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(11) **EP 1 087 058 A2**

(12) **EUROPEAN PATENT APPLICATION**

(43) Date of publication:
28.03.2001 Bulletin 2001/13

(51) Int Cl.7: **D21H 27/16**

(21) Application number: **00308258.3**

(22) Date of filing: **21.09.2000**

(84) Designated Contracting States:
**AT BE CH CY DE DK ES FI FR GB GR IE IT LI LU
MC NL PT SE**
Designated Extension States:
AL LT LV MK RO SI

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(30) Priority: **22.09.1999 JP 26946899**

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(54) **Completely combustible paper**

(57) A paper having an iron compound catalyst in an amount of 0.1 to 20% by weight,

said iron compound catalyst comprising iron oxide hydroxide particles or iron oxide particles having an average particle size of 0.01 to 2.0 μm ; and having a catalytic activity capable of converting not less than 15% by weight of carbon monoxide into carbon dioxide, when 2.8×10^{-4} mole of iron oxide particles obtained by heat-

treating said iron compound catalyst at a temperature of 800°C for 15 minutes in air, are instantaneously contacted with 6.1×10^{-7} mole of carbon monoxide at a temperature of 250°C at a space velocity of 42,400 h^{-1} in an inert gas atmosphere using a pulse catalytic reactor.

The paper of the present invention not only maintains inherent functions as papers, but also can be completely burned and inhibited from generating harmful substances upon incineration thereof after use.

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Description

[0001] The present invention relates to a paper capable of being completely combusted with a less amount of combustion residues or cinders, and being inhibited from generating harmful substances, upon incineration thereof.

[0002] Hitherto, human lives have a close relationship to papers to such an extent that the consumption of papers would indicate a degree of culture of the country. At present, papers have been widely used in various applications such as books, printed matters, wrapping materials, industrial materials or the like. There is a recent tendency that the consumption of papers is more and more increased. With such a tendency, various papers having new functions have been successively developed and marketed. Although various studies have been conducted to improve functions and performances of papers, there are almost no studies for developing those papers suitable for waste disposal after use. Main reasons therefor are that the disposal of papers is not considered to cause any environmental pollution since the papers are inherently made from wood pulps as main raw materials; the papers can be naturally decomposed when buried in the earth for a long period time; and the papers are relatively easy to burn, and free from the generation of harmful substances upon incineration thereof since the papers are made from the substantially same material as wood.

[0003] On the other hand, in waste disposal treatments, especially in waste incineration treatments, the discharge of exhaust gases containing carbon monoxide and/or nitrogen oxides and the generation of dioxins cause significant social problems. In order to solve these problems, earnest studies have been directed to the reduction of wastes and the improvements in incinerators and incineration methods. Simultaneously, various material makers have made efforts for developing materials which show excellent combustion properties and are prevented from causing incomplete combustion and generation of harmful substances upon incineration, from such a viewpoint that these materials must be inevitably incinerated after use. For example, there have been proposed a plastic waste bag made from a thermoplastic resin film containing specific iron oxide particles having a good combustion-promoting capability in an amount of 0.1 to 20.0% by weight (Japanese Patent Application Laid-Open (KOKAI) No. 7-257594(1995)). However, such proposals have been limited to plastic waste bags only, and there have not been studied any method of improving combustion properties of papers which occupy a considerable part of wastes treated in an incinerator.

[0004] Thus, conventionally, the problems caused upon incineration of papers have not positively been studied for such a reason that the papers are considered to be identical in nature to that of natural wood materials. However, when many stacked papers, e.g., books, pamphlets, catalogues or the like are incinerated, there is a tendency that only the surface portion of the stacked papers can be burned and the inside portion thereof remains unburned. In such a case, there also arises the problem due to incomplete combustion. Further, in the case where chlorine-based compounds used in paper making processes or lignin contained in wood pulps still remain in papers, there might be caused such a problem that precursors of dioxin are produced from these substances upon incineration of the papers.

[0005] As a result of the present inventors' earnest studies for solving the above problems, it has been found that by coating a paper with an iron compound catalyst comprising iron oxide hydroxide particles or iron oxide particles having an average particle size of 0.01 to 2.0 μm , the obtained paper can be completely combusted with a less amount of combustion residues or cinders and inhibited from generating harmful substances upon incineration thereof. The present invention has been attained on the basis of this finding.

[0006] It is an object of the present invention to provide a paper capable of not only maintaining inherent functions as papers, but also being completely burned and inhibited from generating harmful substances upon incineration thereof after use.

[0007] To accomplish the aim, in a first aspect of the present invention, there is provided a paper having an iron compound catalyst in an amount of 0.1 to 20% by weight,

said iron compound catalyst comprising iron oxide hydroxide particles or iron oxide particles having an average particle size of 0.01 to 2.0 μm ; and having a catalytic activity capable of converting not less than 15% by weight of carbon monoxide into carbon dioxide, when 2.8×10^{-4} mole of iron oxide particles obtained by heat-treating said iron compound catalyst at a temperature of 800°C for 15 minutes in air, are instantaneously contacted with 6.1×10^{-7} mole of carbon monoxide at a temperature of 250°C at a space velocity (SV) of 42,400 h^{-1} in an inert gas atmosphere using a pulse catalytic reactor.

[0008] The present invention will now be described in detail below.

[0009] The iron compound catalyst used in the present invention has an average particle size of usually 0.01 to 2.0 μm , preferably 0.05 to 1.0 μm , more preferably 0.05 to 0.5 μm .

[0010] When the average particle size of the iron compound catalyst is less than 0.01 μm , the iron compound catalyst be difficult to handle, and further the yield of paper-making process may be deteriorated. When the average particle size of the iron compound catalyst is more than 2.0 μm , the obtained paper may be deteriorated in surface smoothness. In addition, since a specific surface area of such an iron compound catalyst becomes small, the oxidation activity thereof for converting carbon monoxide into carbon dioxide may be also deteriorated, thereby failing to attain the aimed effect of causing the obtained paper to be completely combusted.

[0011] The iron compound catalyst used in the present invention has a BET specific surface area value of usually

0.2 to 200 m²/g, preferably 0.5 to 200 m²/g, more preferably 0.5 to 100 m²/g.

[0012] The iron compound catalyst used in the present invention comprises at least one material selected from the group consisting of iron oxide hydroxide particles such as goethite particles, akaganeite particles and lepidocrocite particles, and iron oxide particles such as hematite particles, maghemite particles and magnetite particles. Among these materials, goethite particles is preferred.

[0013] The shape of the iron oxide hydroxide particles or iron oxide particles constituting the iron compound catalyst used in the present invention is a granular shape, a spherical shape, a spindle shape, an acicular shape or the like. Among these particles, the use of spindle-shaped particles is preferred.

[0014] The iron compound catalyst used in the present invention has a phosphorus content of usually 0.0001 to 0.02% by weight, preferably 0.0001 to 0.01% by weight, more preferably 0.0001 to 0.005% by weight. When the phosphorus content is more than 0.02% by weight, the catalyst poison ability of the phosphorus may become large, so that the oxidation activity for converting carbon monoxide into carbon dioxide may be deteriorated, thereby failing to obtain the aimed effect of causing the obtained paper to be completely combusted. Meanwhile, although the phosphorus content is preferably as low as possible, it is difficult to industrially produce such an iron compound catalyst having a phosphorus content of less than 0.0001% by weight.

[0015] The iron compound catalyst used in the present invention has a sulfur content of usually 0.001 to 0.3% by weight, preferably 0.001 to 0.1% by weight, more preferably 0.001 to 0.07% by weight. When the sulfur content is more than 0.3% by weight, the catalyst poison ability of the sulfur may become large, so that the oxidation activity for converting carbon monoxide into carbon dioxide may be deteriorated, thereby failing to obtain the aimed effect of causing the obtained paper to be completely combusted. Meanwhile, although the sulfur content is preferably as low as possible, it is difficult to industrially produce such an iron compound catalyst having a sulfur content of less than 0.001% by weight.

[0016] The iron compound catalyst used in the present invention has a sodium content of usually 0.001 to 0.3% by weight, preferably 0.001 to 0.2% by weight, more preferably 0.001 to 0.15% by weight. When the sodium content is more than 0.3% by weight, the catalyst poison ability of the sodium may become large, so that the oxidation activity for converting carbon monoxide into carbon dioxide may be deteriorated, thereby failing to obtain the aimed effect of causing the obtained paper to be completely combusted. Meanwhile, although the sodium content is preferably as low as possible, it is difficult to industrially produce such an iron compound catalyst having a sodium content of less than 0.001% by weight.

[0017] The iron compound catalyst used in the present invention exhibits a catalytic activity capable of converting usually not less than 15% by weight, preferably not less than 18% by weight, more preferably not less than 20% by weight of carbon monoxide into carbon dioxide, when 2.8×10^{-4} mole of iron oxide particles obtained by heat-treating said iron compound catalyst at a temperature of 800°C for 15 minutes in air, are instantaneously contacted with 6.1×10^{-7} mole of carbon monoxide at a temperature of 250°C at a space velocity (SV) of 42,400 h⁻¹ in an inert gas atmosphere using a pulse catalytic reactor. The upper limit of the catalytic activity for converting the carbon monoxide into carbon dioxide is 50% by weight.

[0018] As the paper used in the present invention, a paper produced by the followings may be exemplified.

[0019] Ground pulps, alkaline pulps, sulfite pulps, chemical ground pulps and semi-chemical ground pulps are obtained from plant-based raw materials such as wood chips, straws, reed, bagasse or bamboo, waste papers or rags, which wood chips may be optionally subjected to pre-treatments such as bleaching treatment with chlorine, hypochlorite, sodium chlorite, chlorine dioxide, hydrogen peroxide, sodium peroxide or the like, and screening treatment. At least one material selected from the above-described pulps is appropriately hammer-crushed (after being mixed together, if two or more pulp materials are used) in order to vary or modify a fiber shape and colloidal property thereof. The hammer-crushed materials are further mixed with colorants, sizing agents or fillers to prepare a paper material. The paper material is then subjected to a paper-making process and then to finishing treatment, thereby producing the paper.

[0020] The paper of the present invention has the above iron compound catalyst in an amount of usually 0.1 to 20.0% by weight, preferably 0.5 to 10.0% by weight, more preferably 0.5 to 5.0% by weight based on the weight of the paper of the present invention. When the amount of the iron compound catalyst is less than 0.1% by weight, it may be difficult to obtain the aimed effect of the present invention. When the amount of the iron compound catalyst is more than 20.0% by weight, the obtained paper may be considerably deteriorated in coloring property, strength and surface smoothness.

[0021] The paper of the present invention has a combustion velocity of preferably not more than 3.0 minutes, more preferably not more than 2.5 minutes; an amount of the residual combustible components of preferably not more than 10.0 % by weight, more preferably not more than 9.0 % by weight based on the weight of the initially charged combustible components; and a low-temperature combustion property of preferably not more than 530°C, more preferably not more than 520°C, which are measured by the combustion test described in the Example 1.

[0022] Next, the process for producing the iron compound catalyst used in the present invention is described.

[0023] Among the iron compound catalysts used in the present invention, the goethite particles may be produced, for example, by passing an oxygen-containing gas such as air through a suspension containing a ferrous iron-containing

precipitate such as hydroxides of iron or iron carbonates which are produced by reacting a ferrous salt with at least one compound selected from the group consisting of alkali hydroxides, alkali carbonates and ammonia. In case of using ferrous sulfate, the obtained goethite particles are desired to be subject to purifying treatment, i.e. sufficient washing treatment with water in order to lessen the sulfur content.

[0024] Examples of the ferrous salts may include ferrous nitrate, ferrous acetate, ferrous oxalate, ferrous sulfate or the like. Among these ferrous salts, the use of ferrous nitrate, ferrous acetate, ferrous oxalate, etc. which are free from elements acting as catalyst poisons such as phosphorus, sulfur or the like, is preferred.

[0025] Examples of the alkali hydroxides may include sodium hydroxide, potassium hydroxide, calcium hydroxide, magnesium hydroxide or the like. Examples of the alkali carbonates may include sodium carbonate, potassium carbonate or the like.

[0026] Among the iron compound catalysts used in the present invention, the hematite particles may be produced, for example, by heat-dehydrating or heat-treating the above obtained goethite particles at a temperature of usually 200 to 800°C in air; the magnetite particles may be produced, for example, by heat-reducing the above obtained hematite particles at a temperature of usually 300 to 800°C in a reducing atmosphere; and the maghemite particles may be produced, for example, by heat-oxidizing the above obtained magnetite particles at a temperature of usually 200 to 600°C in air.

[0027] In the production of the iron compound catalyst used in the present invention, it is necessary to lessen the contents of phosphorus, sulfur and sodium as catalyst poisons to not more than predetermined amounts. More specifically, the contents of phosphorus, sulfur and sodium should be reduced by avoiding the use of sodium hexameta-phosphate usually added as a sintering preventive upon heat-calcination step, and by removing sulfur ions derived from the raw ferrous materials or sodium ions derived from alkali hydroxides or the alkali carbonates by means of purification treatments such as washing with water or the like.

[0028] The paper of the present invention has the above iron compound catalyst in an amount of usually 0.1 to 20.0% by weight, preferably 0.5 to 10.0% by weight, more preferably 0.5 to 5.0% by weight based on the weight of the paper of the present invention. When the amount of the iron compound catalyst is less than 0.1% by weight, it may be difficult to obtain the aimed effect of the present invention. When the amount of the iron compound catalyst is more than 20.0% by weight, the obtained paper may be considerably deteriorated in coloring property, strength and surface smoothness.

[0029] The paper of the present invention can be produced by the following method.

[0030] As the raw material of the paper, there may be usually used, for example, at least one material selected from the group consisting of ground pulps, alkaline pulps, sulfite pulps, chemical ground pulps, semi-chemical ground pulps and the like. These pulps are obtained from plant-based raw materials such as wood chips, straws, reed, bagasse or bamboo, waste papers or rags, which wood chips may be optionally subjected to pre-treatments such as bleaching treatment with chlorine, hypochlorite, sodium chlorite, chlorine dioxide, hydrogen peroxide, sodium peroxide or the like, and screening treatment. At least one material selected from the above-described pulps is appropriately hammer-crushed (after being mixed together, if two or more pulp materials are used) in order to vary or modify a fiber shape and colloidal property thereof. The hammer-crushed materials are further mixed with colorants, sizing agents or fillers to prepare a complete paper material. The paper material is then subjected to a paper-making process and then to finishing treatment, thereby producing the paper. The iron compound catalyst may be introduced into the paper material by replacing a part or whole of the colorants and/or fillers used.

[0031] Alternatively, in the case where the aimed paper is a coated paper, the iron compound catalyst may be added to a coating solution or an adhesive, and then the obtained coating solution or adhesive is coated on the paper to produce a coated paper. Further, in the case of the laminated paper, the iron compound catalyst may be incorporated into plastic films to be laminated on the paper. However, the present invention is not limited to those papers produced by the above-specified processes, and may be extensively applied to various kinds of papers and various application fields.

[0032] The reason why the paper having the iron compound catalyst according to the present invention can exhibit excellent combustion properties, is considered by the present inventors as follows.

[0033] The combustion mechanism of the present invention is considered as follows. That is, iron atoms present on the surface of each iron compound catalyst particle are initially kept stable by surface hydroxyl groups capable of dissociating and adsorbing water. When these hydroxyl groups undergo dehydration by the heating in the combustion process, coordination-unsaturated iron ions and oxygen ions are produced. Then, the thus produced coordination-unsaturated active sites can activate oxygen due to the oxygen adsorption thereto caused during the combustion process, and show a good catalytic activity in a series of reaction steps such as dehydrogenation reaction from organic substances or the like, thereby exhibiting a good combustion-promoting effect.

[0034] The paper having the iron compound catalyst according to the present invention has the following advantages. That is, when the paper is incinerated after its inherent use as paper, the iron compound catalyst can exhibit a good combustion-promoting effect such that the paper can be completely combusted with less amounts of combustion residues (cinders) and residual ashes without significant deterioration of combustion efficiency even in the case where

an incinerator is operated under low-temperature and low-oxygen concentration conditions which are considered to be effective for reducing the amount of NO_x and preventing damage to the incinerator.

[0035] Further, even when chlorine-based compounds used in paper-making process or lignin contained in wood pulps still remain in the paper, it is possible to inhibit the production of dioxin precursors upon incineration.

EXAMPLES

[0036] The present invention is described in more detail by Examples and Comparative Examples, but the Examples are only illustrative and, therefore, not intended to limit the scope of the present invention.

[0037] Various properties were measured by the following methods.

[0038] (1) The average particle size of the iron compound catalyst was expressed by the average of values measured from an electron micrograph. The specific surface area of the iron compound catalyst was expressed by the value measured by a BET method.

[0039] (2) The contents of phosphorus and sodium contained in particles constituting the iron compound catalyst were expressed by the values measured by an inductively coupled plasma atomic emission spectrometer (SPS-4000 Model, manufactured by Seiko Denshi Kogyo Co., Ltd.).

[0040] (3) The content of sulfur contained in particles constituting the iron compound catalyst was expressed by the value measured by a Carbon-Sulfur Analyzer (EMIA-2200 Model, manufactured by Horiba Seisakusho Co., Ltd.).

[0041] (4) The catalytic activity of the iron compound catalyst was measured by the following method.

[0042] That is, 2.8×10^{-4} mole of iron oxide particles (α -Fe₂O₃) obtained by heat-treating the iron compound catalyst at a temperature of 800°C for 15 minutes in air, were instantaneously contacted with 6.1×10^{-7} mole of carbon monoxide at a temperature of 250°C at a space velocity (SV) of 42,400 h⁻¹ in an inert gas atmosphere using a pulse catalytic reactor. The catalytic activity of the iron compound catalyst was expressed by the percentage (conversion) of carbon monoxide converted into carbon dioxide in the above process.

[0043] Here, the space velocity (SV) represents the value obtained by dividing a flow rate of the reaction gas by a volume of the catalyst, and is expressed by a unit of an inverse number of time (h⁻¹).

[0044] The pulse catalytic reactor used comprises a reactor portion and a gas chromatography portion which is constituted by Gas Chromatography GC-16A (manufactured by Shimadzu Seisakusho Co., Ltd.).

[0045] Meanwhile, the above evaluation method was conducted by referring to methods described in the literatures (e.g., R. J. Kobes, et al, "J. Am. Chem. Soc.", 77, 5860(1955) or "Experimental Chemistry II-Reaction and Velocity" edited by Chemical Society of Japan and published by Maruzen, Tokyo (1993)).

Example 1:

<Production of paper>

[0046] A filter paper (weight: 0.50 g; No.2, manufactured by Advantec Toyo Co., Ltd.) for laboratory chemical experiments was coated with a coating solution prepared by dispersing 1.0 g of goethite particles as iron compound catalyst A (average particle size: 0.25 μm; phosphorus content: 0.002 % by weight; sulfur content: 0.05 % by weight; sodium content: 0.08 % by weight; BET specific surface area: 85 m²/g; catalytic activity by the above evaluation method: 20 %) in 100 ml of water, and then dried in air at a temperature of 60°C for 1 hour, thereby obtaining a paper having 5.0 mg of the iron compound catalyst (1.0 % by weight).

<Combustion test>

[0047] 10 mg of the thus obtained paper having the iron compound catalyst was weighed, and heated at a temperature rise rate of 10°C/minute in air supplied at a flow rate of 300 ml/minute. The change in weight of the paper during the heating treatment was measured by a thermogravimetric analyzer (manufactured by Seiko Denshi Kogyo Co., Ltd.). The combustion properties of the paper were evaluated by measuring the combustion velocity, amount of residual combustible components and low-temperature combustion property.

[0048] Here, the combustion velocity represents the time required from the initiation of abrupt reduction in weight up to the termination thereof (the combustion of the paper is considered to take place during such a time).

[0049] The amount of residual combustible components (it is considered that the lower the value, the smaller the amount of combustion residues (cinders) or residual ashes after the incineration), is expressed by the residual weight percentage (wt%) calculated as the percentage of combustible components which still remain unburned upon the termination of abrupt reduction in weight, based on the weight of initially charged combustible components.

[0050] The low-temperature combustion property (which is considered to be a temperature at which organic substances are completely burned out) is expressed by the temperature at which the reduction in weight is no longer

caused.

[0051] The combustion velocity was 2.1 minutes; the amount of residual combustible components was 7.5 % by weight; and the low-temperature combustion property was 495 °C.

Examples 2 to 7 and Comparative Examples 1 to 3:

[0052] The same procedure as defined in Example 1 was conducted except that the filter paper for laboratory chemical experiments was surface-coated with coating solutions respectively containing the iron compound catalysts shown in Table 1, thereby obtaining papers each containing the predetermined amount of the iron compound catalyst in the filter paper.

[0053] The thus obtained papers were subjected to the combustion test by the same method as defined in Example 1. The combustion properties of the obtained papers are shown in Table 2.

[0054] As is apparent from Table 2, the iron compound catalyst-containing papers of the present invention obtained in Examples 2 to 6 were excellent in all of combustion velocity, amount of residual combustible components and low-temperature combustion property as compared to those of the paper obtained in Comparative Example 1 containing no iron compound catalyst. This indicates that the papers of the present invention can be completely combusted with a less amount of combustion residues or cinders upon incineration. Further, both of the papers obtained in Comparative Examples 2 and 3 containing the iron compound catalyst out of the range specified by the present invention, failed to exhibit improved combustion properties.

Table 1

Iron compound catalyst	Properties of iron compound catalyst		
	Kind	Average particle size (μm)	BET specific surface area (m ² /g)
Iron compound A	Acicular goethite	0.25	85
Iron compound B	Spindle-shaped goethite	0.25	83
Iron compound C	Spindle-shaped hematite	0.30	80
Iron compound D	Acicular maghemite	0.40	40
Iron compound E	Spindle-shaped goethite	0.30	71
Iron compound F	Acicular goethite	0.30	54

Table 1 (continued)

Iron compound catalyst	Properties of iron compound catalyst			
	Phosphorus content (wt. %)	Sulfur content (wt. %)	Sodium content (wt. %)	Catalytic activity (conversion of CO) (%)
Iron compound A	0.002	0.05	0.08	20
Iron compound B	0.0004	0.01	0.05	25
Iron compound C	0.002	0.01	0.07	22
Iron compound D	0.005	0.02	0.05	25
Iron compound E	0.49	0.08	0.18	0.5
Iron compound F	0.01	0.01	0.60	0.2

Table 2

Examples and Comparative Examples	Iron compound catalyst	
	Kind	Amount (wt. %)
Example 2	B	1.0
Example 3	C	1.0
Example 4	D	1.0
Example 5	B	0.5
Example 6	B	5.0
Comparative Example 1	-	None
Comparative Example 2	E	1.0
Comparative Example 3	F	1.0

Table 2 (continued)

Examples and Comparative Examples	Combustion properties		
	Combustion velocity (min.)	Amount of residual combustible components (wt. %)	Low-temperature combustion property (°C)
Example 2	1.95	7.2	493
Example 3	2.00	7.3	498
Example 4	1.98	7.1	492
Example 5	2.34	8.2	517
Example 6	1.62	2.7	400
Comparative Example 1	3.21	10.9	545
Comparative Example 2	3.05	10.0	530
Comparative Example 3	3.09	10.2	532

Claims

1. Paper having an iron compound catalyst in an amount of 0.1 to 20% by weight, said iron compound catalyst comprising particles which severally may be of the same or different composition, each particle comprising iron oxide hydroxide and/or iron oxide; the particles having an average particle size of 0.01 to 2.0 μm , and having a catalytic activity such that not less than 15% by weight of carbon monoxide is converted into carbon dioxide when 2.8×10^{-4} mole of iron oxide-based particles, obtained by heat-treating said catalyst at a temperature of 800°C for 15 minutes in air, are contacted with 6.1×10^{-7} mole of carbon monoxide at a temperature of 250°C at a space velocity of 42,400 h^{-1} using a pulse catalytic reactor.
2. Paper according to claim 1, wherein said iron compound catalyst further has a BET specific surface area of 0.2 to 200 m^2/g .
3. Paper according to claim 1 or 2, wherein said iron compound catalyst comprises at least one of goethite particles, akaganeite particles, lepidocrocite particles, hematite particles, maghemite particles and magnetite particles.
4. Paper according to claim 1, 2 or 3 wherein said iron compound catalyst has a phosphorus content of 0.0001 to 0.02%, a sulfur content of 0.001 to 0.3% and a sodium content of 0.001 to 0.3%, each by weight based on the

weight of the catalyst.

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