WHAT IS CLAIMED IS:

1. A method of preparing a phthalonitrile-based compound, comprising:
(a) preparing a mixture consisting of a phthalic acid-based compound and a nitrile-based compound; and
(b) reacting the mixture,
wherein step (b) is performed under supercritical conditions of the nitrile-based compound,
the phthalic acid-based compound is isophthalic acid, terephthalic acid, or a mixture thereof,
the nitrile-based compound is one or more selected from the group consisting of hydrogen cyanide, acetonitrile, acrylonitrile, butyronitrile, isobutyronitrile, pivalonitrile, succinonitrile, fumaronitrile, crotonitrile, and benzonitrile, and
in step (b), the purity of the product in which two or more carboxyl groups in the phthalic acid-based compound are substituted with nitrile groups is 60% or more.

2. The method of claim 1, wherein the nitrile-based compound is acetonitrile.

3. The method of claim 1, wherein in step (a), a content of the nitrile-based compound is 1 to 500 parts by weight based on 1 part by weight of the phthalic acid-based compound.

4. The method of claim 1, wherein in step (a), a water content of the mixture is less than 6,000 ppm.

5. The method of claim 1, wherein step (b) is performed under conditions of 260 to 350 °C and 40 to 200 bar.

6. The method of claim 1, wherein step (b) is performed for 1 to 500 minutes.

7 The method of claim 1, further comprising (c) separating the product of step (b) after step (b).

8. The method of claim 7, wherein a residual compound separated in step (c) is reused in step (a).