



# Integrated Chlorine Dioxide Process for an Efficient Pulp and Paper Industry

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**Abstract.** Chlorine dioxide ( $\text{ClO}_2$ ) plays an important role in the pulp and paper industry to produce pulp with high purity, yellowing is not desired and paper with high quality. Bleaching pulp using chlorine dioxide ( $\text{ClO}_2$ ) is widely chosen because more environmentally friendly and does not produce dioxins, furans, or adsorbable organic halogens (AOX) compounds. The most widely process to produce chlorine dioxide ( $\text{ClO}_2$ ) in pulp and paper industry are integrated and non-integrated process. This study aims to compare the integrated and non-integrated process, in terms of the raw materials used, waste generated, production costs required and costs required to bleach pulp in pulp and paper industry. The results showed that the integrated process is more efficient and profitable to use in the pulp and paper industry in remote areas due to lower activity for transportation and handling of raw materials. Lower hazardous activity regarding raw material handling and storage, and no solid waste as by-product are some advantages that offered by integrated process. Production costs in the integrated process are also lower than the non-integrated process. But under certain conditions, the non-integrated process is still needed as an option to produce chlorine dioxide ( $\text{ClO}_2$ ) where the integrated process cannot run optimally.

**Keywords:** Sodium chlorate, Chlorine dioxide, Elemental chlorine free, Integrated process, Non-integrated process.

## 1 INTRODUCTION

The pulp and paper industry is highly diversified in terms of products, raw materials, product qualities, distribution channels, and end uses. The demands for pulp and paper are driven by the reliance of society on paper, paperboard, tissue and related products for human welfare and prosperity [1]. Pulp as an intermediate product of the pulp and paper industry is experiencing continuous growth and now dominates the world bleached chemical pulp market to produce a variety of derivative products. In 2021, it was reported that around 191.6 million tons of pulp were produced globally, and around 79% originating from the chemical pulp process [2]. Furthermore, research on the application of derivative cellulose also continuously growth [3].

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The chemical pulp process involves cooking raw materials such as wood chips using aqueous chemical solutions at elevated temperature and pressure to extract pulp fibers. The most widely used chemical pulp process for bleaching pulp is elemental chlorine free (ECF) using chlorine dioxide ( $\text{ClO}_2$ ) as the active chemical [4]. Bleached pulps create papers that are whiter, brighter and softer than unbleached pulps. Bleached pulps are used for products where a high purity is required, and yellowing is not desired (for example papers for printing and writing). Unbleached pulp is typically used to produce boxboards, linerboards, and grocery bags [5]. Initially, chlorine ( $\text{Cl}_2$ ) and oxygen ( $\text{O}_2$ ) have proven to be effective chemicals in bleaching pulp in a sequential step. However, concerns about the generation of chlorinated organic compounds during the pulp bleaching process led to the replacement of chlorine ( $\text{Cl}_2$ ) with chlorine dioxide ( $\text{ClO}_2$ ) in the pulp bleaching process. The use of chlorine dioxide ( $\text{ClO}_2$ ) produces strong, bright white fibers without the production of dioxins, furans, or adsorbable organic halogens (AOX) associated with chlorine. This combination of superior performance and environmental benefit has driven the substantial conversion of pulp bleaching practice to the chlorine dioxide ( $\text{ClO}_2$ ) process [6].

Besides environmental considerations, chlorine dioxide ( $\text{ClO}_2$ ) is widely used because of nature as an oxidizer that is selective to lignin, does not attack cellulose fibers and preserves the mechanical properties of bleached pulp. However, chlorine dioxide ( $\text{ClO}_2$ ) is unstable and cannot be shipped by conventional transport or stored for a long time. Therefore, the chlorine dioxide ( $\text{ClO}_2$ ) production process will always be adjacent to pulp and paper mills and continuously running. For big scale industry such as pulp and paper, the main raw materials to produce chlorine dioxide ( $\text{ClO}_2$ ) are sodium chlorate ( $\text{NaClO}_3$ ) as acid solution and hydrochloric acid ( $\text{HCl}$ ) or methanol ( $\text{CH}_3\text{OH}$ ) as reducing agent [7].

Mostly pulp and paper industry produces prehydrolysis kraft dissolving pulp (DP) and bleached hardwood kraft pulp (BHKP) as a raw material for papermaking with superior quality. Both products require large and continuous quantities of chlorine dioxide ( $\text{ClO}_2$ ) to produce pulp with a brightness that meets market demand. Pulp and paper industry has two different processes for producing chlorine dioxide ( $\text{ClO}_2$ ), an integrated chlorine dioxide plant and a non-integrated chlorine dioxide plant. The two plant are different in terms of the overall processes, raw materials used, by-products or waste produced and the purity of the chlorine dioxide ( $\text{ClO}_2$ ) produced. However, there is quantitative report yet, to evaluate which one is more efficient. This study aims to compare the integrated and non-integrated chlorine dioxide ( $\text{ClO}_2$ ) processes in terms of raw materials used and total production costs, as well as the total daily cost of chlorine dioxide ( $\text{ClO}_2$ ) required to bleach pulp produced.

## 2 METHODOLOGY

Pulp and paper industry has a department called the chemical plant. The department continuously produces chlorine dioxide ( $\text{ClO}_2$ ) for bleaching process, other chemicals needed in the pulp mill operations. There are several plants within the chemical plant department that specifically aim to produce chlorine dioxide ( $\text{ClO}_2$ ). However, there are only two distinct processes within these plants, the integrated chlorine dioxide ( $\text{ClO}_2$ ) process and the non-integrated chlorine dioxide ( $\text{ClO}_2$ ) process. All process

parameters, consumption and production data coming from field instrumentation equipment in the plant. The data will be processed in the distributed control system (DCS) database and recorded in a software called PI ProcessBook. Historical data will be seen as information about plant and saved in PI ProcessBook. The data will be utilized as back up data for trouble shooting, equipment performance evaluation and also some data will be extracted into Microsoft Excel to be used as a daily report production. The data that has been extracted into Microsoft Excel will be used as a calculation of production costs between the integrated chlorine dioxide ( $\text{ClO}_2$ ) process and the non-integrated chlorine dioxide ( $\text{ClO}_2$ ) process. Figure 1 shows data collection and processing in this study while the overall process comparison is obtained from observations of daily activities carried out in the integrated chlorine dioxide ( $\text{ClO}_2$ ) process and the non-integrated chlorine dioxide ( $\text{ClO}_2$ ) process.

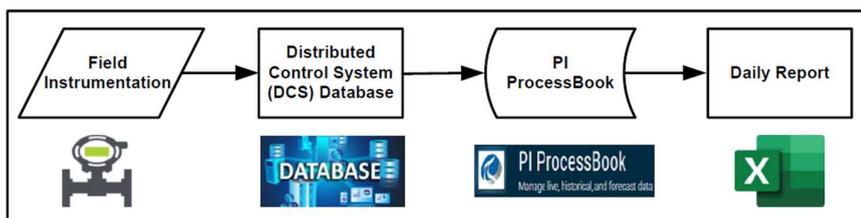


Fig. 1. Data Collection and Processing.

### 3 RESULTS AND DISCUSSIONS

#### 3.1 Process Differences between Integrated and Non-Integrated Process

In the chemical plant department, there are generally two types of chlorine dioxide ( $\text{ClO}_2$ ) manufacturing processes, integrated chlorine dioxide process or non-integrated chlorine dioxide process. Figure 2 shows a typical schematic of integrated chlorine dioxide process and non-integrated chlorine dioxide process.

Integrated chlorine dioxide process only requires salt (or chlorine gas) and electricity as the main raw materials. This process is generally integrated along with the Chlor Alkali Plant, which is a plant with salt solution as the main raw material and caustic soda ( $\text{NaOH}$ ), hydrogen ( $\text{H}_2$ ) gas and chlorine ( $\text{Cl}_2$ ) gas as the main products. There are three main processes in the integrated chlorine dioxide process, the first process is the formation of sodium chlorate ( $\text{NaClO}_3$ ) through the electrolysis of sodium chloride ( $\text{NaCl}$ ) solution. The second process is the combustion reaction of hydrogen ( $\text{H}_2$ ) gas and chlorine ( $\text{Cl}_2$ ) gas (strong chlorine and weak chlorine) to produce hydrochloric acid ( $\text{HCl}$ ). The last process is the reaction between sodium chlorate ( $\text{NaClO}_3$ ) and hydrochloric acid ( $\text{HCl}$ ) to produce chlorine dioxide ( $\text{ClO}_2$ ) which takes place in a chlorine dioxide generator made of titanium. Whereas in the non-integrated chlorine dioxide process, the main chemicals crystal sodium chlorate ( $\text{NaClO}_3$ ), methanol ( $\text{CH}_3\text{OH}$ ) and sulfuric acid ( $\text{H}_2\text{SO}_4$ ) must be imported or purchased from overseas or domestic continuously and stored in tanks at the factory area. During the process, the three main chemicals will be added continuously to produce chlorine

dioxide (ClO<sub>2</sub>), and by-product from this process is sodium sesquisulphate (Na<sub>3</sub>H(SO<sub>4</sub>)<sub>2</sub>).

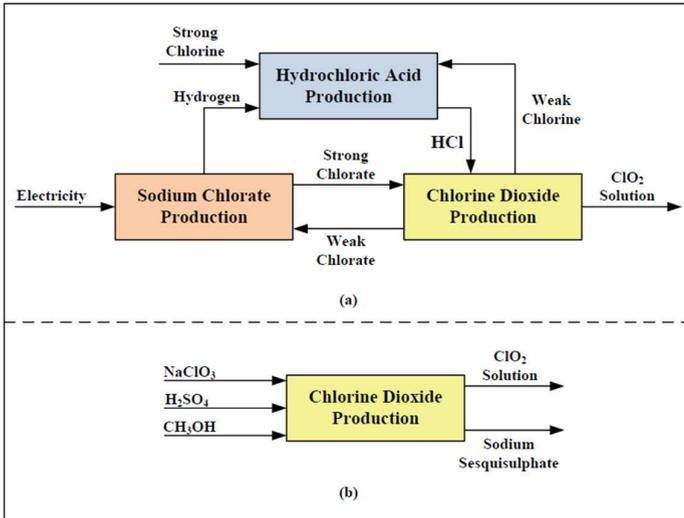
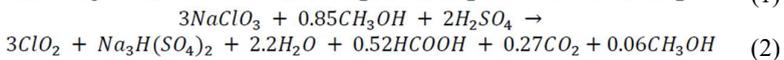
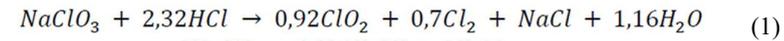
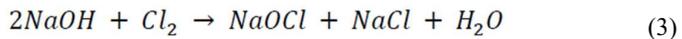


Fig. 2. (a) Typical Schematic of Integrated Chlorine Dioxide Process and (b) Typical Schematic of Non-Integrated Chlorine Dioxide Process.

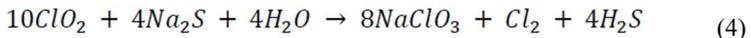
According to the process differences in integrated and non-integrated process, Table 1 summarizes the comparison of integrated and non-integrated process for production of chlorine dioxide (ClO<sub>2</sub>). In overall, integrated process has advantages in the term of transportation of raw materials. It does not increase the biological oxygen demand (BOD) content of the effluent system in the pulp bleaching process, does not generate solids by-products so no periodic boil outs are required, and does not produce solid waste. However, the limitation of integrated process, when it is compared with the non-integrated process, is quite high electrical energy usage in the sodium chlorate (NaClO<sub>3</sub>) production process. This is due to theoretically to produce 1 ton of sodium chlorate (NaClO<sub>3</sub>) requires electrical energy around 4.5 - 5.4 MWh. The purity of the chlorine dioxide (ClO<sub>2</sub>) produced is also lower compared with the non-integrated process. As shown in Equation (1), the reaction between sodium chlorate (NaClO<sub>3</sub>) and hydrochloric acid (HCl) still produces chlorine gas (Cl<sub>2</sub>). Some of the chlorine gas (Cl<sub>2</sub>) will continue to be carried away and dissolved in the chlorine dioxide (ClO<sub>2</sub>) solution produced. Whereas in Equation (2), the reaction of chlorine dioxide (ClO<sub>2</sub>) formation in a non-integrated process does not produce chlorine gas (Cl<sub>2</sub>), so there is no chlorine (Cl<sub>2</sub>) dissolved in chlorine dioxide (ClO<sub>2</sub>) solution produced.



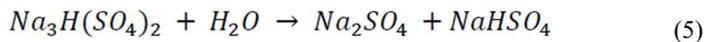
The integrated process produces hydrogen gas (H<sub>2</sub>) and chlorine gas (Cl<sub>2</sub>) which need to be treated before discharging to the atmosphere. For this, sodium hydroxide (NaOH) solution is used as the absorbent. Hydrogen gas (H<sub>2</sub>) will naturally dilute in free air, while chlorine gas (Cl<sub>2</sub>) will react with sodium hydroxide (NaOH) solution to produce sodium hypochlorite (NaOCl) solution as in Equation (3). The sodium hypochlorite (NaOCl) solution or HYPO solution will be stored in a storage tank for used in pulp and paper operations. HYPO solution used as a disinfectant in the water treatment process for internally used at pulp and paper operations or in residential areas. The cooling tower also uses HYPO solution as a moss growth inhibitor. Other plant such us a rayon fiber producer also uses HYPO solution in operational processes.



Meanwhile, the non-integrated process uses white liquor solution as the absorbent to treat off – gas. White liquor is a mixture of chemical compounds used in the chip cooking process in the digester to produce pulp. The main chemicals in white liquor are sodium sulfide (Na<sub>2</sub>S) and sodium hydroxide (NaOH). White liquor was chosen because of ability to absorb chlorine dioxide (ClO<sub>2</sub>) gas that is not completely absorbed in the chlorine dioxide (ClO<sub>2</sub>) absorption tower using chilled water. The reaction that occurs between white liquor and chlorine dioxide (ClO<sub>2</sub>) gas is as shown in equation (4).



The non-integrated process also produces a solid waste as by-product in the form of sodium sesquisulphate (Na<sub>3</sub>H(SO<sub>4</sub>)<sub>2</sub>). Sodium sesquisulphate (Na<sub>3</sub>H(SO<sub>4</sub>)<sub>2</sub>) must be reacted with hot water as in Equation (5) to produce sodium sulfate (Na<sub>2</sub>SO<sub>4</sub>) and sodium bisulfate (NaHSO<sub>4</sub>) in equipment called metathesis filters. This waste can be discharged directly into the mill effluent system after pH adjustment or used in the pulp mill chemical recovery system to replace the sulfur lost in the pulp mill.



**Table 1.** Comparison Integrated and Non-Integrated Process

Integrated Process	Non-Integrated Process	Effect
Using electrical energy and chlorine gas (Cl <sub>2</sub> ) as raw materials to produce chlorine dioxide (ClO <sub>2</sub> ). Electrical energy and chlorine gas (Cl <sub>2</sub> ) are obtained from factories that are still in the same industrial area	Must purchase, transport and handle sodium chlorate (NaClO <sub>3</sub> ), sulfuric acid (H <sub>2</sub> SO <sub>4</sub> ) and methanol (CH <sub>3</sub> OH) as raw materials to produce chlorine dioxide (ClO <sub>2</sub> )	This condition causes significant production costs in non-integrated process, especially in remote areas or areas with high raw material prices. The risk of contaminated raw materials is also one of the disadvantages of the non-integrated process, which does not occur in the integrated process
No organic compounds used as raw material	Using organic compound as raw material (methanol)	Organic compounds will produce organic by-products that will continue to be carried over into the pulp bleaching process. This will increase the BOD content of the effluent discharge system

Integrated Process	Non-Integrated Process	Effect
Chlorine dioxide (ClO <sub>2</sub> ) solution still contains dissolve chlorine around 1.5 gram per liters	Chlorine dioxide (ClO <sub>2</sub> ) solution almost no dissolve chlorine (lower than 0.2 grams per liters)	This dissolved chlorine value is still meets for bleaching process requirements, but if a mill required a lower concentration of chlorine, additional processing is needed and available for the integrated process
Does not require boil out due to no solid produced from reaction	Requires periodic boil out to remove solid deposits on the surface of the heat exchanger	Heat transfer is not good due to deposits on the surface of the heat exchanger and improper boil out will cause corrosion of the titanium heat exchanger tubes in non-integrated process
Using sodium hydroxide (NaOH) for absorbing waste gas	Using white liquor (a mixture of sodium sulfide and sodium hydroxide) for absorbing waste gas	The reaction results in an integrated process produces sodium hypochlorite (NaOCl) and can be used in pulp and paper operation. The reaction results in the non-integrated process are directly disposed to mill effluent system
No solid waste	Produces solid waste as a by-product in the form of sodium sesquisulfate (Na <sub>2</sub> H(SO <sub>4</sub> ) <sub>2</sub> )	This waste can be disposed of directly in the mill effluent system or used in the pulp mill's chemical recovery system

### 3.2 Production Cost Comparison

Production costs, especially cost for raw material and utility consumption for every one ton of chlorine dioxide (ClO<sub>2</sub>) for integrated and non-integrated process, are then compared. Table 2 shows that in the integrated chlorine dioxide process, 72.12% of the production cost comes from the utilities used, which is about 929.31 USD/ton of chlorine dioxide (ClO<sub>2</sub>) produced. The use of power is the largest among all utilities, reaching 67.28% of the total production cost. The largest power consumption in the integrated chlorine dioxide process is in the process of electrolysis of sodium chloride (NaCl) into sodium chlorate (NaClO<sub>3</sub>), which is 4.5 – 5.4 MWh/ton sodium chlorate (NaClO<sub>3</sub>) [8]. To produce one ton of chlorine dioxide (ClO<sub>2</sub>), it takes about 1.76 ton of sodium chlorate (NaClO<sub>3</sub>), so the total power required in the cell electrolyze is about 7.92 – 9.5 MWh

**Table 2.** Raw Material and Utility Cost Comparison Between Integrated and Non-Integrated Process.

Input Operation	Unit	Integrated ClO <sub>2</sub> Plant			Non-Integrated ClO <sub>2</sub> Plant		
		Qty per ton ClO <sub>2</sub>	Price (USD)	Cost (USD)	Qty per ton ClO <sub>2</sub>	Price (USD)	Cost (USD)
<b>Raw Material</b>							
Chlorine	ton/ton ClO <sub>2</sub>	0.98	310	303.8	0	0	0
Caustic Soda	ton/ton ClO <sub>2</sub>	0.11	500	55	0.09	500	45
Sodium Dichromate	kg/ton ClO <sub>2</sub>	0.12	3.15	0.38	0	0	0
Sodium Chlorate	kg/ton ClO <sub>2</sub>	0	0	0	1680	0.88	1478.40
Sulfuric Acid	kg/ton ClO <sub>2</sub>	0	0	0	900	0.13	117
Methanol	kg/ton ClO <sub>2</sub>	0	0	0	160	0.52	83.20
<b>Utility</b>							
Process Water	m <sup>3</sup> /ton ClO <sub>2</sub>	145.25	0.05	7.26	200	0.05	20
Demin Water	m <sup>3</sup> /ton ClO <sub>2</sub>	4.20	0.24	1.01	0	0.24	0
Power	MWh/ton ClO <sub>2</sub>	10.20	84.99	866.90	3.50	84.99	296.03
LP Steam	ton/ton ClO <sub>2</sub>	9.40	5.76	54.14	13.50	5.76	74.52
MP Steam	ton/ton ClO <sub>2</sub>	0	9.57	0	3	9.57	27.48
<b>Total Cost</b>				<b>1288.49</b>			<b>2137.54</b>

It is important to mention that the electricity to power the electrochemical cell is provided from combustion of black liquor from pulp mill, combustion of biomass and

solar cell installed. While the production costs associated with the use of chemicals are only about 27.87% or about 359.18 USD/ton of chlorine dioxide ( $\text{ClO}_2$ ) produced. This is because during normal running, the only chemical added continuously is strong chlorine ( $\text{Cl}_2$ ) gas in the hydrochloric acid ( $\text{HCl}$ ) formation process. Strong chlorine ( $\text{Cl}_2$ ) gas is a product of the Chlor Alkali Plant which is integrated along with the chlorine dioxide plant.

Whereas in the non-integrated process, the largest production cost comes from the chemicals used, which reaches 80.63% or around 1723.60 USD/ton of chlorine dioxide ( $\text{ClO}_2$ ) produced. The three main chemicals, sodium chlorate ( $\text{NaClO}_3$ ), sulfuric acid ( $\text{H}_2\text{SO}_4$ ) and methanol ( $\text{CH}_3\text{OH}$ ), must be added continuously during the process. Sodium chlorate ( $\text{NaClO}_3$ ) is the most widely used chemical to produce chlorine dioxide ( $\text{ClO}_2$ ). Sodium chlorate ( $\text{NaClO}_3$ ) used is crystal form with purity above 99%. This chemical must be imported from abroad, because in Indonesia there is no factory that produces commercially. Therefore, the production cost generated from the use of sodium chlorate ( $\text{NaClO}_3$ ) to produce chlorine dioxide ( $\text{ClO}_2$ ) in the non-integrated process reaches 69.16% of the overall total, or around 1478.4 USD/ton of chlorine dioxide ( $\text{ClO}_2$ ) produced. Meanwhile, sulfuric acid ( $\text{H}_2\text{SO}_4$ ) and methanol ( $\text{CH}_3\text{OH}$ ) are still available in the Indonesian market. The resulting, the production cost of the integrated chlorine dioxide process is lower than the non-integrated chlorine dioxide process. The integrated chlorine dioxide process is also more suitable for pulp and paper mills located in remote areas, as there is no need to spend excessive effort to think about the process of purchasing, transporting and storing the main raw materials in the chlorine dioxide formation process.

Mostly pulp and paper industry produces two types of pulp every day with a total production of around 11,000 ton. Prehydrolysis kraft dissolving pulp (DP) produces around 4,000 ton and bleached hardwood kraft pulp (BHKP) around 7,000 tons. The chlorine dioxide ( $\text{ClO}_2$ ) requirement for dissolving pulp is 10.3 kg per ton of pulp and 15.2 kg per ton of pulp for kraft pulp. The production cost of bleaching the pulp using the non-integrated process would be 315,500 USD per day, or about 1.66 times the total cost of using the integrated process as shown in Table 3. The integrated chlorine dioxide process is preferred for use in the pulp bleaching process at pulp and paper industry because it has a lower economic value and is a more profitable process in terms of purchasing, storing and transporting raw materials. However, an integrated process has own challenges in terms of usage of utility systems, as it will always be related to the environmental impact of the huge consumption of electrical energy.

**Table 3.** Chlorine Dioxide Cost for Bleaching Process.

Pulp Type	Production per Day (ton)	ClO <sub>2</sub> Needed		Cost ClO <sub>2</sub> Production (USD/ton)		Cost ClO <sub>2</sub> for Pulp Production (USD/day)	
		(kg/ton)	Total (ton)	Integrated	Non-Integrated	Integrated	Non-Integrated
DP	4.000	10,3	41.2				
BHKP	7.000	15,2	106.4	1.288,49	2.137,54	190.181,20	315.500,17
<b>Total</b>	<b>11.000</b>		<b>147.6</b>				

#### 4 CONCLUSION

Integrated and non-integrated chlorine dioxide manufacturing processes differ in many aspects, starting from raw material usage, overall process and characteristic chlorine dioxide (ClO<sub>2</sub>) produce. The integrated process is more beneficial to bleached pulp producers that are situated in remote areas which, as a result, may experience supply chain challenges in receiving chemicals needed by the non-integrated process configurations. Lower manufacturing cost, lower challenges associated with importation and transportation of raw materials, lower hazardous activity regarding raw material handling and stored, no solid waste as by-product and more environmental benefit is some advantages that offer by integrated chlorine dioxide process. Under certain conditions, the non-integrated process is still needed as an option to produce chlorine dioxide (ClO<sub>2</sub>) where the integrated process cannot run optimally. However, non-integrated process produced chlorine dioxide gas product purer from chlorine content compared with integrated process, with chlorine content less than 1% in non-integrated process and 17-19% in integrated process.

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