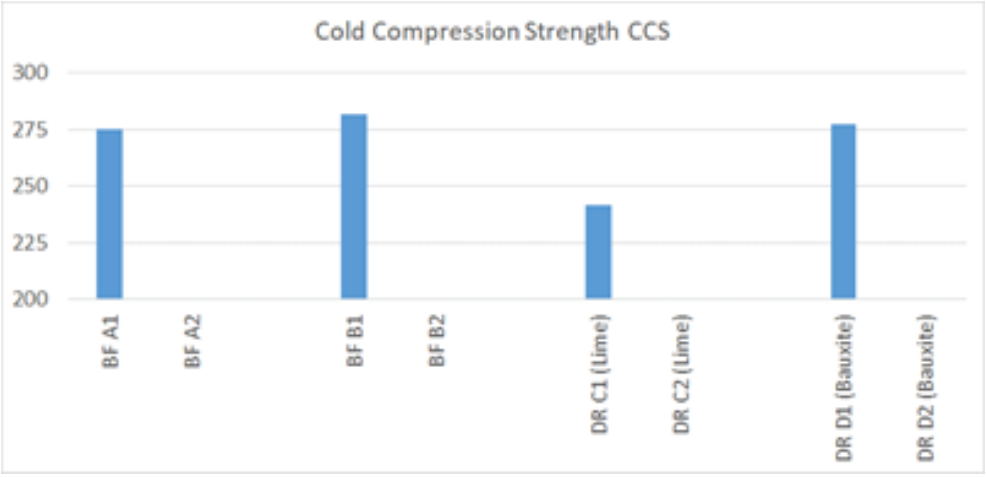


Figure 10. Cold Compression Strength of BF and DR grade pellets.

3.6 Swelling Index

Swelling properties of fired pellets are very critical to both BF as well as DR process. Some pellets have strong tendency to swell enormously in a reducing atmosphere at 700-1000°C. Swelling or volume increase of pellets can lead to sticking of pellets or to loss of their strength and collapse which negatively affects the smooth operation of BF and other shaft furnaces. Currently, such test is only possible in BF conditions with gas consisting of 100% CO at constant temperature of 900 °C. Total reduction time was ~90 minutes. The testing facility of swelling index is shown in Figure 11.

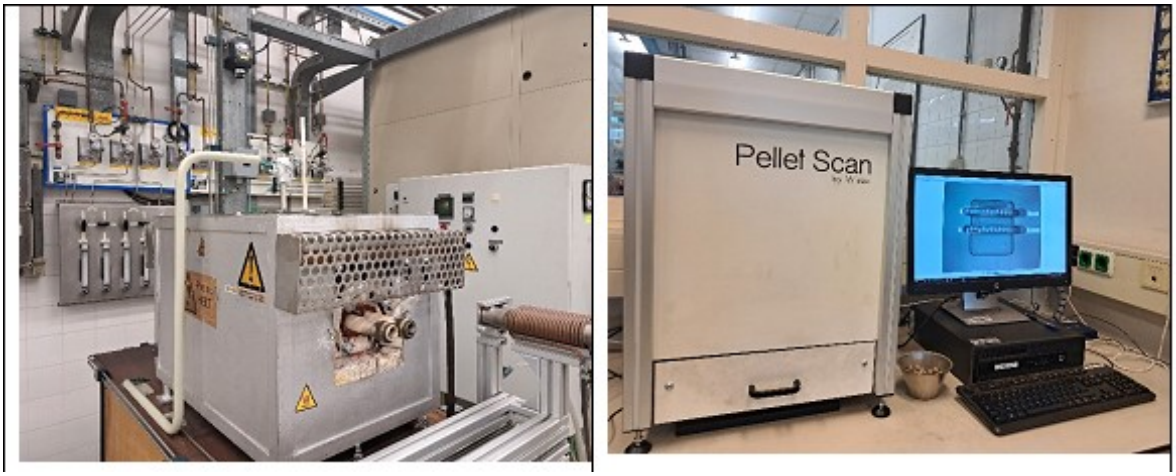


Figure 11. Testing facility of swelling index.

Swelling index of both BF and DR grade pellets were analyzed in BF condition. Results confirmed that BF grade pellet B (B1, B2) has very high swelling index up to 24% (see Figure 12). High swelling index was observed in BF grade pellet B due to excessive swelling of few pellets (red circle) highlighted in Figure 13, whereas other pellets (A, C and D) show relatively low swelling index ~13%.

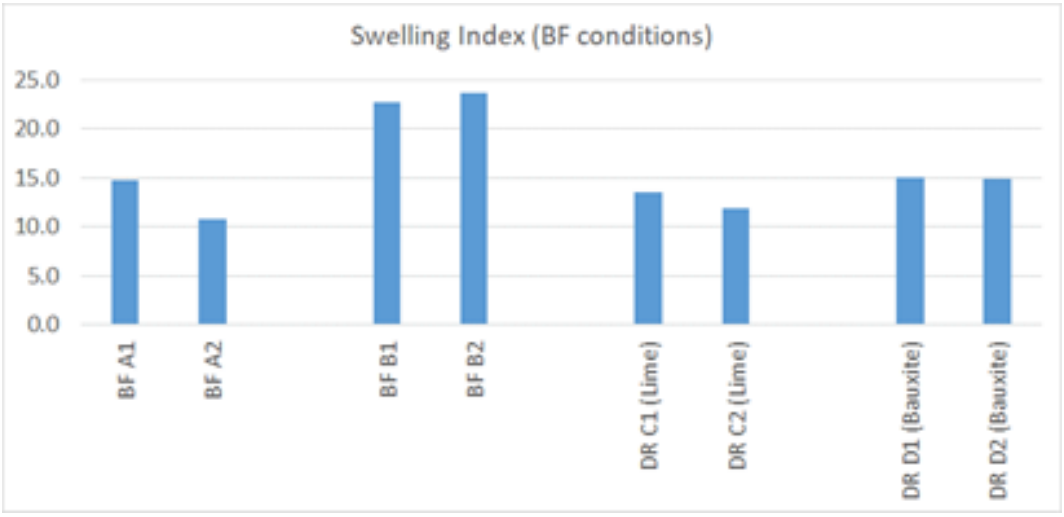


Figure 12. Swelling index of BF and DR grade pellets.

Figure 13. Swelling observation on BF grade pellet B1 (10-13 mm).

In high hydrogen condition, it is important to study swelling behavior of pellet, however testing of swelling behavior under high hydrogen conditions is not possible at IJmuiden.

3.7 Q-HOSIM

The Q-HOSIM test characterizes degradation behavior of the pellets in upper shaft in BF conditions. The Q-HOSIM follows a set time/temperature profile, the reducing gas composition is based on the reduction degree that is monitored during the test by the weighing device. The Q-HOSIM test is stopped when a reduction degree of O/Fe = 1.33 (magnetite) is reached and the time to reach this reduction degree, as well as reduction disintegration index after tumbling, are determined (see Table 8). During phase transition from hematite to magnetite, most reduction disintegration takes place.

Table 8. Overview of Q-HOSIM.

Parameter	Q-HOSIM
Sample size	500 g
Maximum reduction degree (O/Fe)	1.33
Mainly used for	Pellets (10–13 mm) Pellets (13-15 mm)
Charging Temperature	400-550°C
Changing Gas	CO, CO2, N2, H2
Reduction disintegration after tumbling	%<3.15 mm %<6.35 mm

The Q-HOSIM test was performed on both BF and DR grade pellets and the results are summarized in Table 9. Results confirmed that very high degree of degradation was observed for BF grade pellet B (B1, B2) with and without tumbling. After partial reduction (once O/Fe ratio of 1.33 reached), pellet B fines generation (<6.35 mm, %) was as high as 6-7% whereas other pellets B, C, D generated fines below 3.3% before tumbling. After tumbling of the partially reduced pellets, fines generation (<6.35 mm, %) was as high as 19-21% in case of BF grade pellet B (B1, B2) whereas other pellets B, C, D (both sizes) generated fines below 8%. Out of two DR grade pellets (C, D), bauxite coated pellet D (D1, D2) generated more fines below 8% whereas lime coated pellet C (C1, C2) generated slightly less fines up to 4-6%. Possible reasons for high disintegration of BF Pellet B are higher CaO content in pellet, and higher porosity. Hematite to magnetite transition was relatively

fast for DR grade pellet (bauxite coated, 13-15 mm) D which took about 30-33 minutes whereas other pellets took up to 40 minutes (see Figure 14).

Table 9. Q-HOSIM test results of BF and DR grade pellets.

Pellet	Q-Hosim after reaction before tumbling, % <6.35 mm	Q-Hosim after reaction after tumbling, % <3.15 mm
A1	1.18	4.44
A2	0.94	4.42
B1	6.86	19.3
B2	5.75	20.9
C1	0.87	3.85
C2	2.02	6.00
D1	3.24	7.41
D2	2.84	7.86

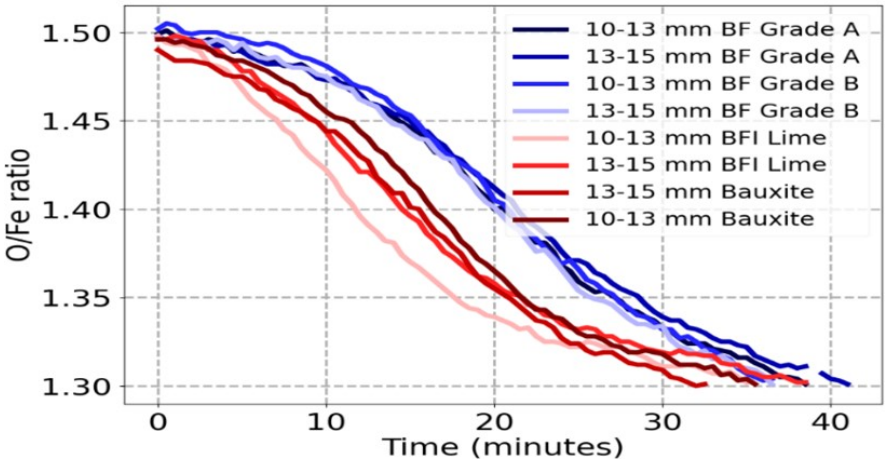


Figure 14. Reduction speed of BF and DR grade pellets under BF conditions.

3.8 Sticking properties

3.8.1 Investigations related to the characterization of material flow properties at UNISA

The work at UNISA concerns the determination of the flow properties of iron ore pellets and sinter in the reduction process occurring in the DR plant. Due to chemical reactions, the composition of fired raw materials changes over time while moving downwards in the furnace; therefore, raw materials, intermediates and products of the reduction process will be materials of interest for experimental testing. The experimental procedure conceived is to replicate the DR of iron ores at a laboratory scale using hydrogen at DR temperatures and by applying compression stresses in the industrial process range to observe the corresponding flow properties. From the top of the DRP, environmental conditions, solid phase composition and compressive stress change along the depth. Consequently, a combined set of operating conditions must be tested. Using standard equipment for shear testing would require quite a large equipment size. Considering the particulate

size of around 1 cm, conventional shear testing equipment would require cell sizes in the order of meters and applied loads of tenths of tons. Such conditions are incompatible with laboratory equipment that can work at process conditions. Therefore, the aim is to use the experimental shear tests with data analysis developed with the help of DEM modelling to calibrate the model with the relevant parameters that can replicate particle friction, softening and cohesion in the different regions of the DRP.

In order to carry out relevant shear tests, the typical stress conditions to which the particles are subjected along the DR plant were determined. In these preliminary calculations, the process temperature and the gas composition give the relevant conditions, while the compressive stress must be estimated. To estimate the magnitude of vertical stress,σ_{zz} of vertical stress the Janssen approach is followed to estimate the maximum stress.

Considering a conservatively high value of the bulk density of 6 000 000 kg m-3 and a furnace internal diameter of 10 m, the overestimated value of the vertical stress component is about 600 kPa.

To summarize, the parameters needed to reproduce in the experimental setup the DR BF conditions are:

- Temperatures up to 1000°C.
- Hydrogen concentrations between 55%w/w and 100%w/w of H_2.
- Compressive stress up to 600 kPa

The design of the experimental setup and procedure has to address the main issues related to these conditions:

- Hydrogen, under DR conditions, can react rapidly with oxygen in the atmosphere, making a flame outside the visible spectrum. Therefore, gas leakage must be avoided. In addition, monitoring the hydrogen concentration in the outlet gas provides information about reactions.
- Working with high temperatures and high hydrogen concentrations challenges maintaining the device integrity over time. Defining a structure of materials able to resist hydrogen attacks and preserve their mechanical properties at high temperatures has been the principal issue.
- Preventing oxygen from entering the system is necessary to avoid the reaction between O_2 and H_2, which results in hydrogen losses due to unwanted reactions, and to prevent further reactions between the oxygen and the pellets.

The work carried out in the first months can be summarized in two main actions:

I. Conduction of preliminary experimental tests to determine the specifications of the testing equipment and the appropriate experimental procedures that will allow a reliable measure even with the coarse material on small cells.
II. Design of experimental testing rig and definition of the whole set of sensors and actuators to be acquired from the market.

3.8.2 Conduction of preliminary experimental tests

The Schulze Ring Shear Tester was used as a reference device to define better our device's structure and test the applicability of a two-step procedure for the iron ore particulates. As previously mentioned, such tests are necessary because the shear tester and the shear testing procedure were developed to characterize particulates with particles having an equivalent diameter of less than a few millimeters, whereas the range of pellets and sinter grains to be analyzed is between 10 to 15 mm.

For all the tests conducted at ambient temperature, a standard Schulze cell (given by the lid and trough) is used; its geometric parameters are reported below:

- Trough : inner diameter 100 mm ; outer diameter 200 mm ; height 40 mm.
- Standard lid: inner diameter 102 mm; outer diameter 198 mm.20 fins of 1(t)x10(h)x48(w) are arranged radially in an equidistant manner.
- Modified lid: inner diameter 102 mm; outer diameter 198 mm.10 fins of 1(t)x12(h)x48(w) mm.

The modified lid was used with larger particles. For these particles the fins of the standard lid were too short to ensure proper mobilization. The internal volume of the trough is 940.23 cm3, the fins volume is 11.54 cm^3, while the trough angular velocity is kept equal to 15 mm min^{-1}, according to the Schulze Ring Shear Tester manual.

Tests were conducted using perfectly spherical monodisperse wooden spheres provided by BFI in the Project consortium and with Iron ore pellets. Their properties and the properties of the other materials tested are reported in Table 10.

Table 10. Sample material properties.

Material	Wood	Wood	Iron ore pellets
Diameter [mm]	6	10	10-13
Density [kg m^{-3}]	~700	~700	4000
Size distribution	mono-sized	mono-sized	wide
Sphericity	1	1	~0.9
Number of particles in the cell	~4600	~1000	~760

The pre-shearing and shearing loads used in the experiments are reported in Table 11.

Table 11. Pre-shear loads.

Pre-shearing loads [kg]	Shearing loads [kg]
5.0	3.0; 2.0; 1.0; 0.3; 0.1
2.5	1.5; 1.0; 0.5; 0.3; 0.1
2.5	0.9; 0.7; 0.5; 0.3; 0.1

Typical data time series observed during the test are shown in Figure 15, for wooden spheres where the grey curve represents the compressive stress, the orange curve represents the measured stress, and the blue curve indicates the vertical displacement of the lid during the test. Negative values of the vertical displacement corresponds to an expansion of the sample.

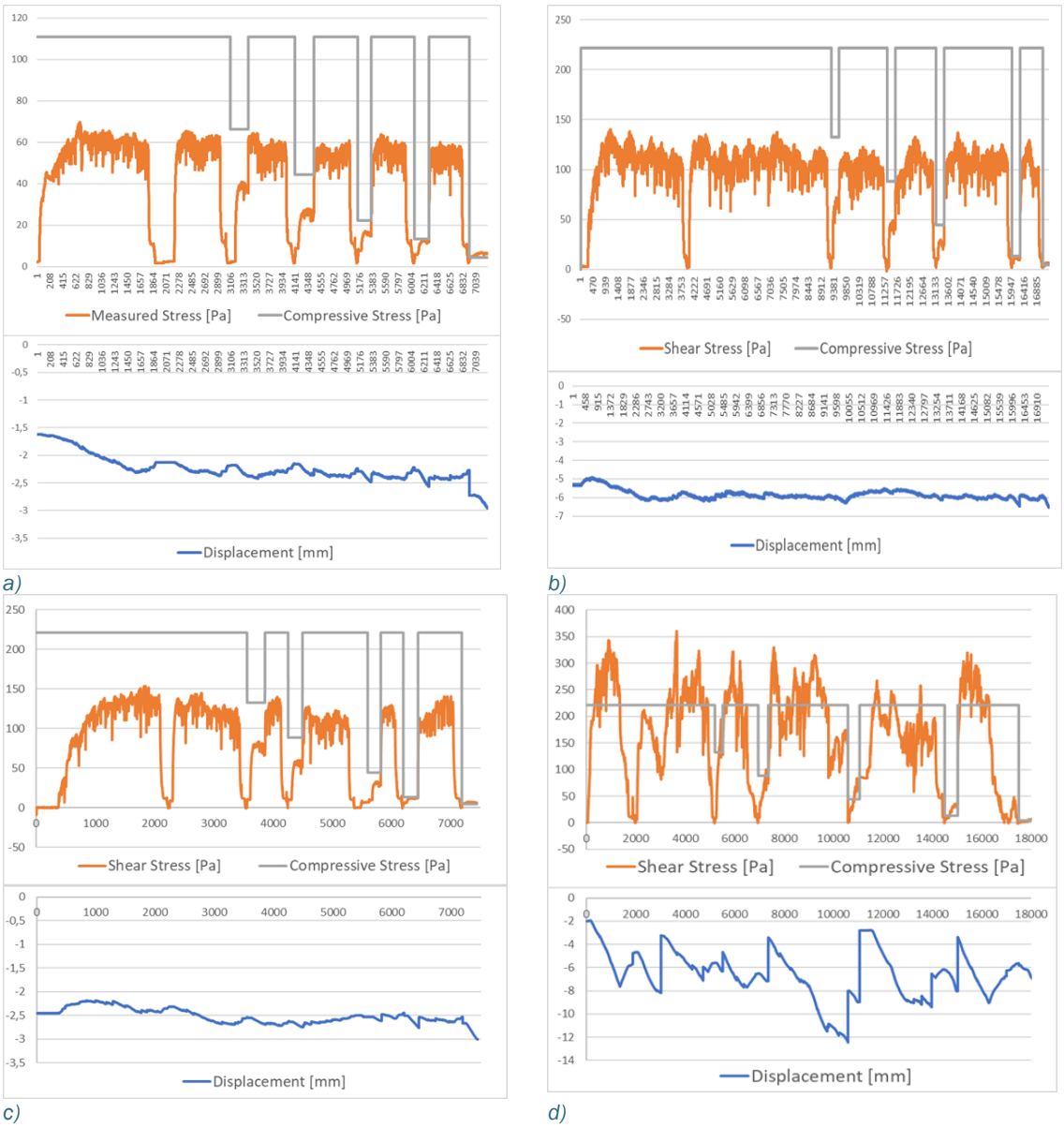


Figure 15. Raw data time series obtained with the Schulze Ring shear tester,5kg pre-shear load: a) 6 mm wooden spheres, standard lid; b) 10 mm wooden spheres, standard lid; c) 10 mm wooden spheres, modified lid; d) Iron ore pellets, modified lid.

Inspection of Figures 15.b and 15.c allows a comparison of the results obtained with the standard lid and the new one with 10 mm monodisperse wooden spheres and applying the pre-shear load of 5 kg. Inspection of the figure reveals that the steady state in the case of the new lid was reached faster than in the case of the standard one. Consequently, the observation time required for testing was significantly reduced, while the oscillations recorded for the measured force and the displacement are comparable. For iron ore pellets Figure 15.d shows intense fluctuations for the measured force and displacement of the lid during the tests. This behavior was associated with the progressive formation and breakage of force chains during the rotation of the trough. These chains can strongly affect the measured value given the reduced number of particles. In addition, the formed force chains progressively ejected the lid from the sample. As a result, the lid reduced the contact surface area with the material, and the measured force value was mainly related to the rubbing of the fins on the sample surface. To solve this issue, a manual twisting of the lid was tried to relocate it within the sample. This action is visible in the blue curve in Figure 15.d, representing

the lid displacement. In this curve, the drastic reduction in the value given by the position sensor, represented by a vertical line, is due to the operator's action.

Figure 16 reports the yield loci (the limiting yield curves on the $\sigma - \tau$ plane) of materials. Experiments were repeated three times and reported in different colors. The data obtained from the two lids for 10 mm wood pellets Figure 16.b and Figure 16.c are comparable, demonstrating the new lid's applicability to perform shear testing. However, with the new lid, there is a significant reduction of the uncertainty of the outputs, suggesting its use with particulates, including particles of about 10 mm in diameter. Despite the problem of the lid ejection and subsequent operator interaction, the data were handled following the same approach for iron ore pellets. The corresponding three yield loci are reported in Figure 16.d. They show higher uncertainty than yield loci obtained with wooden spheres. Indeed, the polydispersity of the iron ore sample and the operator interaction have influenced the results.

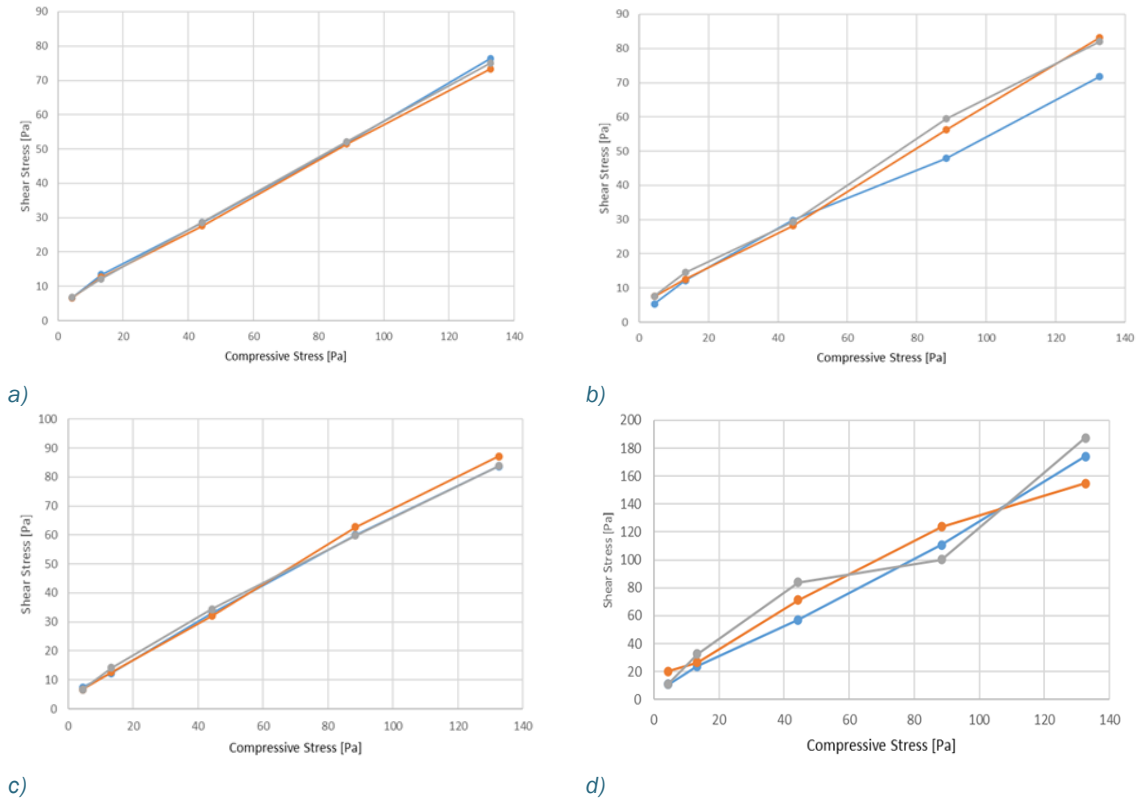


Figure 16. Yield loci at 5.0 kg pre-shear load: a) 6 mm wooden spheres, standard lid; b) 10 mm wooden spheres, standard lid; c) 10 mm wooden spheres, modified lid; d) Iron ore pellets, modified lid.

To improve the procedure and highlight the role of the particle configuration in the system in the chain formation, the experimental procedure was adapted to repeat the procedure but to avoid significant changes in the spatial configuration of the pellet. The steady state in the pre-shear phase was not sought, but the pre-shear step was stopped at the first peak. The outputs obtained are reported in Figure 17 for comparison, while the entire sequence of shear loads is reported in Table 11 for iron ores. Following the orange line in the figure reporting the shear stress, it is possible to identify the three pre-shearing steps, characterized by the highest value of force measured and the three shearing phases. The time associated with the pre-shearing is about 100 s. The first 40 s are required to reach a short 5 s plateau where the measured force can be considered constant. After this time, the system is relaxed by rotating the trough in the opposite direction to reach a zero-shear stress condition. The time associated with sample consolidation ends when the first measured

force plateau value is reached. The counter-rotation also ends when the measured shear stress is close to zero, the load is reduced, and the shearing phase is conducted. Once the failure condition is detected, the system is relaxed again.

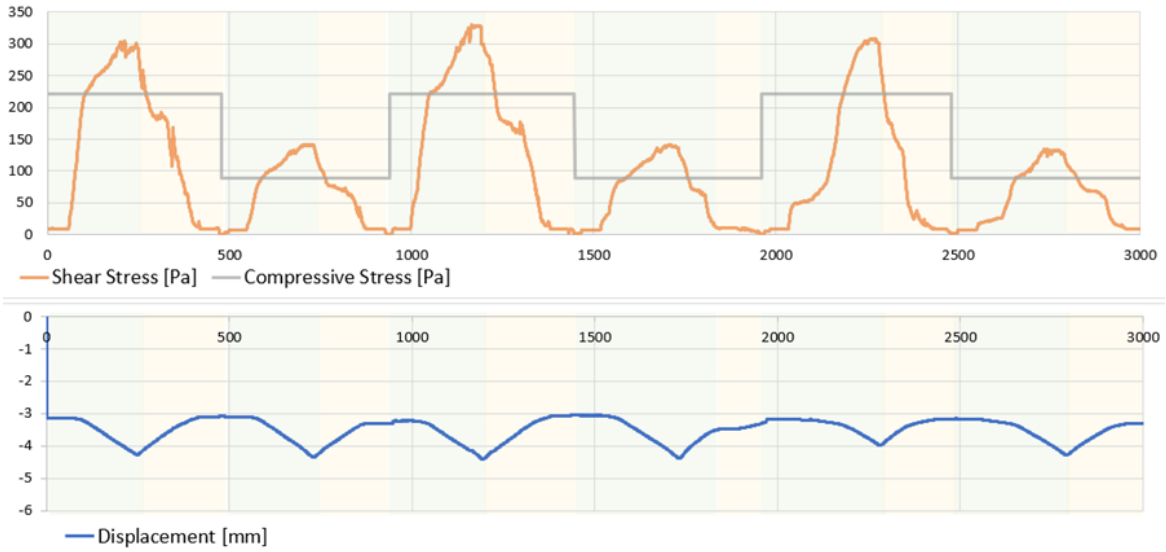


Figure 17. Raw data time series obtained with the Schulze Ring shear tester loaded with iron ore pellets, using the modified lid, using a pre-shearing load of 5 kg and a reduced pre-shear duration of 100 s. Regions characterized by normal trough rotation are green; those characterized by counter rotation are yellow.

By comparing Figure 15.d to Figure 17, the high repeatability of results can be attributed to the limited shear and the relaxation step obtained by the trough counterrotating. The high repeatability of the experimental results includes many details of the time history. It appears that, by relaxing the system, the particles seem to re-establish a similar internal configuration from one step to the next, strongly improving the repeatability of the outputs. The other advantage is related to the shorter time required for the test, which is a clear advantage for DEM simulations.

3.8.3 Design of experimental testing rig

The new device includes a shear cell consisting of a trough and a lid with fins, a heating system to reach temperatures up to 1000°C, an engine system to apply on the lid a normal load of about 800 kPa while the trough is placed in rotation, and a system to feed the hydrogen-rich gas. AISI310 austenitic steel was chosen for the cell and the part in contact with hydrogen for its composition adequate to withstand the hydrogen chemical attack under high temperature.

The lid's main function is to apply the vertical load and effectively drag the particle to promote internal shear in the particulate. Its design is shown in Figure 18.b has a tapered truncated conical shape to offer a wider base of 70 mm diameter to apply the normal stress to the particulate sample. The lid was designed with radially arranged fins on the lower face. Two types of fins are used: 3 fins of 6(t)x10(h)x25(w) mm are alternated at an angle of 60° with 3 shorter fins of 6(t)x10(h)x15(w) mm. The thickness of 6 mm ensures sufficient rigidity during the tests at high normal and tangential stresses and temperatures.

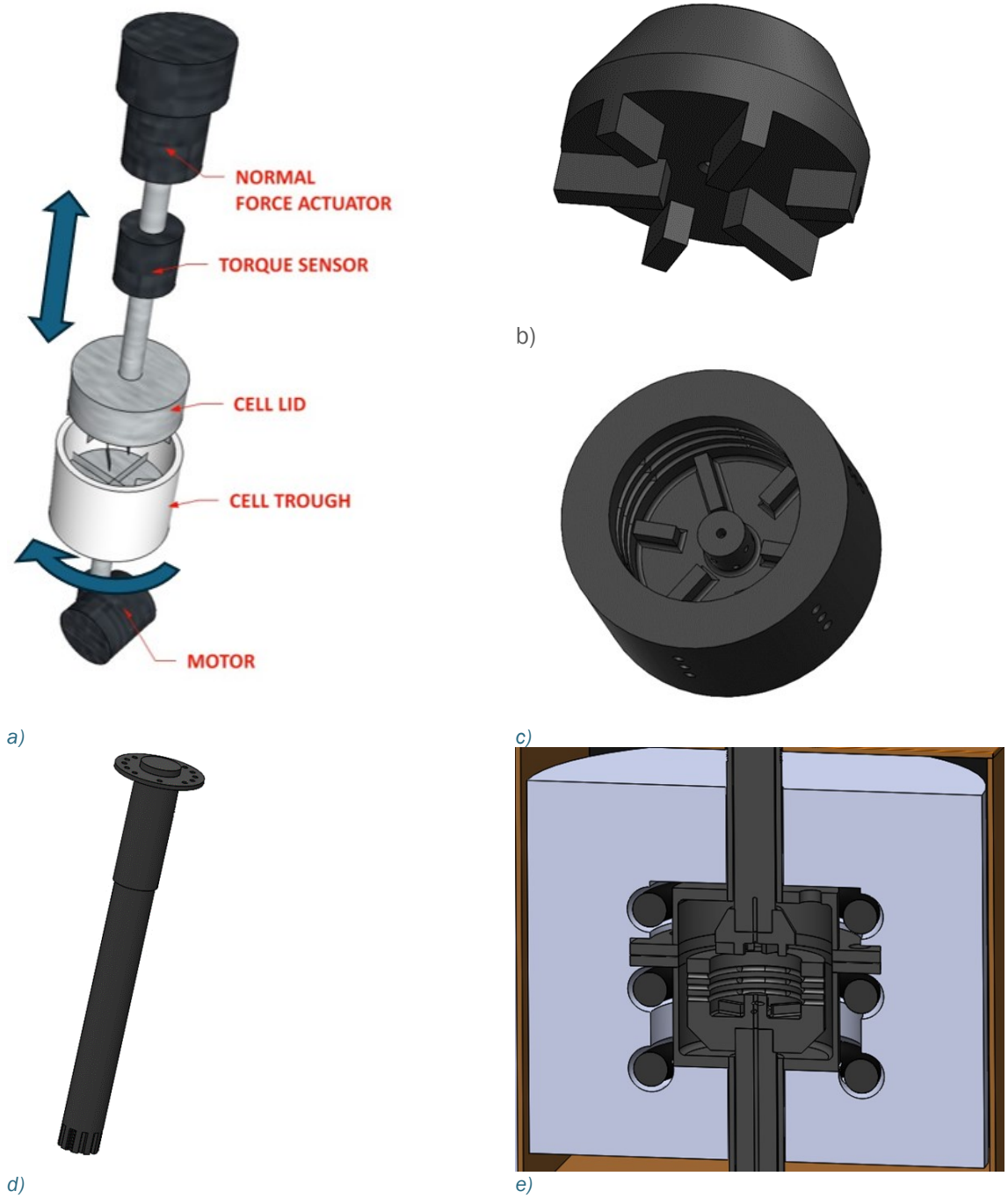


Figure 18. Details of the designed set-up0: a) functional scheme; b) lid; c) trough; d) upper rod; e) measuring chamber in the oven.

The trough of the shear cell is made of AISI 310, represented in Figure 18.c. Its function is to provide a cylindrical space to host the sample and to avoid wall shear of the sample during the experiment. It is a cylindrical cup with an inner diameter of 80 mm, an outer diameter of 120 mm and a cavity height of 40 mm. It is connected to a vertical rod to the motor that forces its rotation. The distributor placed along the cell axis is designed as a cylindrical protrusion to allow the inlet of the gas reagents into the particulate sample. The distributor is provided with a series of pores to spread the gas uniformly within the sample. A system of circular grooves and holes on the cylindrical wall promotes a uniform gas exit from the sample and a uniform distribution among the sample particles. A series of fins, with similar geometry to those on the lid, is located inside the trough to enhance the grip of

pellets on this surface and avoid wall shear. The trough is connected to a rod with a groove at its bottom to hold it in place and to a motor that provides the torque needed to keep the particle moving with an angular speed of 0.016 rpm. This angular speed, similar to the one reported in the Schulze manual, was chosen to simplify the operator's output analysis.

The two rods connect the lid to the vertical actuator, the torque sensor, and the trough to the rotating motor. One of these rods is represented in Figure 19.d. Their cross section was designed to hold compressive forces and avoid buckling phenomena. On the rod tips connected to the chamber, grooves prevent the relative rotation of the lid and the trough with the rods. Flanges ensure attachment with components on the opposite side of the cell. The bottom rod has an internal channel to feed the gas to the trough.

A special oven composed of heating gloves and insulation in an aluminium silicate wall is required to reach high temperatures. An AISI310 stainless steel chamber is used to confine the hydrogen atmosphere to the cell to avoid the interaction between hydrogen and the oven and better control its composition. Figure 19.e represents the oven with the cell in section. The oven opens vertically to access the cell. The chamber is composed of two hollow cylinders linked by two flanges. This connection must be opened before and after each test to fill and empty it. During the test, the chamber prevents oxygen ingress and hydrogen leakage. The gas flows from the channel inside the lower rod, then it passes through the distributor located in the center of the trough; it moves radially in the sample, crosses the trough wall by the purposely designed groves and holes, and finally exits the system through the hole located at the top of the chamber that is connected to a discharging tube not represented in the figure.

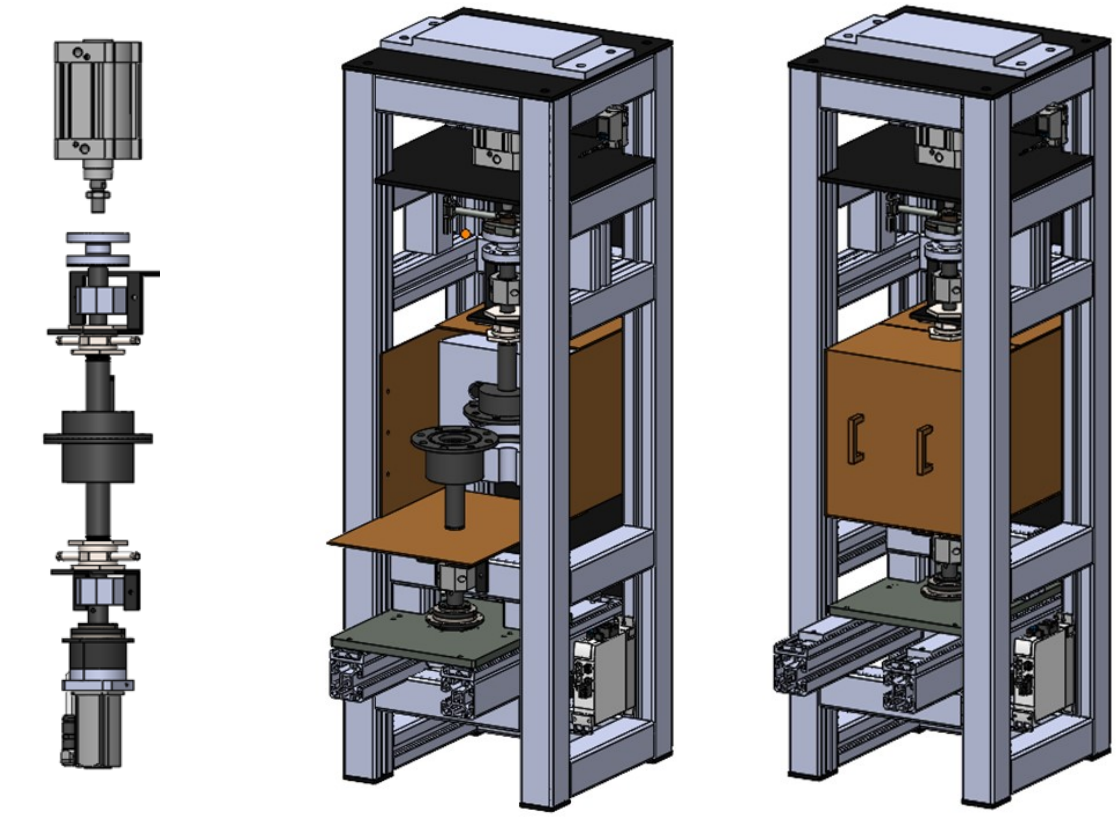
The core of the device operates at very high temperatures, and the heat fed at the center can be dissipated by conduction in the rods. Consequently, it may reach the sensors and the engine located far from the oven. To avoid overheating these parts, a cooling system is designed to cool down the rods. It is fixed at the end of the chamber. Analyses were conducted to estimate the temperature profile of the rod.

Different actuators and sensors were selected on the market. An ISO DSBC-125-100-PPVA-N3 pneumatic piston is used to apply the normal stress in the shear testing procedure. This device can reach the maximum required normal load of 5000 N using the lab compressed air. A proportional valve is used to modulates the pressure and, consequently, the pushing force. Using air instead of a liquid allows easier control of the piston force in case of vertical movements of the lid due to the expansion/contraction of the particulate system. A DC motor coupled with a Neugart flanged planetary gearbox ensures up to 70 Nm torque at 0.015 rpm.

A TRX series flanged torsiometer interposed between the linear actuator and the upper rod can measure torque in the range of ±100 Nm. The upper flange of the sensor must be prevented from rotating to measure torque, while the other is used to be connected to the rod attached to the lid. A system of branches connected to two linear guides is used for this purpose. The vertical position of the lid is detected using a position sensor coupled with the fixed surface of the torsiometer on one side, and the frame on the other. The temperature inside the chamber is measured using Rhodium - Platinum S thermocouple coated in TiN (nitrile nitride). Two flowmeters are connected to the chamber to introduce the gas needed for the test and regulate the composition.

The device frame was designed using Bosch profiles with a square cross-section of about 95 mm side.

The frame is also equipped with a runner (see Figure 19) that facilitates the loading and unloading of the material in the cell. Figure 19.b shows the opening phase of the device for particulate loading. Once the device is opened, the material can be placed inside the trough, and once it is closed, as in Figure 19.c, the system can be brought up to temperature to conduct the shear testing.



a) b) c)

Figure 19. The sequence of elements needed to conduct tests; b) the opened device for cell loading; c) the device in operation.

4. Future work

Two important metallurgical tests will be performed on both BF and DR grade pellets under high hydrogen conditions ranging from 55% up to 100% (see Table 12). Direct Reduced Simulator (DRSIM) will be used to perform isothermal and non-isothermal reduction (see Figure 20). In the proposed tests (Test 1, 2 and 3), iso-thermal conditions with fixed gas compositions will be maintained. At the end of the test, once desired Oxygen to Iron (O/Fe) molar ratio of 1.33 will be achieved, the experiment will be stopped. Reduced sample will be tumbled in mini-ASTM to evaluate degradation behavior of the pellets. in the proposed tests (Test-4 & 5), both gas and temperature profile will be changed during the reduction. DRSIM operating line will be derived from thermodynamic and kinetic model (reductor model) from UL. DRSIM test will be interrupted at different temperature (760, 800, 860 °C) corresponding to DRI based ISO test for reduction degradation, reducibility index and clustering and sticking behavior respectively (see Table 12). After completion of each test, mechanical degradation behavior, reduction potential and sticking tendency of pellets (both BF and DR grade) post tumbling in mini-ASTM will be analyzed.

Table 12. High hydrogen metallurgical test conditions for isothermal and non-isothermal tests.

S. N	Test	Purpose	Test conditions	Suitable
1	Test -1 Close to Midrex (Iso thermal test)	Degradation behavior of pellets under high hydrogen conditions are investigated.	Temperature: 800 °C Gas composition: 55 % H2, 45 % CO Gas flow: 18 L/min End of test: till mass loss stabilizes	DRI
2	Test -2 Close to HYL (Iso thermal test)	Degradation behavior of pellets under high hydrogen conditions are investigated.	Temperature: 800 °C Gas composition: 80 % H2, 20 % CO Gas flow: 12.5 L/min End of test: till mass loss stabilizes	DRI
3	Test-3 Close to 100% H2 based DRI. (Iso thermal test)	Degradation behavior of pellets under high hydrogen conditions are investigated.	Temperature: 800 °C Gas composition: 100% H2 Gas flow: 10 L/min End of test: till mass loss stabilizes	DRI
4	Test-4 DR-SIM test (interrupted)	Degradation behavior of pellets under high hydrogen conditions are investigated.	Temperature: Changing with time Gas composition: Changing with time 55% H2 – 45% CO Gas flow: 18 L/min Interrupted at 760, 800 & 850 °C	DRI
5	Test-5 DR-SIM test (interrupted)	Degradation behavior of pellets under high hydrogen conditions are investigated.	Temperature: Changing with time Gas composition: Changing with time 80% H2 – 20% CO Gas flow: 12.5 L/min Interrupted at 760, 800 & 850 °C	DRI

Figure 20. Q-HOSIM/BRASS Test equipment at IJmuiden.

5. Conclusions

The overall conclusions of the work carried so far are described below:

- A wide variation in chemistry was observed between BF and DR grade pellets w.r.t iron percentage , gangue content, basicity and magnesium oxide content. DR grade pellets were found to have lower gangue percentage (3.3-4.1%) and higher iron content (66.4-67.1%).
- The average porosity of both BF and DR grade pellets were 28.9% and 29.6% respectively. Comparatively, bigger pellets (13-15 mm) were more porous than smaller pellets (10-13 mm). The difference between bigger and smaller pellets is 0.7%.
- The envelop density (i.e., bulk density) of all pellets were measured in the standard GeoPyc 1360 device. Bulk density of both BF and DR grade pellets were varied between 3.50-3.73 g/cm^3.
- The true or absolute density of all pellets were measured in the standard AccuPyc II 1340. The average true density of DR grade pellets (~5.16 g/cm^3) is higher than BF grade pellets (~ 4.97 g/cm^3). It can be easily explained by higher iron content in DR grade pellets.
- The Tumbler Index (TI) of both BF and DR grade pellets were varied between 96.6-98.1%. The Abrasion Index (AI) of all pellets were varied between 1.8-3.2%. There is a marginal difference observed between BF and DR grade pellets w.r.t to their TI and AI. Bigger size pellets (13-15mm) generated 0.5% higher fines compared to their smaller size pellets of 10-13 mm.
- The CCS (ISO) of both BF and DR grade pellets were varied between 240-280 daN. Among all pellets, DR grade pellet C1 (lime coated) has lowest CCS up to 241 daN with weak pellet fraction (<120 daN) as high as 10.5%.
- The swelling index of both BF and DR grade pellets were analyzed in BF condition. Results confirms that BF grade pellet B (B1, B2) has very high swelling index up to 24%. Whereas other pellets (A, C and D) show relatively low swelling index ~13%.
- The Q-HOSIM test was performed on both BF and DR grade pellets and the results confirmed that very high degree of degradation was observed for BF grade pellet B (B1, B2) with and without tumbling.
- UNISA has designed an experimental test rig to conduct the shear testing. The test rig is under construction and should be operatable by the end of May 2024.

- In future work, all these pellets will be reduced in DRSIM reactor under high hydrogen conditions. After partial and complete reductions, physical, chemical and metallurgical aspects of pellets will be investigated. For example, metallisation, degradation and swelling behavior of pellets will analysed in Tata Steel.

6. References

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[3] ISO-3271, 2007, Iron ores for blast furnace and direct reduction feedstocks – Determination of the tumble and abrasion indices, (2007).

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7. Acronyms and Abbreviations

AI	Abrasion Index
ASTM	American Society for Testing and Materials
BF	Blast Furnace
CCS	Cold Compression Strength
DEM	Discrete Element Method
DR	Direct Reduced
DRSIM	Direct Reduced Simulator
DRI	Direct Reduced Iron
DRP	Direct Reduction Plant
DI	Decrepitation Index
EU	European Union
ISO	International Standardization for Organisation
LTD	Low Temperature Degradation
O/Fe	Molar ratio of Oxygen to Iron
Q-HOSIM	Quick Hoogoven Simulation
RDI	Reduction Degradation Index
RI	Reducibility Index
RUL	Reduction Under Load
TS	Tata Steel
TI	Tumbler Index