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# PRODUCTION OF ACTIVATED CARBON FROM SUBBITUMINOUS COAL USING ROTARY KILN AND CYCLONE BURNER

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***ABSTRACT***

*Research on the production of activated carbon from coal has been carried out by Research and Development Center for Mineral and Coal Technology since 1980 in the laboratory as well as pilot plant scale. Production of activated carbon from coal using rotary kiln and oil burner at the pilot plant of 1 ton/day has been carried out successfully to produce good quality product. To reduce the dependence upon using oil fuel, the oil fuel burner was replaced by coal fueled cyclone burner. Product quality and economic evaluation of the production of activated carbon using the burners are described in this paper. The coal used was subbituminous coal from Air Laya, South Sumatera. The coal passing 3 cm screen was carbonized at 500-600° C for 2 hours to produce good quality char. The variables of activation process observed were consisted of particle size of char (+6, -6+12 and -12 +20 mesh) and residence time (1.5, 3 and 6 hours). The results showed that the optimum condition for activation process was using particle size of -6+12 mesh and residence time at a minimum of 3 hours. The activated carbon produced showed quality which fairly met the requirement of Indonesian Industrial Standard with iodine number of 600-800 mg/g compared with standard of 750-1200 mg/g and market quality of 400-1200 mg/g. Eventhough the adsorption capacity obtained was at the lowest limit of the Indonesian Industrial Standard, however the pre utilization test showed that the product could be used for treatment of shrimp farms water.*

*Keywords: activated carbon, coal, rotary kiln, cyclone burner*

## INTRODUCTION

Research on production of activated carbon using Indonesian coal has been carried out by Research and Development Center for Mineral and Coal Technology (*tek*MIRA) since 1980's. The results showed that Indonesian low rank coal can be used as raw material for activated carbon. Currently, the research has been developed into pilot plant of 1 ton product/ day using a rotary kiln and steam for activation process. The quality of activated carbon produced has met the requirement of Indonesian Industrial Standard, except hardness and ash content. Eventhough, the product can be utilized for water purification and room deodorizer (Monika, 2008).

To develop the process into commercial scale economically, some modifications in the pilot plant equipment have been done.The energy for carbonization and activation process which formerly used oil burner was substituted by coal fueled cyclone burner. The use of cyclone burner is expected to reduce the production cost. Product quality and economic evaluation of production of activated carbon using oil and coal fueled burner are reported in this paper.

## LITERATURE STUDY

### Activated Carbon

Activated carbon is the most popular adsorption material for liquid and gas phases purification in the various industrial processes. It has cristaline form with large internal pore structures that make it easy to adsorp. There are a lot of carbonaceous materials that can be activated, particularly which has high content of carbon compounds (Monika, 2008). The adsorption surface of activated carbon is measured by its surface area, generally in the

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range of 400-1600 $m^2/g$ (Paul, 2002). Beside, pores of activated carbon also determine the adsorption capacity for the different utilization. Carbon pores consist of :

- Micro pores (small pore): radius of <1 nm or diameters below 20Å to 8 Å.
- Meso pores (medium pores): radius of 1-25 nm or diameters 500 Å down to 20 Å.
- Macro pores (large pores): radius of >25 nm or with diameters of about 500 Å.
- Pores of less than 8 Å are called submicro pores.

Carbon pore is formed during carbonization and activation of carbonaceous material. The size of total pore is determined using the helium and mercury density. For the measurement of pore distribution in macro pore area and a great portion of meso pore area, mercury porosimetry is used. Meso pores and part of the micro pores are measured by the adsorption of vapors using capillary condensation. Micro and sub-micro pores can be measured by the methanol adsorption (Pruss, 1972).

Large pores is used to transport liquid through carbon, and then the adsorption occurs in the medium and small pores. Therefore, surface area and carbon pores are usually used to measure effectiveness of the carbon adsorption (Strand, 2001). The large macro pores act as channel through the carbon to meso and micro pores. Granular activated carbon always has macro pores, conversely macro pores are seldom found in powdered activated carbon since after grinding, the carbon consists of very small particles. The standard practice to decribe the level activity in activated carbon is by quantifying carbon that has become gaseous and left behind space. Thus, high quality of activated carbon contains a lot of empty space; such carbon has many meso and macropores.

### Production of Activated Carbon from Coal

The production process of activated carbon consists of two steps, carbonization and activation. The carbonization process includes drying and then heating coal to separate by-products which include tars and other hydrocarbons, as well as to drive off any gases generated. The carbonization process is completed by heating the carbonaceous material at 400–600°C in an oxygen-deficient atmosphere that cannot support combustion (Chand, 2001). Char as the solid product of carbonization process can be further activated by chemical or physical activation. Both of these processes determine the adsorption capacity of activated carbon. The ability to adsorp a certain substance or substances, depends on the chemical and physical properties of the activated carbon (Strand, 2001).

### Chemical activation

Chemical activation is principally used for the activation of wood-based activated carbon and activated carbon made from stones, e.g. olive stone (Strand, 2001). The raw material is mixed with an activating and dehydrating substance, usually phosphoric acid and zinc chloride. Normally, activation temperature is at 500p C, and it can go up to 800 p C. In chemical activation, the precursor is impregnated with a given chemical agent and after that, is pyrolized. As a result of the pyrolysis process, a much richer carbon content material with much more ordered structure is produced, and once the chemical agent is eliminated after the heat treatment, the porosity is so much developed. Chemical activation is proven to be a very efficient method to obtain carbons with high surface area and narrow micropores distribution (Moon, 2006).

### Physical activation

Physical activation, steam or carbon dioxide method, is generally used for activation of carbon from peat, coal, coconut shell and wood. First, raw material is converted into char by carbonization. When coal is used as material in steam activation, it always consists of small graphite-like plate. Some of carbons are converted into gas, and leave pores (empty space). In steam activation, carbon is consumed because of reaction of steam ($H_2O$) with carbon (Paul, 2002). The form this takes depends largely on the raw material used. Hard material, like coconute shell leaves almost no micro pores. While soft materials like peat always produce a lot of many messo pores. For long period of steam blowing, more carbon turns to gas and leave empty space. In the beginning micro pores will be formed; when the process continues, the surounding carbon also turns into gas and the pores develop into meso pores. If the process is kept further, macro pores will be obtained. Activated carbon made from peat has both of meso and micro pores. Activated carbon made from bituminous coal also has both of meso and micro pores, and therefore has multipurpose character. Activated carbon based on lignite has many meso pores of 1-4 nm in size (Strand, 2001).

## Chemical Reaction in Activated Carbon Production

Chemical reactions occurred in the processes of the production of activated carbon consist of drying, pyrolisis and gasification (Patisson, 1999). The coal charged into the rotary kiln contains moisture, so that pyrolysis is preceded by drying. The pyrolysis of coal leads to the formation of three classes of products: char, tars and gases. Char is solid residue of the transformation process and is richer in carbon than the coal. Tars and gases are volatile matter and represent 4 to 45 % of the weight of the coal, depending on its type.

Coal is a complex natural organic substance; when heated, the weakest chemical bonds begin to break at 300°C to 400°C, producing molecular fragments, in a process termed depolymerization. The by-product of pyrolysis that is not vaporized is called char and consists mainly of fixed carbon and ash. The carbon remaining in the char is either reacted with steam or hydrogen or combusted with air or pure oxygen. Gasification with steam is more commonly called "reforming" and results in a hydrogen and carbon dioxide rich "synthesis" gas (syngas). Typically, the exothermic reaction between carbon and oxygen provides heat energy required to drive the pyrolysis and char gasification reactions (Klein, 2002). The basic gasification reactions that must be considered are as follows:

$C + O_2 \rightarrow CO_2$ -393 kJ/mol (exothermic)
$C + H_2O \rightarrow CO + H_2$ +131 kJ /mol (endothermic)
$C + CO_2 \rightarrow 2CO$ +172 kJ/mol (endothermic)
$C + 2H_2 \rightarrow CH_4$ -74 kJ/mol (exothermic)
$CO + H_2O \rightarrow CO_2 + H_2$ -41 kJ/mol (exothermic)
$CO + 3H_2 \rightarrow CH_4 + H_2O$ -205 kJ/mol (exothermic)
All of these reactions are reversible.

## Cyclone burner

A cyclone burner is a type of coal combustor (Figure 1) developed by *tek*MIRA. It consists of horizontal cylinder at one end of which crushed coal of passing 30 mesh screen is introduced and then picked-up by a tangential stream of air and brought into rapid rotary motion known as cyclone condition.

The coal burning in this burner has combustion characteristics nearly the same as the fuel oil combustion by an oil burner. The burning of coal emits long flame and the combustion rate may be controlled by a variable feeder. It may be shut of and then put on instantaneously as it is practiced in oil burner. Fine adjustment of the coal and air supply may be performed and this burner may be set up in vertical or horizontal position. It can be used to substitute oil burner in various industrial facilities, such as steam boiler, oil heater, rotary dryer, metal smelter and heat exchanger (Sumaryono, 2009). In the production of activated carbon, cyclone burner is used as the combustor and the direct fire is introduced into the rotary kiln.

Figure 1. Cyclone burner

## METHODOLOGY

### Equipment

The equipments used for the production of activated carbon is consisted of:

- 1 unit of rotary kiln of 1 ton/day equipped with feeder, cooler and scrubber (Figure 2); the dimensions of rotary kiln are 8 m length and 0.8 m internal diameter. The feeder is equipped with bucket elevator to transport coal feed into the rotary kiln. Air is used to cool down the product of carbonization or activation. While scrubber is used to separate particulate and tar.
- 1 unit boiler of 200 kg/hour capacity; it can produce steam at 130-140ºC and 5-6 bar pressure.
- Cyclone burner.
- Screen, for selecting suitable size of coal feed.
- Instruments for evaluation the quality of activated carbon.

Figure 2. Rotary kiln

**Materials used**

- The coal used was subbituminous coal obtained from Air Laya Field, PT Bukit Asam, South Sumatera. The coal was screened to obtain 3 cm size for carbonization and – 30 mesh size for cyclone burner. The analysis data of the coal sample is as follows:

  | | |
  |---|---|
  | Moisture, % adb | : 18.60 |
  | Ash, % adb | : 4.80 |
  | Volatile matter, % adb | : 38.60 |
  | Fixed carbon, % adb | : 38.00 |

- Chemicals for evaluation of activated carbon: iodine solution, potassium iodide, hydrochloric acid, sodium thiosulfate and disodium carbonate.

**Procedure**

The coal was firstly carbonized by feeding it at ± 60 kg/hour into the rotary kiln which has been heated by cyclone burner at 500-600ºC. The carbonization time was set for 2 hours by adjusting angle and rotation speed of the rotary kiln at 1 rpm. The flame of cyclone burner was directly contacted with coal, but the combustion was expected to occur only with the coal volatile matter. The carbonization temperature was achieved by manual adjustment of coal feed rate and air supply into the cyclone burner. In this case, the main target of carbonization is the solid product, char. After carbonization, char was screened to obtain variable sizes of +6, -6+12 and -12+20 mesh sizes for activation. Beside, the char is also ground to – 60 mesh size for its characterization consisting of fixed carbon, moisture, ash and volatile matter contents.

Activation process was started by feeding char into the rotary kiln which has been heated using cyclone burner at ± 900ºC. At the same time, steam was also introduced into the rotary kiln at 180 kg/hour. The feeding rate of char was varied at 17.5, 35.0 and 52.5 kg/hour, while the size was varied at +6, -6+12 and -12+20 mesh. The activation time depended on the feed rate of char, varied from 1.5 to 6 hours. Activated carbon produced was weighed to calculate the yield of activation. Beside, it was also sampled for characterization of product. During activation process, the product of activated carbon was sampled every one hour for iodine number determination.

**Characterization of Product**

Analysis of char consists of moisture, ash, volatile matter and fixed carbon. While, the preferred parameter quality of activated carbon analyzed is iodine number. This parameter of activated carbon is used to evaluate the effect of variable conditions to its quality and to determine the optimum process condition. Apart from iodine number, the optimum process condition is also determined using surface area, diameter and volume of pores of the product.

The characterization used to evaluate the product resulted from the optimum condition is based on the requirement of activated carbon according to the Indonesian Industrial Standard consisting of iodine number, methylene blue number, loss on ignition, moisture content, ash content, pure activated carbon content, bulk density and size (to pass 325 mesh screen).

## RESULTS AND DISCUSSION

**Carbonization and activation process**

The analysis results of char resulted from carbonization of Air Laya coal using rotary kiln and cyclone burner are shown in Table 1. As a comparison, it also shows the ideal requirement of charcoal as raw material of activated carbon according to Indonesian Industrial Standard (Standar Industri Indonesia, 1999). This standard (for charcoal) was used, since there is still no standard for char from coal. Table 1 shows that the quality of char met the requirement, especially its volatile matter, fixed carbon and moisture contents i.e.

17.5, 70 and 4.5% respectively compared with those of standard which are in the ranges of 15-20, 70-80 and 3-10% each. However, the ash content of char is higher than that of the requirement. This can be understood because ash content of the raw coal is already higher compared with the ideal requirement of standard. This is the disadvantage of using coal, because it normally contains higher ash than wood or coconut shell which are often generally used as raw material for activated carbon.

Table 1. Quality of char and the ideal requirement according to the standard

| Parameter | Unit | Char | Standard |
|---|---|---|---|
| Volatile matter | % | 17.54 | 15 – 20 |
| Fixed carbon | % | 70.23 | 70 – 80 |
| Moisture | % | 4.51 | 10-Mar |
| Ash | % | 8.57 | 1 – 2 |

The other reason of the high ash content of char is because of the combustion of carbon during carbonization process. It is expected that the energy for carbonization comes from the combustion of volatile matter. However, in direct carbonization process some carbon can also be burnt. In addition, it was more difficult to control carbonization temperature in the rotary kiln using coal fueled cyclone burner compared with the use of oil burner.

The ideal carbonization temperature for producing good char for activated carbon is in the range of 500-600ºC. During the experiment carbonization temperature was controlled by adjusting the coal introduced into the cyclone burner. The increase of temperature to higher than 600ÚC can cause the combustion of carbon which results into the formation of ash. In such conditions, the inverter and the air blower of cyclone burner should be turned off. However this makes the pressure in the kiln is greater than the atmospheric pressure, so that the hot gas goes out of the system and the temperature drops again to <500ºC. Therefore, the adjusment of cyclone burner condition should be done carefully. At the optimum condition(500-600ºC for 2 hours) the yield of carbonization obtained was about 50%.

**Activation Process**

It has been mentioned above that activation process was carried out at 900ºC using steam at 180kg/hour. In the activation process the particle size of char were varied (+6, -6 +12 and -12 + 20 mesh). The feed rate of char affected the residence time of char in the kiln. At the feed rate of 17.5 kg/hour the life time of char was 6 hours. While at the feed rate of 35 and 52.5 kg/hours, 3 and 1.5 hours activation processes were needed respectively. Therefore, the residence times of char i.e. 1.5, 3 and 6 hours were also used to vary the activation process time.

The effect of particle size and feed rate of char to the iodine number are shown in Figure 3. The iodine numbers shown in Figure 3 are the average number resulted from products which were sampled every one hour.

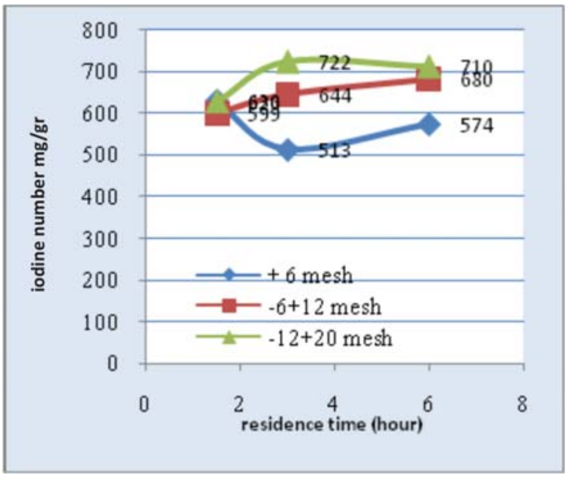


Figure 3. Effect of particle size and residence time of char towards iodine number

It shows that the smaller the particle size, the higher is the iodine number. Similarly, the longer the activation process, the higher is the iodine number. By using the particle size of + 6 mesh, the iodine number reached 722 mg/g at 3 hour residence time and 710 mg/g at 6 hour residence time respectively. While by using the particle size of -6+12 and +6 mesh, the iodine number resulted from 6 hour residence time were only 680 and 574 mg/g respectively. The results show that the best size of char for activation process was at -12+20 mesh. It can be understood that the longer the residence time, the more complete is the reaction between steam and carbon or volatile matter from char. Similarly, the smaller the particle size, the greater is the surface area of char which can react with steam. This condition produces a lot of pores in the product and then resulted in high adsoprtion capacity or iodine number.

Apart from high iodine number, the other parameter needed to be considered in comercializing activated carbon production is yield of activation. The longer the residence time, the higher is the iodine number of the product. However, the longer the residence time, the lower the yield of activation is there should be a balance between iodine number and yield of carbonization. Figure 4 shows the effect of resedence time to yield of activation at various particle size of the char. It can be seen that even at the particle size of -12+20 mesh tended to produce the higest iodine number, although it showed lower yield of activation. In contrast, the particle size of -6+12 mesh showed better yield of activation, although with lower iodine number. Therefore, for economic evaluation, the optimum condition is at -6+12 mesh particle size and 3 hours activation.

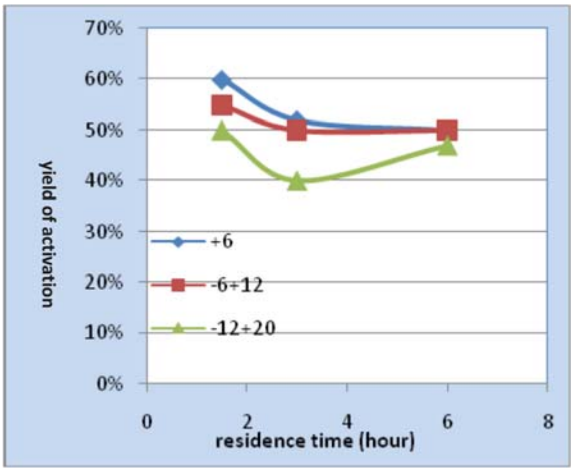


Figure 4. Effect of residence time to yield of activation

## Surface area and porosity of coal activated carbon

To know more specific about adsorption capacity, surface area and porosity of the best products, products made from -12+20 mesh size and 6 hours of residence times were analyzed.The surface area of active carbon is usually determined using Brunauer-Emmet-Teller (BET) method. The technique involves measuring the volume of nitrogen gas adsorbed at liquid nitrogen temperatures (78 K) at various pressures. From this isotherm it is possible to calculate (using the BET equation) the volume of nitrogen adsorbed at monolayer coverage since this area is covered by nitrogen (Juntgen, 1973). The analysis results of surface area, diameter and volume of pore are shown in Table 2.

It shows that the longer the residence time, the higher the surface area and the total volume of pores of product. The greatest surface area and total volume of pore i.e. 528 $m^2/g$ and 0.48 cc/g resulted from the activation of -12+20 mesh size char at 6 hous residence time. Converserly, The activation process of -12+20 mesh size char at 1.5 residence time resulted in smaller surface area and total volume of pores i.e. 452 $m^2/g$ and 0.39 mL/g respectively. These results can explain why adsorption capacity of certain product is high.

Table 3 also shows the diameter of pores ranges from 15.2 to 17.0 Å. which is classified as micro pore. Figure 5 shows the distribution of volume and diameter of pore of activated carbon made from coal (Juntgen, 1973).

The figure shows that the pore volume of these activated carbons is distributed over a wide range of pore diameters. Activated carbon made from bituminous coal characterized by larger micro and submicropore volume than those made from lig-

Table 2. Surface area, diameter and volume of pores analyzed using BET method*

| Residence time, Hour | Surface area, $m^2/g$ | Diameter of average pores, Å | Total volume of pores mL/g |
|---|---|---|---|
| 6 | 528 | 16.3 | 0.48 |
| 3 | 483 | 15.2 | 0.42 |
| 1.5 | 452 | 17 | 0.39 |

* Using -12+20 mesh size feed (char)

Table 3. Quality of product and Indonesian Industrial Standard

| Parameter | Unit | Product of experiment (with cyclone burner) | Product of former experiment (with oil burner)* | Indonesian Industrial Standard |
|---|---|---|---|---|
| Loss on heating at 950°C | % | 6 | 6,0 | 15-25 |
| Moisture | % | 10 | 5 | 15-Apr |
| Ash | % | 8 | 18 | 10-Feb |
| Iodine number | mg/g | 600-800 | 500-750 | 750-1200 (400-1200)** |
| Pure activated carbon | % | 75 | 75 | 60-80 |
| Benzene Adsorption | % | - | - | 25 |
| Methylene blue number | mg/g | 60 | 50 | 60-120 |
| Bulk density | g/ml | 0.52 | 0.53 | 0.30-0.55 |

* injection of steam only from 1 direction
**Quality found in the commercial market

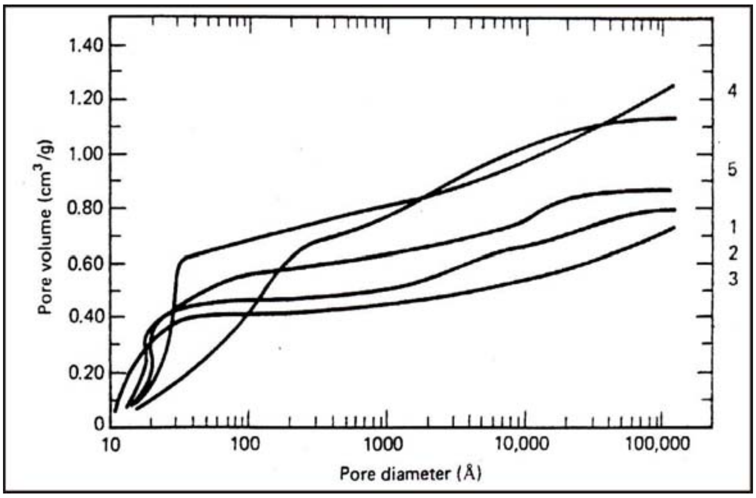


Figure 5. Pore volume distribution and diameter for various activated carbon made from coal

nite. Therefore, Table 2 explains that activated carbon produced from Air laya coal which is belong to subbtuminous coal has distribution of volume and pore type similar with those of bituminous coal.

## Quality of Product and Economic Evaluation

The quality of activated carbon produced using cyclone burner is shown in Table 3. As a comparison, the table also shows the quality of activated carbon resulted from activation using oil burner and the requirement of Indonesian Industrial Standard.

The activated carbon produced using cyclone burner showed a better quality compared with that produced using oil burner, especially the iodine number. Its iodine number was between 600-800 mg/g, while the product of oil burner was between 500-750 mg/g. Benzene adsorption were not tested because no equipment avalaible. Chemical characteristics of product (loss on heating, moisture and pure activated carbon) shows the results of 6, 10 and 75% respectively which met the standard requirement (i.e. 15-25, 4-15 and 60-80% respectively). The bulk density of both products were similar, 0.52 and 0.53 mg/ml each, and met the requirement of standard i.e. 0.30-0.55 mg/ml.

The product of activated carbon using cyclone burner can be utilized for liquid waste treatment. The results of pre utilization test of the product showed that it can be used for the treatment of water from shrimp farms. The product used to reduce the pH from 8.70 down to 7.14, alkalinity from 139 to 74 ppm, and N-$NH_4^+$ from 0.27 ppm to <0.016 ppm. The treated water met the standard of water for shrimp farms.

The economic evaluation showed that the process using cyclone is cheaper than that using oil burner. Activation process of 1 ton char using oil burner at 900ºC consumed 720 liters of oil fuel. While the activation process using cyclone burner at the same condition needed 1,440 kg of coal. Assuming that the oil and coal prices are Rp 7,000/l and Rp 1,250/kg respectively, the saving of fuel for activation process is about Rp 2,820,000 per ton of char.or more than 64%.

## CONCLUSION

– Production of activated carbon from subbituminous coal using rotary kiln and coal fueled cyclone burner for carbonization as well as activation processes has been carried out successfully.

– The Activated carbon produced showed fairly good quality and met the requirement according to Indonesian Industrial Standard, the iodine number was better than that produced using oil burner.

– The use of cyclone burner saved the fuel cost of activation process of up to 64% compared with the use of oil burner.

– Pre utilization test of the product in the laboratory as well as directly in the field showed that it can be used for treatment of shrimp farms water.

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