

Contributed by Jiri Čejka

Verified by R. Ryoo, A. Puškarić

Type Material: (SDA)₄[Si₆₃Ge₁₂B₁O₁₅₂] : w H₂O
(SDA = 7-Ethyl-6-azoniaspiro[5.5]undecane hydroxide)

Method: O. V. Shvets, M. V. Shamzhy, P. S. Yaremov, Z. Musilová, D. Procházková, J. Čejka [1]

Batch Composition: 0.74 SiO₂ : 0.4 GeO₂ : 0.03 B₂O₃ : 0.3 SDAOH/Br : 30 H₂O

Source Materials

deionized water
sodium hydroxide (98 %, NaOH)
1,5-dibromopentane (97 %)
2-ethylpiperidine (98%)
chloroform (99%)
sodium sulfate anhydrous (99 %, Na₂SO₄)
diethyl ether (99%)
Dowex[®] SBR LCNG (Supelco)
boric acid (99.99%, H₃BO₃)
germanium oxide (99.99 % GeO₂)
silica (Degussa Aerosil 200, or Cab-O-Sil M5)

Batch Preparation (for 3.2 g dry product)

- (1) [22.0 g water + 0.89 g sodium hydroxide + 5.11 g 1,5-dibromopentane], stir in a flask
- (2) [(1) + 2.53 g 2-ethylpiperidine], add 2-ethylpiperidine to (1) drop by drop over a period of 30 min under reflux; reflux for 12 h under vigorous stirring^a; cool with an ice bath
- (3) [5.5 g water + 5.5 g sodium hydroxide], stir until dissolved, cool with ice bath
- (4) [(2) + (3)], stir, recover solid by filtration
- (5) [solid (4) + 100 ml chloroform], stir until dissolved
- (6) [(5) + 15 g sodium sulfate anhydrous], stir, left for 30 min, remove solid by filtration, evaporate about 50 ml of chloroform using rotovap
- (7) [(5) + 150 ml diethyl ether], mix, recover solid by filtration, wash with diethyl ether, dry at ambient temperature for 12 h
- (8) [solid (7)^b + 35.9 g water + 30 g Dowex[®]], stir for 2 h, remove Dowex[®] by filtration
- (9) [solution (7) + 0.248 g boric acid], stir until dissolved^c
- (10) [solution (8) + 2.789 g germanium oxide], stir until dissolved^c
- (11) [(9) + 2.96 g Cabosil], stir for 30 minutes^d

Crystallization

Vessel: Teflon-lined stainless steel autoclave
Temperature: 170° C
Time: 11 days

Agitation: 60 rpm

Product Recovery

- (1) Dilute reaction mixture with water
- (2) Filter and wash with water
- (3) Dry at ambient temperature or at 90°C
- (4) Yield: 3.2 g

Product Characterization

XRD: UTL; competing phase: STF (when $B / (Si + Ge + B) > 0.11$ in initial gel)

Elemental analysis: 83 SiO₂ : 16 GeO₂ : 0.5 B₂O₃^e

Crystal size and habit: thin platelet-like crystals

Reference

- [1] O. V. Shvets, M. V. Shamzhy, P. S. Yaremov, Z. Musilová, D. Procházková, J. Čejka, Chem. Mater. 23 (2011) 2573

Notes

- a. The satisfactory mixing of two phases at > 1000 rpm.
- b. 90 % yield of SDA
- c. Clear solution
- d. pH of final gel is 10.096. The required amount of 1M SDAOH solution or 5 M HCl was added to the above mixture under stirring to adjust the pH of the gel.
- e. According to Ref. [1] the chemical composition of the product depends on the pH of the final gel.
- f. An additional aqueous solution of sodium hydroxide could be added into the mixture until SDA was precipitated.