Supplementary Information (SI) for Journal of Materials Chemistry C.

Supplementary Material

MnTi co-doping in $Mg_2Al_4Si_5O_{18}$ ceramics enhances *Q* factor and improves temperature stability with a τ_f value closer to zero

1. The Rietveld refinement results of $Mg_2Al_4Si_{5-x}(Mn_{0.5}Ti_{0.5})_xO_{18}$ ($0 \le x \le 0.16$) ceramics

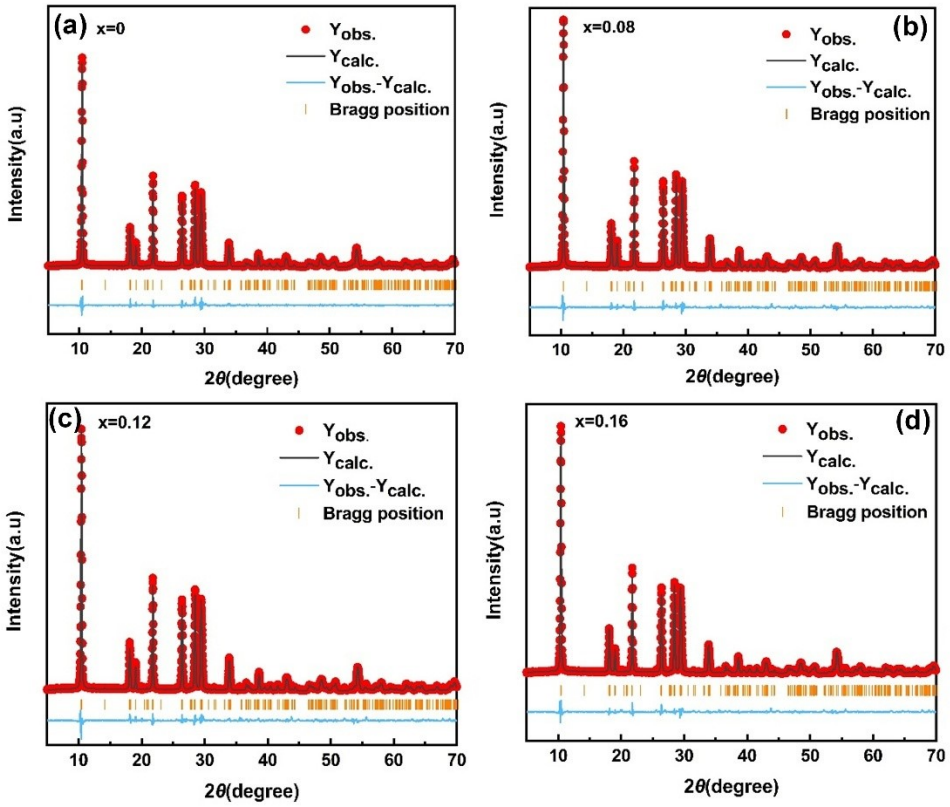

Fig. S1. (a) The refinement pattern of $Mg_2Al_4Si_5O_{18}$ ceramics sintered at the optimum temperature, (b) The refinement pattern of $Mg_2Al_4Si_{4.92}(Mn_{0.5}Ti_{0.5})_{0.08}O_{18}$ ceramics sintered at the optimum temperature, (c) The refinement pattern of $Mg_2Al_4Si_{4.88}(Mn_{0.5}Ti_{0.5})_{0.12}O_{18}$ ceramics sintered at the optimum temperature, (d) The refinement pattern of $Mg_2Al_4Si_{4.84}(Mn_{0.5}Ti_{0.5})_{0.16}O_{18}$ ceramics sintered at the optimum temperature

Table S1. Bond lengths of $Mg_2Al_4Si_{5-x}(Mn_{0.5}Ti_{0.5})_xO_{18}$ ($0 \le x \le 0.16$) ceramics obtained by Rietveld refinement.

x value	x=0	x=0.04	x=0.08	x=0.12	x=0.16
Mg-O1(Å)	2.119	2.1193	2.1385	2.1243	2.1167
Mg-O2(Å)	2.1	2.0943	2.0784	2.0915	2.1023

Mg-O3(Å)	2.125	2.1253	2.1209	2.1267	2.1197
Al1-O1(Å)	1.804	1.7629	1.754	1.7529	1.744
Al1-O3(Å)	1.711	1.7026	1.693	1.7003	1.7171
Al2-O2(Å)	1.754	1.7495	1.763	1.7629	1.7619
Al2-O4(Å)	1.713	1.7416	1.7666	1.7585	1.767
Al2-O5(Å)	1.696	1.6775	1.6937	1.6765	1.6542
Si1-O2(Å)	1.641	1.661	1.6677	1.6534	1.6395
Si2-O1(Å)	1.566	1.6239	1.6091	1.6219	1.6442
Si2-O5(Å)	1.586	1.6348	1.6264	1.634	1.6323
Si2-O6(Å)	1.579	1.5633	1.5707	1.5704	1.568
Si3-O3(Å)	1.66	1.6833	1.6936	1.6845	1.6734
Si3-O4(Å)	1.57	1.5226	1.4977	1.5048	1.5136
Si3-O6(Å)	1.614	1.6225	1.6276	1.635	1.6355

2. P-V-L (Phillips-Van Vechten-Levine) bond theory calculation

The bond ionicity (fi) of each type of bond in the cell can be found according to Eqs. * MERGEFORMAT (S1)-* MERGEFORMAT (S7)below:

$$\varepsilon_{pvl} = \frac{n^2 - 1}{1 - f_i} + 1 \qquad \text{* MERGEFORMAT (S1)}$$

$$f_i = \left(C^{\mu}\right)^2 / \left(E_g^{\mu}\right)^2 \qquad \text{* MERGEFORMAT (S2)}$$

$$\left(E_g^{\mu}\right)^2 = \left(E_h^{\mu}\right)^2 + \left(C^{\mu}\right)^2 \qquad \text{* MERGEFORMAT (S3)}$$

$$E_h^{\mu} = 39.74 / \left(d^{\mu}\right)^{2.48} \qquad \text{* MERGEFORMAT (S4)}$$

$$C^{\mu} = 14.4 b^{\mu} \exp\left(-\frac{k_s^{\mu} d^{\mu}}{2}\right)\left[\left(Z_A^{\mu}\right)^* - \frac{n\left(Z_B^{\mu}\right)^*}{m}\right]\left(\frac{d^{\mu}}{2}\right)^{-1} \text{* MERGEFORMAT (S5)}$$

$$b^{\mu} = \beta\left(N_c^{\mu}\right)^2 \qquad \text{* MERGEFORMAT (S6)}$$

$$N_c^{\mu} = \frac{m}{m+n} N_{CA}^{\mu} + \frac{n}{m+n} N_{CB}^{\mu} \qquad \text{* MERGEFORMAT (S7)}$$

Among these, n is the refractive index ($n_{cordierite}$ = 1.542), E_h^{μ} and C^{μ} represent the homopolarization energy corresponding to the covalent nature of the chemical bond μ and the heteropolarization energy corresponding to the ionic nature (units: e*V*); E_g^{μ} represents the average energy gap (unit: eV), which is the sum of the polarization energies corresponding to the ionic and covalent nature of the chemical bond μ; d^{μ} is the bond length of chemical bond μ; b^{μ} is the structural correction factor(*b*=3.024); $e^{\left(-k_s^{\mu} r_o^{\mu}\right)}$ is the Thomas-Fermi shielding factor; $\left(Z_A^{\mu}\right)^*$ and $\left(Z_B^{\mu}\right)^*$ represent the effective valence electron counts of cation A and anion B, respectively; β is generally approximately equal to 0.089; N_c^{μ} is the average coordination number of the chemical bond μ; N_{CA}^{μ} and N_{CB}^{μ} represent the coordination numbers of cation A and anion B, respectively.

Table S2. Ion activity (f_i) of the bond in the $Mg_2Al_4Si_{5-x}(Mn_{0.5}Ti_{0.5})_xO_{18}$ ($0 \le x \le 0.16$) ceramics.

Bond type	x=0	x=0.04	x=0.08	x=0.12	x=0.16
Mg-O1	74.960%	74.570%	74.736%	74.615%	74.570%
Mg-O2	74.291%	74.350%	74.208%	74.327%	74.444%
Mg-O3	74.509%	74.622%	74.585%	74.636%	74.596%
Al1-O1	63.730%	63.407%	63.306%	63.294%	63.215%
Al1-O3	62.645%	62.679%	62.559%	62.652%	62.889%
Al2-O2	63.164%	63.251%	63.410%	63.410%	63.425%
Al2-O4	38.055%	38.549%	38.851%	38.755%	38.884%
Al2-O5	37.841%	37.742%	37.953%	37.731%	37.462%
Si1-O2	70.186%	70.528%	70.600%	70.450%	70.324%
Si2-O1	69.321%	70.125%	69.960%	70.106%	70.376%
Si2-O5	45.543%	46.351%	46.243%	46.344%	46.350%
Si2-O6	45.446%	45.371%	45.479%	45.476%	45.470%
Si3-O3	70.390%	70.759%	70.865%	70.774%	70.685%
Si3-O4	45.319%	44.771%	44.389%	44.501%	44.664%
Si3-O6	45.925%	46.189%	46.259%	46.357%	46.392%
f_i	**58.755%**	**58.884%**	**58.894%**	**58.895%**	**58.916%**

The lattice energy (U) can be obtained as follows Eqs. * MERGEFORMAT (S8)-* MERGEFORMAT (S11):

$$U = \sum U^{\mu} \qquad \text{* MERGEFORMAT (S8)}$$

$$U^{\mu} = U^{\mu}_{bc} + U^{\mu}_{bi} \qquad \text{* MERGEFORMAT (S9)}$$

$$U^{\mu}_{bc} = \frac{2100 m f^{\mu}_{c} (Z^{\mu}_{+})^{1.64}}{(d^{\mu})^{0.75}} \qquad \text{* MERGEFORMAT (S10)}$$

$$U^{\mu}_{bi} = 1270 \frac{(m+n) Z^{\mu}_{+} Z^{\mu}_{-}}{(d^{\mu})^{0.75}} (1 - \frac{0.4}{d^{\mu}}) f^{\mu}_{i} \qquad \text{* MERGEFORMAT (S11)}$$

The lattice energy (U) is the sum of two components—the covalent lattice energy (Uμbc) and the ionic lattice energy (Uμbi). Zμ− and Zμ+ are the cationic and anionic valence states in a particular bond, respectively, and fc is the bond covalence.

Table S3. Lattice energy (U) of $Mg_2Al_4Si_{5-x}(Mn_{0.5}Ti_{0.5})_xO_{18}$ ($0 \le x \le 0.16$) ceramics.

Bond type	x=0	x=0.04	x=0.08	x=0.12	x=0.16
Mg-O1	2036	2081.8	2066.6	2077.8	2083.7

Mg-O2	2097.9	2101.9	2114.8	2104.1	2095.3
Mg-O3	2077.9	2077	2075.5	2075.9	2081.4
Al1-O1	8464.8	8605.9	8616.9	8640.7	8672.1
Al1-O3	8789.1	8820.2	8825.3	8828.6	8767.9
Al2-O2	8636.2	8652.6	8605.5	8605.9	8609.5
Al2-O4	4092.1	4040.9	3997.6	3991.5	3996.8
Al2-O5	4122.9	4156.4	4126.5	4158.3	4200.1
Si1-O2	15526.4	15572	15509.7	15520.2	15709.6
Si2-O1	15890.8	15909.7	15906.4	15792.8	15679.5
Si2-O5	7002.6	7078.4	7064.4	7050.9	7056.1
Si2-O6	7225.2	7276.6	7252.3	7253.3	7261.1
Si3-O3	15475.3	15432.1	15368.2	15424.7	15494.5
Si3-O4	7254.6	7413.5	7440	7435.1	7444.5
Si3-O6	7113.3	7086.6	7070.7	7047.8	7046.2
U_{avg}	**7720.3**	**7753.7**	**7736.0**	**7733.8**	**7746.6**

The bond energy (E) can be calculated according to Eqs. * MERGEFORMAT (S12)-* MERGEFORMAT (S17) :

$$E = \sum_{\mu} E_b^{\mu} \qquad \text{* MERGEFORMAT (S12)}$$

$$E_b^{\mu} = t_i E_i^{\mu} + t_c E_c^{\mu} \qquad \text{* MERGEFORMAT (S13)}$$

$$E_i^{\mu} = 33200 / d^{\mu} \qquad \text{* MERGEFORMAT (S14)}$$

$$E_c^{\mu} = \frac{(r_{CA} + r_{CB})}{d^{\mu}} \left(E_{A-A} + E_{B-B}\right)^{\frac{1}{2}} \qquad \text{* MERGEFORMAT (S15)}$$

$$t_i + t_c = 1 \qquad \text{* MERGEFORMAT (S16)}$$

$$t_i = \left|\left(S_A - S_B\right) / 2\Delta S_B\right| \qquad \text{* MERGEFORMAT (S17)}$$

Among these, E_i^{μ} and E_c^{μ} represent the energy contributions from ionic and covalent bonds, respectively; t_i and t_c denote the ionic coefficient and covalent coefficient, respectively ; r_{CA} and r_{CB} represent the covalent radii of elements A and B, respectively; E_{A-A} and E_{B-B} denote the bond energies of the A-A and B-B bonds, respectively, which can be obtained from bond energy reference manuals. S_A and S_B

represent the electronegativity of elements A and B, respectively, with ΔS_B being a constant of 3 (B is typically an oxygen element).

Table S4. Bond energies (E) of the $Mg_2Al_4Si_{5-x}(Mn_{0.5}Ti_{0.5})_xO_{18}$ ($0 \le x \le 0.16$) ceramics.

Bond type	x=0	x=0.04	x=0.08	x=0.12	x=0.16
Mg-O1	226.5	232.9	230.8	232.3	233.1
Mg-O2	235	235.6	237.4	236	234.7
Mg-O3	232.2	232.2	232.7	232.1	232.8
Al1-O1	247.3	253	254.3	254.5	255.8
Al1-O3	260.7	262	263.5	262.4	259.8
Al2-O2	254.3	255	254	253	253.2
Al2-O4	260.4	256.1	257.5	253.7	252.5
Al2-O5	263	265.9	267.4	266.1	267.7
Si1-O2	245	242	241.1	243.1	245.2
Si2-O1	246.7	247.6	249.8	247.9	244.5
Si2-O5	243.5	245.9	247.2	246	243.3
Si2-O6	254.6	257.1	255.9	256	256.4
Si3-O3	238.2	238.8	237.4	238.6	240.2
Si3-O4	256.1	264	268.4	267.1	265.6
Si3-O6	249.1	247.8	247	245.9	245.8
$\boldsymbol{E_{avg}}$	**247.5**	**249.1**	**249.6**	**248.9**	**248.7**

The τ_f value correlates with the thermal expansion coefficient (α) through the following relationship Eqs * MERGEFORMAT (S18)-* MERGEFORMAT (S22). The specific values are listed in Table 2.

$$\tau_f = -\left(\tau_\varepsilon / 2 + \alpha\right) \qquad \text{* MERGEFORMAT (S18)}$$

$$\alpha = \sum_u F_{mn}^u \alpha_{mn}^u \qquad \text{* MERGEFORMAT (S19)}$$

$$\alpha_{mn}^u = -3.1685 + 0.8376\gamma_{mn} \qquad \text{* MERGEFORMAT (S20)}$$

$$\gamma_{mn} = kZ_A^u N_{CA}^u \beta_{mn} / U_b^u \Delta_A \qquad \text{* MERGEFORMAT (S21)}$$

$$\beta_{mn} = m(m+n)/2n \qquad \text{* MERGEFORMAT (S22)}$$

Here, F_{mn}^u indicates the proportion of the chemical bond in the crystal, k is the Boltzmann

constant, Δ_A is the correction parameter, and Z_A^u and N_{CA}^u denote the oxidation state and coordination number of the cation, respectively.

Table S5. Thermal expansion coefficient (α 10^{-6}/K) of $Mg_2Al_4Si_{5-x}(Mn_{0.5}Ti_{0.5})_xO_{18}$ ($0 \leq x \leq 0.16$) ceramics.

Bond type	x=0	x=0.04	x=0.08	x=0.12	x=0.16
Mg-O1	13.12	13.15	13.27	13.18	13.13
Mg-O2	13.02	12.99	12.89	12.98	13.04
Mg-O3	13.16	13.19	13.16	13.20	13.15
Al1-O1	3.85	3.74	3.71	3.71	3.69
Al1-O3	3.59	3.57	3.55	3.57	3.61
Al2-O2	3.71	3.70	3.74	3.74	3.74
Al2-O4	3.06	3.14	3.20	3.18	3.21
Al2-O5	2.07	2.03	2.07	2.03	1.98
Si1-O2	1.11	1.15	1.16	1.14	1.11
Si2-O1	0.98	1.08	1.06	1.08	1.12
Si2-O5	1.55	1.65	1.63	1.65	1.65
Si2-O6	1.53	1.50	1.52	1.52	1.51
Si3-O3	1.15	1.19	1.21	1.19	1.17
Si3-O4	1.51	1.41	1.36	1.38	1.39
Si3-O6	1.61	1.63	1.64	1.65	1.65
α_{Mg-O}	**13.11**	**13.11**	**13.11**	**13.12**	**13.11**
α_{Al-O}	**3.22**	**3.24**	**3.25**	**3.25**	**3.24**
α_{Si-O}	**1.35**	**1.37**	**1.37**	**1.37**	**1.37**