(54) Title: METHOD OF MANUFACTURING HIGH-PURITY PENTAERYTHRITOL

(57) Abstract: The present invention relates to a method of manufacturing high-purity pentaerythritol, and, more particularly, to a method of manufacturing high-purity pentaerythritol having a purity of 99.9 wt%, in which a pentaerythritol solution containing impurities is vaporized to recrystallize pentaerythritol, and then the recrystallized pentaerythritol undergoes natural precipitation, filter-pressing, centrifugal separation and drying processes, and then the resulting pentaerythritol is separated according to particle size using a two-stage screen separator to yield high-purity pentAPTERITOL.
— with international search report (Art. 21(3))
Description

METHOD OF MANUFACTURING HIGH-PURITY PENTAERYTHRITOL

Technical Field

The present invention relates to a method of separating and refining pentaerythritol, and, more particularly, to a method of manufacturing high-purity pentaerythritol, in which a pentaerythritol solution containing impurities is vaporized to recrystallize pentaerythritol, and then the recrystallized pentaerythritol undergoes natural precipitation, filter-pressing, centrifugal separation and drying processes, and then the resulting pentaerythritol is separated according to particle size using a two-stage screen separator to obtain high-purity pentaerythritol.

Background Art

Pentaerythritol is represented by the chemical formula C(CH₂OH)₄, and has a molecular weight of 136.15, a specific gravity of 1.395 and a melting point of 260°C. Pentaerythritol is chiefly made from acetaldehyde (CH₃CHO) and formaldehyde (HCHO). That is, pentaerythritol is obtained by condensing one acetaldehyde molecule and four formaldehyde molecules using an alkali catalyst (sodium hydroxide or potassium hydroxide).

Pentaerythritol is used as a raw material of polyurethane foam (foamed plastic), coating agents, adhesives, sealants, flame retardants, explosives, lubricants, cosmetics, medicine, fiber or the like, and thus has various uses. Further, pentaerythritol is increasingly used as a raw material of special chemicals too.

Currently, low-purity pentaerythritol containing 90 - 95% of monopentaerythritol is commercially available. However, in order to convert the low-purity pentaerythritol into high-purity pentaerythritol containing 99.9% or more of monopentaerythritol, there is a problem in that a recrystallization process must be conducted two or three times, so that a manufacturing system becomes complicated and a manufacturing cost is increased.

In order to solve the above problem, Korean Patent Registration No. 0716541, which was filed by the present applicant and is integrated with the present invention, discloses "a method of manufacturing high-purity pentaerythritol", in which pentaerythritol is refined and separated using a vaporizing concentrating crystallizing apparatus, a centrifugal separator, a hot air drier and a two-stage screen separator. As described above, since pentaerythritol has many uses and the demand thereof is increasing, the quality of products is determined according to the improvement of the purity of pentaerythritol and the productivity of pentaerythritol is also directly related.
to the improvement of the purity of pentaerythritol. In particular, high-purity pentaerythritol is required to manufacture chemical reagents or medical supplies. Therefore, it is continuously required to develop methods of remarkably increasing the purity of pentaerythritol at low cost.

Disclosure

Technical Problem

Accordingly, the present invention has been made based on the necessity of improving the purity of pentaerythritol, and an object of the present invention is to provide a method of manufacturing high-purity pentaerythritol, which has high energy efficiency because several recrystallization processes are not performed, and which is economical because additional complicated apparatuses are not required. In particular, the present invention provides a method of manufacturing high-purity pentaerythritol having a purity of 99.9 wt% or more.

Technical Solution

In order to accomplish the above object, an aspect of the present invention provides a method of manufacturing high-purity pentaerythritol, including the steps of: mixing raw pentaerythritol having a monopentaerythritol content of 85 - 90 wt% with pure water in a vaporizing concentrating crystallizing apparatus, dissolving the raw pentaerythritol in the pure water and then vaporizing and concentrating the mixed solution to form a crystalline suspension liquid; slowly stirring the crystalline suspension liquid for a predetermined time to naturally precipitate crystals and then filter-pressing a suspended mother liquid located at an upper portion of the vaporizing concentrating crystallizing apparatus to form a purified mother liquid and then reintroducing the purified mother liquid into the vaporizing concentrating crystallizing apparatus; stirring the crystalline suspension liquid and the purified mother liquid which was reintroduced into the vaporizing concentrating crystallizing apparatus; sending the crystalline suspension liquid to a centrifugal separator while maintaining the crystalline suspension liquid at a temperature of 60 ~ 90°C and then centrifugally separating the crystalline suspension liquid into monopentaerythritol and by-products such as dipentaerythritol and tripentaerythritol; drying the separated monopentaerythritol crystals using a hot air drier to remove water therefrom; and separating and then obtaining the dried monopentaerythritol crystals which were blown on by the hot air drier into monopentaerythritol crystals having a different crystal size from each other using a two-stage screen separator equipped with a primary screen and a secondary screen which differ in mesh size.

Here, the method of manufacturing high-purity pentaerythritol may further include the step of: recirculating the monopentaerythritol crystals which failed to pass through
the primary screen and the monopentaerythritol crystals which passed through the secondary screen to the vaporizing concentrating crystallizing apparatus.

The step of reintroducing the purified mother liquid into the vaporizing concentrating crystallizing apparatus may be performed by transferring the suspended mother liquid located at the upper portion of the vaporizing concentrating crystallizing apparatus to a suspended mother liquid tank, and then transferring the suspended mother liquid from the suspended mother liquid tank to a purified mother liquid tank through a filter press to turn the suspended mother liquid into a purified mother liquid, and then sending the purified mother liquid to the vaporizing concentrating crystallizing apparatus.

The two-stage screen separator may be equipped with the primary screen having a size of 10 meshes and the secondary screen having a size of 30 meshes so that the monopentaerythritol crystals may be separated into monopentaerythritol crystals having a particle size larger than 10 meshes, monopentaerythritol crystals having a particle size of 10 ~ 30 meshes and monopentaerythritol crystals having a particle size smaller than 30 meshes.

Advantageous Effects

According to the present invention, since several recrystallization processes are not required in order to refine pentaerythritol, there is an advantage in that the productivity of pentaerythritol and the energy efficiency are increased.

Further, according to the present invention, since a natural precipitation method and a storage tank and a filter press, which are simple apparatuses, are used in order to improve the purity of pentaerythritol, there are advantages in that it is economical because additional complicated apparatuses are not required and in that high-quality pentaerythritol having a high purity of 99.9 wt% or more can be obtained. Therefore, the high-purity pentaerythritol obtained using the method of the present invention can be suitably used to manufacture medical supplies or chemical reagents.

Brief Description of Drawings

FIG. 1 is a block diagram showing a process of manufacturing high-purity pentaerythritol according to the present invention; and

FIG. 2 is a schematic view showing a concentrating system and a refining system according to the present invention.

< Description of the elements in the drawings >

110: vaporizing concentrating crystallizing apparatus
111: steam heater
112: stirrer
113: motor
115: steam outlet
[21] 116: pure water inlet
[22] 117: control valve
[23] 120: centrifugal separator
[24] 130: hot air drier
[25] 140: screen separator
[26] 141: primary screen
[27] 142: secondary screen
[28] 143: high-purity pentaerythritol outlet
[29] 150: recovery pipe
[31] 170: filter press
[32] 180: purified mother liquid

Best Mode for Carrying out the Invention

[33] Hereinafter, a preferred embodiment of the present invention will be described in detail with reference to the attached drawings. FIG. 1 is a block diagram showing a process of manufacturing high-purity pentaerythritol according to the present invention, and FIG. 2 is a schematic view showing a concentrating system and a refining system according to the present invention.

[34] As shown in FIG. 1, a method of manufacturing high-purity pentaerythritol according to the present invention includes the steps of: dissolving, vaporizing, concentrating and crystallizing dry pentaerythritol (ST1); naturally precipitating the crystal particles in the crystallized pentaerythritol solution and then reintroducing a purified mother liquid obtained by filter-pressing a suspended mother liquid formed at the upper portion of the crystallized pentaerythritol solution into a vaporizing concentrating crystallizing apparatus and then stirring the purified mother liquid to prepare a crystal solution (ST2); separating the crystal solution using a centrifugal separator to obtain monopentaerythritol crystals (ST3); drying the monopentaerythritol crystals using hot air (ST4); selecting desired-sized crystal particles and then circulating residual crystal particles to the dissolving step using a two-stage screen separator equipped with a primary screen and a secondary screen (ST5); carefully selecting and packaging the monopentaerythritol obtained from the screen separator (ST6); and storing and delivering the packaged monopentaerythritol product (ST7).

[35] Hereinafter, a process of manufacturing high-purity pentaerythritol will be described according to the above steps in relation to a specific manufacturing system with reference to FIG. 2. First, a system for manufacturing high-purity pentaerythritol of the present invention largely includes a vaporizing concentrating crystallizing apparatus 110, a suspended mother liquid tank 160, a filter press 170, a purified mother liquid
tank 180, a centrifugal separator 120, a hot air drier 130 and a screen separator 140. The vaporizing concentrating crystallizing apparatus 110, which serves to mix low-purity pentaerythritol, which is a raw material, with pure water and then to dissolve the mixture and then to vaporize, concentrate and crystallize the dissolved mixture, includes a raw material inlet 114 for introducing low-purity pentaerythritol, steam outlet 115, pure water inlet 116, a stirrer 112 and a steam heater 111. The reference numeral 117 designates a control valve for transferring the crystalline suspension liquid vaporized and concentrated in the vaporizing concentrating crystallizing apparatus 110 to the centrifugal separator 120.

[36] The suspended mother liquid tank 160 serves to extract and store the suspension liquid formed at the upper portion of the vaporizing concentrating crystallizing apparatus 110 in which dry pentaerythritol, which is a raw material, is dissolved in pure water and then vaporized, concentrated and crystallized, and then slowly stirred for a predetermined time. The filter press 170 serves to filter the suspended mother liquid. The purified mother liquid tank 180 serves to store the press-filtered suspended mother liquid. The stored purified mother liquid is transferred to the vaporizing concentrating crystallizing apparatus 110 by a pump (P).

[37] The centrifugal separator 120 serves to separate the crystalline suspension liquid transferred from the vaporizing concentrating crystallizing apparatus 110 into monopentaerythritol and a residual product through centrifugal separation. The hot air drier 130 serves to remove water from the monopentaerythritol separated from the centrifugal separator 120 to form dried monopentaerythritol crystals. The screen separator 140, which serves to divide the dried monopentaerythritol crystals by particle size thereof and thus to produce or recirculate, includes a primary screen 141 and a secondary screen 142 having different sized meshes from each other. For example, the screen separator 140 may be a two-stage screen separator including a primary screen 141 having a mesh size of 10 and a secondary screen 142 having a mesh size of 30. It is preferred that each of the primary screen 141 and the secondary screen 142 be a vibrational screen. The reference numeral 150 designates a recovery pipe for recirculating the desired-size monopentaerythritol crystals obtained from the screen separator 140 to the vaporizing concentrating crystallizing apparatus 110. That is, the recovery pipe serves to recirculate the large sized monopentaerythritol crystals not having passed the primary screen 141 and the small sized monopentaerythritol crystals having passed both the primary screen 141 and the secondary screen 142. Therefore, as described in the above example, when the primary screen 141 has a mesh size of 10 and the secondary screen 142 has a mesh size of 30, the monopentaerythritol crystals having a mesh size of 10 or more and the monopentaerythritol crystals having a mesh size of 30 or less are recirculated to the vaporizing concentrating crystallizing
The reference numerals 121, 151 and 161 designate valves, respectively.

Hereinafter, the method of manufacturing high-purity pentaerythritol according to the present invention will be described in order of the steps using the above-mentioned manufacturing system to embody the method.

First, raw pentaerythritol containing 85 ~ 90 wt% of monopentaerythritol is supplied to the vaporizing concentrating crystallizing apparatus 110 and then mixed with pure water. Subsequently, the mixture of raw pentaerythritol and pure water is stirred using a stirrer 112 to completely dissolve the raw pentaerythritol in the pure water. After the raw pentaerythritol is completely dissolved in the pure water, the stirred mixture is vaporized and concentrated in the vaporizing concentrating crystallizing apparatus 110 at a temperature of about 90 ~ 95°C while controlling the concentration rate to form a crystalline suspension liquid. Thereafter, when the crystalline suspension liquid is slowly stirred for a predetermined time to naturally precipitate crystals therefrom, the crystals are settled at the lower portion of the vaporizing concentrating crystallizing apparatus 110, and a saturated liquid containing impurities is located at the upper portion of the vaporizing concentrating crystallizing apparatus 110. In this case, the saturated liquid (herein after, referred to as "a suspended mother liquid") including therein suspended products, located at the upper portion of the vaporizing concentrating crystallizing apparatus 110, is discharged to the suspended mother liquid tank 160. Then, the suspended mother liquid stored in the suspended mother liquid tank 160 is filtered by the filter press 170 to create purified mother liquid, and the purified mother liquid is stored in the purified mother liquid tank 180. The purified mother liquid stored in the purified mother liquid tank 180 is reintroduced into the vaporizing concentrating crystallizing apparatus 110 by a pump (P). As a result of this procedure, most of the impurities included in the suspended mother liquid are removed. Furthermore, this procedure allows the impurities to be more efficiently removed than when the crystalline suspension liquid formed in the vaporizing concentrating crystallizing apparatus 110 is directly transferred to the centrifugal separator 120, with the result that high-quality pentaerythritol can be manufactured.

The filter press 170 may be configured such that several filter plates and filter frames are alternately arranged, with filter cloths interposed therebetween and then strongly fixed. When the suspended mother liquid is forcibly charged into the filter frames, the suspended mother liquid passes through the filter cloths and then flows toward the filter plates, and impurities collect on the filter cloths. The suspended mother liquid which has passed through the filter cloths becomes purified mother liquid, and this purified mother liquid is stored in the purified mother liquid tank 180.

When the purified mother liquid obtained by removing impurities from the
suspended mother liquid is reintroduced into the vaporizing concentrating crystallizing apparatus 110, the purified mother liquid is further stirred using the stirrer 112 in order to mix the purified mother liquid with the crystals settled at the bottom of the vaporizing concentrating crystallizing apparatus 110. In this case, it is preferred that the stirring speed, that is, the revolutions per minute of the stirrer 112, be faster than those of the stirrer 112 in the step of naturally precipitating crystals. For example, in the step of obtaining the suspended mother liquid by naturally precipitating the crystals, it is preferred that the stirring speed of the stirrer be about 3 - 5 rpm, and, in the step of mixing the reintroduced purified mother liquid with the crystalline suspension liquid, it is preferred that the stirring speed of the stirrer be about 30 - 40 rpm.

The crystalline suspension liquid formed through the secondary stirring procedure is sent to the centrifugal separator 120 while being maintained at about 60 - 90°C, and then it is centrifugally separated. In the centrifugal separator 120, the crystalline suspension liquid is separated into monopentaerythritol and other by-products. The by-products include dipentaerythritol, tripentaerythritol and the like. In the centrifugal separation procedure, since dipentaerythritol and tripentaerythritol have a smaller particle size and specific gravity than those of monopentaerythritol, the dipentaerythritol and tripentaerythritol are lighter than the monopentaerythritol, and thus they float upward. In contrast, since the monopentaerythritol has larger specific gravity than that of the dipentaerythritol and tripentaerythritol, the monopentaerythritol is heavier than the dipentaerythritol and tripentaerythritol, and thus it settles downward. Therefore, when an over flow type centrifugal separator is used, the by-products, such as dipentaerythritol, tripentaerythritol and the like, overflow and are thus sent to the suspended mother liquid tank 160, and only the monopentaerythritol recovered from the centrifugal separator is supplied to a subsequent process, that is, a hot air drying process.

Subsequently, the separated monopentaerythritol crystals are dried using the hot air drier 130 and then sent to the screen separator 140. In the screen separator 140, monopentaerythritol crystals having a particle size larger than 10 meshes are first separated by the primary screen 141 having a mesh size of 10, and then the monopentaerythritol crystals having passed the primary screen 141 are separated into monopentaerythritol crystals having a particle size smaller than 30 meshes by the secondary screen 142 having a mesh size of 30, and residual monopentaerythritol crystals pass through the secondary screen 142. Therefore, the monopentaerythritol crystals are separated into monopentaerythritol crystals having a particle size larger than 10, monopentaerythritol crystals having a particle size of 10 - 30 meshes and monopentaerythritol crystals having a particle size smaller than 30 meshes according to the particle size thereof. That is, the monopentaerythritol crystals having a particle size of
10 - 30 meshes are discharged to a high-purity pentaerythritol tank through a high-purity pentaerythritol outlet 143, and the monopentaerythritol crystals having a particle size larger than 10 meshes and the monopentaerythritol crystals having a particle size smaller than 30 meshes are recirculated to the vaporizing concentrating crystallizing apparatus 110 through the recovery pipe 150. Here, the reason why the monopentaerythritol crystals are separated using the screens having meshes differing in size to result in particles of different size is that the purity of pentaerythritol is found to be influenced by the particle size thereof. That is, through the test, it was found that, after the centrifugal separation process, the monopentaerythritol crystals having a particle size larger than 10 and the monopentaerythritol crystals having a particle size smaller than 30 meshes have a monopentaerythritol content of about 98 wt%, but the monopentaerythritol crystals having a particle size of 10 ~ 30 meshes have a monopentaerythritol content of 99 wt% or more. Only the high-purity monopentaerythritol having a monopentaerythritol content of 99 wt% or more is classified as a pure grade and thus can be used to manufacture chemical reagents or medical supplies.

The pentaerythritol product having undergone the above processes is converted into a high-purity pentaerythritol product having a purity of 99.9 wt% or more.

Mode for the Invention

Example

First, 1400 mL of pure water was put into a 5L flask provided therein with a condenser and a stirrer, and then the pure water was heated to 90°C using an electric mantle heater while being stirred using the stirrer. Subsequently, 1200 g of dry pentaerythritol (water content: 8 wt%) having a monopentaerythritol content of 87 wt% was slowly introduced into the flask to be completely dissolved in the pure water to form a pentaerythritol solution. Thereafter, the pentaerythritol solution was vaporized and concentrated until 550 mL of steam condensate was produced while the flask was continuously heated, thus growing crystals. In this case, the concentration rate of the pentaerythritol solution was maintained constant for 90 minutes by controlling the electric mantle heater. Thereafter, the concentrated pentaerythritol solution was aged for 30 minutes and simultaneously naturally cooled at 70°C, and then stirred for 20 minutes at a stirring speed of 3 rpm. Thereafter, 400 mL of a suspended mother liquid located at the upper portion of the flask was removed, and then 400 mL of purified mother liquid (a saturated liquid from which impurities were removed) was charged in the flask and then stirred at a stirring speed of 30 rpm to form a suspended solution. Subsequently, the suspended solution was dewatered using an experimental centrifugal separator equipped with a stainless screen of 30 meshes, and then the dewatered suspended solution was washed with 100 mL of pure water, filtered for 10 minutes,
recovered and then dried for 1 hour using a hot air drier. Subsequently, the resulting product was naturally cooled and then primarily separated using a circular sieve of 10 meshes to obtain 68 g of large sized crystals. Then, the crystals which passed through the circular sieve of 10 meshes were secondarily separated using a circular sieve of 30 meshes to obtain 515 g of middle-sized crystals as a final product. In addition, 226 g of powdered crystals which passed through the circular sieve of 30 meshes were obtained. The amount of the mother liquid recovered after dewatering was 1240 g. The content of pentaerythritol included in the mother liquid was 28.7 wt%, and this mother liquid was recirculated to the recovery process. Analysis of the middle-sized pentaerythritol product of 10 ~ 30 meshes obtained through the above processes showed that such pentaerythritol had a purity of 99.94, a water content of 0.16 wt% and an ash content of 0.08 wt%.

**Industrial Applicability**

Pentaerythritol is used as a raw material of polyurethane foam (foamed plastic), coating agents, adhesives, sealants, flame retardants, explosives, lubricants, cosmetics, medicine, fiber or the like, and thus has many uses. Further, pentaerythritol is increasingly used as a raw material of special chemicals, too. According to the present invention, since high-purity pentaerythritol can be manufactured in large quantities at low cost, the manufacturing cost thereof can be decreased, thus contributing to the advancement in the related industry. Further, since the pentaerythritol manufactured by the method according to the present invention has much more excellent energy efficiency than the pentaerythritol manufactured through conventional repetitive recrystallization processes for producing pentaerythritol having a purity of 99.9 wt% or more, the dependency on exhausted fossil fuel can be decreased.
Claims

[1] A method of manufacturing high-purity pentaerythritol, comprising the steps of: mixing raw pentaerythritol having a monopentaerythritol content of 85 - 90 wt% with pure water in a vaporizing concentrating crystallizing apparatus, dissolving the raw pentaerythritol in the pure water, and then vaporizing and concentrating the mixed solution to form a crystalline suspension liquid; slowly stirring the crystalline suspension liquid for a predetermined time to naturally precipitate crystals and then filter-pressing a suspended mother liquid located at an upper portion of the vaporizing concentrating crystallizing apparatus to form a purified mother liquid and then reintroducing the purified mother liquid into the vaporizing concentrating crystallizing apparatus; stirring the crystalline suspension liquid and the purified mother liquid which was reintroduced into the vaporizing concentrating crystallizing apparatus; sending the crystalline suspension liquid to a centrifugal separator while maintaining the crystalline suspension liquid at a temperature of 60 ~ 90°C and then centrifugally separating the crystalline suspension liquid into monopentaerythritol and by-products such as dipentaerythritol and tripentaerythritol; drying the separated monopentaerythritol crystals using a hot air drier to remove water therefrom; and separating and then obtaining the dried monopentaerythritol crystals which were blown on by the hot air drier into monopentaerythritol crystals having a different crystal size from each other using a two-stage screen separator equipped with a primary screen and a secondary screen which differ in mesh size.

[2] The method of manufacturing high-purity pentaerythritol according to claim 1, further comprising the step of: recirculating the monopentaerythritol crystals which failed to pass through the primary screen and the monopentaerythritol crystals which passed through the secondary screen to the vaporizing concentrating crystallizing apparatus.

[3] The method of manufacturing high-purity pentaerythritol according to claim 1 or 2, wherein the step of reintroducing the purified mother liquid into the vaporizing concentrating crystallizing apparatus is performed by transferring the suspended mother liquid located at the upper portion of the vaporizing concentrating crystallizing apparatus to a suspended mother liquid tank, and then transferring the suspended mother liquid from the suspended mother liquid tank to a purified mother liquid tank through a filter press to turn the suspended mother liquid into a purified mother liquid, and then sending the purified mother liquid to the vaporizing concentrating crystallizing apparatus.
The method of manufacturing high-purity pentaerythritol according to claim 3, wherein the two-stage screen separator is equipped with the primary screen having a size of 10 meshes and the secondary screen having a size of 30 meshes so that the monopentaerythritol crystals are separated into monopentaerythritol crystals having a particle size larger than 10 meshes, monopentaerythritol crystals having a particle size of 10 ~ 30 meshes and monopentaerythritol crystals having a particle size smaller than 30 meshes.
[Fig. 1]

1. DISSOLVING, VAPORIZING, CONCENTRATING AND CRYSTALLIZING DRY PENTAERYTHRITOL
2. NATURAL PRECIPITATION
3. CENTRIFUGAL SEPARATION
4. HOT AIR DRYING
5. SEPARATION AND CIRCULATION
6. SELECTION AND PACKAGING
7. STORING AND DELIVERING
INTERNATIONAL SEARCH REPORT

A. CLASSIFICATION OF SUBJECT MATTER

C07C 29/38(2006.01)1, C07C 29/74(2006.01)1, C07F 9/6574(2006.01)1

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC C07C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
e-KIPASS(KIPO-Internal)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

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<td>KR 10-0716541 B1 (WONJIN HEAVY INDUSTRIES CO, LTD) 10 May 2007 cited in the application See example and claims</td>
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☐ See patent family annex

* Special categories of cited documents
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Date of the actual completion of the international search
28 MAY 2009 (28 05 2009)

Date of mailing of the international search report
28 MAY 2009 (28.05.2009)

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