SUMMARY

In one aspect, the present disclosure refers to a process for preparing a silver-containing catalyst for the epoxidation of olefins comprising the steps of:

(a) providing a porous multimodal support having at least a first set of support pores of a first size range and at least a second set of support pores of a second size range wherein the second size range of the second set of support pores is smaller than the first size range of the first set of support pores;

(b) providing a first silver-containing impregnation solution for impregnating the first silver-containing impregnation solution into the at least first set of support pores and the at least second set of support pores of the porous multimodal support;

(c) impregnating, one or more times, the porous multimodal support with the first silver-containing impregnation solution from step (b) to provide the porous multimodal support with a first amount of silver-containing impregnation solution; and

(d) selectively removing at least a portion of the impregnated first silver-containing impregnation solution from the porous multimodal support to provide the porous multimodal support with a second amount of first silver-containing impregnation solution remaining in the support pores; wherein at least a portion of the impregnated first silver-containing impregnation solution is removed from the at least first set of support pores of the porous multimodal support by subjecting, one or more times, the impregnated porous multimodal support after the impregnation step (c) to a removal means for a time sufficient to selectively remove at least a portion of the impregnated first silver-containing impregnation solution contained in the at least first set of support pores of the porous multimodal support, wherein the removal means is centrifugation.

One example of the present disclosure is directed to a process for preparing a silver-containing catalyst for the oxidation of ethylene to ethylene oxide (EO) including the steps of:

(a) providing a porous multimodal support having at least a first set of support pores of a first size range and at least a second set of support pores of a second size range wherein the second size range of the second set of support pores is smaller than the first size range of the first set of support pores;

(b) providing an impregnation solution, such as an impregnation solution comprising a silver component, for impregnating the impregnation solution into the at least first set of support pores and the at least second set of support pores of the porous multimodal support;

(c) impregnating, at least one time (i.e., at least once), the porous multimodal support with a least a portion of the impregnation solution from step (b);

(d) selectively removing the impregnation solution prior to fixing the silver upon the carrier at least the first set of support pores of the porous multimodal catalyst support by subjecting, at least once, the impregnated porous multimodal support after the impregnation step (c) to a removal means for a time sufficient to selectively remove at least a portion of impregnated silver impregnation solution contained in the at least first set of support pores of the porous multimodal catalyst support.

In another example, the process for preparing a silver-containing catalyst may include the additional step of:

(e) roasting (i.e. calcining), at least once, the impregnated catalyst support member from step (d) for a time and temperature sufficient to form a silver-containing catalyst useful for the epoxidation of olefins.

In still another example, the selective removal means in step (d) may include a centrifuge for centrifuging the impregnated catalyst support member to remove at least a portion of impregnated solution contained in the at least first set of support pores.

In an optional example, the impregnation step (c) or the removing step (d) may be repeated, at least one more time, for a time sufficient to remove further the Ag impregnation solution contained in the catalyst support member.

In further example, the removing step (d) may be carried out at least two times before the roasting step (e). The centrifugation step (d) is advantageously performed at a centrifugal force and for a period of time sufficient to remove the Ag solution from the desired pores and to provide the support with a more evenly distributed Ag particle size in the multimodal pores of the catalyst support after roasting.

In even yet other examples, any one or more of the above steps (a)-(e) can optionally be repeated any number of times as desired and sufficiently to control the amount of Ag deposited in the pores of the catalyst support.

Also provided in the present application is a reaction system for producing ethylene oxide from a feed comprising ethylene and oxygen over an ethylene oxide catalyst prepared by the process of the present disclosure.

BRIEF DESCRIPTION OF THE DRAWINGS

The drawings show non-limiting embodiments of the present invention wherein:

Figure 1 is graphical illustration of the effect of Ag particle size on ethylene oxide selectivity taken from Goncharova *et al.*, in "Size effects in ethylene oxidation on silver catalysts. Influence of support and Cs promoter." Applied Catalysis A: General, 126(1), 67-84. The symbols illustrated in Figure 1 represent the following: $Ag/\alpha\text{-}Al_2O_3$ (☆); Ag/SiO_2 (★) and Ag powder (■).The epoxidation conditions of the process includes the following: 230 °C, 1 bar, 2 % C_2H_4, 7 % O_2 in N_2.

Figure 2 is a schematic flow diagram showing a synthesis of a catalyst according to the process of the present invention.

Figure 3 is another schematic flow diagram showing a synthesis of a catalyst according to the process of the present invention providing for two centrifugation steps.

Figure 4 is another schematic flow diagram showing a synthesis of a catalyst according to the process of the present invention providing for a series of one impregnation step, one centrifugation step, and one calcination step followed in sequence by a second series of a second impregnation step, a second centrifugation step, and a second calcination step.

WHAT IS CLAIMED IS:

1. A process for preparing a silver-containing catalyst for the epoxidation of olefins comprising the steps of:

(a) providing a porous multimodal support having at least a first set of support pores of a first size range and at least a second set of support pores of a second size range wherein the second size range of the second set of support pores is smaller than the first size range of the first set of support pores;

(b) providing a first silver-containing impregnation solution for impregnating the first silver-containing impregnation solution into the at least first set of support pores and the at least second set of support pores of the porous multimodal support;

(c) impregnating, one or more times, the porous multimodal support with the first silver-containing impregnation solution from step (b) to provide the porous multimodal support with a first amount of silver-containing impregnation solution; and

(d) selectively removing at least a portion of the impregnated first silver-containing impregnation solution from the porous multimodal support to provide the porous multimodal support with a second amount of first silver-containing impregnation solution remaining in the support pores; wherein at least a portion of the impregnated first silver-containing impregnation solution is removed from the at least first set of support pores of the porous multimodal support by subjecting, one or more times, the impregnated porous multimodal support after the impregnation step (c) to a removal means for a time sufficient to selectively remove at least a portion of the impregnated first silver-containing impregnation solution contained in the at least first set of support pores of the porous multimodal support, wherein the removal means is centrifugation.

2. The process of claim 1, wherein the selectively removing step (d) removes at least 20 percent of the impregnated first silver-containing impregnation solution contained in the at least first set of support pores of the porous multimodal support.

3. The process of claim 1, including step (e) roasting, one or more times, the impregnated porous multimodal support produced in step (d) for a time and temperature sufficient to form a silver-containing catalyst useful for the epoxidation of olefins.

4. The process of claim 1, wherein the selectively removing step (d) is carried out, one or more times, by one or more centrifugation means.

5. The process of claim 1, wherein the impregnation step (c) is carried out two or more times; or wherein the removing step (d) is carried out two or more times and under two or more different process conditions.

6. The process of claim 4, wherein the impregnating step (c) is carried out two or more times prior to the removal step (d).

7. The process of claim 3, wherein the selective removal step (d) is carried out two or more times prior to the roasting step (e) or wherein the roasting step (e) is carried out two or more times.

8. The process of claim 1, wherein the first pore size range of the first set of pores of the support is from 3 microns to 200 microns; and wherein the second pore size range of the second set of pores of the support is from 0.01 micron to 3 microns, wherein pore size is determined by mercury porosimetry.

9. The process of claim 1, wherein the first silver-containing impregnation solution further comprises one or more promoters.

10. The process of claim 1, wherein the support comprises alpha-alumina.

11. The process of claim 1, wherein a silver loading of at least 10 weight percent is provided on the porous multimodal support, based on the total weight of the porous multimodal support.

12. The process of claim 3, wherein the process includes the following steps in sequence:

(f) providing a second silver-containing impregnation solution for impregnating the second silver-containing impregnation solution into the at least first set of support pores and the at least second set of support pores of the porous multimodal support;

(g) impregnating, one or more times, the porous multimodal support obtained in step (e) with at least a portion of the second silver-containing impregnation solution from step (f);

(h) selectively removing, one or more times, the impregnated second silver-containing impregnation solution from the at least first set of support pores of the porous multimodal catalyst support by subjecting, at least once, the impregnated porous multimodal support after the impregnation step (g) to a removal means for a time sufficient to selectively remove a portion of the impregnated second silver-containing impregnation solution contained in the at least first set of support pores of the porous multimodal catalyst support;

(i) roasting, one or more times, the impregnated porous multimodal support from step (h) for a time and temperature sufficient to form a silver-containing catalyst useful for the epoxidation of olefins;

(j) providing a third silver-containing impregnation solution for impregnating the third silver-containing impregnation solution into the at least first set of support pores and the at least second set of support pores of the porous multimodal support;

(k) impregnating, one or more times, the silver-containing catalyst from step (i) with at least a portion of the third silver-containing impregnation solution from step (j);

(l) selectively removing, one or more times, the impregnated third impregnation solution from the at least first set of support pores of the impregnated roasted porous multimodal catalyst support from step (i) by subjecting, one or more times, the impregnated roasted porous multimodal support after the impregnation step (k) to a removal means for a time sufficient to selectively remove at least a portion of the impregnated third silver-containing impregnation solution contained in the at least first set of support pores of the impregnated roasted porous multimodal catalyst support; and

(m) roasting, one or more times, the impregnated catalyst support member from step (l) for a time and temperature sufficient to form a silver-containing catalyst useful for the epoxidation of olefins.