Electronic Supplementary Information

Low temperature synthesis of photoconductive BaSi<sub>2</sub> films by

mechanochemically assisted close-spaced evaporation

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(a) Ground with a pestle and mortar





(b) Milled for 60 s



Figure S1: BE images and EDX maps of (a) the BaAl<sub>4</sub>–Ni powder ground with a pestle and mortar (used in K. O. Hara, et al., Mater. Sci. Semicond. Process. 113, 105044 (2020)) and (b) that milled at 50 Hz for 60 s. After milling for 60 s, Ba, Al, and Ni are homogeneously mixed.



Figure S2: Cross-sectional SE images of the CSE samples prepared at 900, 800 and 700 °C for 5 min using the BaAl<sub>4</sub>–Ni powder milled for 60 s.



Figure S3: XRD patterns of the BaAl<sub>4</sub>–Ni powder milled for different time together with that ground with a pestle and mortar.



Figure S4: XRD patterns of the powders milled for 60, 150, and 1200 s, mixed with Si as an internal standard. Calculated powder patterns of BaAl<sub>4</sub>, Ni, and Si are also displayed.

Table S1: Lattice parameters of Ni calculated from the XRD patterns in Fig. S4 by whole pattern fitting

Milling time (s)	Lattice parameter of Ni (Å)
60	3.523
150	3.526
1200	3.523



Figure S5: XRD patterns of the sources after CSE at 800 °C for 5 min. When using a 60 s milled source, Ni<sub>3</sub>Al is produced together with NiAl, Ni<sub>2</sub>Al<sub>3</sub>, and BaAl<sub>2</sub>O<sub>4</sub>. With increasing milling time, Ni<sub>3</sub>Al peaks become weak and undetectable after 300 s. BaAl<sub>2</sub>O<sub>4</sub> peaks also become undetectable after 600 s milling. This observation can be interpreted as the phase transformation from Ni-dissolved BaAl<sub>4</sub> to Ni–Al–Ba alloy with the NiAl structure. Mechanochemically induced solid solution of Ni in BaAl<sub>4</sub> (mechanical alloying) therefore prevents the Ba gas generation reaction route.



Figure S6: XRD patterns of the 60 s milled sources after CSE at 700 °C for 5 and 50 min. BaAl<sub>4</sub> and Ni remain after 50 min of CSE.



Figure S7: 2D XRD patterns of the CSE films synthesized at 700 °C for (a) 2.5, (b) 5, (c) 7.5, (d) 10, (e) 20, and (f) 30 min.