Supplementary Materials for: Complexions and stoichiometry of the 60°//[100](011) symmetrical tilt grain boundary in Mg_2SiO_4 forsterite: a combined empirical potential and first-principles study

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We present here additional material obtained from our calculations: the distribution in energies and excess volumes for the different non-stoichiometric GBs, and the total energies and excess volumes for the non-stoichiometric GBs with the lowest segregation energy.

1. Dispersion of the non-stoichiometric GBs

The segregation energy ΔE (see Eq.5) and excess volume of the non-stoichiometric GB complexions obtained by MD relaxation using the methodology presented in section 4 and from the six low energy GB complexions introduced in section 3.3 are presented in figure S1.
As for the E1 complexion presented in the manuscript, the insertion of a MgO vacancy pair at the GBs is almost always energetically favorable (i.e. negative segregation energy). In few cases it also permits to decrease the GB excess volume.

2. Non-stoichiometric GBs with the lowest segregation energy

Table S1 presents the total energies and excess volumes, computed using MD and DFT, for the non-stoichiometric complexions with the lowest segregation energy (highlighted by the green star in Fig.S1). Both methods are in good agreement concerning the excess volumes. The ordering of total energies is consistent for the two methods for almost all complexions.

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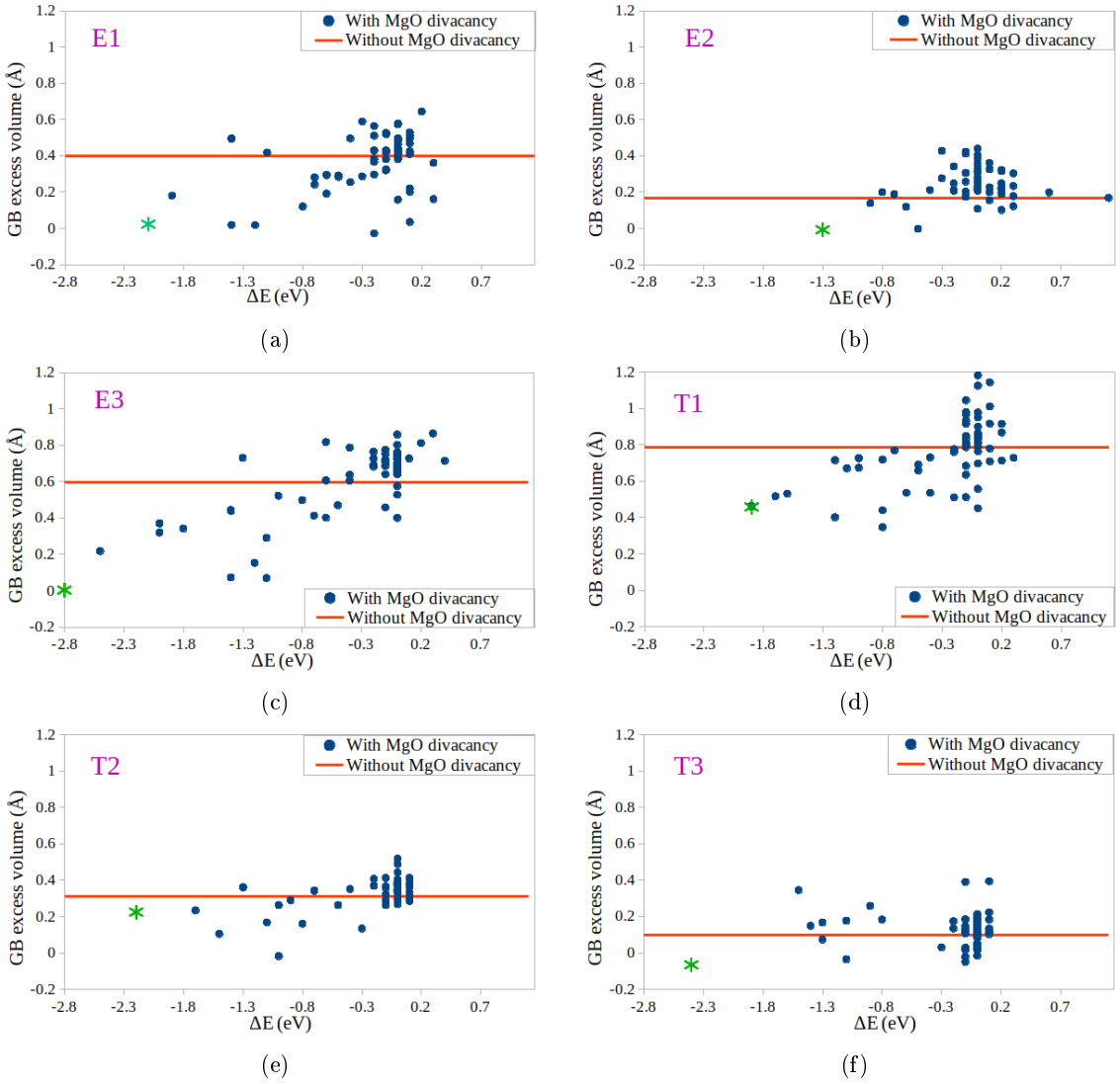

Figure S1: Segregation energy ΔE (see Eq.5) and excess volume of the different non-stoichiometric GBs, the excess volume of the parent stoichiometric GBs (see Fig.4) are represented by the orange lines. The green stars highlight the complexions presented in figure 7.

GB label	Total energy (eV)		Excess volume ($\AA$)	
	MD	DFT	MD	DFT
E1n	-12083.92	-2897.89	0.36	0.33
E2n	-12084.01	-2896.67	0.34	0.52
E3n	-12081.31	-2895.91	0.33	0.39
T1n	-12081.52	-2893.61	0.80	0.88
T2n	-12082.28	-2893.43	0.58	0.84
T3n	-12082.85	-2895.97	0.28	0.23

Table S1: Summary of the total energies and excess volumes for the lowest energy non-stoichiometric complexions obtained by MD and DFT.