Supplementary Information

Structural and Thermoelectric Properties of $Mn(Si_{1-x}Al_x)_{1.75}$: A Commensurate Phase Mixture Approximation

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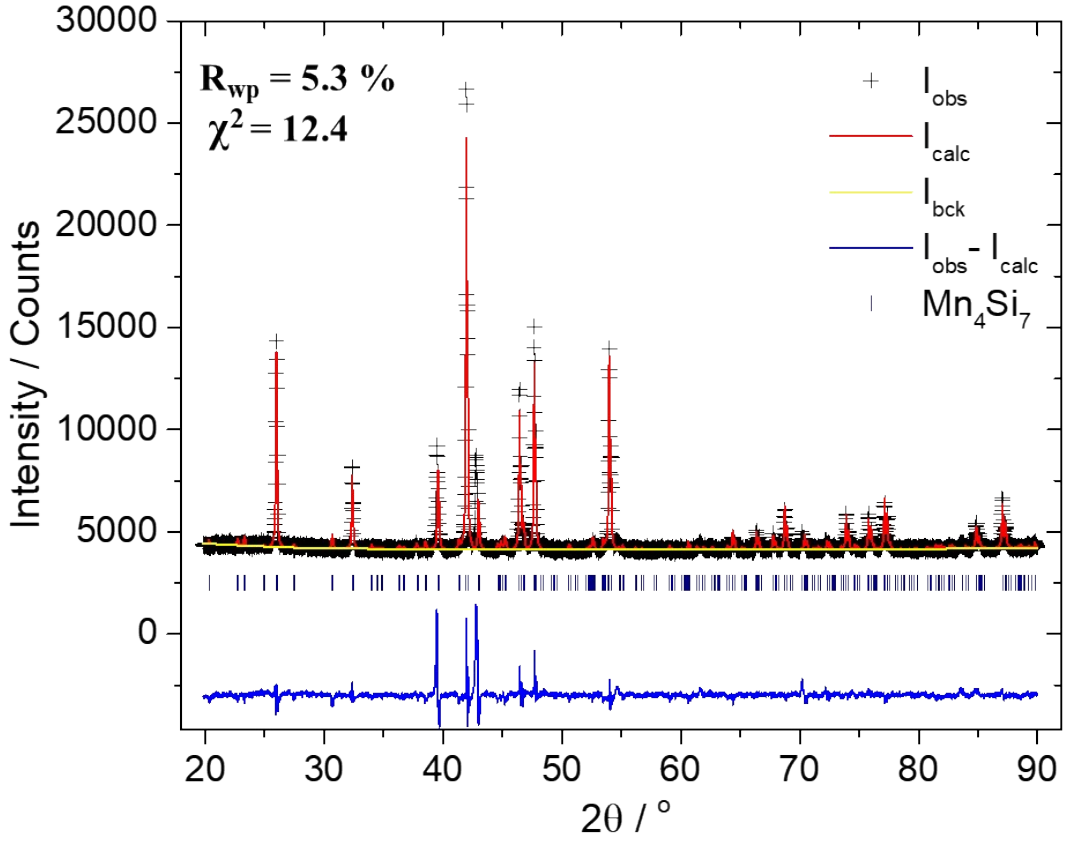

Figure S1 Powder XRD Rietveld refinement profile of $MnSi_{1.75}$ for the single phase $P\bar{4}c2$ Mn_4Si_7 model. Final observed (black crosses), calculated (red solid line), calculated background (yellow line) and difference (blue line).

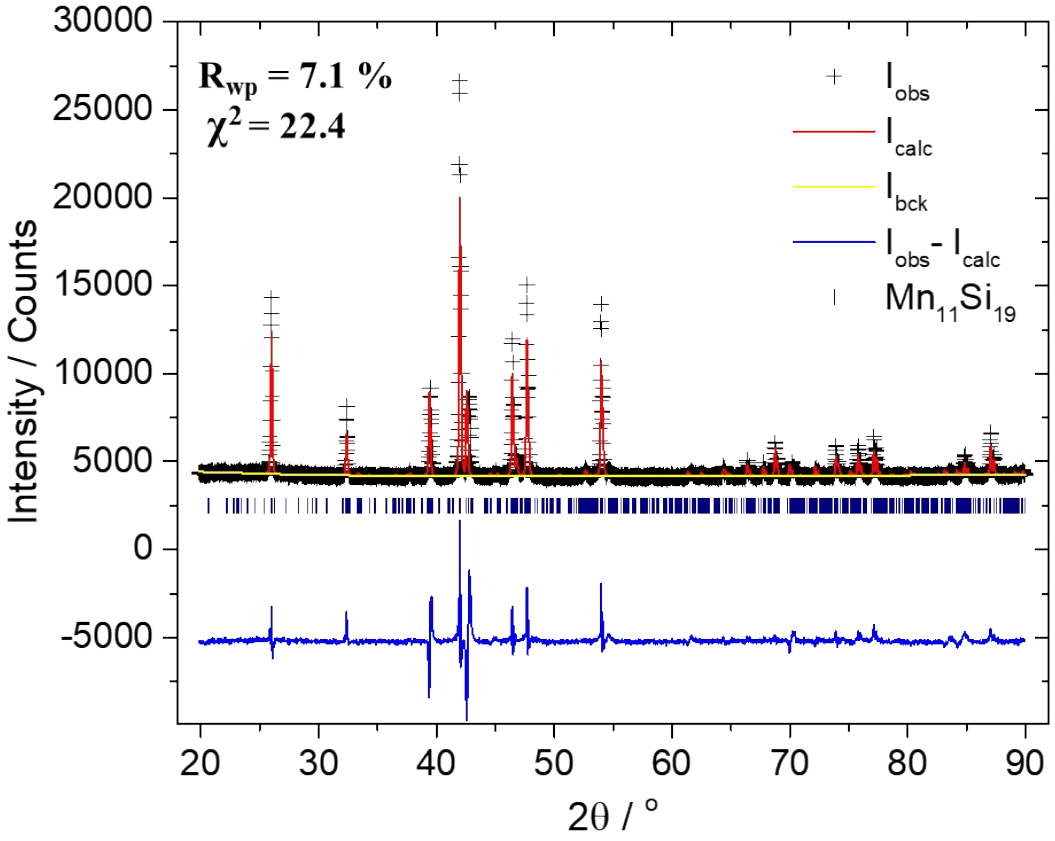

Figure S2 Powder XRD Rietveld refinement profile of $MnSi_{1.75}$ for the single phase $P\bar{4}n2$ $Mn_{11}Si_{19}$ model. Final observed (black crosses), calculated (red solid line), calculated background (yellow line) and difference (blue line).

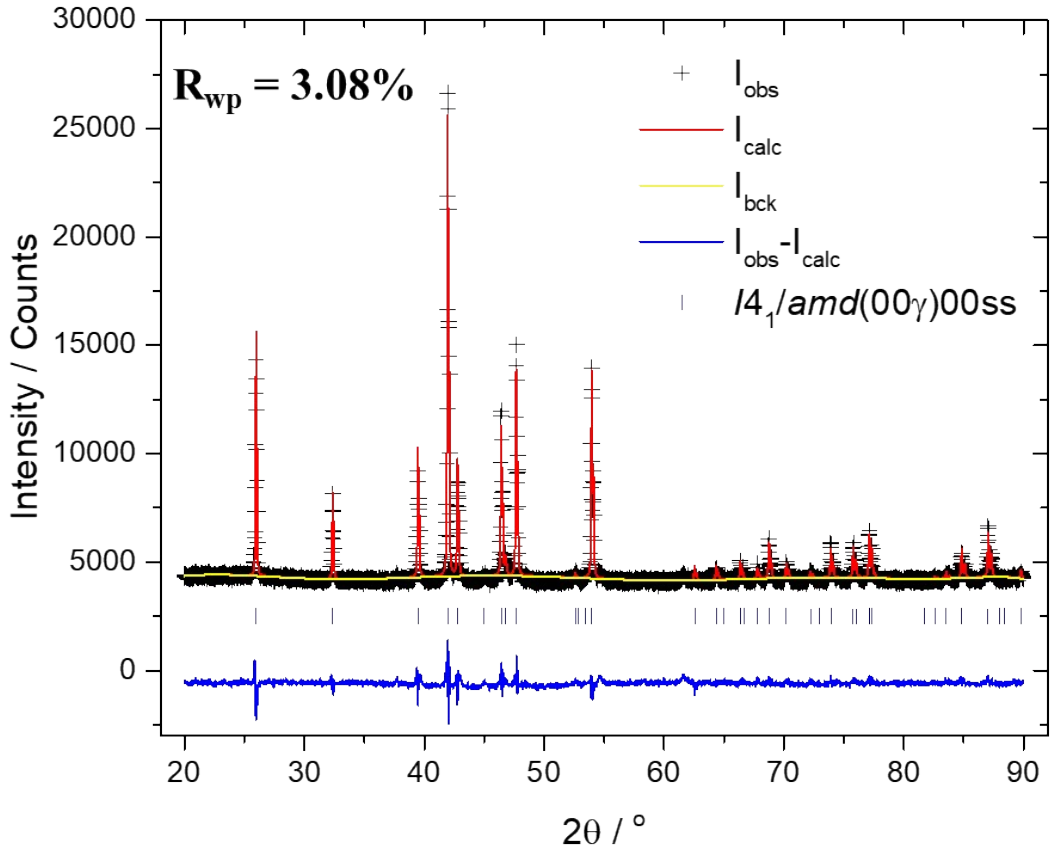

Figure S3 Le Bail refinement profile of $MnSi_{1.75}$ for the incommensurate structural model with the (3+1)D superspace group $I4_1/amd(00\gamma)00ss$ (*gof* = 2.04). Final observed (black crosses), calculated (red solid line), calculated background (yellow line) and difference (blue line).

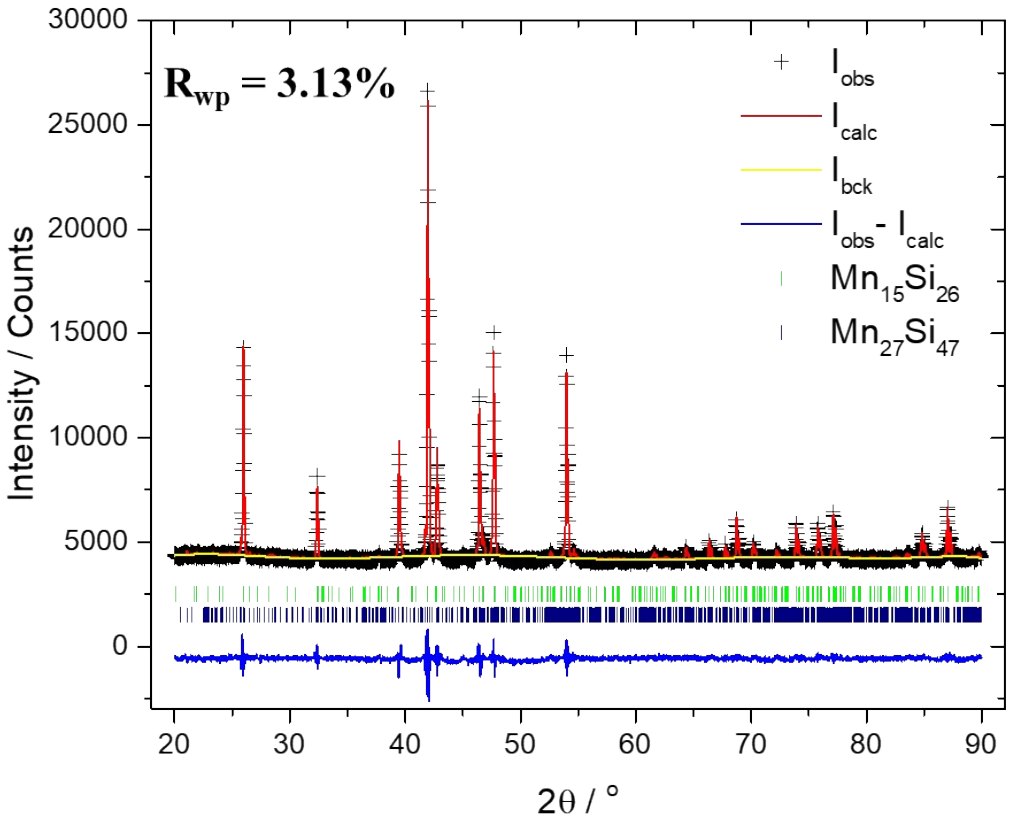

Figure S4 Le Bail refinement profile of $MnSi_{1.75}$ for the commensurate phase mixture model of $P\bar{4}n2$ $Mn_{27}Si_{47}$ and $I\bar{4}2d$ $Mn_{15}Si_{26}$ (*gof* = 2.07). Final observed (black crosses), calculated (red solid line), calculated background (yellow line) and difference (blue line).

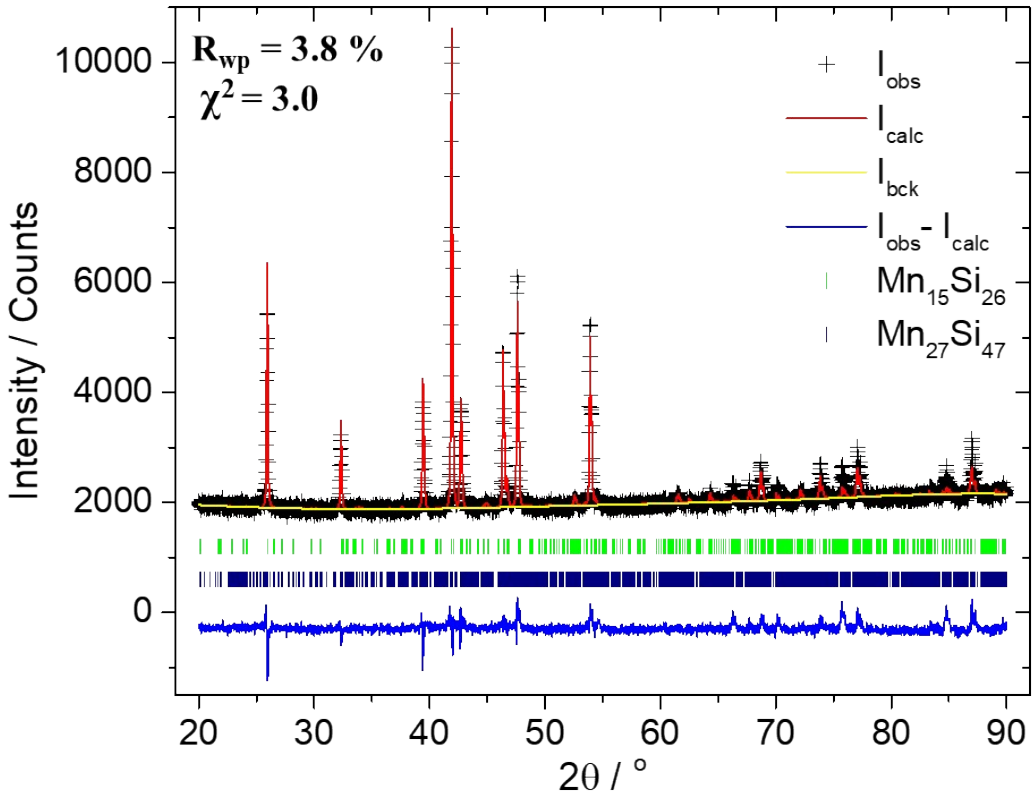

Figure S5 Powder XRD Rietveld refinement profile of $Mn(Si_{0.98}Al_{0.02})_{1.75}$ for the phase mixture structural model. Final observed (black crosses), calculated (red solid line), calculated background (yellow line) and difference (blue line). Reflection positions for the two phases, $Mn_{15}Si_{26}$ and $Mn_{27}Si_{47}$, are marked with green and navy color respectively.

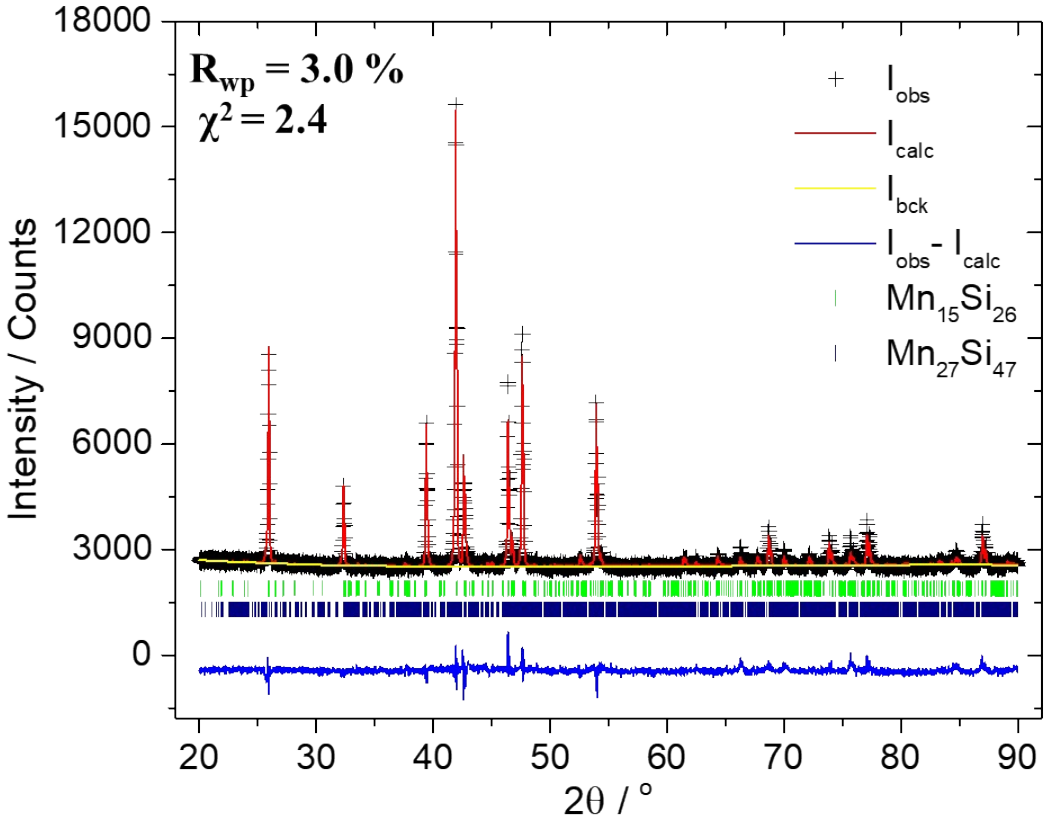

Figure S6 Powder XRD Rietveld refinement profile of $Mn(Si_{0.975}Al_{0.025})_{1.75}$ for the phase mixture structural model. Final observed (black crosses), calculated (red solid line), calculated background (yellow line) and difference (blue line). Reflection positions for the two phases, $Mn_{15}Si_{26}$ and $Mn_{27}Si_{47}$, are marked with green and navy color respectively.

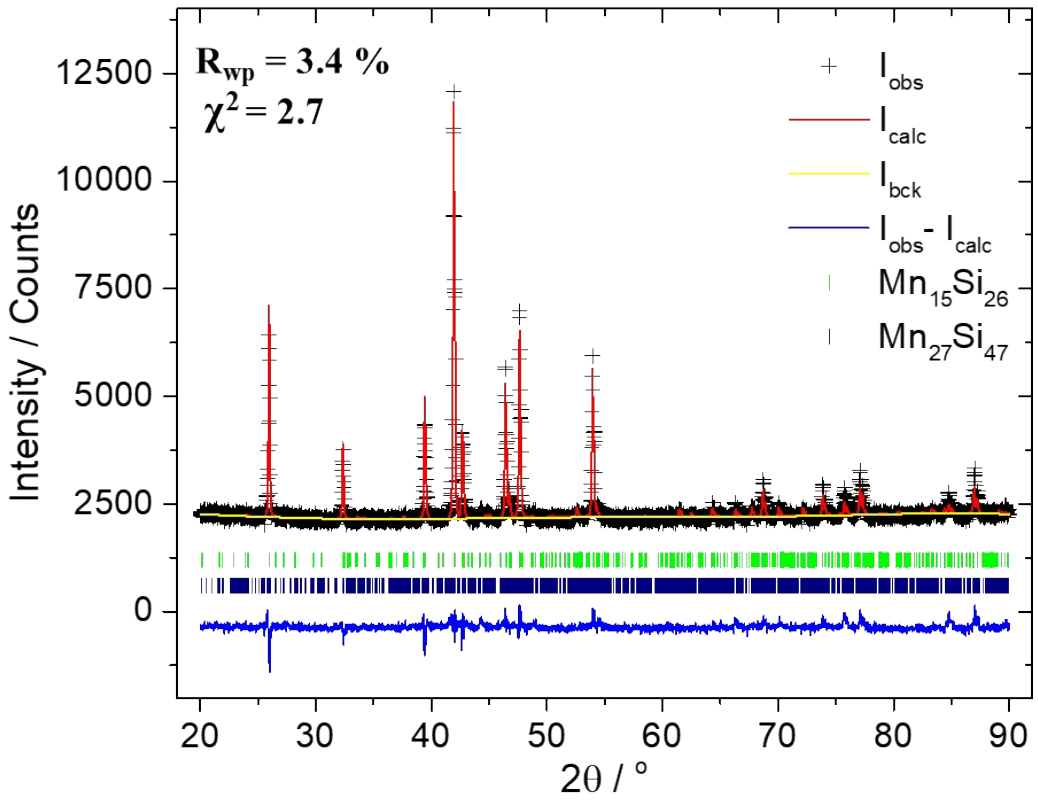

Figure S7 Powder XRD Rietveld refinement profile of $Mn(Si_{0.97}Al_{0.03})_{1.75}$ for the phase mixture structural model. Final observed (black crosses), calculated (red solid line), calculated background (yellow line) and difference (blue line). Reflection positions for the two phases, $Mn_{15}Si_{26}$ and $Mn_{27}Si_{47}$, are marked with green and navy color respectively.

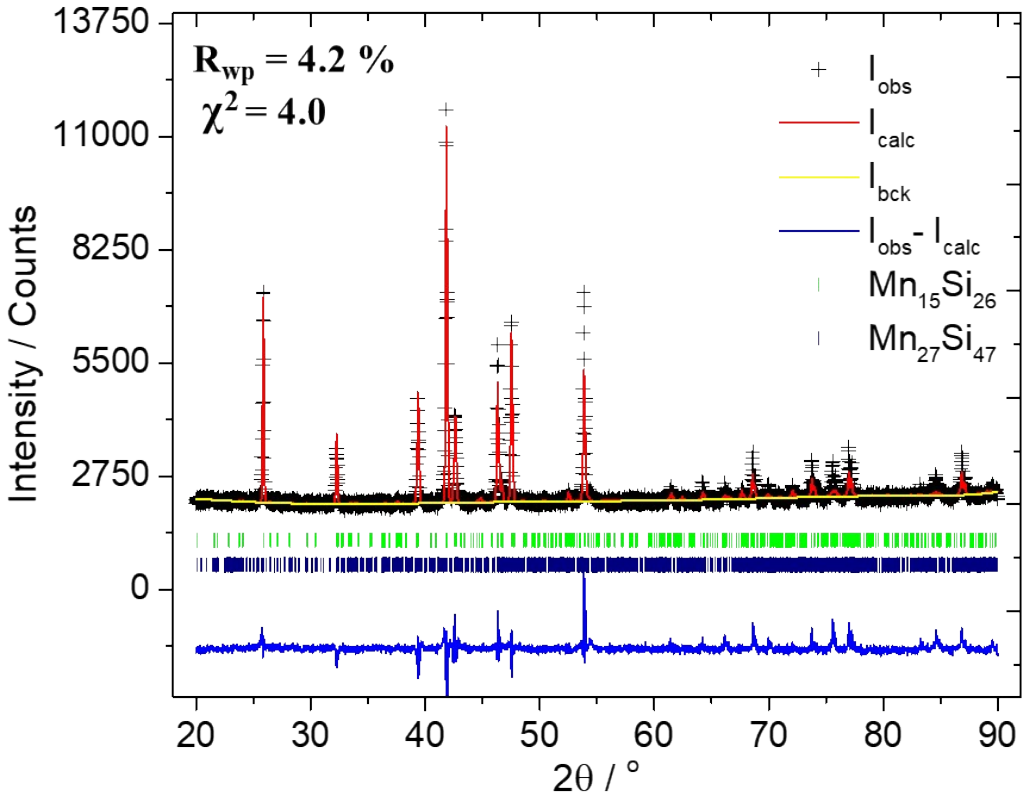

Figure S8 Powder XRD Rietveld refinement profile of $Mn(Si_{0.96}Al_{0.04})_{1.75}$ for the phase mixture structural model. Final observed (black crosses), calculated (red solid line), calculated background (yellow line) and difference (blue line). Reflection positions for the two phases, $Mn_{15}Si_{26}$ and $Mn_{27}Si_{47}$, are marked with green and navy color respectively.

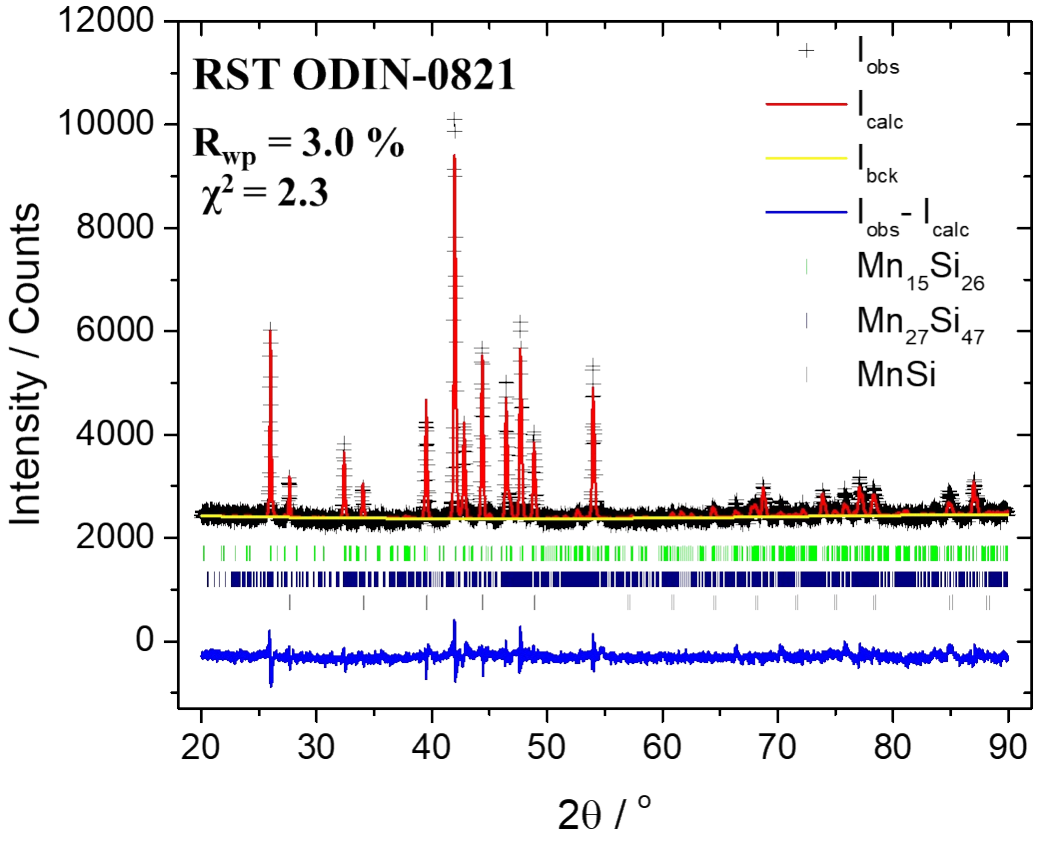

Figure S9 Powder XRD Rietveld refinement profile of $Mn(Si_{0.975}Al_{0.025})_{1.75}$ based on RST ODIN-0821 silicon kerf for the phase mixture structural model. Final observed (black crosses), calculated (red solid line), calculated background (yellow line) and difference (blue line). Reflection positions for the $Mn_{15}Si_{26}$, $Mn_{27}Si_{47}$ and MnSi phases are marked with green, navy and gray color respectively.

Table S1 Refined lattice parameters and modulation vector extracted from Le Bail refinement of incommensurate model with the (3+1)D superspace group $I4_1/amd(00\gamma)00ss$.

γ	a / Å	c_{Mn} / Å	c_{Si} / Å
1.739	5.52519(6)	4.36513(7)	2.51014(7)

Table S2 Densities of fabricated $Mn(Si_{1-x}Al_x)_{1.75}$ ($0 \leq x \leq 0.04$) pellets: experimental and theoretical values calculated by Rietveld analysis.

x	Experimental / g cm^{-3}	Theoretical / g cm^{-3}
0	4.95	5.17
0.02	4.98	5.16
0.025	4.95	5.15
0.03	5.03	5.16
0.04	5.06	5.15
0.025, RST 1-2	5.15	5.28
0.025, RST ODIN-0821	5.13	5.29

Table S3 Estimations of VEC for the two Al-doped $Mn_{27}Si_{47}$ ($\gamma = 1.74$) and $Mn_{15}Si_{26}$ ($\gamma = 1.73$) in the phase mixture according to the formula: $VEC = 7 + (4(1 - x) - 3x)\gamma$

x	$Mn_{27}Si_{47}$	$Mn_{15}Si_{26}$
0	13.96	13.92
0.02	13.7164	13.6778
0.025	13.6555	13.61725
0.03	13.5946	13.5567
0.04	13.4728	13.4356

Table S4 Refined parameters from Rietveld analysis of powder XRD data for $Mn(Si_{0.975}Al_{0.025})_{1.75}$ phases based on the two Si kerfs, RST ODIN-0821 and RST 1-2.

Phase Mixture	$Mn_{27}Si_{47}$			$Mn_{15}Si_{26}$			MnSi
Si-kerf	wt. %	*a* / Å	*c* / Å	wt. %	*a* / Å	*c* / Å	wt. %
RST 1-2	50.8(4)	5.5315(3)	117.97(1)	29.3(4)	5.5286(2)	65.530(3)	19.9(1)
RST ODIN-0821	52.5(4)	5.5301(2)	117.987(6)	27.9(5)	5.5305(4)	65.525(7)	19.6(2)