## METANHAURCHCANLINJECTION THECHNOLOGY STEATHSTHCANLANAVAYSIS OF JEXTDERIMENTES

WITH EVIEWS

DR-DR- KASIMASKER HASAN

## **Metallurgical Injection Technology**

Statistical Analysis of Experiments with EViews System to Increase the Cost-Effectiveness of Pig Iron Desulturization Plant

Editorial La Espada Rota

Metallurgical Injection Technology- Statistical Analysis of Experiments with EViews System to Increase the Cost-Effectiveness of Pig Iron Desulfurization Plant ©Dr. Dr. Kasim Asker Hasan ©Editorial La Espada Rota

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#### Thanks

Mainly my wife Suad and my children. Thanks for the support supreme and patience for all these years.

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Key words: Metallurgical injection technology, Pig iron desulfurization, Statistic Analyse System, Desulfurization Cost

#### **INTRODUTION**

Two phases in contact with each other share a common surface, the so-called phase boundary interface the bringing together of two phases at a common phase boundary interface is often done with the intention of exchanging mass, heat and/or momentum between the phases. The duration of these exchange processes is important for the technical feasibility of the process and its economic success; it is inversely proportional to the size of the phase interface. For this reason, most of the processes aim for a maximum of the phase interface/volume ratio in order to accelerate the desired exchange process. Depending on the physical state of the phases involved, there are various methods of increasing the contact areas. The method generally used in liquid /gas systems to enlarge the reaction area is the fine distribution (dispersion) of the gas phase in the liquid phase.

This method of dispersing the gas phase into the liquid phase by means of immersed lances, nozzles, perforated plates or sieve stones is very effective despite its simple apparatus design and operation due to the large phase interface/phase volume ratio.

As the interest in such processes grew, so did the desire for more detailed knowledge of these exchanges on the individual parameters that influence the profitability of such processes. The movement of the phases is at least as important as the chemical reactions and therefore the dynamics of the gas phase in metallurgical dispersion systems, especially the lack of precise knowledge about the formation, separation, rise, expansion and deformation of the gas bubbles with simultaneous mass transfer at high temperatures and under vacuum, must be investigated more precisely. The typical example<sup>1</sup> represent the processes of interaction in the system metal – slag in course the pig iron desulphurisation, in course of refining in the refining reactor, in course of the ladle metallurgy. Some of them can be recorded in form of the following equations: /Me/ + / S/ = (MeS) ... (1) (CaO) + /S/ = (CaS) + /O/ ...(2) where (CaO), (CaS) and (MeS) are the components dissolved in slag, and / S/ and /O/ are components of metal. It is possible to express the thermodynamic sulphur partition coefficient applying the equilibrium constant of the considered reactions. In case of the steel slag the value of the sulphur partition coefficient usually rises with the increased activity of CaO, which represents the exact measure of the slag basicity, and with the decreasing oxygen activity.

The increasing requirements for the production of lowand ultralow-sulfur steel grades have been highlighted<sup>2</sup>. The reasons for increased sulfur input to the Blast Furnace (BF) have also been given. Extension of the desulfurization work in the the Blast Furnace (BF) does not seem to be an acceptable method, with regard to performance and productivity losses of the BF. The concept of "alkali-oriented Blast Furnace (BF) slag operation" has been introduced and discussed. As a consequence of this BF operation, elevated Hot Metal (HM) sulfur contents are unavoidable and the efforts for sulfur removal in the steelmaking shop have to be enhanced. But the softening of the sulfur limitations of the blast furnace is opening a huge potential for cost savings. Brief descriptions and discussions of the different metallurgical facilities in the Basic Oxygen Furnace (BOF) shop have been given with respect to their capability to remove sulfur. The combined utilization of BF, HMDS, BOF and LF as desulfurization facilities, with suitable and coordinated desulfurization degrees of all units, is the key to managing high sulfur contents of the HM successfully.

Besides the economical balance between savings at the BF and rising expenses in the BOF shop, the "alkali- oriented BF slag operation" seems to be especially interesting for all plants suffering from HM shortage or that want to increase their HM output without adding new melting facilities.

Since technical measurements and evaluations are not feasible in practice, a model and a Statistical Analysis System<sup>3</sup> is developed and verified which describes the entire behaviour of the gas phase during the dispersion process as accurately as possible, at least theoretically. In the practical part of this study are an analysis of the own cold experiment in laboratory of Institue of Metallurgy - RWTH Aachen Germany and own industrial data (hot experimente) for injection of calcium carbide (CAC2) and Magnisum. The Eviews program is used to nalayse the data of experimentes. It is possible to predict the behavior of the variables involved in the process, resulting in savings of time and money.

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<sup>&</sup>lt;sup>3</sup> Kasim Asker Hasan, Diploma thesis 1991, University of Applied Sciences and Arts. Dortmund- Germany.

## THEORETICAL CONTEXT

#### Chapter 1. Regression analysis with the Eview system

The command Estimate Equation sur is available for performing a multilinear regression analysis, in which the linear relationship between an interval-scaled dependent characteristic and one or more interval-scaled (or binary independent characteristics) is to be examined retrieve the quality of a linear fit and the decision criteria as to whether the assumption of linearity is justified at all and is transferable to the population represented by the present sample.

#### Description of linear relationship and adjustment criteria.

Provided that there is a linear relationship between a variable marked as dependent and one or more variables marked as independent, a regression analysis can be retrieved with the following REGRESSION command:

Proc reg; Model  $\gamma = \chi 1, \chi 2, \chi 3 \dots$  etc: run;

As a result, the regression coefficients  $\beta 0$  (regression constant),  $\beta 1$ , ... bn are output, for which the regression coefficient defined by the regression relationship

 $\gamma' = \beta 0 + \beta 1 * \chi 1 + ... Bn * xn$  determined Y' value

the best prediction for the dependent variable Y by representing that the value of the adjustment criterion<sup>4,5</sup>, sum ( $\gamma'$ - $\gamma$ ) is minimal among all possible coefficient values.

The hypotheses have been tested using multilinear regression addressing the issues of ordinary least square assumptions.

$$\gamma = \alpha + \beta 1 \chi 1 + \beta 2 \chi 2 + \beta 3 \chi 3 + \varepsilon$$

Where  $\gamma$  the dependent variable gas bubble volume is,  $\alpha$  is the intercept of  $\gamma$  .

 $\beta$ 1,  $\beta$ 2 and  $\beta$ 3 are the slope coefficients and  $\chi$ 1,  $\chi$ 2 and  $\chi$ 3 are the independents variables, physical parameters such as kinetic viscosity, density and surface tension.

The error term is denoted as  $\in$ .

The result of the regression analysis is presented in Appendices 2, 3 and 4.

<sup>&</sup>lt;sup>4</sup> Hartung, E.; curso básico de estadística. Munich 1987.p.159.

<sup>&</sup>lt;sup>5</sup> Dürr, W.; Mayer, H.; Probability calculation and shooting statistics. Carl Hanser Publisher Munich Vienna 1987.

#### Chapter 2. Sources of Sulfur in Iron and Steelmaking<sup>6</sup>

Sulfur in liquid hot metal (HM) is included in burden materials like limestone and ore, and primary fuel such as coke, oil and pulverized coal used in the blast furnace (BF) for ironmaking (Table 1).<sup>7</sup> Coke and oil are by far the largest sources. Coke and coal contain approximately 0.8–1.2% sulfur, depending on the mine source. Oil and heavy oil contain 1.3–2.0%. This adds up to a sum of 95% of the total input. Due to the reducing atmosphere in the ironmaking process in the BF, more than 80% of the total input is removed through the slag, so that there is only a balance of approximately 12%, which remains in the liquid hot metal. In the balance in Table 1, this equals to a hot metal sulfur content of 0.055%.

S input (	4.40 kg	output %		
Sinter	5.1	6.2 (sum of sinter,	Slag	82.4
Pellets	1.0	pellets	Hot metal	12.6
Additives	0.1	and additives)	BF top gas	3.6
Coke	60.2		BF dust	0.7
Oil	33.6		BF sludge	0.5
Total	100		Casthouse dust	0.2
			Total	100

Table 1: Sulfur Balance of a German Blast Furnace

<sup>6</sup> Buğra Şener steel plant manager, İsdemir, Iskenderun Demir Ve Celik A.S., Iskenderun, Turkey bsener@isdemir.com.tr

<sup>&</sup>lt;sup>7</sup> Altland, R.; Beckmann, B.; and Stricker, K-P., "Verfahrensoptimierung am Hochofen durch kontrollierte Alkaliund Schlackenbedingungen" ("Process optimization of the blast furnace by controlled alkali and slag conditions"), Stahl und Eisen, Vol. 119, No. 11, 1999.

#### Chapter 3. Sulfur in in Iron and steel<sup>8</sup>

Sulfur (S) may dissolve in liquid iron (Fe) at any concentration. However solubility of sulfur in solid iron is limited: 0.002% in  $\alpha$ -iron at room temperature and 0.013% in  $\gamma$ -iron at 1000°C.

When a liquid steel cools down and solidifies the solubility of sulfur drops and it is liberated from the solution in form of iron sulfide (**FeS**) forming an eutectic with the surrounding iron. The eutectic is segregated at the iron grain boundaries. The eutectic temperature is relatively low - about 988°C.

Fe-FeS eutectic weakens the bonding between the grains and causes sharp drop of the steel properties (brittleness) at the temperatures of hot deformation (Rolling, Forging etc.).

Brittleness of steel at hot metal forming operations due to the presence of low-melting iron sulfides segregated at grain boundaries is called **hot shortness**.

In order to prevent formation of low-melting iron sulfide manganese (**Mn**) is added to steel to a content not less than 0.2%.

Manganese actively reacts with iron sulfides during solidification of steel transforming FeS to MnS according to the reaction:

#### (FeS) + [Mn] = (MnS) + Fe

(square brackets [] - signify concentration in steel, round brackets () signify concentration in slag)

The melting temperature of manganese sulfide is relatively high - about 1610°C therefore the steels containing manganese may be deformed in hot state (no hot shortness). Unfortunately MnS inclusions are:

- Brittle (less ductile than steel);
- They may have sharp edges;
- They are located between the steel grains.

All these factors determine negative influence of sulfide inclusions on the mechanical properties. Cracks may be initiated at brittle sharp edge inclusions. Sulfide inclusions especially arranged in a chain form also make easier the cracks propagation along the grain boundaries.

The negative effect of sulfur on the steel properties becomes more significant in large ingots and castings, some zones of which are enriched by sulfur (macrosegregation of sulfur).

#### The properties negatively affected by sulphur:

- Ductility;

- Impact toughness;

- corrosion resistance;
- Weldability.

<sup>&</sup>lt;sup>8</sup> Dr. Dmitri Kopeliovich, Director of Research & Development of King Engine Bearings. http://substech.com/dokuwiki/doku.php?id=dmitri\_ kopeliovich

Transition of sulfur from steel to slag may be presented by the chemical equation:

[S] + (CaO) = (CaS) + [O]

The equilibrium constant K<sub>s1</sub> of the reaction is:

$$K_{S1} = a_{[O]}^{*} a_{(CaS)} / a_{[S]}^{*} a_{(CaO)}$$

Where:

 $a_{[O]}^{}$ ,  $a_{[s]}^{}$  - activities of oxygen and sulfur in the liquid steel;  $a_{(CaS)}^{}$ ,  $a_{(CaO)}^{}$  - activities of CaS and CaO in the slag.

The same reaction in ionic form:

 $[S] + (O^{2-}) = (S^{2-}) + [O]$ 

The equilibrium constant  $K_{s_2}$  of the reaction is:

$$\begin{split} \mathbf{K}_{\text{S2}} &= \mathbf{a}_{[\text{O}]}^{*} \mathbf{a}_{(\text{S2-})} / \mathbf{a}_{[\text{S}]}^{*} \mathbf{a}_{(\text{O2-})} \\ \text{Where:} \\ \mathbf{a}_{(\text{S2-})}, \, \mathbf{a}_{(\text{O2-})} - \text{activities of } \text{S}^{2-} \text{ and } \text{O}^{2-} \text{ in the slag.} \end{split}$$

Capability of a slag to remove sulfur from steel is characterized by the **distribution coefficient of sulfur**:

 $L_{s} = (S)/[S]$ 

Where:

(S) - concentration of sulfur in slag; [S] - concentration of sulfur in steel;

As appears from the above equations desulfurization is effective in deoxidized (low (O)) basic (high (CaO)) slags. Therefore ability of Basic Oxygen Process (BOP) to remove sulfur is low due to its highly oxidized slag.

Desulfurization may be effectively conducted in the reducing slag stage of the steel making process in Electric-arc furnace. At this stage the oxidizing slag is removed and then lime flux is added to form basic slag with high CaO content.

Deep desulfurization by slags may be achieved in ladle:

- The refining (desulfurizing) slag with high content of CaO and no FeO is prepared and placed in an empty ladle.

- The molten steel is poured into the ladle filled with the refining slag.

- Energy of the falling steel stream causes mixing the slag with the steel, during which sulfur is removed from the

steel to slag phase.

Effect of desulfurization may be enhanced by additional stirring, for which electromagnetic (induction) stirrers or argon bubbling are used.

#### Chapter 4. Injection system and lances

The design of the injection system is also important, pressure vessels with a steep sided cone, fitted with a fluidization cone or nozzles at the bottom to assist with an even continuous flow of reagent into the nitrogen carrier gas stream. In treatment ladles normally a refractory coated lance is lowered vertically into the molten iron to within 500 mm of the ladle bottom to maximize the injection depth. Co-injection systems are available for injecting more than one reagent simultaneously into the hot metal. Different lances are available original straight bottom discharge, T- lance with the discharge from 2 holes on the side of the lance parallel to the ladle bottom, 4 hole lance and dual pipe lances have all been tried. Their use is dependent on the individual customer's system. Lance life is mostly a function of heat stresses and cooling, and shorter injection times increases lance life. Improved refractory designs with lower conductivity, increasing the thickness and addition of stainless steel fibres to the refractory covering on the lance have extended lance life. Cleaning of the lance outlets, especially on multi port lances, improves lance life by reducing clogging.<sup>9,10</sup>

<sup>&</sup>lt;sup>9</sup> SALINAS, A. DS-Lances for hot metal desulphurization, State of the art and new developments. HTMK, Almamet and Polysius. The VIII. International Symposium for Desulphurization of Hot Metal and Steel, September 20–24, 2004, Nizhny, Russia, p. 33.

<sup>&</sup>lt;sup>10</sup> CHANDRA, S., PATAK, S., MATHEW, K.S., KUMAR, R., MISHRA, A., SEN, S., and MUKHERJEE, T. Improvements in External Desulphurization of Hot Metal at Tata Steel, Jameshepur India. Almamet GmbH: The VII. International Symposium for Desulphurization of Hot Metal and Steel, September 26–27, 2002 in Anif/ Austria, p. 34.

#### Chapter 5. Desulphization of Steel and Pig Iron <sup>10,11</sup>

Metallurgical slag qualities must be defined by the whole complex of physico - chemical characteristics, such as an oxidative ability, optical basicity, sulphide capacity up to slag fluidity, its surface tension etc. The understanding of regulation of basic physico - chemical qualities of molten metals and slag depending on a chemical structure and a temperature has its importance at the level of the metallurgical process control. Slag design (Thermodynamic Calculation) The post-desulphurisation slag is mainly from the following sources: (1) The blast furnace slag carryover with major components of CaO, SiO2, Al2O3 and MgO. A normalised blast furnace slag composition (wt%) is 42.29%CaO, 36.78%SiO2, 12.46%Al2O3 and 7.63%MgO. (2) The injected CaO/Mg and their reaction products.

The amount of CaO/Mg injected is decided by the initial sulphur content in the hot metal and the sulphur specification of the steel grades to be made. (3) The residual from previous heat(s) in the hot metal ladle which can be effectively reduced by clean ladle practice. The formation of MgO and the injected CaO increase the slag basicity (%CaO/%SiO2) and MgO content, and consequently increase the melting temperature and viscosity of the post-desulphurisation slag. Therefore, without modification the post-desulphurisation slag in the hot metal ladle can be considered as a quaternary CaO-SiO2-Al2O3-MgO slag with higher basicity and MgO content compared to the blast furnace slag, which results in the increase in the melting temperature and viscosity.

#### Important issues in desulphurization of hot metal <sup>12</sup>

- During the desulphurizing process, the generation of slag is proportional to the amount of reagent added to the hot metal. Also during the process, some hot metal gets trapped in the slag and gets pulled out of transfer ladle during the slag rimming. This amount is around 1 % for the co-injection process. Desulphurization slag contains about 50 % iron.
- The loss of heat during the desulphurizing process is an important factor since it reduces the sensible heat of the hot metal sent to the converters. The three primary sources of heat loss are radiation from the surface of the hot metal, addition of cold reagents and introduction of cold injection lances into the hot metal. The largest temperature loss occurs during injection rather than skimming. A temperature loss of 30 deg °C is expected during the desulphurization process.
- Desulphurizing process donot have any major effect on the refractory lining life of the hot metal ladle since the treatment time is small.
- Both reagent injection and slag skimming operation generate fumes which are to be collected and dedusted prior to their release to the environment. The captured fumes are typically cleaned in a pulse jet type bag house designed for metallurgical operations.

<sup>&</sup>lt;sup>10</sup> Zushu Li, Mick Bugdol1 and Wim Crama, Tata Steel RD&T, Swinden Technology Centre Rotherham, United Kingdom

<sup>&</sup>lt;sup>11</sup> Tata Steel RD&T, Swinden Technology Centre, Rotherham, IJmuiden, the Netherlands

<sup>&</sup>lt;sup>12</sup> J. Kijac, Faculty of Metallurgy, Technical University of Kosice, Slovak Republic. https://www.ispatguru.com/desulphurization-of-hot-metal/

#### Chapter 6. Theoretical aspects of slag emulsification

Liquid steel desulphurization by slag-metal reaction is an exchange reaction between two non-miscible phases, thermodynamically governed by the sulphur partition ratio between the two phases, and kinetically governed by the inter phase exchange area and sulphur transfer driving force. The kinetic aspects can be suitably influenced by fluid dynamics effects. Since proper bath stirring induced by gas injection affects the relative velocity at the interface of metal and slag, it, in turn, affects the solute mass transport coefficient. High interface velocities can even cause slag emulsification, resulting in a great increase of exchange surface area. Moreover, efficient metal mixing in the liquid steel bath makes faster the attainment of the desired final sulphur level in the liquid steel, resulting in a decrease of the duration of desulphurization operation. Knowledge is hence needed of in-ladle fluid-dynamics induced by gas injection, in order to reach the best conditions in terms of suitable flow field namely (i) at the metal-slag interface, to favour emulsification and, in turn, acceleration of the chemical kinetics, and (ii) in the ladle bath, in order to allow the mixing of just desulphurized steel at the interface with liquid steel in the ladle bulk up to reaching perfect mixing throughout all the ladle at the target sulphur level. On the other hand, too intense stirring actions which are related to high operational costs and bath temperature losses, are to be avoided. Mechanical energy is transferred to the bath by means of (i) bottom blowing, from one or more porous plugs, (ii) top blowing, through lance, or (iii) combined blowing. Several investigations have been performed on the effects of gas stirring on in-ladle fluid-dynamics, focused on the gas path or on slagmetal interface processes, involving desulphurization reaction as well. The most relevant aspects arising from these investigations are the following <sup>13</sup>. In several steelmaking processes in which bath stirring is concerned, mixing times depend on the power transferred to the bath in the ladle at the power of 0.3 - 0.4.

- The mono-plug bottom blowing stirring which ensure the shortest mixing times for a fixed bath and a fixed gas flow rate supply is achieved with plug eccentrical with respect to the ladle. A position between quarter and half ladle radius is generally desired.
- Multiple porous plugs stirring in the ladle is to be set carefully in order to have the relative velocity at the interface slag-metal to favour emulsification. Asymmetrical plug positions proved to be of maximum efficiency in reducing mixing times. With symmetrical plug positions, flow recirculations are induced in the ladle with zones which have counteracting flows destroying their stirring effects. Lance blowing is beneficial for emulsification, whereas bottom stirring is beneficial for ladle mixing. A suitable combined blowing merges the two desired effects.
- Studies carried out on the effect of the slag properties on the emulsification phenomenon show that there are critical conditions which are required to be met for steel velocity at the interface with the slag and flow rate of gas blown from the plugs to allow emulsification onset. These relationships take into account slag physical properties such as viscosity and density.

- Among the parameters used to define improved conditions for mixing in the ladle and mass transfer at the slag metal interface, of great importance are the ratio between ladle diameter (D) and the bath height (H). Normally, the ratio D/H is not far from 1.
- Data available in literature on the effect of gas flow rate injection on desulphurization rate show that the most interesting aspect is that an onset gas flow rate is to be found for enhancing significantly desulphurization rate.

<sup>&</sup>lt;sup>13</sup> Satyendra, July 30, 2016... https://www.ispatguru.com/desulphurization-ofliquid-steel/

#### Chapter 7. Desulfurization Cost

The desulfurization costs were calculated many times for single operations. Figure 1 shows an example for hot metal De-S. Generally, the cost increases with increasing the hot metal initial sulfur SI. This is basically caused by the consumption of De-S agents, but all other parameters like maintenance, lances and nitrogen increase as well. It can be estimated that temperature losses, slag (%Fe) losses and slag processing costs increase as well.

Only the skimming losses for iron remain constant in operations. They are more related to the efficiency of the deslagging process than to the sulfur level in the hot metal.

The total cost for hot metal desulfurization from the blast furnace level of 0.060% down to 140 ppm are at almost \$US6.00/tHM with 50% being the De-S agents, 25% being the skimming losses and 25% other cost factors. A cost addition of approximately \$US2.50/tHM for labor; services; selling, general and administrative expenses; and capital cost has to be taken into account on top of that. A benchmark comparison of blast furnace, hot metal and steel desulfurization is given in Table 2 for a sulfur decrease of 100 ppm in the hot metal or steel.



Figure 1 Hot metal desulfurization operation cost structure. Source: Iron & Steel Technology , A Publication of the Association for Iron & Steel Technology page 6, AIST.org April 2013

F	3last furnace	Hot metal desulfurization	Steel desulfurization	
Parameter	(US\$/t)	(US\$/t)	(US\$/t)	
Fluxes	0.48			
Fuel	1.01	_	_	
Granulated BF	slag –0.05	_	_	
De-S agents		0.48		
Consumables		0.07		
Maintenance		0.07		
Transport	_	0.066	_	
Slag and yield	losses —	0.129		
Argon stirring	_	_	0.48	
Aluminum cor	nsumption —	_	0.12	
Total	1.44	0.82	0.60	

#### Table 2 : Desulfurization Cost Benchmark for 100 ppm of Sulfur

Source: Iron & Steel Technology, A Publication of the Association for Iron & Steel Technology page 6, AIST.org April 2013

# Chapter 8. Modeling and Simulation of Hot Metal Desulfurization by Powder Injection<sup>14</sup>

The equation of continuity in cylindrical coordinates<sup>15</sup> can be employed to mathematically represent the desulfurization process:

```
\begin{aligned} ds:dt+vr^* ds:dr+v\theta^* ds:r^* d\theta+vz * ds:dz &= DS [1/r * d:dr(r^* ds:dr) \\ &+ 1:r^{2*} d^2s:d\theta^2+d^2s:dz^2] + rs & \dots 1 \end{aligned}
```

where S is the sulfur concentration in hot metal, t is time, and r,  $\theta$  and z are the radial, the angular and the axial components of the cylindrical coordinates, respectively; vr, v $\theta$  and vz are the components of the velocity vector in the r,  $\theta$  and z directions. Besides, rS is the rate of disappearance of S by chemical reaction and DS is the diffusion coefficient of sulfur in the hot metal. Given that the bubbles of the carrier gas are mainly moving in the axial direction, one can assume that the desulfurization phenomenon is predominant in that direction; then Equation (1) is reduced to

```
ds:dt+vz^*ds:dz=Ds^*d^2s:dz^2+rs \qquad \dots 2
```

Furthermore, considering that: 1) the diffusion of sulfur is small compared to its convective transport and 2) the vessel is a well stirred tank with small concentration gradients, then Equation (2) is simplified to an ordinary differential equation:

ds:
$$dt = rs$$
 ... 3

The desulfurization reactions are first order chemical reactions whose kinetics can be expressed as follows [16]:

$$rs = -k (S-Se) \dots 4$$

where k is the reaction rate constant and Se is the value of the sulfur concentration at thermodynamic equilibrium. The reaction rate constant is a temperature-dependent term which is well represented by Arrhenius's law<sup>16</sup>:

$$k(T) = k0^* e^{-Ea/(RT)}$$
 ... 5

where k0 is the pre-exponential constant, Ea is the reaction activation energy, R is the universal gas constant and T is the absolute temperature. Substituting Equation (4) into Equation (3) and integrating one obtains

$$S(t) = Se + (So-Se) e^{-kt} \qquad \dots 6$$

where S0 is the initial concentration of S

<sup>&</sup>lt;sup>14</sup>Miguel A. Barron, Isaias Hilerio, Dulce Y. Medina Departmento de Materiales, Universidad Autonoma Metropolitana Azcapozalco, Mexico City, Mexico Email: bmma@correo.azc.uam.mx, dyolotzin@correo.azc.uam. mx, ihc@correo.azc.uam.mx

<sup>&</sup>lt;sup>15</sup> Bird, R., Stewart, W.E. and Lightfoot, E.N. (2002) Transport Phenomena. 2nd Edition, John Wiley and Sons, NewYork.

<sup>&</sup>lt;sup>16</sup> Oeters, F. (1989) Metallurgy of Steelmaking. Springer-Verlag, Berlin.

#### **REVIEW OF LITERATURE**

Considerable efforts have been made during the past four decades to investigate gas injection operations in steelmaking ladles. Towards these, numerous physical and mathematical model studies embodying aqueous as well as full scale systems have been reported. On the basis of an extensive literature search, a summary, discussion and analysis of these are now presented. For the sake of convenience and clarity of presentation, studies have been categorised into three major groups<sup>17,18</sup>: (1) physical modelling studies, (2) combined physical and mathematical modelling studies and (3) mathematical modelling studies. In each of these categories, a great number of publications on various phenomena, such as gas-liquid interactions, turbulent fluid flow, mixing, solid-liquid mass transfer, etc. have been reported. Accordingly, and as discussed in the text, considerable improvements have resulted in our understanding of the various gas injection induced phenomena in ladle metallurgy operations. Coupled with these, extensive mathematical modelling studies have also lead to a reasonably accurate framework for carrying out engineering design and process calculations. Nonetheless, some obscurities and uncertainties still remain and these are pointed out, together with those areas where further work is needed.

Modeling of Gas-Particle Jets The behavior of the gasparticle jet is closely coupled with the performance of hot metal desulfurization, and consequently, plenty of research has been dedicated to studying different aspects of the associated phenomena ranging from heat-up of the jet in the lance to its penetration into the metal bath. A summary of studies on injection of powders into liquid metals is presented in Table 3,4. An excellent synthesis of the fluid flow behavior of gas-particle jets relevant for hot metal desulfurization has been given by Farias and Irons.<sup>[22]</sup> They noted

that with high particle loadings fine particles generally travel at velocities corresponding to that of the gas flow and labeled this behavior as coupled flow. An uncoupled flow was described as a flow in which the particle loading is low and the boundary layers of the particles no longer overlap. A further distinction was made to describe the penetration behavior. When a coupled gas-particle flow penetrates into a liquid, the resulting flow regime is called jetting, whereas in the case of an uncoupled flow, the resulting fluid flow regime is labeled as bubbling. The term transition regime refers to a fluid flow regime that has characteristics of both jetting and bubbling regimes. Heat-up of gas-particle jets: Along with the change in pressure, the heat-up of the gas can have a significant effect on the carrier gas velocity, and thereby affect the velocity of the particles conveyed by the carrier gas. Due to the difficulties in direct measurement, virtually no experimental information exists on the heat-up of the gas-particle mixture in the lance. Consequently, physical and numerical modeling has been used for estimating the heatup in industrial ladles. Irons<sup>[24]</sup> studied heat transfer during submerged injection by injecting silica particles into liquid lead using nitrogen as the carrier gas. In these experiments, the inner heat transfer coefficient was found to be dependent on the particle loading, but the outer heat transfer coefficient was not affected by it. By making use of a 1D two-phase heat-up model,

Irons<sup>[24]</sup> estimated that under industrial conditions the temperature of the gas-particle mixture should be less than 500  $^{\circ}$ C at the lance exit.

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<sup>&</sup>lt;sup>18</sup>Roderick I. L. Guthrie McGill Metals Processing Centre, Department of Mining & Metallurgical Engineering, McGillUniversity.https://www. jstage.jst.go.jp/article/isijinternational1989/35/1/35\_1\_1/\_article

Year Carrie	Study er gas	Mod	eling	Liquida)	Lanceb)	Particlesc)	
	Ph	ysical	Numeri	cal			
1979 Air	Engh et al.[19]	Yes	No	H2O	S	Polystyrene,	
					]	polythene, Fe spikes, sugar	
1982 Ar	Ghosh and	Yes	No	H2O, et	hanol; S	Na2CO3, SiO2,	
	Lange[20]			glycerine, gly	vcerine gr	aphite powders	
				+20% H2O,	1,1,2,2-		
				tetrabromoe	ethane		
1983 N2	Irons and Tu[21]	Yes	No	Pb	S	Coarse sand, SiO2 flour	
1985 N2	Farias and Irons[22]	Yes	No	H2O	S	SiO2 sand, hollow or He	
						glass particles	
1986 N2	Irons and Farias[23]	Yes	No	Pb	S or HS .	Al(OH)3,SiO2	
1987 N2	Irons[24]	Yes	No	Pb	S SiC	02 sand, SiO2 flour	
1994	Zhao and Irons[25]	Yes	No	HM	S	CaD N2 or CO2	
2010 Ar	Sun et al.[26]	No	Yes	HM	S Gra	anulated Mg	
2016 None* Tripat Air*)	Nakano and Ito[27] )/CaC2**) –2017 Yes thi et al.28] /N2**	Yes Yes	Yes H	H2O*) / I2O*) / HM**)	Fe**) – Poly S, T,	ystyrene*) / CaO**)	
,			T-45, T-15, TT,				
	C-45, H-45						
2018 Ar	Matsuzawa et al.[29]	Yes	No	H2O	S Pol	ypropylne	
2019 N2	Ma et al.[30]	No	Yes	HM	T 60% I	Mg + 40% CaO	

# Table 3. Modeling studies on the injection of powders intoliquid metals.

a) HM ¼ hot metal; b)C-45 ¼ two curved ports in mutually opposite directions, HS ¼ hockey-stick, H-45 ¼ two spiral ports in mutually opposite directions, S ¼ straight, T ¼ T-lance, T-15 ¼ two ports at 15° angle in mutually opposite directions, T-45 ¼ two ports at 45° angle in mutually opposite directions, TT ¼ four ports in mutually perpendicular directions; c)CaD ¼ calcium diamide (commercial CaC2 powder); \*) In physical model; \*\*) In CFD model.

**Source**: Steel research int. 2020, 91, 1900454 1900454 (16 of 25) © 2019 The Authors. Published by WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

Study	Liquida)	Particles or powd	erb) (	Carrier g	gas Particle	s penetrating [%]	Basis of
deduction							
Irons and Farias[31]	Pb	Al(OH)3, SiO2		N2		30 H	Ieat losses
Chiang et al.	[32] HM	CaC2		Ar	30-5	0 Reactio	n rates
Zhao and Iro	ons[33]HM	I CaD	N2 or	CO2	30*), 50**)	Heat	losses
Vargas-Rami	rez HM	I CaO-SiO2-		N2	23	Fitting	of a
et al.[3 mathematica	34] Il model	CaF2-FeO-	Na2O				
Jin et al.[35]	HM	CaO- and Mg-	based	N2	98.05	5 Force bal	lance
Scheepers et	al.[36] HN	I CaC2		N2	30	Fitting of a mathe model	ematical
Ma et al.[37]	HN	A CaO + Mg		N2	36.39	Fitting of a mat model	hematical

Table 4. Estimates for the share of particles penetrating.

a) HM ¼ hot metal; b) CaD ¼ calcium diamide (commercial CaC2 powder); \*) Loading below 60 kg Nm3 ; \*\*) Loading above 60 kg Nm3

**Source**: Steel research int. 2020, 91, 1900454 1900454 (18 of 25) © 2019 The Authors. Published by WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

Although there have been a numerous number of studies on mathematical model of hot metal desulfurization by deep injection of calcium carbide, the research field as a whole is not well integrated. This study presents a model that takes into account the kinetics, thermodynamics, and transport processes to predict the sulfur levels in the hot metal throughout a blow. The model could be utilized to assess the influence of the treatment temperature, time of injection, and initial concentration of sulfur on the desulfurization kinetics.

## CONCLUSION

The parameters influencing the investigation are both constructive and operational, e.g. nozzle shape, diameter, orientation, gas flow rate, liquid properties, immersion depth, etc. The statistical evaluation results with the Eviews are as follows:

The results of cold experiment (Appendix I) were obtained from data analysis. It shows that coefficient of Determination Adj.- R<sup>2</sup> 0.8833 which means the independent variable Viscosity, Density and Surface tension are explaining the Gas Bubble Volume in Experiment I by 88.33%. The P – value of F-Statistic is 0.000 indicates the model is fit for the overall population. It is ensured that the independent variable Viscosity is significantly influencing the dependent variable Gas Bubble Volume with p-value of 0.000 smallar than 5%. Hence the null hypothesis H0 is rejected. But the independent variable Density is not significantly influencing the dependent with p-value of 0.0809 larger than 5%. The independent Variable Surface tension is significantly influencing the dependent variable Gas Bubble Volume with p-value of 0.0172 smallar than 5%. The P – Value of Breusch – Godfrey Serial Correlation LM Test is 0.603. It is larger than 5% and the null hypothesis H01 is rejected, which means the data series is not suffering from serial correlation.

**The results of cold experiment (Appendix II)** were obtained from data analysis. It shows that coefficient of Determination Adj.–  $R^2 0.6980$  which means the independent variable Viscosity, Density and Surface tension are explaining the Gas Bubble Volume in **Experiment II** by 69.80 %. The P – value of F-Statistic is 0.0000 indicates the model is fit for the overall population. It is ensured that the independent variable Viscosity is significantly influencing
the dependent variable Gas Bubble Volume with p-value of 0.000 smallar than 5%. Hence the null hypothesis H0 is rejected. But the independent variables Density and Surface tension are not significantly influencing the dependent with p-value of 0.6897 and 0.1423 larger than 5%. The P – Value of Breusch – Godfrey Serial Correlation LM Test is 0.0051. It is smaller than 5% and the null hypothesis H01 is not rejected, which means the data series is suffering from serial correlation. The P – Value of Obs \*R- Squared: Heteroskedasticity: Breusch – Godfrey Test 0.057 is understood that the heteroskedasticity is not found since the p-value is larger than 5% and null hypothesis H02 is not rejected. The P – Value of Jarque-Bera – Normality Test 0.5188 is larger than 5%, hence the null hypothesis is H03 is not rejected which means that the residuals are normally distributed.

The results of hot metall experiment (Appendix III) were obtained from data analysis. It shows that coefficient of Determination R<sup>2</sup> 0.3588 which means the independent variable Pig iron weight, Temperature before desulphurization, Temperature after desulphurization, Initial sulphur content in pig iron, Desulphurization agent calcium carbide, Desulphurization agent Magnesium, The time of the desulphurization process are explaining the Final sulphur content in Experiment by 35.88 %. The P – value of F-Statistic is 0.0000 indicates the model is fit for the overall population. It is ensured that the independent variables nitial sulphur content in pig iron, Desulphurization agent calcium carbide and Temperature after desulphurization are significantly influencing the dependent variable Final sulphur content with p-values of 0.0296, 0.0000 and 0.0337 smallar than 5%. Hence the null hypothesis H0 is rejected. But the independent variables Pig iron weight, Temperature before desulphurization, Desulphurization agent Magnesium and

the Time of the desulphurization process are not significantly influencing the dependent with p-values of 0.1319, 0.0765, 0.6483 and 0.8031 larger than 5%. The P – Value of Breusch – Godfrey Serial Correlation LM Test is 0.069. It is larger than 5% and the null hypothesis H01 is rejected, which means the data series is not suffering from serial correlation. The P – Value of Obs \*R- Squared: Heteroskedasticity: Breusch – Godfrey Test 0.223 is understood that the heteroskedasticity is not found since the p-value is larger than 5% and null hypothesis H02 is not rejected. The P – Value of Jarque-Bera – Normality Test 0.0002 is smaller than 5%, hence the null hypothesis is H03 is not rejected which means that the residuals are normally distributed.

# The statistical evaluation results with the Eviews in comparison with operating results for the desulphurization of pig iron are consistent. Conclusions can now be drawn on the economical use of desulphurization agents (CAC2, Mg), which increases the economic efficiency of the desulphurization plant.

Furthermore, it has been proven that with the help of the statistical evaluation with the Eviews a realistic simulation of the dynamic interplay of the physical-metallurgical processes in metallurgical quantities is possible and thus the prerequisite of the process sequence and indications for the apparatus setup for the optimization of the entire process become possible without having to carry out a large number of costly and time-consuming development experiments.

# PRACTICAL PART

### Data & Methodology

In the present study the primery data was used to analyze **a cost** effective desulphurization of pig iron with an extremely low sulphur content. Various desulphurization agents were blown into the lades filled with pig iron. The data has been got from own cold and hot experiments. The analysis was performed by using software econometric views (E-Views9). The hypotheses have been tested using simple linear and multiple regression addressing the issues of ordinary least square assumptions.

$$\gamma = \alpha + \beta 1 \chi 1 + \beta 2 \chi 2 + \beta 3 \chi 3 + \varepsilon$$

Where  $\gamma$  the dependent variable gas bubble volume is,  $\alpha$  is the intercept of  $\gamma$  .

 $\beta$ 1,  $\beta$ 2 and  $\beta$ 3 are the slope coefficients and  $\chi$ 1,  $\chi$ 2 and  $\chi$ 3 are the independents variables, physical parameters such as kinetic viscosity, density and surface tension .

The error term is denoted as  $\in$ . The result of the regression analysis is presented in appendices No. 1 and No.2.

Also the hypotheses have been tested using multi linear regression addressing the issues of ordinary least square assumptions.

 $\gamma = \alpha + \beta 1 \chi 1 + \beta 2 \chi 2 + \beta 3 \chi 3 + \beta 4 c \chi 4 + \beta 5 \chi 5 + \beta 6 \chi 6 + \beta 7 \chi 7 + \varepsilon$ 

Where  $\gamma$  the dependent variable is,  $\alpha$  is the intercept of  $\gamma$  .

 $\beta$ 1,  $\beta$ 2,  $\beta$ 3,  $\beta$ 4,  $\beta$ 5,  $\beta$ 6 and  $\beta$ 7 are the slope coefficients and  $\chi$ 1,  $\chi$ 2,  $\chi$ 3,  $\chi$ 4,  $\chi$ 5,  $\chi$ 6 and  $\chi$ 7 are the independents variables.

χ1: Pig iron weight (ton)
χ2:Temperature before desulphurization (C)
χ3:Temperature after desulphurization (C)
χ4:Initial sulphur content in pig iron (ppm - part per million)
χ5: Desulphurization agent calcium carbide (CAC2) in kilograms
χ6: Desulphurization agent Magnesium (Mg) in kilograms
χ7:The time of the desulphurization process (t)
γ:Final sulphur content (ppm - part per million) in pig iron

The method of desulpharization of pig iron by blowing solid matter into the ladle or into the torpedo (injection metallurgy) has been known for long time. The following equation is used to determine the consumption of desulpharization agent.

Kg ( desulpharization agent)/ t (pig iron) = 1.33 ln SE/SA .... (1)

In the reality, however, the consumption of desulpharization agents dependes on sevrel factors:

Ladle design, injection system and lances, gas and solid flow rates, sulphur levels, reagents, temperature and slag affecting the desulphurization, nozzel form, nozzle type, nozzle diameter, nozzle orientaion, kind of gas throughtput, nozzle depth, pig iron weight, temperature before desulphurization ,temperature after desulphurization, initial sulphur content in pig iron, the time of the desulphurization process, final sulphur content in pig iron, ...etc

The mathematical equation which takes most of these factors into account runs as follows:

 $\gamma = f(\chi_1, \chi_2, \chi_3, \chi_4, \chi_5, \chi_6, \chi_7 ... \chi_n)$  ... (2)

Where  $\chi_1$ : Pig iron weight (ton)

χ2:Temperature before desulphurization (C)
χ3:Temperature after desulphurization (C)
χ4:Initial sulphur content in pig iron (ppm - part per million)
χ5: Desulphurization agent calcium carbide (CAC2) in kilograms
χ6: Desulphurization agent Magnesium (Mg) in kilograms
χ7:The time of the desulphurization process (t)
χ:Final sulphur content (ppm - part per million) in pig iron
χn: Other factorsconstructive and operative types (Residuls)

Appendix No. 1: Experimental part of the gas liquid/ dispersion experiments <sup>38</sup>

A) Development of experiment equipment (cold experiment systems)

The cold experiment equipment consists of (Figure 2):

-Argon and nitrogen gas

-Micro flow meter and controller

-High resolution camera

-Beaker with silicone oils<sup>39</sup>

-Capillaries

### Working method

Extensive test used to investigate the fundamental principles of bubble formation, bubble seperation and rise. To this end a measuring cell (figure 2) is built which makes it possible to observe bubble volume, retention time of bubbles and the formation and rise of bubbles with capillaries of various designs as a function of physical proporties of the surrounding medium. For this tests several silicone oils and water used. Inorder to determine bubble growth, time of seperation, time of rise and time of retention a hight resolution fotos were taken and the variation in pressure were measured.



Figure 2: Simulation Model for Formation, Detachmente & Ascent of Individual Dubbles in Silicone Oil

<sup>&</sup>lt;sup>38</sup> El Gammal, T., International Symposium on state of the Art of production and use of DRI

S. 3 (28-31) April 1981 El-Tabbin Cario-ARE)

<sup>&</sup>lt;sup>39</sup> Wacker Siliconöle, Siliconöle AK, notification and manufacture by Wacker-Chemie GmbH Munich

B) The hot experiment equipment consists of :

Metallurgical injection technology<sup>40</sup> - Pig iron desulfurization

### Working method

The entire system (short description) consists of two separate Desulfurization storage and weighing, into which the pig iron pans and the slag containers are moved on ferries (Figure 3,4)

There are two conveyor systems at each desulfurization stand, from which two desulphurizing agents or alternatively any mixture of both substances can be blown into the pig iron.

In order to investigate a cost effectiveness desulphurization of pig iron and also reduction of iron loss with an extremely low sulphur content various desulphurization agents were blown into the lades filled with pig iron





Figure 4: Flow Diagram of a Typical Injection Unit

<sup>40</sup> Brdicka, R " Fundamentals of physical chemistry. 14th edition Berlin 1972

# Appendix No. 2: Statistical Data Analyzes of Experiment I

Statistical analyzes of the photographically determined cold test results with various silicone oils.

Test Results and Discussion for Results of the cold tests with silicone oils Experiment I.

Tointerpret the results of simple linear regression, Breusch-Godfrey Serial Correlation LM Test, Heterosked asticity: Breusch Test and Jaque-Bera-Normality Test, Stability Test and draw a conclusion

The hypotheses have been tested using multilinear regression addressing the issues of ordinary least square assumptions.

$$\gamma = \alpha + \beta 1 \chi 1 + \beta 2 \chi 2 + \beta 3 \chi 3 + \varepsilon$$

Where  $\gamma$  the dependent variable gas bubble volume is,  $\alpha$  is the intercept of  $\gamma$  .

 $\beta$ 1,  $\beta$ 2 and  $\beta$ 3 are the slope coefficients and  $\chi$ 1,  $\chi$ 2 and  $\chi$ 3 are the independents variables, physical parameters such as kinetic viscosity, density and surface tension .

The error term is denoted as  $\in$ . The result of the regression analysis is presented below.

### Testable Hypothesis

H0= The independents variables such as kinematics viscosity (v), density ( $\rho$ ) and surface tension ( $\sigma$ ) are not significantly influencing the dependent variable gas bubble volume (V).

Ha= The independent variables are significantly influencing the dependent variable gross domestic production.

H01= No serial correlation in residuals
Ha1= There is serial correlation in residuals
H02= No heteroskedasticity is found in residuals
H0a= Heteroskedasticity is found in residuals
H03= The residuals of data series are normally distributed
Ha3= The residuals of data series are not normally distributed
H04= There is no cointegration between dependent and independent variables
Ha4= There is cointegration between dependent and

independent variables

### The Regression Equations:

- The variables Gas Bubble Volume, Kinematical Viscosity, Density and Surface tension are not stationary (they have unit roots) at level

		7olume= 222970.51+ 0.0168 Viscosity − 43662.69 Density + 907.46 Surface tension +€	Э
--	--	--	---

Significant Significant Significant

- The variables , DGas BubbleVolume, DKinematical Viscosity, DDensity and DSurface tension are stationary (they have no unit roots) at 1<sup>st</sup> difference.

DVolume= -33.374+ 0.0218 DViscosity- 24382.05 DDensity + 818.02 DSurface tension\_ +C

Significant Not significant Significant

#### Results and Discussion 1st difference.

Coefficient of Determination Adj- R<sup>2</sup>=0.8833

- P Value of "F" Statistic = 0.0000
- P Value of Viscosity Independent Variable=0.000
- P Value of Density Independent Variable=0.0809
- P Value of Surface tension Independent Variable=0.0172
- P Value of Obs \*R- Squared: Breusch Godfrey Serial Correlation LM Test= 0.603
- P Value of Obs \*R- Squared: Heteroskedasticity: Breusch Test =0.855
- P Value of Jarque-Bera Normality Test = 0.0001

The above results were obtained from data analysis. It shows that coefficient of Determination Adj.- R<sup>2</sup>0.8833 which means the independent variable Viscosity, Density and Surface tension are explaining the Gas Bubble Volume in Experiment I by 88.33%. The P – value of F-Statistic is 0.000 indicates the model is fit for the overall population. It is ensured that the independent variable Viscosity is significantly influencing the dependent variable Gas Bubble Volume with p-value of 0.000 smallar than 5%. Hence the null hypothesis H0 is rejected. But the independent variable Density is not significantly influencing the dependent with p-value of 0.0809 larger than 5%. The independent Variable Surface tension is significantly influencing the dependent variable Gas Bubble Volume with p-value of 0.0172 smallar than 5%. The P -Value of Breusch - Godfrey Serial Correlation LM Test is 0.603. It is larger than 5% and the null hypothesis H01 is rejected, which means the data series is not suffering from serial correlation.

The P – Value of Obs\*R-Squared: Heteroskedasticity: Breusch – Godfrey Test 0.855 is understood that the heteroskedasticity is not found since the p-value is larger than 5% and null hypothesis H02 is not rejected.

The P – Value of Jarque-Bera – Normality Test 0.0001 is smaller than 5%, hence the null hypothesis is H03 is rejected which means that the residuals are not normally distributed.

	VOLUME	VISCOSITY	DENSITY	TENSION
1	81	150	0.96	21
2	76	150	0.96	21
3	140	250	0.96	21
4	137	250	0.96	21
5	161	1000	0.965	21.2
6	153	1000	0.965	21.2
7	94	1000	0.965	21.2
8	87	1000	0.965	21.2
9	161	2000	0.965	21.3
10	159	2000	0.965	21.3
11	175	2000	0.965	21.3
12	165	2000	0.965	21.3
13	905	30000	0.965	21.5
14	839	30000	0.965	21.5
15	905	30000	0.965	21.5
16	807	30000	0.965	21.5
17	1307	60000	0.97	21.5
18	1274	60000	0.97	21.5
19	1023	60000	0.97	21.5
20	918	60000	0.97	21.5

The four variables Gas Bubble Volume, Kinematical Viscosity, Density and Surface tension are not stationary (they have unit roots) at level. But they became stationary after first difference. Variables (Gas Bubble Volume, Kinematical Viscosity, Density and Surface tension) are cointegrated.

# <u>Regression Analysis –</u> Gas Bubble Volume, Kinematical Viscosity, Density and Surface tension

### 1. Method: Least Squares

- The variables Gas Bubble Volume, Kinematical Viscosity, Density and Surface tension are not stationary (they have unit roots) at level

Dependent Variable: VOLUME Method: Least Squares Date: 06/16/20 Time: 22:13 Sample: 1 20 Included observations: 20

Variable	Coefficient	Std. Error	t-Statistic	Prob.
VISCOSITY	0.016804	0.001587	10.58542	0.0000
DENSITY	-43662.69	12841.17	-3.400211	0.0037
TENSION	907.4693	211.8809	4.282921	0.0006
С	22970.51	10667.32	2.153355	0.0469
R-squared Adjusted R-squared S.E. of regression Sum squared resid Log likelihood F-statistic Prob(E-statistic)	0.965432 0.958951 91.35532 133532.7 -116.4425 148.9526 0.000000	Mean depend S.D. depende Akaike info cri Schwarz crite Hannan-Quini Durbin-Watso	ent var nt var iterion rion n criter. n stat	478.3500 450.9007 12.04425 12.24339 12.08312 1.186010

- The variables, DGas BubbleVolume, DKinematical Viscosity, DDensity and DSurface tension are stationary (they have no unit roots) at 1<sup>st</sup> difference.

#### Dependent Variable: DVOLUME Method: Least Squares Date: 06/16/20 Time: 22:31 Sample (adjusted): 2 20 Included observations: 19 after adjustments

Variable	Coefficient	Std. Error	t-Statistic	Prob.
DVISCOSITY	0.021820	0.002278	9.578298	0.0000
DDENSITY	-24382.05	13170.74	-1.851228	0.0839
DTENSION	818.0282	305.4077	2.678480	0.0172
С	-33.37469	19.08374	-1.748855	0.1007
R-squared	0.902824	Mean depend	lent var	44.05263
Adjusted R-squared	0.883388	S.D. depende	ent var	219.6329
S.E. of regression	75.00127	Akaike info cr	iterion	11.65755
Sum squared resid	84377.86	Schwarz crite	rion	11.85638
Log likelihood	-106.7467	Hannan-Quin	n criter.	11.69120
F-statistic	46.45277	Durbin-Watso	n stat	1.622546
Prob(F-statistic)	0.000000			

### 2. Breusch-Godfrey Serial Correlation LM Test: H0= No Serial Correlation, while p>5%

Breusch-Godfrey Serial Correlation LM Test:							
F-statistic	0.365388	Prob. F(2,13)	0.7008				
Obs*R-squared	1.011214	Prob. Chi-Square(2)	<mark>0.6031</mark>				

Test Equation: Dependent Variable: RESID Method: Least Squares Date: 06/16/20 Time: 22:34 Sample: 2 20 Included observations: 19 Presample missing value lagged residuals set to zero.

Variable	Coefficient	Std. Error	t-Statistic	Prob.
	0.000040	0.000504	0.004070	0.0040
DVISCOSITY	-0.000213	0.002524	-0.084370	0.9340
DDENSITY	-5642.876	17801.01	-0.316998	0.7563
DTENSION	-69.32613	330.8878	-0.209515	0.8373
С	-1.997163	20.10330	-0.099345	0.9224
RESID(-1)	0.176330	0.290716	0.606538	0.5546
RESID(-2)	0.446503	0.749289	0.595902	0.5615
R-squared	0.053222	Mean depend	ent var	-1.50E-15
Adjusted R-squared	-0.310924	S.D. depende	nt var	68.46648
S.E. of regression	78.39109	Akaike info cri	iterion	11.81339
Sum squared resid	79887.12	Schwarz crite	rion	12.11163
Log likelihood	-106.2272	Hannan-Quin	n criter.	11.86386
F-statistic	0.146155	Durbin-Watso	n stat	2.162335
Prob(F-statistic)	0.977709			

### 3. Heteroskedasticity Test: Breusch-Pagan-Godfrey No Heteroskedasticity, Breusch p > 5%

Heteroskedasticity	Test:	Breusch-Pagan-God	frey
--------------------	-------	-------------------	------

F-statistic	0.213159	Prob. F(3,15)	0.8857
Obs*R-squared	0.776884	Prob. Chi-Square(3)	0.8550
Scaled explained SS	1.358677	Prob. Chi-Square(3)	0.7152

Test Equation: Dependent Variable: RESID^2 Method: Least Squares Date: 06/16/20 Time: 22:34 Sample: 2 20 Included observations: 19

Variable	Coefficient	Std. Error	t-Statistic	Prob.
C	5461.405	2950.480	1.851023	0.0840
DVISCOSITY	-0.084407	0.352205	-0.239653	0.8138

DDENSITY	-431921.3	2036289.	-0.212112	0.8349
DTENSION	-20035.74	47218.16	-0.424323	0.6774
	0.040000			
R-squared	0.040889	Mean depend	ent var	4440.940
Adjusted R-squared	-0.150934	S.D. depende	nt var	10808.68
S.E. of regression	11595.72	Akaike info criterion		21.73932
Sum squared resid	2.02E+09	Schwarz criterion		21.93815
Log likelihood	-202.5236	Hannan-Quin	n criter.	21.77297
F-statistic	0.213159	Durbin-Watso	n stat	2.193426
Prob(F-statistic)	0.885718			

# 4. Jarque-Bera Normality test . Result p=0.5552 . Residals are normal distributed



### 5. Residuals Stability test/ Resulat : stabil

**6.** The Variables (DGDP, DFDI and DPERC\_INVEST and DEMP) are cointegrated (long run assocation according **Johansen cointegrationtest**-Pedroni Residual Cointegration Test ).

We can not run restricted VAR (VECM –Model) because of observation number (20) and We have to run Dynamic Least Squares (FOLS) Model

Date: 06/16/20 Time: 22:56 Sample (adjusted): 4 20 Included observations: 17 after adjustments Trend assumption: Linear deterministic trend Series: DVOLUME DVISCOSITY DDENSITY DTENSION Lags interval (in first differences): 1 to 1

#### Unrestricted Cointegration Rank Test (Trace)

Hypothesized No. of CE(s)	Eigenvalue	Trace Statistic	0.05 Critical Value	Prob.**
None *	0.880879	68.58978	47.85613	<mark>0.0002</mark>
At most 1 *	0.733179	32.42031	29.79707	0.0244
At most 2	0.343991	9.960282	15.49471	0.2838
At most 3	0.151528	2.793399	3.841466	0.0947

Trace test indicates 2 cointegrating eqn(s) at the 0.05 level

\* denotes rejection of the hypothesis at the 0.05 level

\*\*MacKinnon-Haug-Michelis (1999) p-values

Unrestricted Cointegration Rank Test (Maximum Eigenvalue)

Hypothesized No. of CE(s)	Eigenvalue	Max-Eigen Statistic	0.05 Critical Value	Prob.**
None *	0.880879	36.16947	27.58434	0.0031
At most 1 *	0.733179	22.46003	21.13162	0.0323
At most 2	0.343991	7.166883	14.26460	0.4696
At most 3	0.151528	2.793399	3.841466	0.0947

Max-eigenvalue test indicates 2 cointegrating eqn(s) at the 0.05 level \* denotes rejection of the hypothesis at the 0.05 level

\*\*MacKinnon-Haug-Michelis (1999) p-values

Unrestricted Cointegrating Coefficients (normalized by b'*S11*b=I):					
	DVISCOSITY	DDENSITY	DTENSION		
-0.016092	0.000371	213.4465	13.75745		
0.014574	-0.000291	937.7141	-34.46983		
0.004724	7.93E-05	-405.5835	-9.090820		
-0.027859	0.000554	-1116.182	12.90106		
Unrestricted Adjus	tment Coefficient	ts (alpha):			
	-144 8202	76 63408	-100 6615	20 02442	
	-6572 501	3521 328	-1689 038	-137 /822	
	-0.001553	0 000436	0.000325	-0.000170	
D(DTENSION)	-0.038975	0.053956	0.002973	0.018228	
/					
1 Cointegrating Eq	uation(s):	Log likelihood	-145.2257		
1 Cointegrating Eq	uation(s):	Log likelihood	-145.2257		
1 Cointegrating Eq	uation(s):	Log likelihood	-145.2257 r in parenthese	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME	uation(s): grating coefficient DVISCOSITY	Log likelihood ts (standard error DDENSITY 13264 00	-145.2257 r in parenthese DTENSION	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124)	Log likelihood ts (standard erro DDENSITY -13264.00 (7265 83)	-145.2257 r in parenthese DTENSION -854.9160 (172 645)	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124)	Log likelihood ts (standard erro DDENSITY -13264.00 (7265.83)	-145.2257 r in parenthese DTENSION -854.9160 (172.645)	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000 Adjustment coeffici	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124) ients (standard el	Log likelihood ts (standard error DDENSITY -13264.00 (7265.83) rror in parenthes	-145.2257 r in parenthese DTENSION -854.9160 (172.645) es)	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000 Adjustment coeffici D(DVOLUME)	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124) ients (standard en 2.330616	Log likelihood ts (standard error DDENSITY -13264.00 (7265.83) rror in parenthes	-145.2257 r in parenthese DTENSION -854.9160 (172.645) es)	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000 Adjustment coeffici D(DVOLUME)	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124) ients (standard et 2.330616 (1.06801)	Log likelihood ts (standard erro DDENSITY -13264.00 (7265.83) rror in parenthes	-145.2257 r in parenthese DTENSION -854.9160 (172.645) es)	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000 Adjustment coeffici D(DVOLUME) D(DVISCOSITY	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124) ients (standard et 2.330616 (1.06801) 105.7658	Log likelihood ts (standard erro DDENSITY -13264.00 (7265.83) rror in parenthes	-145.2257 r in parenthese DTENSION -854.9160 (172.645) es)	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000 Adjustment coeffici D(DVOLUME) D(DVISCOSITY	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124) ients (standard et 2.330616 (1.06801) 105.7658 (45.2095)	Log likelihood ts (standard erro DDENSITY -13264.00 (7265.83) rror in parenthes	-145.2257 r in parenthese DTENSION -854.9160 (172.645) es)	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000 Adjustment coeffici D(DVOLUME) D(DVISCOSITY D(DDENSITY)	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124) ients (standard el 2.330616 (1.06801) 105.7658 (45.2095) 2.50E-05	Log likelihood ts (standard erro DDENSITY -13264.00 (7265.83) rror in parenthes	-145.2257 r in parenthese DTENSION -854.9160 (172.645) es)	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000 Adjustment coeffici D(DVOLUME) D(DVISCOSITY D(DDENSITY)	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124) ients (standard er 2.330616 (1.06801) 105.7658 (45.2095) 2.50E-05 (5.0E-06)	Log likelihood ts (standard erro DDENSITY -13264.00 (7265.83) rror in parenthes	-145.2257 r in parenthese DTENSION -854.9160 (172.645) es)	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000 Adjustment coeffici D(DVOLUME) D(DVISCOSITY D(DDENSITY) D(DTENSION)	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124) ients (standard er 2.330616 (1.06801) 105.7658 (45.2095) 2.50E-05 (5.0E-06) 0.000627	Log likelihood ts (standard erro DDENSITY -13264.00 (7265.83) rror in parenthes	-145.2257 r in parenthese DTENSION -854.9160 (172.645) es)	es)	
1 Cointegrating Eq Normalized cointeg DVOLUME 1.000000 Adjustment coeffici D(DVOLUME) D(DVISCOSITY D(DDENSITY) D(DTENSION)	uation(s): grating coefficient DVISCOSITY -0.023069 (0.00124) ients (standard et 2.330616 (1.06801) 105.7658 (45.2095) 2.50E-05 (5.0E-06) 0.000627 (0.00039)	Log likelihood ts (standard erro DDENSITY -13264.00 (7265.83) rror in parenthes	-145.2257 r in parenthese DTENSION -854.9160 (172.645) es)	25)	

2 Cointegrating Equation(s):

Log likelihood -133.9957

Normalized cointegrating coefficients (standard error in parentheses)					
DVOLUME	DVISCOSITY	DDENSITY	DTENSION		
1.000000	0.000000	567518.6	-12157.52		
		(103661.)	(2549.04)		
0.000000	1.000000	25176311	-489955.8		
		(4630484)	(113864.)		
Adjustment coeffici	ents (standard err	or in parenthes	ses)		
D(DVOLUME)	3.447443	-0.076085			
	(1.35074)	(0.02936)			
D(DVISCOSITY)	157.0839	-3.465516			
	(56.4704)	(1.22732)			
D(DDENSITY)	3.13E-05	-7.04E-07			
	(6.2E-06)	(1.3E-07)			
D(DTENSION)	0.001414	-3.02E-05			
	(0.00039)	(8.4E-06)			

3 Cointegrating Equ	ation(s):	Log likelihood	-130.4123
Normalized cointegr	ating coefficien	ts (standard error	in parentheses)
DVOLUME	DVISCOSITY	DDENSITY	DTENSION

1.000000	0.000000	0.000000	-2422.265
			(1119.45)
0.000000	1.000000	0.000000	-58079.68
			(49848.7)
0.000000	0.000000	1.000000	-0.017154
			(0.00444)

Adjustment coefficients (standard error in parentheses)

		,
2.929379	-0.084780	85424.47
(1.17098)	(0.02522)	(55007.4)
134.9276	-3.837365	3801283.
(48.5054)	(1.04454)	(2278566)
3.29E-05	-6.78E-07	-0.053849
(5.9E-06)	(1.3E-07)	(0.27913)
0.001428	-2.99E-05	41.06999
(0.00039)	(8.5E-06)	(18.5496)
	2.929379 (1.17098) 134.9276 (48.5054) 3.29E-05 (5.9E-06) 0.001428 (0.00039)	2.929379         -0.084780           (1.17098)         (0.02522)           134.9276         -3.837365           (48.5054)         (1.04454)           3.29E-05         -6.78E-07           (5.9E-06)         (1.3E-07)           0.001428         -2.99E-05           (0.00039)         (8.5E-06)

7. There is long and short run causalty running from independet variables to dependent variable. We have to Use Panel Fully Modified Least Squares (FMOLS)

Dependent Variable: DVOLUME Method: Fully Modified Least Squares (FMOLS) Date: 06/20/20 Time: 10:11 Sample (adjusted): 3 20 Included observations: 18 after adjustments Cointegrating equation deterministics: C Long-run covariance estimate (Bartlett kernel, Newey-West fixed bandwidth = 3.0000)

Variable	Coefficient	Std. Error	t-Statistic	Prob.
DVISCOSITY DENSITY DTENSION C	0.022255 -17697.94 550.5141 17053.58	0.001312 3882.139 178.0937 3748.102	16.96508 -4.558812 3.091148 4.549923	0.0000 0.0004 0.0080 0.0005
R-squared Adjusted R-squared S.E. of regression Long-run variance	0.930858 0.916041 65.38911 1954.077	Mean depend S.D. depende Sum squared	ent var nt var resid	46.77778 225.6696 59860.30

# Appendix No. 3: Statistical Data Analyzes of Experiment II.

Statistical analyzes of the photographically determined cold test results with various silicone oils.

Test Results and Discussion for Results of the cold tests with silicone oils Experiment II.

To interpret the results of simple linear regression, Breusch-Godfrey Serial Correlation LM Test, Heteroskedasticity: Breusch Test and Jarque-Bera-Normality Test, Stability Test and draw a conclusion

The hypotheses have been tested using multilinear regression addressing the issues of ordinary least square assumptions.

$$\gamma = \alpha + \beta 1 \chi 1 + \beta 2 \chi 2 + \beta 3 \chi 3 + \varepsilon$$

Where  $\gamma$  the dependent variable gas bubble volume is, a is the intercept of  $\gamma$  .

 $\beta$ 1,  $\beta$ 2 and  $\beta$ 3 are the slope coefficients and  $\chi$ 1,  $\chi$ 2 and  $\chi$ 3 are the independents variables, physical parameters such as kinetic viscosity, density and surface tension.

The error term is denoted as  $\in$ . The result of the regression analysis is presented below.

### **Testable Hypothesis**

H0= The independents variables such as kinematics viscosity (v), density ( $\rho$ ) and surface tension ( $\sigma$ ) are not significantly influencing the dependent variable gas bubble volume (V).

Ha= The independent variables are significantly influencing the dependent variable gross domestic production.

H01= No serial correlation in residuals
Ha1= There is serial correlation in residuals
H02= No heteroskedasticity is found in residuals
H0a= Heteroskedasticity is found in residuals
H03= The residuals of data series are normally distributed
Ha3= The residuals of data series are not normally distributed
H04= There is no cointegration between dependent and independent variables
Ha4= There is cointegration between dependent and

independent variables

### The Regression Equations:

- The variables Gas Bubble Volume, Kinematical Viscosity, Density and Surface tension are not stationary (they have unit roots) at level

Volume= 1319.93+ 0.021	Viscosity -	34727.27 Density +	1530.68 Surface tension	n +€

Significant Significant Significant

# - The variables , DGas BubbleVolume, DKinematical Viscosity, DDensity and DSurface tension are stationary (they have no unit roots) at 1<sup>st</sup> difference.

DVolume= 45.01+ 0.0177 DViscosity +11529.47 DDensity + 1032.92 DSurface tension\_ +C

Significant Not significant Not Significant

### **Results and Discussion for 1st difference.**

Coefficient of Determination Adj- R<sup>2</sup>=0.6980

- P Value of "F" Statistic = 0.0000
- P Value of Viscosity Independent Variable=0.000
- P Value of Density Independent Variable=0.6897
- P Value of Surface tension Independent Variable=0.1423
- P Value of Obs \*R- Squared: Breusch Godfrey Serial Correlation LM Test= 0.0051
- P Value of Obs \*R- Squared: Heteroskedasticity: Breusch Test =0.057
- P Value of Jarque-Bera Normality Test = 0.5188

The above results were obtained from data analysis. It shows that coefficient of Determination Adj.–  $R^2$ 0.6980 which means the independent variable Viscosity, Density and Surface tension are explaining the Gas Bubble Volume in **Experiment I** by 69.80 %. The P – value of F-Statistic is 0.0000 indicates the model is fit for the overall population. It is ensured that the independent variable Viscosity is significantly influencing the dependent variable Gas Bubble Volume with p-value of 0.000 smallar than 5%. Hence the null hypothesis H0 is rejected. But the independent variables Density and Surface tension are not significantly influencing the dependent with p-value of 0.6897 and 0.1423 larger than 5%.

The P – Value of Breusch – Godfrey Serial Correlation LM Test is 0.0051. It is smaller than 5% and the null hypothesis H01 is not rejected, which means the data series is suffering from serial correlation.

The P – Value of Obs\*R-Squared: Heteroskedasticity: Breusch – Godfrey Test 0.057 is understood that the heteroskedasticity is not found since the p-value is larger than 5% and null hypothesis H02 is not rejected.

The P – Value of Jarque-Bera – Normality Test 0.5188 is larger than 5%, hence the null hypothesis is H03 is not rejected which means that the residuals are normally distributed.

	VOLUME	VISCOSITY	DENSITY	TENSION
1	121	150	0.96	21
2	93	150	0.96	21
3	100	150	0.96	21
4	99	150	0.96	21
5	172	250	0.96	21
6	150	250	0.96	21
7	157	250	0.96	21
8	136	250	0.96	21
9	229	1000	0.965	21.2
10	239	1000	0.965	21.2
11	382	1000	0.965	21.2
12	327	1000	0.965	21.2
13	338	2000	0.965	21.3
14	382	2000	0.965	21.3
15	524	2000	0.965	21.3
16	508	2000	0.965	21.3
17	1563	30000	0.965	21.5
18	1406	30000	0.965	21.5
19	1332	30000	0.965	21.5
20	1124	30000	0.965	21.5
21	1988	60000	0.97	21.5
22	1732	60000	0.97	21.5
23	1791	60000	0.97	21.5
24	1715	60000	0.97	21.5

The four variables Gas Bubble Volume, Kinematical Viscosity, Density and Surface tension are not stationary (they have unit roots) at level. But they became stationary after first difference. Variables (Gas Bubble Volume, Kinematical Viscosity, Density and Surface tension) are cointegrated.

<u>Regression Analysis</u> – Gas Bubble Volume, Kinematical Viscosity, Density and Surface tension

#### 1. Method: Least Squares

- The variables Gas Bubble Volume, Kinematical Viscosity, Density and Surface tension are not stationary (they have unit roots) at level

Dependent Variable: VOLUME Method: Least Squares Date: 06/20/20 Time: 22:59 Sample: 1 24 Included observations: 24

Variable	Coefficient	Std. Error	t-Statistic	Prob.
VISCOSITY	0.021093	0.001614	13.06894	0.0000
DENSITY	-34727.27	13358.80	-2.599581	0.0171
TENSION	1530.687	220.8111	6.932113	0.0000
С	1319.439	10365.30	0.127294	0.9000
R-squared Adjusted R-squared S.E. of regression Sum squared resid Log likelihood	0.981058 0.978217 99.05016 196218.7 -142.1617	Mean dependent var S.D. dependent var Akaike info criterion Schwarz criterion Hannan-Quinn criter.		692.0000 671.1148 12.18014 12.37648 12.23223
F-statistic Prob(F-statistic)	345.2905 0.000000	Durbin-Watso	n stat	2.138378

# - The variables, DGas BubbleVolume, DDKinematical Viscosity, DDensity and DSurface tension are stationary (they have no unit roots) at 1<sup>st and</sup> 2<sup>nd</sup> difference.

Dependent Variable: DVOLUME Method: Least Squares Date: 06/20/20 Time: 23:14 Sample (adjusted): 3 24 Included observations: 22 after adjustments

Variable	Coefficient	Std. Error	t-Statistic	Prob.
DDVISCOSITY DDENSITY DTENSION C	0.017727 11529.47 1032.928 45.01097	0.003227 28415.58 673.2319 39.38833	5.493499 0.405745 1.534283 1.142749	0.0000 0.6897 0.1423 0.2681
R-squared Adjusted R-squared S.E. of regression Sum squared resid Log likelihood F-statistic Prob(F-statistic)	0.741222 0.698093 167.3987 504402.1 -141.6576 17.18591 0.000016	Mean depende S.D. depender Akaike info crit Schwarz criter Hannan-Quinr Durbin-Watsor	ent var nt var terion ion n criter. n stat	73.72727 304.6599 13.24160 13.43997 13.28833 1.635795

### 2. Breusch-Godfrey Serial Correlation LM Test: H0= No Serial Correlation is rejected. P=0.0051 Ha= Has Serial Correlation, while p<5%

Breusch-Godfrey Serial Correlation LM Test:

F-statistic	7.370378	Prob. F(2,16)	0.0054
Obs*R-squared	10.54940	Prob. Chi-Square(2)	0.0051

Test Equation: Dependent Variable: RESID Method: Least Squares Date: 06/20/20 Time: 23:20 Sample: 3 24 Included observations: 22 Presample missing value lagged residuals set to zero.

Variable	Coefficient	Std. Error	t-Statistic	Prob.
DDVISCOSITY	0.007004	0.003260	2.148402	0.0473
DDENSITY	-18188.18	22930.75	-0.793179	0.4393
DTENSION	140.4999	519.6775	0.270360	0.7903
С	5.488121	30.28509	0.181215	0.8585
RESID(-1)	0.607433	0.250548	2.424414	0.0275
RESID(-2)	-0.771924	0.211554	-3.648834	0.0022
R-squared	0.479518	Mean depend	lent var	4.52E-15
Adjusted R-squared	0.316868	S.D. depende	ent var	154.9811
S.E. of regression	128.0947	Akaike info cr	iterion	12.77042
Sum squared resid	262532.0	Schwarz crite	rion	13.06797
Log likelihood	-134.4746	Hannan-Quin	n criter.	12.84051
F-statistic	2.948151	Durbin-Watso	n stat	1.565864
Prob(F-statistic)	0.045008			

# 3. Heteroskedasticity Test: Breusch-Pagan-Godfrey No Heteroskedasticity, Breusch p > 5%

Heteroskedasticity Test: Breusch-Pagan-Godfrey

F-statistic	3.096397	Prob. F(3,18)	0.0530
Obs*R-squared	7.488760	Prob. Chi-Square(3)	0.0578
Scaled explained SS	4.042527	Prob. Chi-Square(3)	0.2569

Test Equation: Dependent Variable: RESID^2 Method: Least Squares Date: 06/20/20 Time: 23:21 Sample: 3 24 Included observations: 22

Variable	Coefficient	Std. Error	t-Statistic	Prob.
С	15181.60	6151.377	2.468000	0.0238
DDVISCOSITY	-0.558104	0.503947	-1.107465	0.2827
DDENSITY	4002003.	4437735.	0.901812	0.3791
DTENSION	260773.8	105140.3	2.480245	0.0232
R-squared	0.340398	Mean dependent var		22927.37
Adjusted R-squared	0.230465	S.D. dependent var		29801.81
S.E. of regression	26143.09	Akaike info criterion		23.34352
Sum squared resid	1.23E+10	Schwarz criterion		23.54189
Log likelihood	-252.7788	Hannan-Quinn criter.		23.39025
F-statistic	3.096397	Durbin-Watson stat		0.976564
Prob(F-statistic)	0.052959			



# 4. Jarque-Bera Normality test . Result p=0.518. Residals are normal distributed

5. Residuals Stability test/ Resulat : Stabil



66

**6.** The Variables (DGDP, DFDI and DPERC\_INVEST and DEMP) are cointegrated (long run assocation according **Johansen cointegrationtest**-Pedroni Residual Cointegration Test ).

### We can not run restricted VAR (VECM –Model) because of observation number (20) and We have to run Dynamic Least Squares (FOLS) Model

Date: 06/20/20 Time: 23:29 Sample (adjusted): 5 24 Included observations: 20 after adjustments Trend assumption: Linear deterministic trend Series: DVOLUME DDVISCOSITY DDENSITY DTENSION Lags interval (in first differences): 1 to 1

Unrestricted Cointegration Rank Test (Trace)

Hypothesized No. of CE(s)	Eigenvalue	Trace Statistic	0.05 Critical Value	Prob.**
None *	0.845550	72.70019	47.85613	<mark>0.0001</mark>
At most 1 *	0.537470	35.34254	29.79707	0.0104
At most 2 *	0.510600	19.92168	15.49471	0.0101
At most 3 *	0.245355	5.630161	3.841466	0.0176

Trace test indicates 4 cointegrating eqn(s) at the 0.05 level

\* denotes rejection of the hypothesis at the 0.05 level

\*\*MacKinnon-Haug-Michelis (1999) p-values

Hypothesized No. of CE(s)	Eigenvalue	Max-Eigen Statistic	0.05 Critical Value	Prob.**
None *	0.845550	37.35765	27.58434	<mark>0.0020</mark>
At most 1	0.537470	15.42086	21.13162	0.2605
At most 2 *	0.510600	14.29152	14.26460	0.0495
At most 3 *	0.245355	5.630161	3.841466	0.0176

Unrestricted Cointegration Rank Test (Maximum Eigenvalue)

Max-eigenvalue test indicates 1 cointegrating eqn(s) at the 0.05 level

\* denotes rejection of the hypothesis at the 0.05 level

\*\*MacKinnon-Haug-Michelis (1999) p-values

7. There is long and short run causalty running from independet variables to dependent variable. We have to Use Panel Fully Modified Least Squares (FMOLS).

Dependent Variable: DVOLUME

Method: Fully Modified Least Squares (FMOLS) Date: 06/20/20 Time: 23:32 Sample (adjusted): 4 24 Included observations: 21 after adjustments Cointegrating equation deterministics: C Long-run covariance estimate (Bartlett kernel, Newey-West fixed bandwidth = 3.0000)

Variable	Coefficient	Std. Error	t-Statistic	Prob.
	0.047000	0.00070		
DDVISCOSITY	0.017399	0.002973	5.852009	0.0000
DDENSITY	6293.588	26187.52	0.240328	0.8130
DTENSION	1036.725	621.1534	1.669033	0.1134
C	49.22388	37.30178	1.319612	0.2045
R-squared	0.740401	Mean dependent var		76.90476
Adjusted R-squared	0.694589	S.D. dependent var		311.8097
S.E. of regression	172.3185	Sum squared resid		504792.4
Long-run variance	23740.65			

# Appendix No. 4: Statistical Data Analyzes of Experiment (Hot Metal) III .

Statistical analyzes of the hot metal (pig iron) desulphurization results test.

To interpret the results of mutiple linear regression, Breusch-Godfrey Serial Correlation LM Test, Heteroskedasticity: Breusch Test and Jaque-Bera-Normality Test, Stability Test and draw a conclusion

The hypotheses have been tested using multilinear regression addressing the issues of ordinary least square assumptions.

 $\gamma = \alpha + \beta 1 \chi 1 + \beta 2 \chi 2 + \beta 3 \chi 3 + \beta 4 \chi 4 + \beta 5 \chi 5 + \beta 6 \chi 6 + \beta 7 \chi 7 + \varepsilon$ 

Where  $\gamma$  the dependent variable is,  $\alpha$  is the intercept of  $\gamma$ .

 $\beta$ 1,  $\beta$ 2,  $\beta$ 3,  $\beta$ 4,  $\beta$ 5,  $\beta$ 6 and  $\beta$ 7 are the slope coefficients and  $\chi$ 1,  $\chi$ 2,  $\chi$ 3,  $\chi$ 4,  $\chi$ 5,  $\chi$ 6 and  $\chi$ 7 are the independents variables.

χ1: Pig iron weight (ton)
χ2:Temperature before desulphurization (C)
χ3:Temperature after desulphurization (C)
χ4:Initial sulphur content in pig iron (ppm - part per million)
χ5: Desulphurization agent calcium carbide (CAC2) in kilograms
χ6: Desulphurization agent Magnesium (Mg) in kilograms
χ7:The time of the desulphurization process (t)
γ:Final sulphur content (ppm - part per million) in pig iron

The error term is denoted as  $\in$ . The result of the regression analysis is presented below.

Testable Hypothesis

H0= The independents variables such as Pig iron weight, Temperature before desulphurization, Temperature after desulphurization, Initial sulphur content, Desulphurization agent calcium carbide, Desulphurization agent Magnesium and The time of the desulphurization process are not significantly influencing the dependent variable Final sulphur content.

Ha= The independent variables are significantly influencing the dependent variable gross domestic production.

H01= No serial correlation in residuals

Ha1= There is serial correlation in residuals

H02= No heteroskedasticity is found in residuals

- H0a= Heteroskedasticity is found in residuals
- H03= The residuals of data series are normally distributed
- Ha3= The residuals of data series are not normally distributed
- H04= There is no cointegration between dependent and independent variables
- Ha4= There is cointegration between dependent and independent variables

# The Regression Equations:

- The variables, Pig iron weight, Temperature before desulphurization, Temperature after desulphurization, Initial sulphur content in pig iron, Desulphurization agent calcium carbide, Desulphurization agent Magnesium, The time of the desulphurization process and Final sulphur content (they have unit roots) at level  $\chi$ 1: Pig iron weight (ton)

 $\chi$ 2:Temperature before desulphurization (C )

χ3:Temperature after desulphurization (C)

χ4:Initial sulphur content in pig iron (ppm - part per million)

 $\chi$ 5: Desulphurization agent calcium carbide (CAC2) in kilograms

 $\chi$ 6: Desulphurization agent Magnesium (Mg) in kilograms

 $\chi$ 7:The time of the desulphurization process (t)

y:Final sulphur content (ppm - part per million) in pig iron

### **Results and Discussion**

Coefficient of Determination Adj- R<sup>2</sup>=0.3588

- P Value of "F" Statistic = 0.0000
- P Value of Pig iron weight Independent Variable=0.1393
- P Value of Temperature before desulphurization Independent Variable=0.0765
- P Value of Temperature after desulphurization Independent Variable=0.0296
- P Value of Initial sulphur content in pig iron Independent Variable=0.0000
- P Value of Desulphurization agent calcium carbide Independent Variable=0.0337
- P Value of Desulphurization agent Magnesium Independent Variable=0.6483
- P Value of The time of the desulphurization process Independent Variable=0.8031
- P Value of Obs \*R- Squared: Breusch Godfrey Serial Correlation LM Test= 0.069

- P Value of Obs \*R- Squared: Heteroskedasticity: Breusch Test =0.223
- P Value of Jarque-Bera Normality Test = 0.0002

The above results were obtained from data analysis. It shows that coefficient of Determination R<sup>2</sup> 0.3588 which means the independent variable Pig iron weight, Temperature before desulphurization, Temperature after desulphurization, Initial sulphur content in pig iron, Desulphurization agent calcium carbide, Desulphurization agent Magnesium, The time of the desulphurization process are explaining the Final sulphur content in Experiment by 35.88 %. The P – value of F-Statistic is 0.0000 indicates the model is fit for the overall population. It is ensured that the independent variables nitial sulphur content in pig iron, Desulphurization agent calcium carbide and Temperature after desulphurization are significantly influencing the dependent variable Final sulphur content with p-values of 0.0296, 0.0000 and 0.0337 smallar than 5%. Hence the null hypothesis H0 is rejected. But the independent variables Pig iron weight, Temperature before desulphurization, Desulphurization agent Magnesium and the Time of the desulphurization process are not significantly influencing the dependent with p-values of 0.1319, 0.0765, 0.6483 and 0.8031 larger than 5%.

The P – Value of Breusch – Godfrey Serial Correlation LM Test is 0.069. It is larger than 5% and the null hypothesis H01 is rejected, which means the data series is not suffering from serial correlation.

The P – Value of Obs\*R-Squared: Heteroskedasticity: Breusch – Godfrey Test 0.223 is understood that the heteroskedasticity is
not found since the p-value is larger than 5% and null hypothesis H02 is not rejected.

The P – Value of Jarque-Bera – Normality Test 0.0002 is smaller than 5%, hence the null hypothesis is H03 is not rejected which means that the residuals are normally distributed.

	Y	X1	X2	X3	X4	X5	X6	X7
1	5	174.4	1339	1339	86	502	113	22
2	22	179.1	1252	1250	103	500	117	23
3	6	172	1302	1301	54	276	71	12
4	12	176	1296	1287	42	206	47	11
5	6	176	1355	1345	80	395	89	18
6	17	173.2	1318	1315	48	230	52	11
7	11	178.3	1312	1308	77	357	80	16
8	12	170	1374	1369	86	393	88	17
9	4	176.1	1358	1351	36	356	80	15
10	9	170.8	1364	1351	93	413	93	18
11	4	169	1397	1388	41	271	61	18
12	8	167.4	1334	1323	49	185	42	10
13	12	169.5	1360	1354	48	292	66	19
14	12	166.8	1394	1377	59	238	57	12
15	17	170	1337	1300	72	470	106	23
16	12	175.8	1344	1342	92	421	95	19
17	3	175.9	1387	1374	70	457	102	14
18	4	175	1347	1343	80	495	111	23
19	3	177.3	1327	1322	82	506	113	23
20	4	175.9	1345	1340	34	241	54	12
21	6	175.2	1381	1373	62	422	95	19
22	8	176.9	1358	1349	78	389	87	18
23	9	178.7	1377	1367	65	340	76	19
24	3	177.8	1318	1306	64	403	90	18
25	5	178.2	1346	1342	67	382	86	17
26	16	177.7	1341	1335	68	271	61	13
27	10	166.1	1353	1346	54	234	53	11
28	21	164.1	1382	1372	65	314	72	16
29	3	171.3	1327	1315	67	407	92	21
30	15	167	1332	1312	30	107	24	9

31	5	163.6	1383	1368	47	348	79	13
32	7	163.7	1346	1342	58	311	70	15
33	3	163.2	1366	1363	35	372	83	17
34	7	173.6	1320	1318	31	227	51	11
35	4	165.2	1351	1344	35	230	52	11
36	11	162.8	1319	1317	26	157	36	8
37	3	164.6	1361	1351	50	441	99	19
38	2	164.1	1354	1349	35	320	72	14
39	12	161	1339	1326	52	334	76	16
40	6	161.7	1344	1344	35	265	61	13
41	11	165.4	1386	1378	25	155	15	8
42	12	160.5	1346	1345	63	302	68	13
43	3	165	1353	1343	40	559	126	18
44	8	166.7	1367	1360	39	246	56	11
45	4	165	1389	1385	45	326	73	14
46	14	165.2	1400	1400	50	256	58	11
47	5	171	1375	1366	41	319	72	15
48	6	166.6	1359	1357	42	223	50	10
49	3	166.5	1309	1305	33	279	63	13
50	6	166.4	1370	1354	96	496	112	23
51	13	165.3	1371	1365	26	159	34	9
52	10	167.2	1337	1333	84	468	106	20
53	14	166.5	1364	1355	82	466	106	22
54	14	166	1327	1327	40	164	64	8
55	7	171.1	1341	1335	30	300	68	9
56	6	172.4	1351	1349	48	258	58	12
57	11	164.9	1344	1341	73	348	78	16
58	8	169.2	1322	1320	80	304	68	14
59	6	165.7	1359	1355	32	294	68	7
60	9	163	1360	1353	44	410	92	19
61	5	166.7	1348	1339	45	296	67	14
62	3	163.3	1345	1340	46	420	94	20
63	2	170.6	1327	1321	38	399	90	19
64	4	177.9	1345	1340	41	340	78	4
65	4	162.7	1338	1334	34	247	56	13
66	11	164	1337	1333	38	202	46	10
67	10	165.6	1335	1330	36	197	45	10
68	4	167.1	1364	1356	71	383	86	22
69	7	161.6	1317	1289	11	227	51	10
70	12	161.4	1377	1371	39	202	46	11
71	7	161.1	1363	1350	55	317	71	15
72	9	165.1	1343	1339	37	200	45	9

	-							
73	8	166.9	1335	1327	37	202	46	11
74	10	168.5	1377	1369	56	287	65	13
75	11	165.5	1341	1339	82	380	85	18
76	7	161	1329	1321	45	361	82	18
77	10	168.8	1318	1303	54	312	71	16
78	9	165.3	1382	1375	62	306	69	14
79	9	165.5	1370	1366	61	302	68	14
80	6	165.7	1343	1330	36	338	107	17
81	9	167.4	1333	1331	76	364	82	17
82	10	163.8	1309	1304	43	238	54	12
83	10	166.8	1320	1315	78	370	83	16
84	12	170.6	1318	1309	40	219	49	11
85	5	166.4	1370	1366	39	209	47	11
86	6	171.7	1350	1341	58	401	80	19

## **Regression Analysis**

### Three ways to examine the stationary of variables

- 1. Graphical analysis
- 2. Correlation
- 3. unit root test



1. Graphics











#### 2. Correlogram

Date: 06/27/20 Time: 16:43 Sample: 1 86 Included observations: 86

Autocorrelation	Partial Correlation		AC	PAC	Q-Stat	Prob
I	1 1	1	0.008	0.008	0.0059	0.939
I	I I I	2	0.234	0.234	4.9507	0.084
1	I I I	3	-0.164	-0.177	7.3931	0.060
1 1	1 🛛 1	4	-0.005	-0.056	7.3956	0.116
1 🗖 1	1 1	5	-0.103	-0.023	8.3892	0.136
1 1	1 1	6	-0.002	-0.012	8.3895	0.211
1 🗖 1	1 🛛 1	7	-0.098	-0.084	9.3015	0.232
1	1 🛛 1	8	-0.041	-0.059	9.4649	0.305
1 🗖 1	1 🗖 1	9	-0.138	-0.111	11.324	0.254
1 🗖 1	1 1	10	-0.123	-0.140	12.827	0.234
I 🛛 I	1 [ 1	11	-0.055	-0.021	13.131	0.285
1	1 1	12	0.097	0.116	14.086	0.295
I 🔲 I	1 1	13	0.152	0.122	16.482	0.224
I 🔲 I	I I I I	14	0.116	0.028	17.888	0.212
1 🛛 1	1	15	-0.039	-0.114	18.051	0.260
I ] I	I I I I	16	0.038	0.032	18.206	0.312
		17	-0.204	-0.185	22.753	0.157
I ] I	I I	18	0.036	-0.005	22.898	0.195
1 🗖 1	101	19	-0.146	-0.075	25.314	0.150
I 🛛 I	1 1	20	0.069	0.008	25.866	0.170
1 🛛 1	I I I I	21	-0.034	0.047	25.997	0.207
1 🔲 1	1	22	-0.085	-0.126	26.849	0.217
1	1 1	23	-0.061	0.001	27.302	0.243
		24	0.261	0.352	35.624	0.060
1 1		25	0.022	-0.056	35,686	0.076

#### 3. Method: Least Squares

- The variables, Pig iron weight, Temperature before desulphurization, Temperature after desulphurization, Initial sulphur content in pig iron, Desulphurization agent calcium carbide, Desulphurization agent Magnesium, The time of the desulphurization process and Final sulphur content stationary (they have no unit roots) at level

#### $Y = 66.05 - 0.122 \chi 1 + 0.120 \chi 2 - 0.147 \chi 3 + 0.177 \chi 4 - 0.039 \chi 5 + 0.033 \chi 6 - 0.043 \chi 7 + C$

#### Dependent Variable: Y

Method: Least Squares Date: 06/27/20 Time: 15:31 Sample: 1 86 Included observations: 86

Variable	Coefficient	Std. Error	t-Statistic	Prob.
X1 X2 X3 X4 X5 X6 X7	-0.122796 0.120335 -0.147726 0.177380 -0.039334 0.033432 -0.043793	0.082217 0.067037 0.066657 0.028824 0.018194 0.073012 0.175038	-1.493568 1.795054 -2.216223 6.153875 -2.161866 0.457897 -0.250193	0.1393 0.0765 0.0296 0.0000 0.0337 0.6483 0.8031
С	66.05238	27.34023	2.415941	0.0180
R-squared Adjusted R-squared S.E. of regression Sum squared resid Log likelihood F-statistic Prob(F-statistic)	0.411664 0.358865 3.410276 907.1388 -223.3345 7.796760 0.000000	Mean depend S.D. depende Akaike info cri Schwarz crite Hannan-Quini Durbin-Watso	ent var nt var terion rion n criter. <mark>n stat</mark>	8.244186 4.259069 5.379872 5.608183 5.471756 2.405619

#### Y= 25.23 -0.094 χ1 + 0.168 χ4 -0.044 χ5 + 0.055 χ6 +€

Dependent Variable: Y Method: Least Squares Date: 06/27/20 Time: 15:32 Sample: 1 86 Included observations: 86

Variable	Coefficient	Std. Error	t-Statistic	Prob.
X1	-0 094389	0.081015	-1 165072	0 2474
X4	0.168043	0.027668	6.073513	0.0000
X5	-0.044284	0.017242	-2.568334	0.0121
X6	0.055722	0.073271	0.760487	0.4492
С	25.23868	13.21402	1.909993	0.0597
R-squared Adjusted R-squared S.E. of regression Sum squared resid Log likelihood F-statistic	0.355086 0.323238 3.503747 994.3755 -227.2827 11.14952	Mean depend S.D. depende Akaike info cr Schwarz crite Hannan-Quin Durbin-Watso	lent var ent var iterion rion n criter. <mark>n stat</mark>	8.244186 4.259069 5.401924 5.544618 5.459352 2.424356
Prob(F-statistic)	0.000000			

#### Without χ2:Temperature before desulphurization (C) χ3:Temperature after desulphurization (C)

Υ = 24.61 -0.091 χ1 + 0.165 χ4 - 0.045 χ5 + 0.055 χ6 + 0.047 χ7 +€

#### Dependent Variable: Y

Method: Least Squares Date: 06/27/20 Time: 15:32 Sample: 1 86 Included observations: 86

Variable	Coefficient	Std. Error	t-Statistic	Prob.
X1 X4 X5	-0.091150 0.165586 -0.045842	0.082371 0.029293 0.018287	-1.106572 5.652680 -2.506813	0.2718 0.0000
X6	0.055831	0.073695	0.757586	0.4509

X7	0.047552	0.177090	0.268522	0.7890
C	24.61357	13.49270	1.824214	0.0719
R-squared	0.355666	Mean depend	ent var	8 244186
Adjusted R-squared	0.315395	S.D. depende	4.259069	
S.E. of regression	3.523989	Akaike info criterion		5.424279
Sum squared resid	993.4801	Schwarz criter	rion	5.595512
Log likelihood	-227.2440	Hannan-Quini	n criter.	5.493192
F-statistic	8.831855	Durbin-Watso	n stat	2.413871
Prob(F-statistic)	0.000001			

- As we can see from the equation the consumption of desulpharization agent is only dependent on three factors. Dies factors are RE (pig iron), SA (initial sulphur content and SE (final sulphur content)

 $\chi 5=\alpha +\beta 1\chi 1 +\beta 2\gamma +\beta 4\chi 4 +\varepsilon$ 

 $\chi 5 = 244.14 - 0.334 \chi 1 - 9.48 \chi + 3.91 \chi 4 + \varepsilon$ 

χ1: Pig iron weight (ton)
χ4:Initial sulphur content in pig iron (ppm - part per million)
χ5: Desulphurization agent calcium carbide (CAC2) in kilograms
γ:Final sulphur content (ppm - part per million) in pig iron

Dependent Variable: X5 Method: Least Squares Date: 06/28/20 Time: 16:23 Sample: 1 86 Included observations: 86

Variable	Coefficient	Std. Error	t-Statistic	Prob.
X1	-0.334955	1.405715	-0.238281	0.8123
Y	-9.483367	1.593001	-5.953147	0.0000
X4	3.914264	0.377801	10.36064	0.0000

С	244.1486	230.9586	1.057110	0.2936	
Derward	0.000450			200 4047	
R-squared	0.033152	wean depend	ent var	320.1047	
Adjusted R-squared	0.619731	S.D. depende	97.83444		
S.E. of regression	60.33053	Akaike info cri	Akaike info criterion		
Sum squared resid	298461.4	Schwarz criter	ion	11.19710	
Log likelihood	-472.5668	Hannan-Quinr	n criter.	11.12889	
F-statistic	47.17535	Durbin-Watso	n stat	2.233822	
Prob(F-statistic)	0.000000				

#### $\chi 6= 60.124 - 0.103 \chi 1 - 2.032 \chi + 0.869 \chi 4 + \varepsilon$

x1: Pig iron weight (ton)

 $\chi$ 4:Initial sulphur content in pig iron (ppm - part per million)  $\chi$ 6: Desulphurization agent Magnesium (Mg) in kilograms  $\gamma$ :Final sulphur content (ppm - part per million) in pig iron

Dependent Variable: X6 Method: Least Squares Date: 06/28/20 Time: 16:45 Sample: 1 86 Included observations: 86

Variable	Coefficient	Std. Error	t-Statistic	Prob.
×4	0 100710	0.040700	0.000040	0 7000
×1	-0.103740	0.342783	-0.302640	0.7629
Y	-2.032301	0.388452	-5.231792	0.0000
X4	0.869678	0.092127	9.440036	0.0000
C	60.12404	56.31909	1.067561	0.2889
Dequered	0 594602	Maan danand	optvor	70 67440
R-squared	0.564695	wean depend	ent var	12.01442
Adjusted R-squared	0.569499	S.D. depende	nt var	22.42186
S.E. of regression	14.71156	Akaike info cri	iterion	8.260539
Sum squared resid	17747.25	Schwarz crite	rion	8.374695
Log likelihood	-351.2032	Hannan-Quin	n criter.	8.306481
F-statistic	38.48149	Durbin-Watso	n stat	2.079145
Prob(F-statistic)	0.000000			

## 4. Breusch-Godfrey Serial Correlation LM Test: H0= No Serial Correlation, while p>5%

Breusch-Godfrey Serial Correlation LM Test:

F-statistic	2.516516	Prob. F(2,76)	0.0874
Obs*R-squared	5.341534	Prob. Chi-Square(2)	0.0692

Test Equation: Dependent Variable: RESID Method: Least Squares Date: 06/27/20 Time: 15:34 Sample: 1 86 Included observations: 86 Presample missing value lagged residuals set to zero.

Variable	Coefficient	Std. Error	t-Statistic	Prob.
X1	-0.005169	0.080785	-0.063987	0.9491
X2	-0.016552	0.066512	-0.248857	0.8041
X3	0.018179	0.066201	0.274604	0.7844
X4	0.009539	0.028757	0.331725	0.7410
X5	-0.006347	0.018093	-0.350804	0.7267
X6	0.032928	0.073495	0.448030	0.6554
X7	-0.024016	0.174170	-0.137889	0.8907
С	-1.715755	26.85585	-0.063888	0.9492
RESID(-1)	-0.205451	0.120589	-1.703729	0.0925
RESID(-2)	0.128026	0.116095	1.102773	0.2736
R-squared	0.062111	Mean depend	ent var	-1 23E-15
Adjusted R-squared	-0.048955	S D depende	nt var	3 266836
S.F. of regression	3.345845	Akaike info cri	iterion	5.362260
Sum squared resid	850.7956	Schwarz crite	rion	5.647649
Log likelihood	-220.5772	Hannan-Quin	n criter.	5.477116
F-statistic Prob(F-statistic)	0.559226 0.826035	Durbin-Watso	n stat	1.977485

## 5. Heteroskedasticity Test: Breusch-Pagan-Godfrey No Heteroskedasticity, while p > 5%

Heteroskedasticity Test: Breusch-Pagan-Godfrey

F-statistic	1.370709	Prob. F(7,78)	0.2296
Obs*R-squared	9.420252	Prob. Chi-Square(7)	0.2239
Scaled explained SS	12.22323	Prob. Chi-Square(7)	0.0935

Test Equation: Dependent Variable: RESID^2 Method: Least Squares Date: 06/27/20 Time: 15:36 Sample: 1 86 Included observations: 86

Variable	Coefficient	Std. Error	t-Statistic	Prob.
		1		
С	161.6562	148.8258	1.086211	0.2807
X1	-0.116021	0.447544	-0.259240	0.7961
X2	0.159348	0.364915	0.436672	0.6636
X3	-0.268763	0.362843	-0.740716	0.4611
X4	0.270653	0.156904	1.724962	0.0885
X5	-0.036343	0.099040	-0.366956	0.7146
X6	-0.002857	0.397439	-0.007189	0.9943
X7	0.761446	0.952817	0.799153	0.4266
R-squared	0 109538	Mean depend	ent var	10 5/813
Adjusted D squared	0.103030		not vor	10.04010
Aujusteu R-squareu	0.029023	S.D. depende		10.04490
S.E. of regression	18.56374	Akaike info cr	iterion	8.768706
Sum squared resid	26879.78	Schwarz crite	rion	8.997017
Log likelihood	-369.0544	Hannan-Quin	n criter.	8.860591
F-statistic	1.370709	Durbin-Watso	n stat	2.101615
Prob(F-statistic)	0.229579			

### 6. Jarque-Bera Normality test . Result p=0.0002 . Residals are not normal distributed



#### 7. Residuals Stability test/ Resulat : stabil



**8.** The Variables ( $\chi 1, \chi 2, \chi 3, \chi 4, \chi 5, \chi 6$  and  $\chi 7$ ) are cointegrated (long run assocation according **Johansen cointegrationtest**-Pedroni Residual Cointegration Test).

We can run restricted VAR (VECM –Model) and We We have to Use Method Dynamic Least Squares (DOLS)

Date: 06/27/20 Time: 15:43 Sample (adjusted): 4 86 Included observations: 83 after adjustments Trend assumption: Linear deterministic trend Series: Y X1 X2 X3 X4 X5 X6 X7 Lags interval (in first differences): 1 to 2

Unrestricted Cointegration Rank Test (Trace)

Hypothesized		Trace	0.05 Critical	
No. of CE(s)	Eigenvalue	Statistic	Value	Prob.**
	C			
None *	0.520684	245.3221	159.5297	0.0000
At most 1 *	0.425139	184.2843	125.6154	0.0000
At most 2 *	0.384568	138.3333	95.75366	0.0000
At most 3 *	0.304829	98.04249	69.81889	0.0001
At most 4 *	0.276990	67.86395	47.85613	0.0002
At most 5 *	0.214937	40.94442	29.79707	0.0018
At most 6 *	0.191872	20.85913	15.49471	0.0070
At most 7	0.037556	3.177196	3.841466	0.0747

Trace test indicates 7 cointegrating eqn(s) at the 0.05 level

\* denotes rejection of the hypothesis at the 0.05 level

\*\*MacKinnon-Haug-Michelis (1999) p-values

Unrestricted Cointegration Rank Test (Maximum Eigenvalue)

Hypothesized		Max-Eigen	0.05 Critical	
No. of CE(s)	Eigenvalue	Statistic	Value	Prob.**
None *	0.520684	61.03779	52.36261	0.0052
At most 1	0.425139	45.95102	46.23142	0.0535
At most 2 *	0.384568	40.29080	40.07757	0.0473

At most 3	0.304829	30.17855	33.87687	0.1298
At most 4	0.276990	26.91953	27.58434	0.0606
At most 5	0.214937	20.08529	21.13162	0.0695
At most 6 *	0.191872	17.68194	14.26460	0.0139
At most 7	0.037556	3.177196	3.841466	0.0747

Max-eigenvalue test indicates 1 cointegrating eqn(s) at the 0.05 level

\* denotes rejection of the hypothesis at the 0.05 level

\*\*MacKinnon-Haug-Michelis (1999) p-values

					-
Y	X1	X2	X3	X4	X5
-0.065811	0.052348	0.263151	-0.272008	-0.021390	-0.002867
-0.573091	-0.082920	0.023582	-0.005758	0.170035	-0.027706
-0.096558	-0.198518	0.079790	-0.106732	0.119737	0.062892
0.261619	0.040712	0.043822	0.001604	0.015937	0.010130
0.003224	-0.157275	-0.063512	0.102553	0.007959	-0.019552
-0.199052	-0.097629	0.208085	-0.201625	0.067378	-0.018657
-0.049195	-0.128689	-0.016720	-0.009024	0.077050	-0.056849
-0.012127	0.169538	0.057655	-0.047598	0.031871	-0.013644

Unrestricted Cointegrating Coefficients (normalized by b'\*S11\*b=I):

Coefficients c1 has negative sign but not significant. It means independent variables and dependent variable have no long run association. There is no long run causality running from seven independent variables to dependent variable. Meaning that  $\chi$ 1: Pig iron weight,  $\chi$ 2:Temperature before desulphurization,  $\chi$ 3:Temperature after desulphurization,  $\chi$ 4:Initial sulphur content in pig iron,  $\chi$ 5: Desulphurization agent calcium carbide,  $\chi$ 6: Desulphurization agent Magnesium and  $\chi$ 7:The time of the desulphurization process are not influence the dependent variable such as  $\gamma$ :Final sulphur content. The Null hypothesis of no cointegration is accepted.

Note: The signs of coefficients are reversed in long-run.

According to Wald test there is short run association between  $\chi$ 1: Pig iron weight,  $\chi$ 2:Temperature before desulphurization,  $\chi$ 3:Temperature after desulphurization,  $\chi$ 4:Initial sulphur content in pig iron,  $\chi$ 5: Desulphurization agent calcium carbide,  $\chi$ 6: Desulphurization agent Magnesium and  $\chi$ 7:The time of the desulphurization process are not influence the dependent variable such as  $\gamma$ :Final sulphur content. Meaning that there is no short run causality running from three independent variables to dependent variable.

**RESULT:** There is no long but there is short run casualty running from independet variables to dependent variable. We have to Use Method Dynamic Least Squares (DOLS)

 $Y = -53.31 - 0.79 \chi 1 - 0.3.99 \chi 2 + 4.13 \chi 3 + 0.32 \chi 4 + 0.04 \chi 5 - 0.20 \chi 6 + 10.12 \chi 7 + C$ 

Dependent Variable: D(Y) Method: Least Squares (Gauss-Newton / Marquardt steps) Date: 06/27/20 Time: 15:48 Sample (adjusted): 4 86 Included observations: 83 after adjustments D(Y) = C(1)\*(Y(-1) - 0.795418166916\*X1(-1) - 3.99856429112\*X2(-1) + 4.1331448244\*X3(-1) + 0.325022722213\*X4(-1) + 0.0435632039388 \*X5(-1) - 2.09668202534\*X6(-1) + 10.1253906583\*X7(-1) -53.3137408875 ) + C(2)\*D(Y(-1)) + C(3)\*D(Y(-2)) + C(4)\*D(X1(-1)) + C(5)\*D(X1(-2)) + C(6)\*D(X2(-1)) + C(7)\*D(X2(-2)) + C(8)\*D(X3(-1)) + C(9)\*D(X3(-2)) + C(10)\*D(X4(-1)) + C(11)\*D(X4(-2)) + C(12)\*D(X5( -1)) + C(13)\*D(X5(-2)) + C(14)\*D(X6(-1)) + C(15)\*D(X6(-2)) + C(16) \*D(X7(-1)) + C(17)\*D(X7(-2)) + C(18)

	Coefficient	Std. Error	t-Statistic	Prob.
C(1)	-0.032011	0.033809	-0.946804	0.3472
C(2)	-0.557968	0.162556	-3.432464	0.0010
C(3)	0.042375	0.152931	0.277089	0.7826
C(4)	-0.177146	0.148986	-1.189008	0.2388

C(5)	0.053472	0.168258	0.317800	0.7517
C(6)	0.052770	0.122099	0.432189	0.6670
C(7)	-0.012512	0.095686	-0.130757	0.8964
C(8)	-0.048307	0.122272	-0.395081	0.6941
C(9)	0.007444	0.093379	0.079723	0.9367
C(10)	-0.001527	0.049211	-0.031028	0.9753
C(11)	-0.020093	0.046802	-0.429314	0.6691
C(12)	0.022873	0.024429	0.936304	0.3526
C(13)	0.011505	0.023786	0.483668	0.6302
C(14)	-0.162983	0.100134	-1.627648	0.1084
C(15)	-0.086196	0.092536	-0.931483	0.3551
C(16)	0.340698	0.329155	1.035068	0.3045
C(17)	0.303719	0.253027	1.200339	0.2344
C(18)	-0.157802	0.516745	-0.305376	0.7611
R-squared	0.430011	Mean depend	lent var	0.000000
Adjusted R-squared	0.280937	S.D. depende	nt var	5.519367
S.E. of regression	4.680289	Akaike info cr	iterion	6.113878
Sum squared resid	1423.832	Schwarz crite	rion	6.638446
Log likelihood	-235.7260	Hannan-Quin	n criter.	6.324620
F-statistic	2.884549	Durbin-Watso	n stat	1.997947
Prob(F-statistic)	0.001112			

According to Wald test there is short run association between  $\chi_1$ : Pig iron weight and  $\gamma$ :Final sulphur content C(4)=C(5)=0 are zero. There is short run casualty running from  $\chi_1$ : Pig iron weight to  $\gamma$ :Final sulphur content

Wald Test: Equation: Untitled			
Test Statistic	Value	df	Probability
F-statistic Chi-square	1.041981 2.083963	(2, 65) 2	0.3586 0.3528

Null Hypothesis: C(4)=C(5)=0 Null Hypothesis Summary:		
Normalized Restriction (= 0)	Value	Std. Err.
C(4) C(5)	-0.177146 0.053472	0.148986 0.168258

Restrictions are linear in coefficients.

According to Wald test there is short run association between  $\chi 2$ :Temperature before desulphurization and  $\gamma$ :Final sulphur content C(6)=C(7)=0 are zero. There is short run casualty running from  $\chi 2$ :Temperature before desulphurization to  $\gamma$ :Final sulphur content

Wald Test:			
Equation: Untitle	d		
Test Statistic	Value	df	Probability
F-statistic	0.191249	(2, 65)	0.8264
Chi-square	0.382498	2	0.8259
Null Hypothesis: Null Hypothesis \$	C(6)=C(7)=0 Summary:		
Normalized Rest	riction (= 0)	Value	Std. Err.
C(6)		0.052770	0.122099
C(7)		-0.012512	0.095686

Restrictions are linear in coefficients.

According to Wald test there is short run association between  $\chi$ 3:Temperature after desulphurization and  $\gamma$ : Final sulphur content C(8)=C(9)=0 are zero. There is short run 90

# casualty running from $\chi$ 3:Temperature after desulphurization to $\gamma$ : Final sulphur content

Wald Test:			
Equation: Untitle	d		
Test Statistic	Value	df	Probability
F-statistic	0.148435	(2, 65)	0.8623
Chi-square	0.296870	2	0.8621
Null Hypothesis: Null Hypothesis	C(8)=C(9)=0 Summary:		
Normalized Rest	riction (= 0)	Value	Std. Err.
C(8) C(9)		-0.048307 0.007444	0.122272 0.093379

Restrictions are linear in coefficients.

According to Wald test there is short run association between  $\chi$ 4:Initial sulphur content in pig iron and  $\gamma$ :Final sulphur content C(10)=C(11)=0 are zero. There is short run casualty running from  $\chi$ 4:Initial sulphur content in pig iron to  $\gamma$ :Final sulphur content

Wald Test: Equation: Untitled			
Test Statistic	Value	df	Probability
F-statistic Chi-square	0.169644 0.339289	(2, 65) 2	0.8443 0.8440

Null Hypothesis: C(10)=C(11)=0

Null Hypothesis Summary:		
Normalized Restriction (= 0)	Value	Std. Err.
C(10) C(11)	-0.001527 -0.020093	0.049211 0.046802

Restrictions are linear in coefficients.

According to Wald test there is short run association between  $\chi 5$ : Desulphurization agent calcium carbide and  $\gamma$ :Final sulphur content C(12)=C(13)=0 are zero. There is short run casualty running from  $\chi 5$ : Desulphurization agent calcium carbide to  $\gamma$ :Final sulphur content

Wald Test: Equation: Untitle	d		
Toot Statiatia	Value	df	Drobobility
	value	u	Probability
F-statistic	0.439146	(2, 65)	0.6465
Chi-square	0.878291	2	0.6446
Null Hypothesis: Null Hypothesis \$	C(12)=C(13)=0 Summary:		
Normalized Rest	riction (= 0)	Value	Std. Err.
C(12) C(13)		0.022873 0.011505	0.024429 0.023786

Restrictions are linear in coefficients.

According to Wald test there is short run association between  $\chi$ 6: Desulphurization agent Magnesium and  $\gamma$ :Final sulphur content C(14)=C(15)=0 are zero. There is short run casualty running from  $\chi$ 6: Desulphurization agent Magnesium to  $\gamma$ :Final sulphur content

Wald Test: Equation: Untitled

Test Statistic	Value df		Probability	
F-statistic	1.325329	(2, 65)	0.2728	
Chi-square	2.650657	2	<mark>0.2657</mark>	

Null Hypothesis: C(14)=C(15)=0 Null Hypothesis Summary:

Normalized Restriction (= 0)	Value	Std. Err.	
C(14)	-0.162983	0.100134	
C(15)	-0.086196	0.092536	

Restrictions are linear in coefficients.

According to Wald test there is short run association between  $\chi$ 7:The time of the desulphurization process and  $\gamma$ :Final sulphur content C(16)=C(17)=0 are zero. There is short run casualty running from  $\chi$ 7:The time of the desulphurization process to  $\gamma$ :Final sulphur content

Wald Test: Equation: Untitled Test Statistic Value df Probability F-statistic 0.754464 (2, 65)0.4743 Chi-square 1.508928 2 0.4703 Null Hypothesis: C(16)=C(17)=0 Null Hypothesis Summary: Normalized Restriction (= 0) Value Std. Err.

C(16)	0.340698	0.329155
C(17)	0.303719	0.253027

Restrictions are linear in coefficients.

7. There is long and short run causalty running from independet variables to dependent variable. We have to Use Method Dynamic Least Squares (DOLS).

Υ= 31.53 - -0.199 χ1+ 0.024 χ2 -0.016χ3 + 0.277 χ4 -0.038 χ5 + 0.021 χ6 -0.33 χ7 +€

Dependent Variable: Y Method: Dynamic Least Squares (DOLS) Date: 06/27/20 Time: 16:03 Sample (adjusted): 3 85 Included observations: 83 after adjustments Cointegrating equation deterministics: C Fixed leads and lags specification (lead=1, lag=1) Long-run variance estimate (Bartlett kernel, Newey-West fixed bandwidth

Variable	Coefficient	Std. Error	t-Statistic	Prob.
X1	-0.199142	0.112602	-1.768543	0.0826
X2	0.024913	0.130301	0.191192	0.8491
X3	-0.016867	0.135096	-0.124854	0.9011
X4	0.277004	0.054203	5.110526	0.0000
X5	-0.038665	0.032007	-1.207988	0.2323
X6	0.021637	0.132331	0.163507	0.8707
X7	-0.330633	0.403709	-0.818987	0.4164
C	31.53362	40.51347	0.778349	0.4398
R-squared	0.629970	Mean depend	lent var	8.144578
Adjusted R-squared	0.438103	S.D. depende	ent var	4.036820
S.E. of regression	3.025991	Sum squared	resid	494.4575
Long-run variance	7.194418	•		

= 4.0000)

## **ABBREVIATIONS**

BF	– Blast Furnace
BOF	– Basic Oxygen Furnace
CaC2	– Calicum Carbide Powder
D	– Diameter
DOLS	– Dynamic Least Squares
FMOLS	<ul> <li>Fully Modified Least Squares</li> </ul>
(Fe)	– Liquid Iron
(FeS)	– Iron Sulfide
Н	– Hight
HM	– Hot Metal
HMDS	- Hot Metal Desulfurization
(S)	<ul> <li>– concentration of sulfur in slag</li> </ul>
[S]	- concentration of sulfur in steel
χ1	– Pig iron weight (ton)
χ2	– Temperature before desulphurization (C)
χ3	– Temperature after desulphurization (C)
χ4	<ul> <li>Initial sulphur content in pig iron (ppm - part per million)</li> </ul>
χ5	– Desulphurization agent calcium carbide
	(CAC2) in kilograms
χ6	<ul> <li>Desulphurization agent Magnesium (Mg) in kilograms</li> </ul>
χ7	<ul> <li>The time of the desulphurization process (t)</li> </ul>
γ	<ul> <li>Final sulphur content (ppm - part per million) in pig iron</li> </ul>

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Metallurgical Injection Technology- Statistical Analysis of Experiments with EViews System to Increase the Cost-Effectiveness of Pig Iron Desulfurization Plant

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