Supplementary Information

Scaling up Si-C composite synthesis from recycled graphite for high-energy-density and low-environmental-impact batteries

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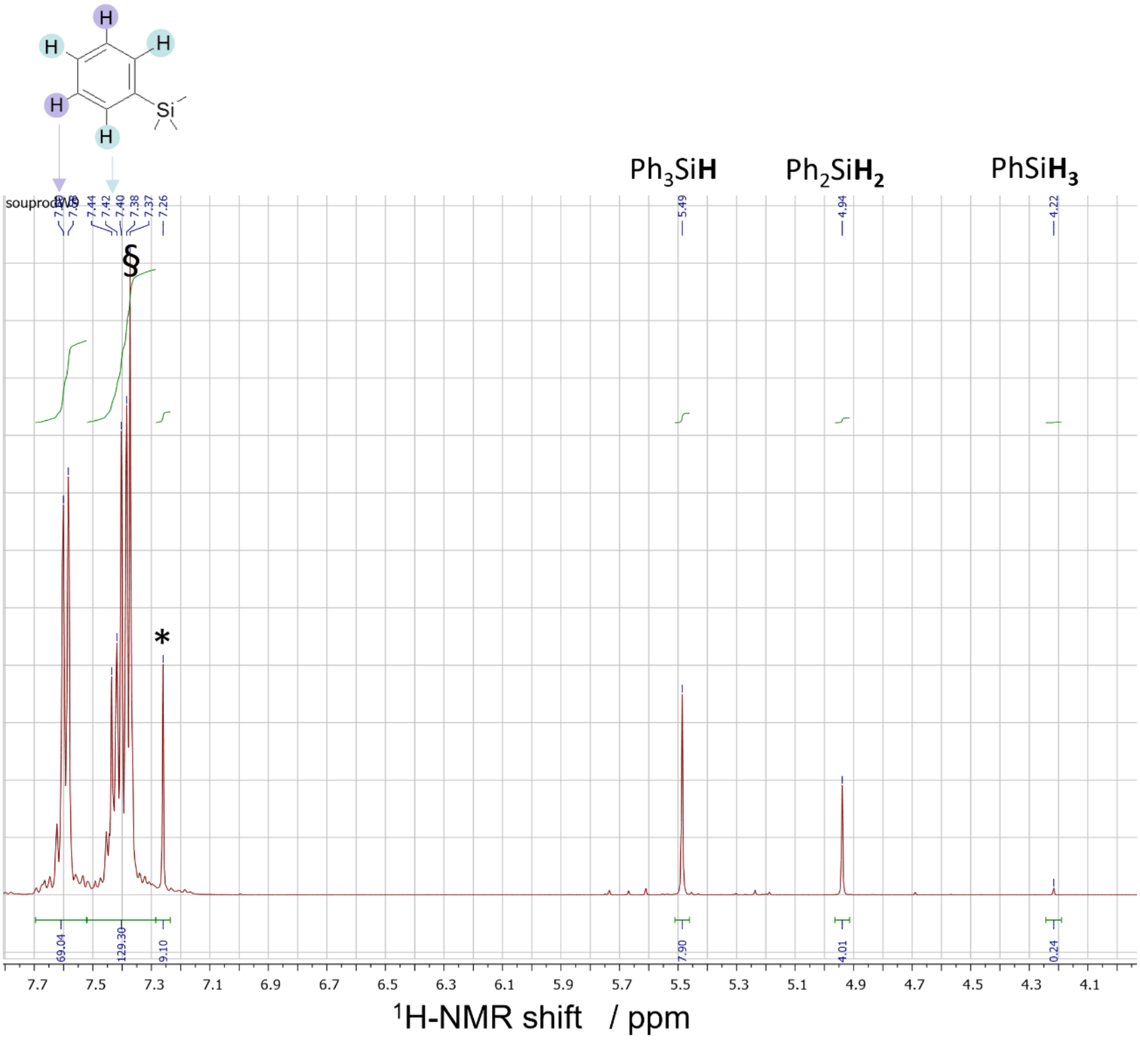


Figure S1. ^{1}H-NMR spectrum of a diphenylsilane sample after incubation at 400°C. Relaxation time T1 was set to 1 minute which was found long enough for the complete relaxation of all protons in the mixture. Phenylsilane, diphenylsilane and triphenylsilane were quantified from the integral of their characteristic Si-H peaks at 4.2, 4.9 and 5.4 ppm respectively. The expected intensity of the aromatic peaks from these species was calculated from this quantitation and subtracted from the measured integrals of the aromatic cluster at 7.65 ppm. This value was used to quantify tetraphenylsilane. The expected integral of the aromatic peaks in the cluster at 7.4 ppm for all silane species was subtracted from the measured integral. This value was used to quantify the amount of benzene, which gives a single peak at 7.37 ppm (§). The presence of benzene was further checked by ^{13}C NMR, on which the benzene signal does not super-impose with the peaks from the silane phenyls. (*) $CHCl_3$.

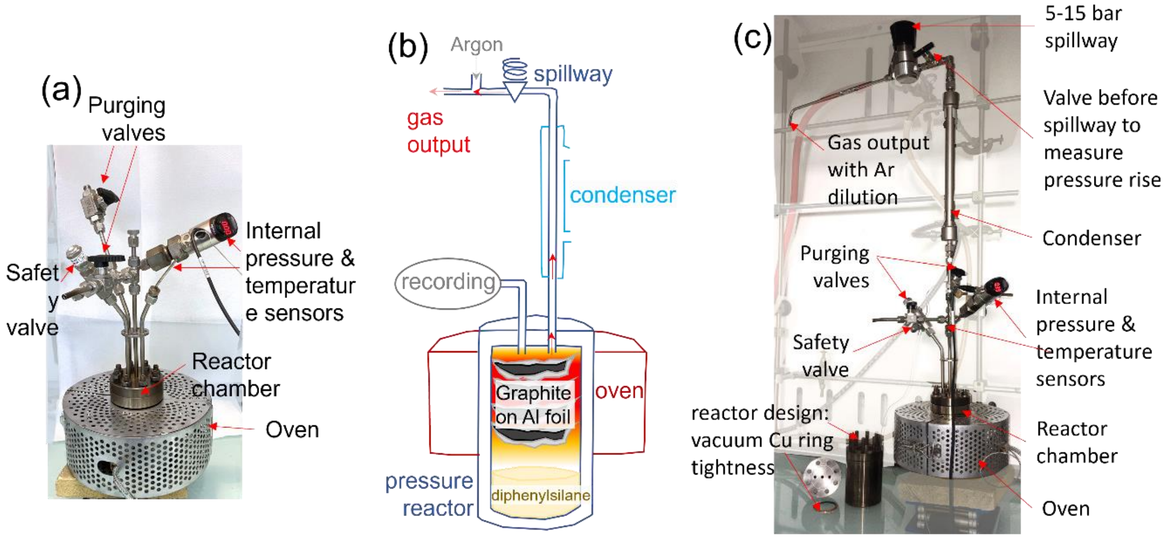


Figure S2: Pressure reactor scheme and photographs: (a) closed reactor, (b,c) half-open reactor. (b) Scheme of the half-open reactor design for GtSi synthesis, including the reaction chamber, connections to the pressure and temperature recording, a pipe to the exhaust through a water-cooled condenser and a spillway. The exhaust gas is diluted with a neutral gas before escaping to the hood venting. The stock of diphenylsilane at the bottom of the chamber lays below the heated zone. The graphite with SnS seeds is introduced on aluminum foil in the heated zone.

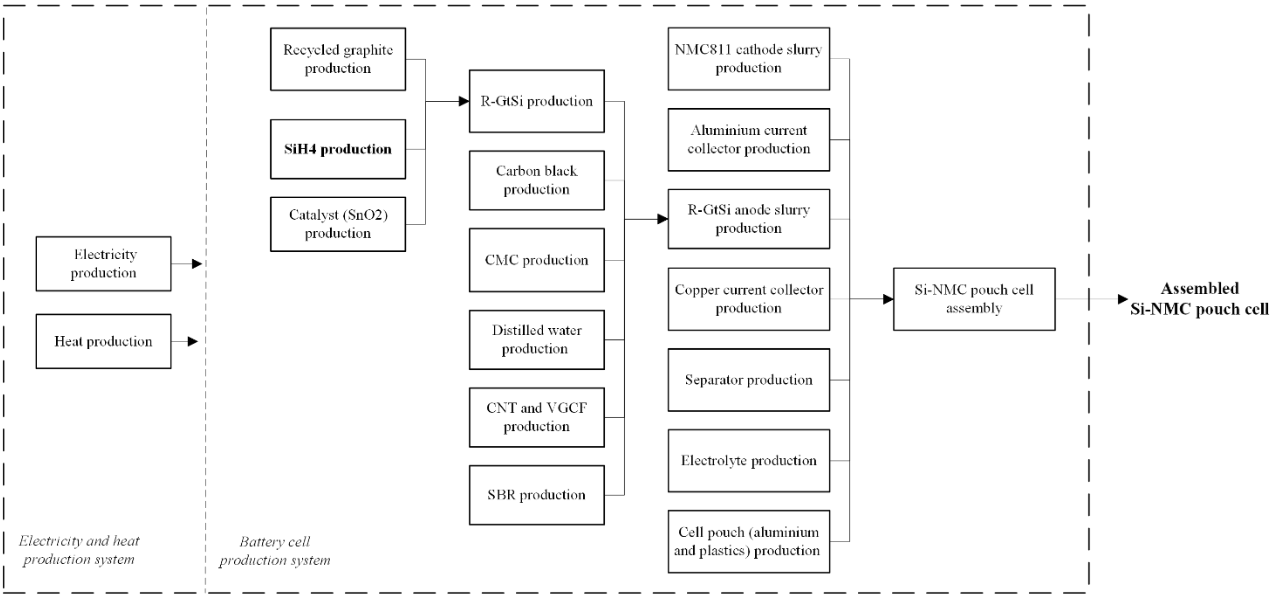


Figure S3. System boundary for the Life Cycle Assessment (LCA) of the 40 mAh Si–NMC single-layer pouch cell.

Table S1: elemental analysis by quantitative Emission Dispersion Spectroscopy (EDS) Gt and Rec-Gt

Weight %	C	O	Al	Si	Ni	Co	Na	Fe
Recycled graphite	94.7	4.1	0.13	0.02	0.85	0.05	0.05	0.01

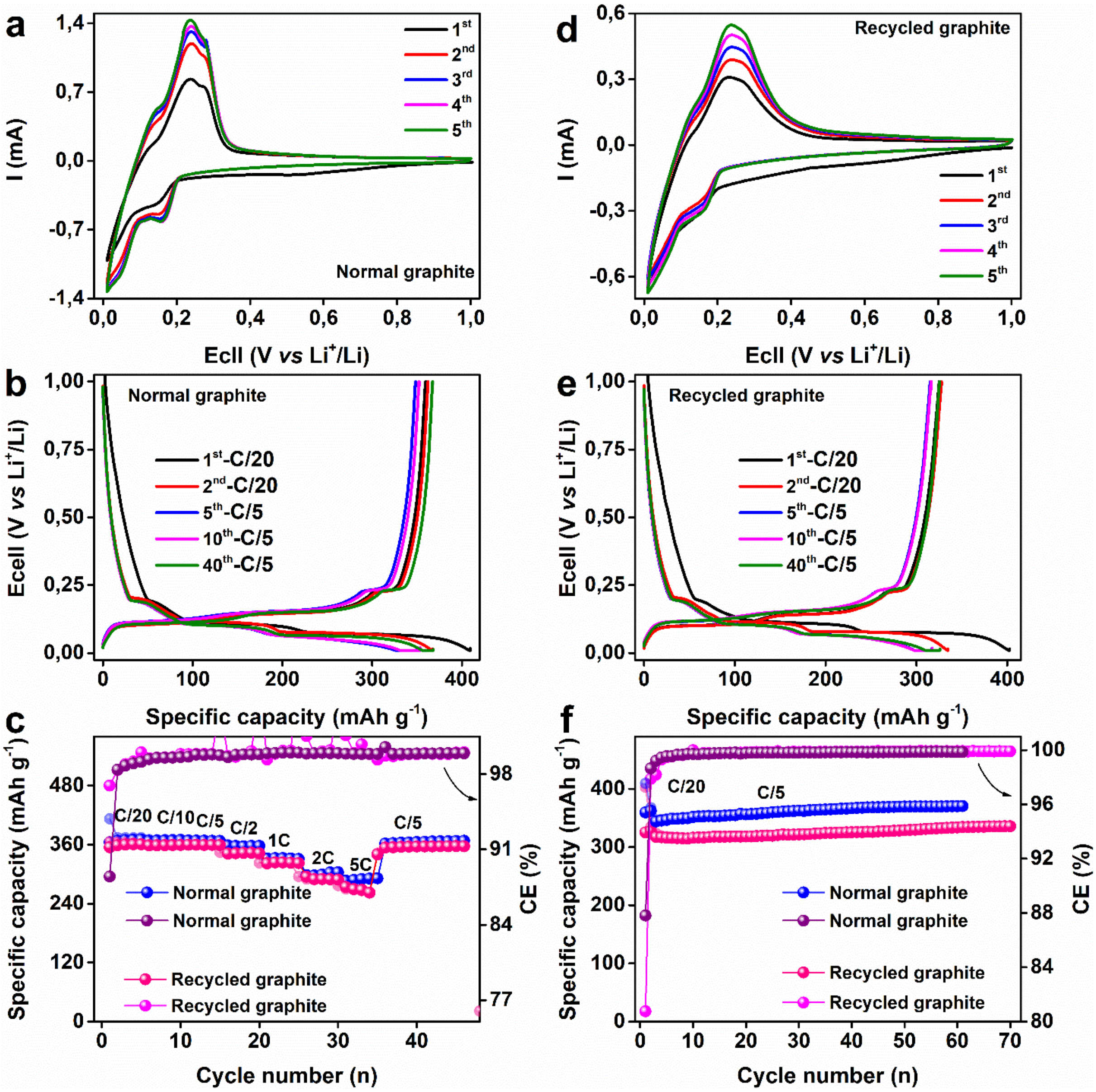


Figure S4: Electrochemical performance: (a, b) and (d, e) represent CV and discharge/charge curves of normal graphite and recycled graphite, respectively; (c) represents C-rate and (e) long cycle behaviors at C/5 with 3 initial cycles at C/20 of normal graphite and recycled graphite.

Aqueous slurry preparation: 80% active material, 10% super P, and 10% of Na-CMC ; Mass loading: graphite cell (b, f) = 2.16 mg.cm^{-2} (c) = 2.10 mg.cm^{-2}; Recycled graphite cell (e, f) = 2.23 mg.cm^{-2} (c) = 2.32 mg.cm^{-2}; Working electrode: disk of 14 mm diameter ; Electrolyte: 1 M of $LiPF_6$ in EC/DEC (1:1); Separator: Celgard 2500, disk of 18 mm diameter;; Potential window: 0.01-1 V

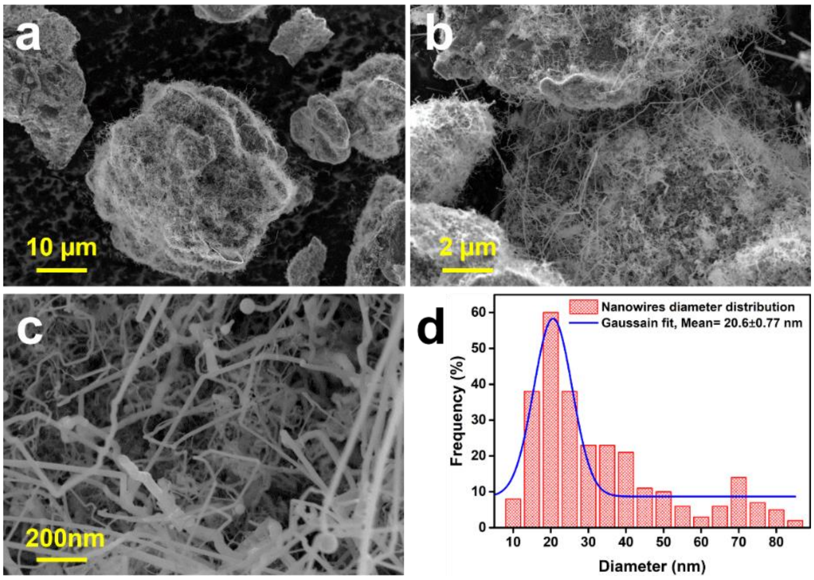


Figure S5: Scanning electron microscopy (SEM) images of GtSi composites at different magnification (a, b, c). (d) Diameter distribution of Si nanowires, showing a preferential diameter of 20 +/- 1 nm.

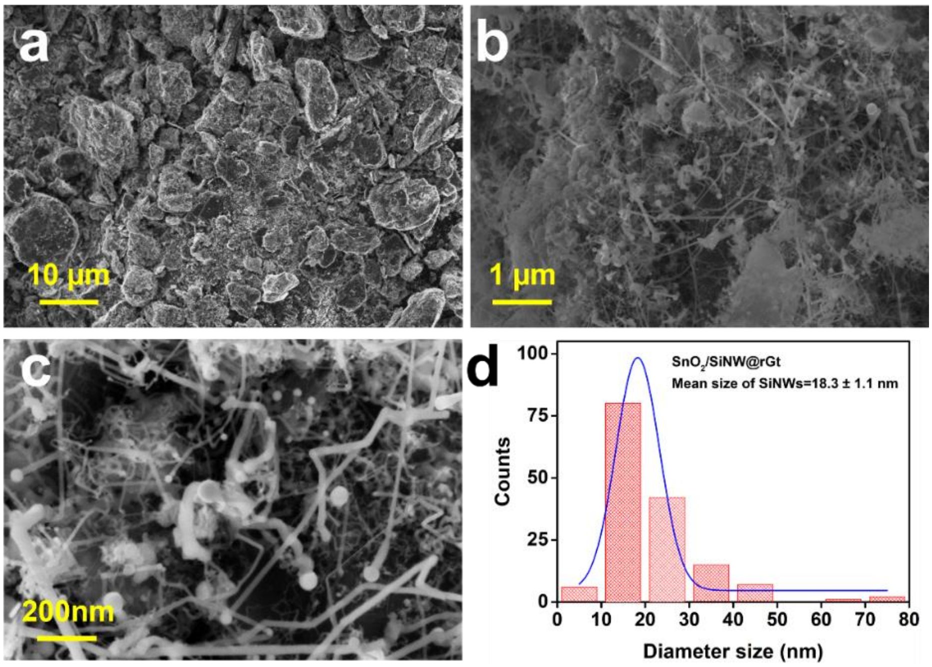


Figure S6: SEM images of Rec-GtSi composites at different magnification (a, b, c). (d) Diameter distribution of Si nanowires, showing a preferential diameter of 18 +/- 1 nm.

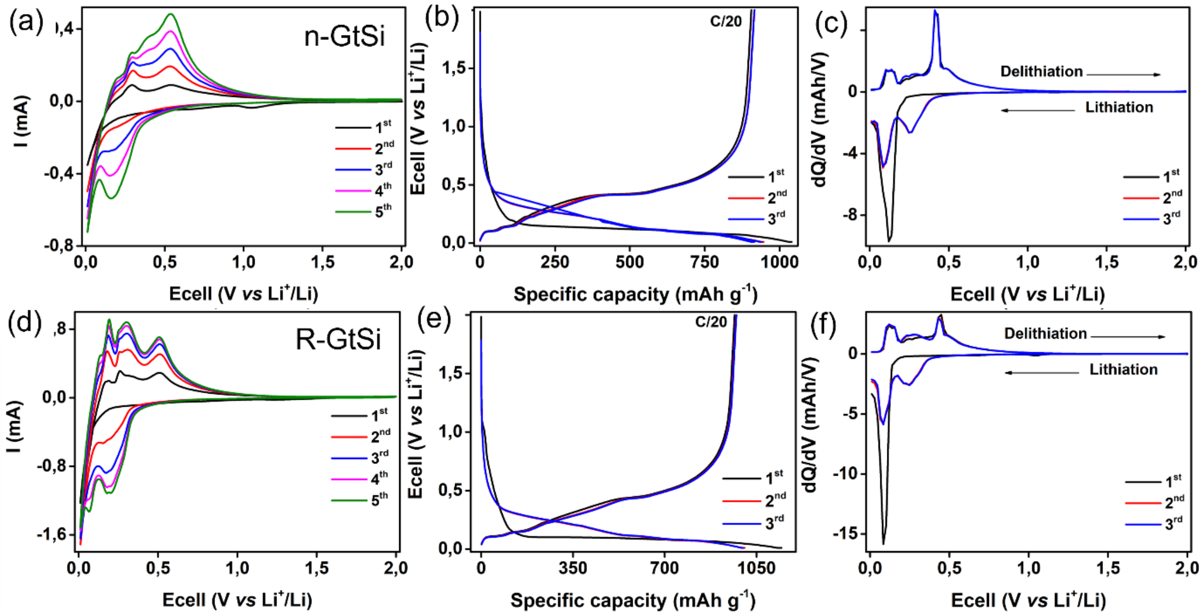


Figure S7. Electrochemical characterization of n-GTSi (a-c) and R-GtSi (d-f) anode materials in coin cells in half-cell configuration: cyclic voltammograms (a,d), specific capacity vs potential (b,e) and corresponding incremental capacity (c,f). The electrolyte was 1 M of $LiPF_6$ in EC/DMC/EMC 1:1:1 + 10% FEC. 14 mm diameter electrode disks were prepared from a slurry 80% active material, 10% super P, and 10% of CMC in deionized water. Mass loading of active material was 2.23 mg.cm^{-2}. Potential window 0.01 to 2 V vs Li^+/Li.

Table S2. Composition in molar percentage of samples of **diphenylsilane** after thermal decomposition at 300°C and 400°C as a function of time, as determined by ^{1}H-NMR. The value for SiH_4 is an estimate using mass conservation law.

hours at **300°C**	Ph_4Si	Ph_3SiH	Ph_2SiH_2	$PhSiH_3$	SiH_4	benzene
0	- 0.5 ± 0.5	0	100.5 ± 1	0	0	0
1	1.1 ± 0.1	9.5 ± 0.4	79.2 ± 1	9.5 ± 0.2	0.7 ± 2	0.8 ± 1
3	0.6 ± 0.1	13.2 ± 0.5	73.4 ± 1	12.7 ± 0.3	0.1 ± 2	1.5 ± 1
6.8	1.2 ± 0.1	15.2 ± 0.6	68.9 ± 1	14.2 ± 0.3	0.5 ± 2	2.3 ± 1
25	3.6 ± 0.4	32.2 ± 0.9	44.7 ± 0.6	20.9 ± 0.4	-1.3 ± 2	20.9 ± 2
40	5.6 ± 0.4	42.2 ± 0.9	22.0 ± 0.4	17.6 ± 0.4	12.6 ± 3	12.2 ± 2
hours at **400°C**						
0.5	2.5 ± 0.2	27.4 ± 0.8	45.6 ± 1	19.8 ± 0.5	4.7 ± 2	3.0 ± 1
1	3.0 ± 0.3	31.0 ± 0.9	39.7 ± 1	19.6 ± 0.5	6.7 ± 3	4.0 ± 1
3	7.5 ± 0.6	37.6 ± 0.9	24.9 ± 0.7	11.7 ± 0.4	18.2 ± 3	4.5 ± 1
8.5	13.8 ± 0.6	35.9 ± 0.9	18.2 ± 0.6	6.7 ± 0.4	25.5 ± 3	5.9 ± 1

Table S3. Composition in molar percentage of samples of **phenylsilane** after thermal decomposition at 300°C and 400°C as a function of time, as determined by ^{1}H-NMR. The value for SiH_4 is an estimate using mass conservation law.

hours at **300°C**	Ph_4Si	Ph_3SiH	Ph_2SiH_2	$PhSiH_3$	SiH_4	benzene
0	0	0	0	99.9 ± 1	0	0
2	0.2 ± 0.1	0.4 ± 0.1	3.5 ± 0.1	90.3 ± 0.9	5.5 ± 2	0.6 ± 1
8	0.3 ± 0.1	1.4 ± 0.1	12.9 ± 0.2	66.5 ± 0.7	18.9 ± 2	2.7 ± 1
16	0.9 ± 0.1	2.5 ± 0.1	15.5 ± 0.3	56.9 ± 0.6	24.2 ± 2	1.3 ± 1
31	0.6 ± 0.1	2.9 ± 0.1	16.0 ± 0.3	55.5 ± 0.5	25.0 ± 2	1.8 ± 1
65	0.6 ± 0.1	4.4 ± 0.2	18.5 ± 0.4	46.4 ± 0.4	30.2 ± 3	1.6 ± 1
hours at **400°C**						
0.5	1.3 ± 0.1	4.7 ± 0.2	22.9 ± 0.4	32.7 ± 0.3	38.8 ± 2	3.0 ± 1
1	1.7 ± 0.2	5.9 ± 0.3	22.2 ± 0.4	29.6 ± 0.3	40.8 ± 4	1.9 ± 1
3	2.5 ± 0.2	12.8 ± 0.5	18.4 ± 0.4	11.3 ± 0.1	55.4 ± 5	4.5 ± 1
8.5	1.4 ± 0.1	16.4 ± 0.6	16.8 ± 0.3	8.4 ± 0.1	57.6 ± 5	4.6 ± 1

Table S4. Composition in molar percentage of samples of **triphenylsilane** after thermal decomposition at 300°C and 400°C as a function of time, as determined by ^{1}H-NMR. The value for SiH_4 is an estimate using mass conservation law.

hours at **300°C**	Ph_4Si	Ph_3SiH	Ph_2SiH_2	$PhSiH_3$	SiH_4	benzene
0	0.6 ± 0.5	99.4 ± 1	0	0	0	0
2	2.8 ± 0.2	96.8 ± 1	1.1 ± 0.1	0.0 ± 0.1	-0.7 ± 2	0.6 ± 1
16	2.3 ± 0.2	96.9 ± 1	1.5 ± 0.1	0.0 ± 0.1	-0.7 ± 2	1.3 ± 1
31	5.8 ± 0.5	90.5 ± 0.9	3.8 ± 0.2	0.2 ± 0.1	-0.3 ± 2	1.8 ± 1
65	16.4 ± 0.8	74.0 ± 0.7	7.0 ± 0.3	0.8 ± 0.1	1.8 ± 2	1.6 ± 1
hours at **400°C**						
1	3.8 ± 0.3	93.2 ± 1.0	2.7 ± 0.1	0.1 ± 0.05	0.2 ± 2	0.8 ± 1
3	21.8 ± 0.7	63.4 ± 0.8	11.1 ± 0.3	1.5 ± 0.1	2.3 ± 2	1.7 ± 1
8.5	33.5 ± 0.9	48.0 ± 0.6	11.3 ± 0.3	2.4 ± 0.1	4.8 ± 2	6.3 ± 1
15	39.7 ± 1	44.7 ± 0.6	8.1 ± 0.2	1.2 ± 0.1	6.2 ± 3	21.1 ± 2

Note S1. **Determination of rate constants of phenyl/hydride exchange between phenylsilane species**

The expected reactions for the thermal decomposition of mono-, di- and tri-phenylsilane are the following equilibria of phenyl and hydride exchange:

Reactions				Equilibrium constant	Forward reaction rate
2 $SiPh_2H_2$	⇔	$SiPhH_3$ + $SiPh_3H$	(3)	K_2	p_2
$SiPhH_3$ + $SiPh_2H_2$	⇔	SiH_4 + $SiPh_3H$	(4)	$K_{12} = K_1K_2$	p_{12}
$SiPhH_3$ + $SiPh_3H$	⇔	SiH_4 + $SiPh_4$	(5)	$K_{13} = K_1K_2K_3$	p_{13}
2 $SiPhH_3$	⇔	SiH_4 + $SiPh_2H_2$	(6)	K_1	p_1
$SiPh_2H_2$ + $SiPh_3H$	⇔	$SiPh_4$ + $SiPhH_3$	(7)	$K_{23} = K_2K_3$	p_{23}
2 $SiPh_3H$	⇔	$SiPh_4$ + $SiPh_2H_2$	(8)	K_3	p_3

To simplify notations, we introduce short names for the five species as follows: the concentrations in SiH_4, $SiPhH_3$, $SiPh_2H_2$, $SiPh_3H$ and $SiPh_4$ are noted C_0, C_1, C_2, C_3, and C_4 respectively. They are expected to follow the rate laws as:

$$\frac{dC_0}{dt} = p_{12}C_1C_2 - \frac{p_{12}}{K_{12}}C_0C_3 + p_1C_1^2 - \frac{p_1}{K_1}C_0C_2 + p_{13}C_1C_3 - \frac{p_{13}}{K_{13}}C_0C_4$$

$$\frac{dC1}{dt} = p_2C_2^2 - \frac{p2}{K2}C_1C_3 - p_{12}C_1C_2 + \frac{p12}{K12}C_0C_3 - 2p_1C_1^2 + 2\frac{p1}{K1}C_0C_2 + p_{23}C_2C_3 - \frac{p23}{K23}C_1C_4 - p_{13}C_1C_3 + \frac{p13}{K13}C_0C_4$$

$$\frac{dC2}{dt} = -2p_2C_2^2 + 2\frac{p2}{K2}C_1C_3 - p_{12}C_1C_2 + \frac{p12}{K12}C_0C_3 + p_1C_1^2 - \frac{p1}{K1}C_0C_2 - p_{23}C_2C_3 + \frac{p23}{K23}C_1C_4 + p_3C_3^2 - \frac{p3}{K3}C_2C_4$$

$$\frac{dC3}{dt} = p_2C_2^2 - \frac{p2}{K2}C_1C_3 + p_{12}C_1C_2 - \frac{p12}{K12}C_0C_3 - p_{23}C_2C_3 + \frac{p23}{K23}C_1C_4 - 2p_3C_3^2 + 2\frac{p3}{K3}C_2C_4 - p_{13}C_1C_3 + \frac{p13}{K13}C_0C_4$$

$$\frac{dC4}{dt} = p_{23}C_2C_3 - \frac{p23}{K23}C_1C_4 + p_3C_3^2 - \frac{p3}{K3}C_2C_4 + p_{13}C_1C_3 - \frac{p13}{K13}C_0C_4$$

The 15 concentration profiles over time (5 species, 3 initial conditions) were simulated with a time step of 1 minute, and fitted simultaneously to the 15 experimental concentration profiles. The 9 parameters K1, K2, K3, p2, p12, p13, p1, p23 and p3 were optimized for the best fit. The corresponding rate constants are given in Table 1 and Table S4. The fitted concentration profiles are displayed in Figure S7.

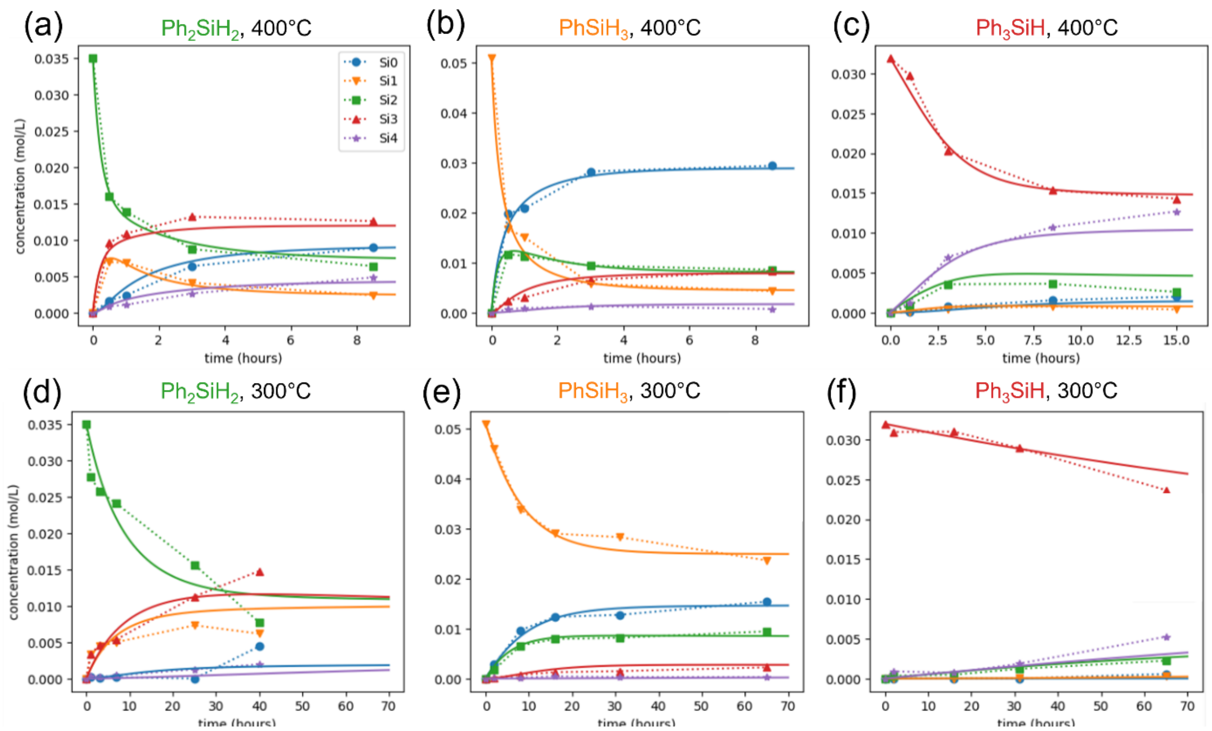


Figure S8. Concentration profiles of SiH_4 (Si0), $PhSiH_3$ (Si1), Ph_2SiH_2 (Si2), Ph_3SiH (Si3) and PH_4Si (Si4) as a function of incubation time at 400°C (a, b, c) and 300°C (d, e, f) for samples of initially pure diphenylsilane (a,d), phenylsilane (b,e) and triphenylsilane (c,f). The lines show best fits for the chemical dynamics model as detailed in Note S1. Best fitting parameters are disclosed in Table S5.

Table S5. Rate constants of forward and reverse reactions (3) to (8) from best fit with experimental concentration profiles over time. (*ND*: could not be determined).

400°C		Species giving phenyl and gaining hydride			
	($L.mol^{-1}.min^{-1}$)	$PhSiH_3$	Ph_2SiH_2	Ph_3SiH	Ph_4Si
Species giving hydride and gaining phenyl	SiH_4		0.043	0.041	0.14
	$PhSiH_3$	0.51		1.1	2.3
	Ph_2SiH_2	0.26	0.53		1.5
	Ph_3SiH	0.19	0.27	0.033	

300°C		Species giving phenyl and gaining hydride			
	($L.mol^{-1}.min^{-1}$)	$PhSiH_3$	Ph_2SiH_2	Ph_3SiH	Ph_4Si
Species giving hydride and gaining phenyl	SiH_4		0.05	0.07	*ND*
	$PhSiH_3$	0.010		0.19	*ND*
	Ph_2SiH_2	0.012	0.020		*ND*
	Ph_3SiH	*ND*	*ND*	0.001	

Note S2. Estimation of non-ideal behavior of phenylsilane and diphenylsilane vapors in the growth conditions.

The ideal gas law, that describes the behavior of gases in the limit of non-interacting molecules, must be corrected at high pressure and temperature, in particular for large molecules. The van der Waals equation for real gases takes into account the effect of intermolecular interactions and molecular size through parameters *a* and *b*, respectively, as follows:

$$\left(P + a\frac{n^2}{V^2}\right)(V - nb) = nRT \qquad (9)$$

where *P* is the pressure, *V* the volume, *T* the temperature and *n* the number of moles of gas molecules. Parameters *a* and *b* are tabulated for many gases. Although no data are available for phenylsilane and diphenylsilane, approximate values can be found from neighboring compounds like toluene and diphenylmethane. Table S6 reports *a* and *b* values for the species present in the reactor, or similar compounds. The expected value for *n* were calculated from equation (9) at 390°C and 20 bars (as in the closed reactor) and 10 bars (as in the half-open reactor) as if it was the only species in the reactor.

The ratio *PV/nRT* is reported in Table S6 as a measurement of ideality: if the molecular interactions are negligible, *PV/nRT* is close to 1. Small molecules like hydrogen and silane behave as ideal gases in these pressure and temperature conditions, but larger molecules deviate significantly. The non-ideality of toluene, considered here as a model for the reagent phenylsilane, is significant, but with a twice lower deviation at 10 bars than 20 bars. The predictions for diphenylsilane (approximated by diphenylmethane) are so far away from ideality that a more accurate model than the van der Waals equation would be required.

Table S6. Van der Waals constants *a* and *b* for gas species present in the reactor during growth or for model species (from Reference table A8 in https://chem.libretexts.org), and calculated ratio *PV/nRT* as an estimate of ideality at 390°C and 20 or 10 bar.

	a	*b*	*PV/nRT*	
	L^2.bar/mol^2	L/mol	@ 20 bar	@ 10 bar
H_2	0.24	0.0265	1.01	1.00
SiH_4	4.38	0.058	0.99	1.00
benzene	18.24	0.1193	0.92	0.96
toluene	24.38	0.1463	0.88	0.94
diphenylmethane	60.46	0.2798	0.17	0.09

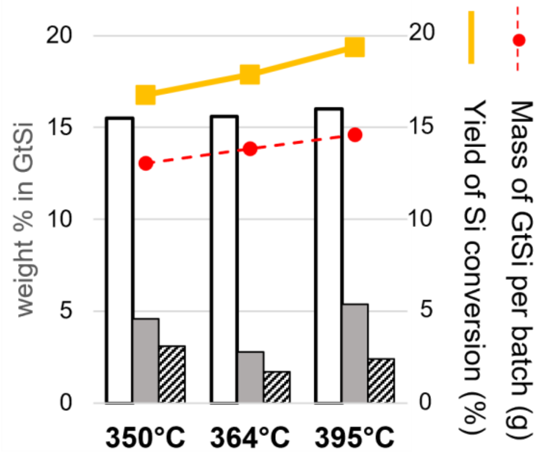


Figure S9. Comparison of n-GtSi synthesis mass production, yield of Si conversion and product elemental composition as a function of growth temperature in the closed reactor.

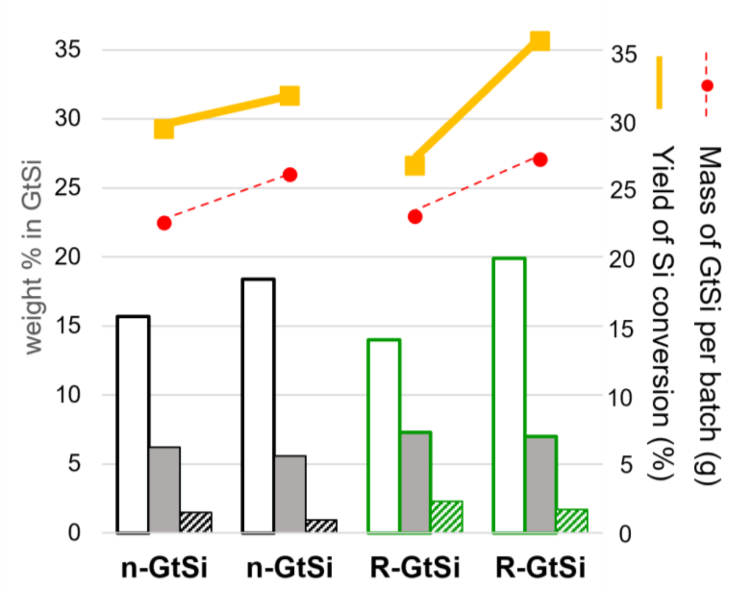


Figure S10. Comparison of n-GtSi and R-GtSi synthesis in conditions 7 and 9 described in Figure 4.

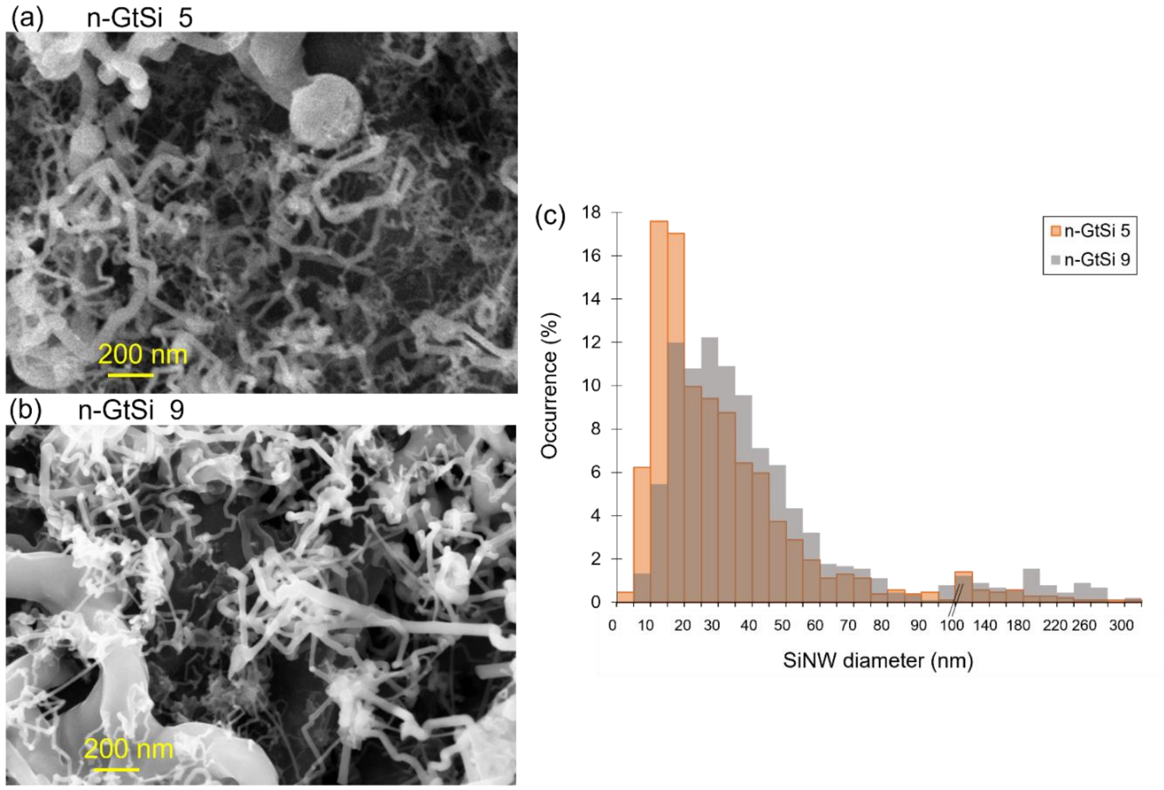


Figure S11: (a, b) Scanning electron microscopy (SEM) images of n-GtSi composites from syntheses n°5 (constant temperature growth) and 9 (scaled-up, temperature-ramped growth) as described in Figure 4, and (c) corresponding SiNW diameter distributions (>1000 counts each).

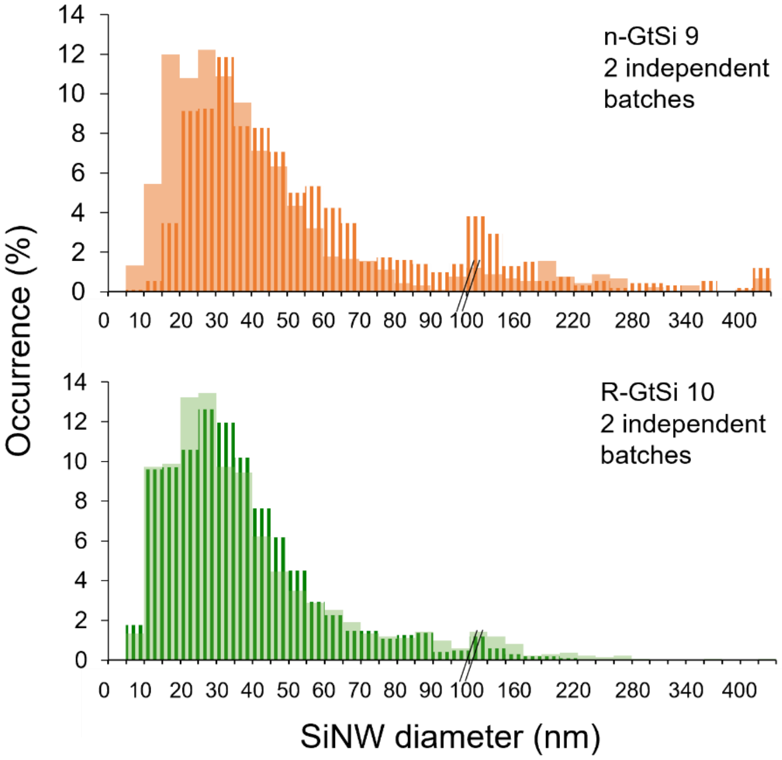


Figure S12: Diameter distribution (>1000 counts each) of Si nanowires in n-GtSi and R-GtSi of 4 independent batches obtained from identical synthesis conditions (columns 9 and 10 in Figure 4).

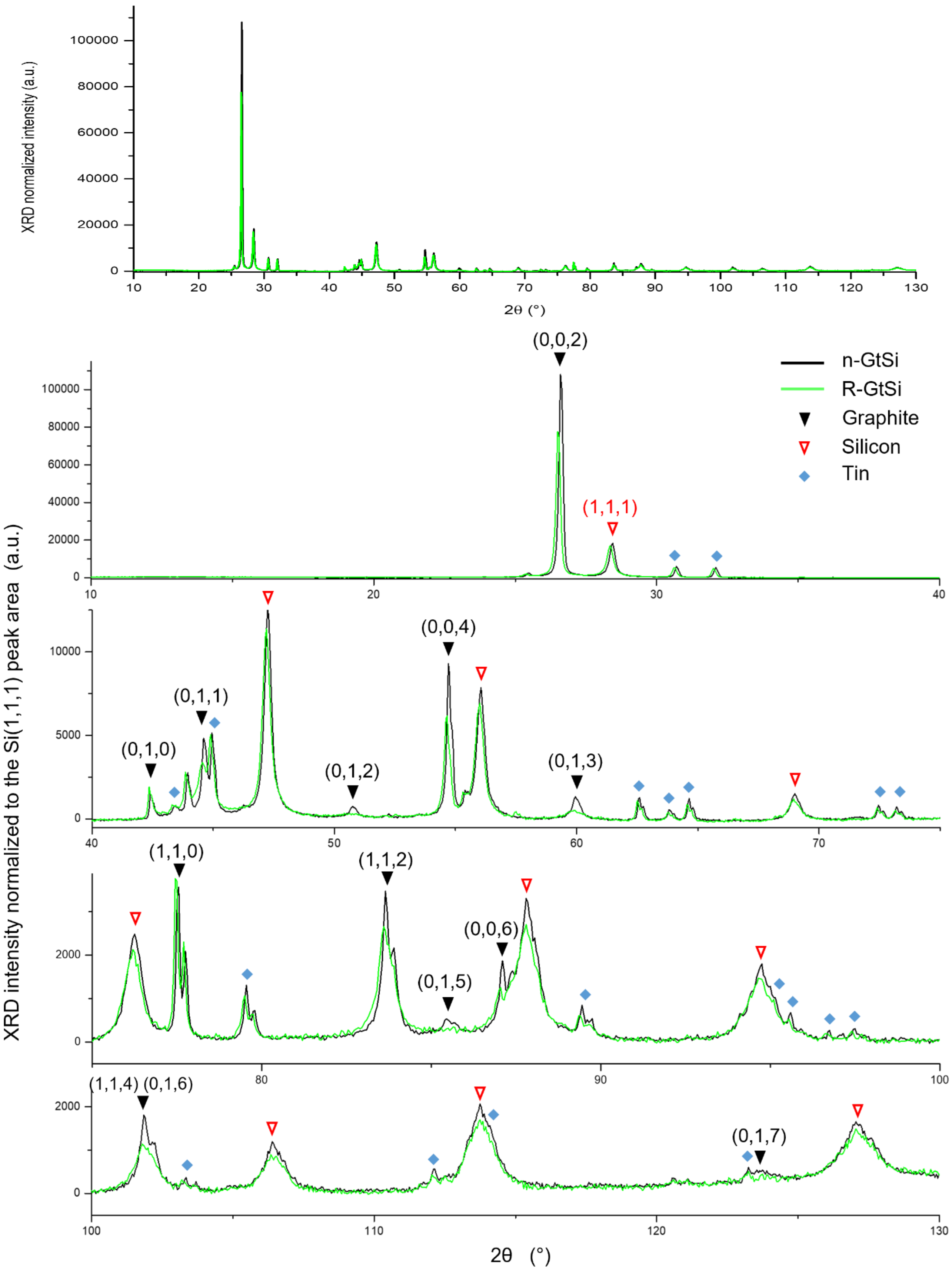


Figure S13: X-ray diffractogram of n-GtSi and R-GtSi composites. The intensity was normalized to the area of the Si (1,1,1) reflection. The Lebail refinement data are reported in Table S8 and Figures S16, S17 and S18. Reference ICDD files: Tin 00-004-0673 ; Silicon 00-027-1402 ; graphite 00-056-0159.

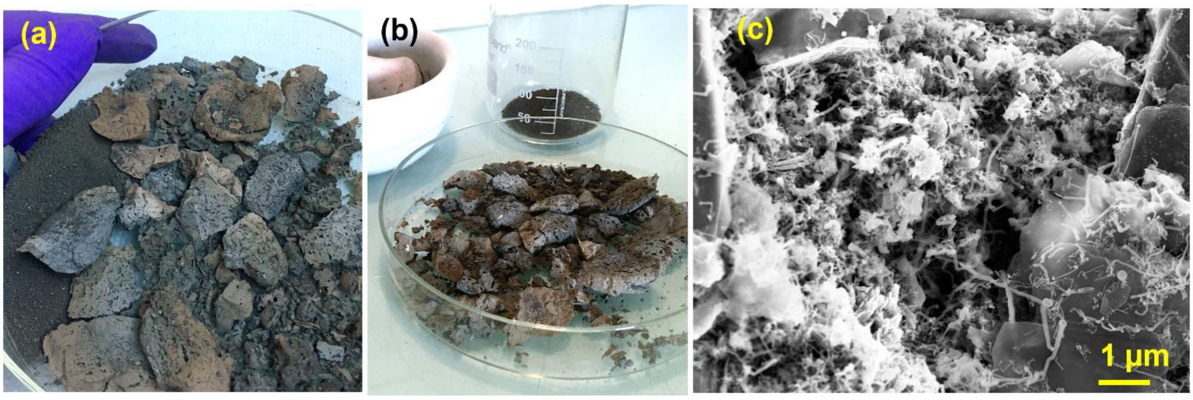


Figure S14. (a,b) Photographs of the GtSi raw product; (c) SEM image of GtSi composite showing intertwined SiNWs between neighboring graphite grains.

Table S7. Parameters for the calculation of the cell core energy density.

Mean value on 3 cells (cathode of 10.24 cm^2)	**Pouch cell Energy discharged at the 1st cycle (mWh)**	**Cathode thickness* (µm)**	**Anode thickness* (µm)**	**Separator thickness (µm)**	**cell electrochemical core thickness (µm)**	**Cell core energy density (Wh/L)**
n-GtSi	137.1	75	41	20	136	984
R-GtSi	128.5	75	39	20	134	936

* Considering one active layer and half of the current collector thickness (Al and Cu foils of 20 and 10µm).

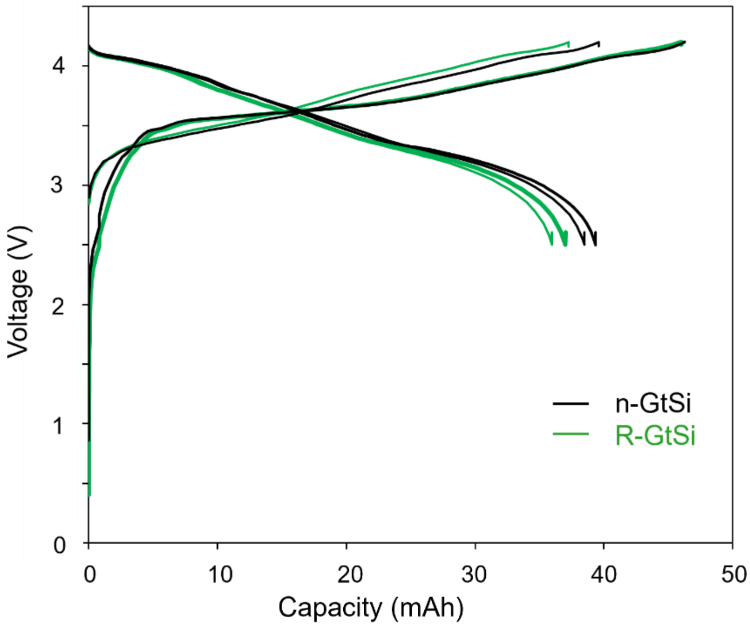


Figure S15. Voltage-capacity profiles of the 2 first cycles of SLP cells.

Table S8. Le Bail Refinement parameters for samples n-GtSi (W14) and R-GtSi (W16).

	W14	**W16**
Graphite	a=b=2,4619±0,0001 Å	a=b=2,4609±0,0001 Å
194	c=6,7125±0,0002 Å	c=6,7144±0,0004 Å
Hexagonal 6/mmm	α=β=90° et γ=120°	α=β=90° et γ=120°
Si	a=b=c=5,4466±0,0001 Å	a=b=c=5,4440±0,0002 Å
227	α=β=γ=90°	α=β=γ=90°
Cubic m-3m		
Sn	a=b=5,8326±0,0002 Å	a=b=5,8307±0,0003 Å
141	c=3,1821±0,0002 Å	c=3,1809±0,0002 Å
Tetragonal 4/mmm	α=β=γ=90°	α=β=γ=90°

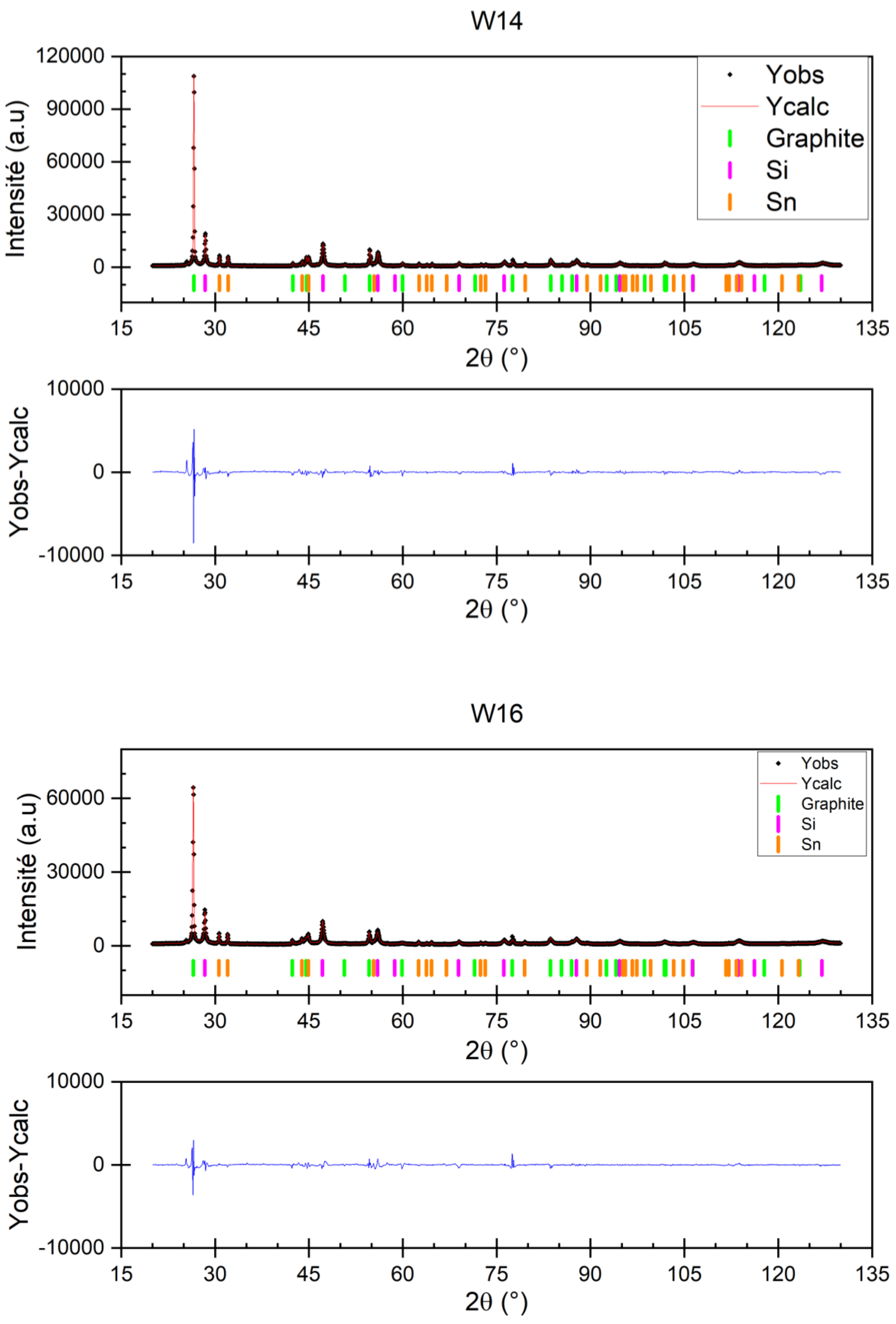
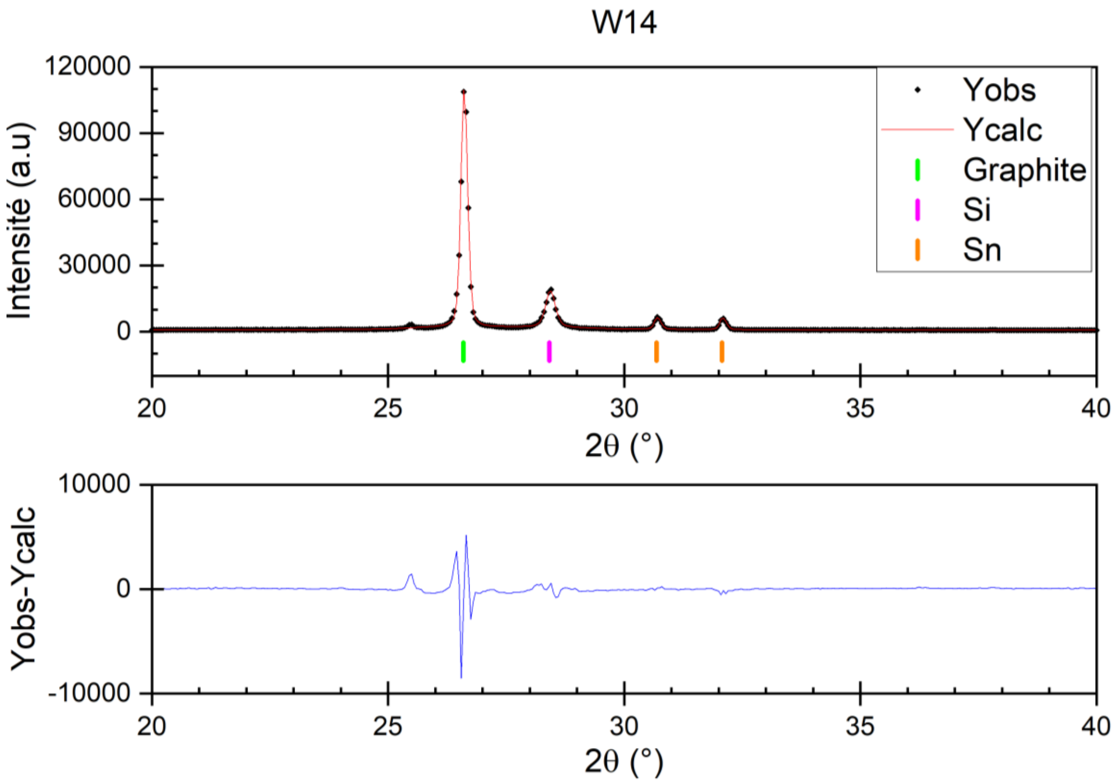
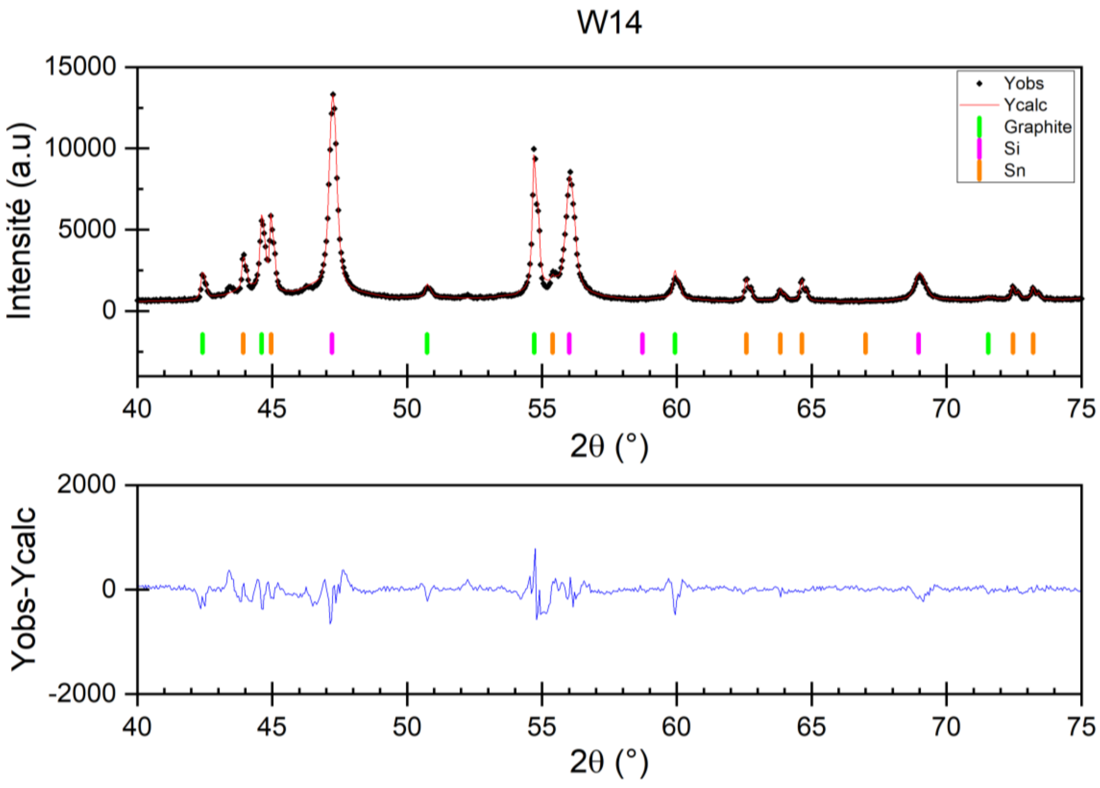


Figure S16. Le Bail refinement fitting for samples n-GtSi (W14) and R-GtSi (W16): global views.


W14
Intensité (a.u)
120000
90000
60000
30000
0
Yobs
Ycalc
Graphite
Si
Sn
20
25
30
35
40
2θ (°)
Yobs-Ycalc
10000
0
-10000
20
25
30
35
40
2θ (°)


W14
Intensité (a.u)
15000
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5000
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Yobs
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Graphite
Si
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40
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50
55
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70
75
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Yobs-Ycalc
2000
0
-2000
40
45
50
55
60
65
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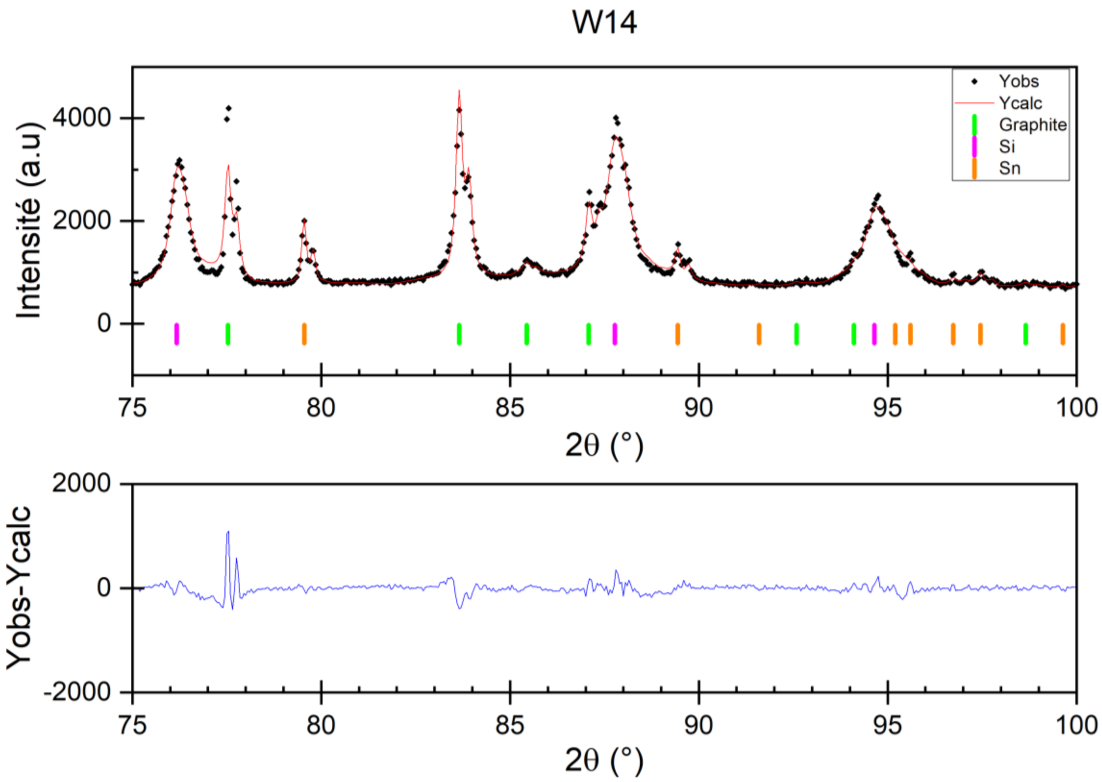
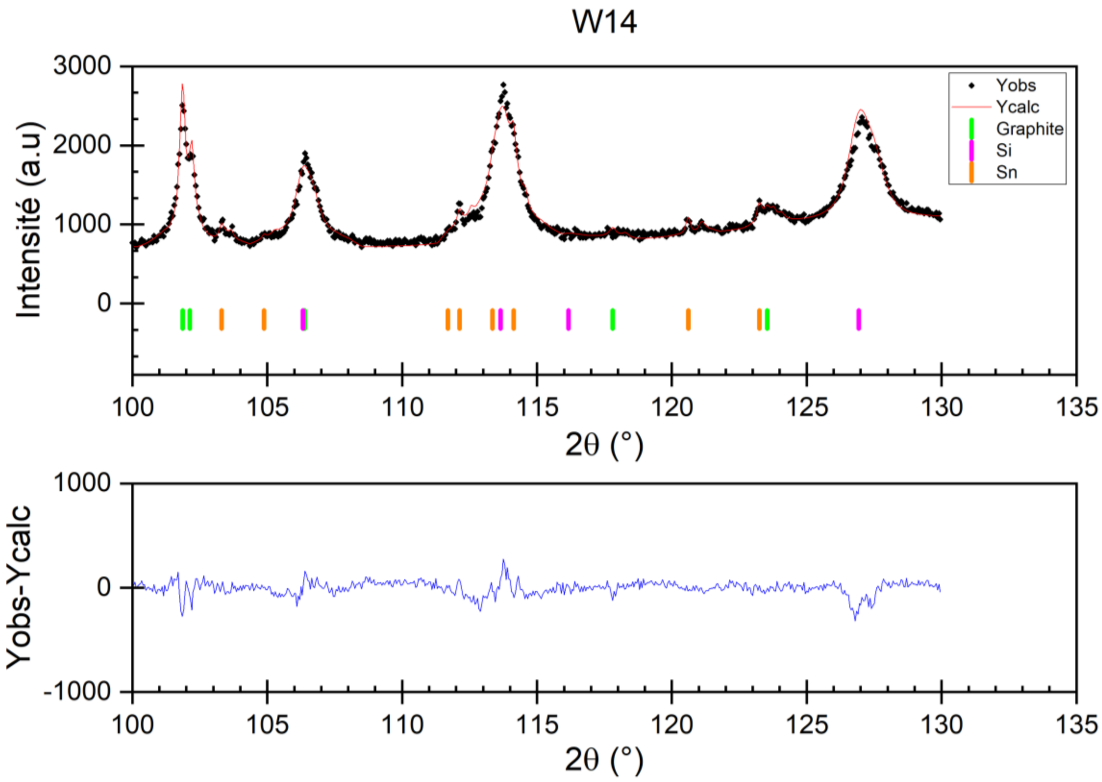
75
2θ (°)





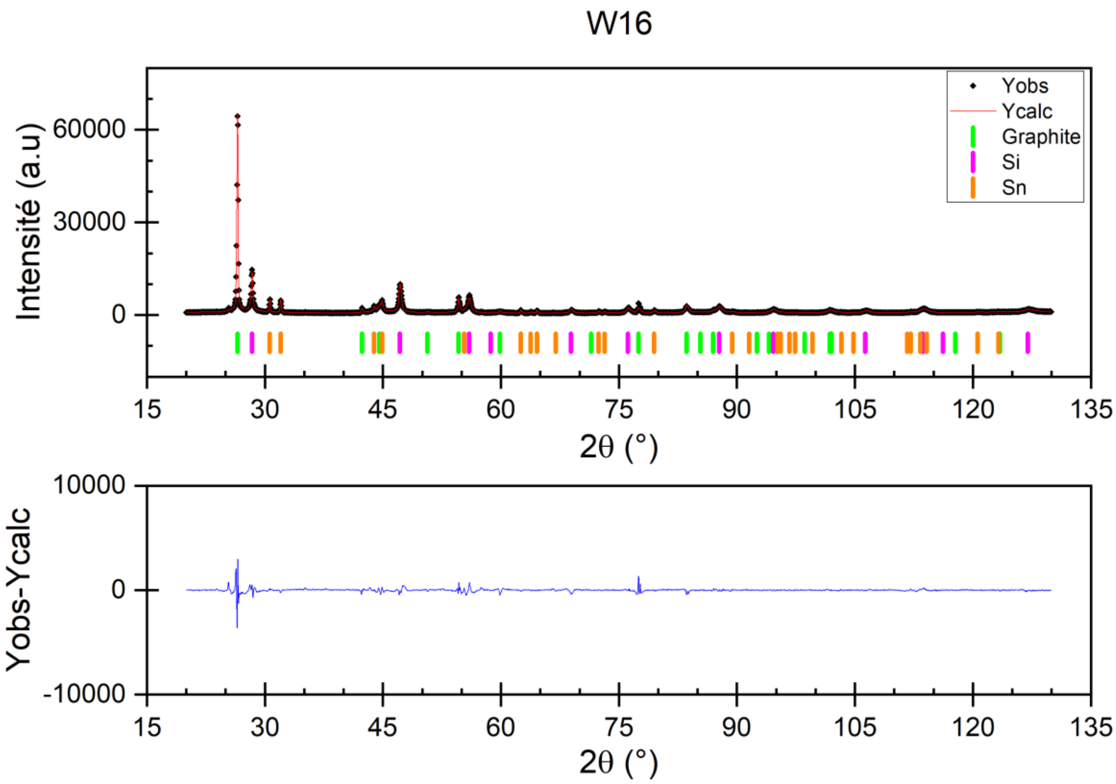
No.	2θ (°)	FWHM (°)	Area calc.	Assignment	h	k	l	Height [cts]	Shape Left
1	26,5915	0,1443	14879,05	Graphite	0	0	2	82664(512)	0,454
2	26,6586	0,1443	7439,53	Graphite	0	0	2	41320(278)	0,455

3	28,4139	0,2208	3689,17	Si	1	1	1	12198(281)	0,691
4	28,4859	0,221	1844,59	Si	1	1	1	6093(146)	0,692
5	30,6864	0,1271	757,63	Sn	0	2	0	4512(133)	0,603
6	30,7644	0,1271	378,81	Sn	0	2	0	2256(78)	0,603
7	32,0694	0,1273	783,99	Sn	0	1	1	4659(163)	0,605
8	32,1511	0,1273	391,99	Sn	0	1	1	2329(92)	0,605
9	42,4134	0,1501	283,75	Graphite	0	1	0	1440(123)	0,586
10	42,5237	0,1502	141,88	Graphite	0	1	0	719(70)	0,587
11	43,9241	0,1294	323,62	Sn	2	2	0	1879(104)	0,622
12	44,0388	0,1294	161,81	Sn	2	2	0	939(61)	0,622
13	44,6075	0,1514	805,59	Graphite	0	1	1	4024(134)	0,604
14	44,7242	0,1515	402,8	Graphite	0	1	1	2010(71)	0,605
15	44,952	0,1296	722,91	Sn	1	2	1	4187(115)	0,624
16	45,0696	0,1296	361,46	Sn	1	2	1	2093(81)	0,624
17	47,2136	0,2836	3589,05	Si	0	2	2	9138(303)	0,718
18	47,3379	0,2841	1794,52	Si	0	2	2	4561(164)	0,718
19	50,7391	0,1558	152,21	Graphite	0	1	2	724(84)	0,655
20	50,874	0,1559	76,11	Graphite	0	1	2	361(56)	0,656
21	54,7028	0,1592	1655,56	Graphite	0	0	4	7600(199)	0,688
22	54,8499	0,1594	827,78	Graphite	0	0	4	3795(112)	0,689
23	55,3857	0,1324	80,18	Sn	0	3	1	452(39)	0,639
24	55,535	0,1325	40,09	Sn	0	3	1	226(19)	0,639
25	56,0019	0,3198	2445,06	Si	1	1	3	5493(238)	0,73
26	56,1532	0,3204	1222,53	Si	1	1	3	2741(134)	0,73
27	58,7256	0,3319	13,12	Si	2	2	2	28(30)	0,734
28	58,8857	0,3326	6,56	Si	2	2	2	14(16)	0,734
29	59,9287	0,1645	375,78	Graphite	0	1	3	1640(120)	0,731
30	60,0927	0,1647	187,89	Graphite	0	1	3	818(79)	0,733
31	62,5758	0,135	253,21	Sn	1	1	2	1393(102)	0,649
32	62,7487	0,1351	126,61	Sn	1	1	2	696(68)	0,65
33	63,833	0,1355	128,25	Sn	0	4	0	702(77)	0,651
34	64,0102	0,1356	64,12	Sn	0	4	0	351(50)	0,652
35	64,6317	0,1358	237,27	Sn	2	3	1	1296(94)	0,652
36	64,8117	0,1359	118,64	Sn	2	3	1	648(68)	0,653
37	66,9973	0,1369	12,75	Sn	0	2	2	69(40)	0,656
38	67,1857	0,1369	6,37	Sn	0	2	2	34(26)	0,656
39	68,9583	0,3821	698,27	Si	0	0	4	1303(174)	0,748
40	69,1538	0,3831	349,13	Si	0	0	4	650(96)	0,748
41	71,5404	0,1798	43,25	Graphite	0	1	4	166(62)	0,828
42	71,7455	0,1801	21,63	Graphite	0	1	4	83(40)	0,83
43	72,4584	0,1395	143,4	Sn	2	4	0	759(83)	0,664
44	72,667	0,1396	71,7	Sn	2	4	0	379(56)	0,664
45	73,2111	0,1399	135,54	Sn	1	4	1	715(78)	0,665
46	73,4226	0,14	67,77	Sn	1	4	1	357(59)	0,665

47	76,1725	0,4224	1021,45	Si	1	3	3	1717(185)	0,758
48	76,3956	0,4237	510,73	Si	1	3	3	856(96)	0,758
49	77,5318	0,1898	575,83	Graphite	1	1	0	2043(130)	0,878
50	77,7604	0,1902	287,91	Graphite	1	1	0	1019(87)	0,88
51	79,5482	0,1437	232,88	Sn	1	3	2	1192(100)	0,674
52	79,7852	0,1438	116,44	Sn	1	3	2	595(74)	0,674
53	83,6541	0,2019	1058,48	Graphite	1	1	2	3451(190)	0,929
54	83,9089	0,2024	529,24	Graphite	1	1	2	1719(113)	0,931
55	85,4372	0,2058	67,42	Graphite	0	1	5	214(65)	0,944
56	85,7001	0,2064	33,71	Graphite	0	1	5	107(41)	0,946
57	87,0827	0,2095	362,53	Graphite	0	0	6	1124(95)	0,957
58	87,3534	0,2102	181,26	Graphite	0	0	6	560(41)	0,959
59	87,7675	0,4981	1463,42	Si	2	2	4	2071(191)	0,774
60	88,0414	0,5001	731,71	Si	2	2	4	1031(120)	0,774
61	89,4425	0,1511	49,12	Sn	3	4	1	238(35)	0,689
62	89,4425	0,1511	49,11	Sn	0	5	1	238(35)	0,689
63	89,7245	0,1514	24,56	Sn	0	5	1	119(27)	0,689
64	89,7246	0,1514	24,56	Sn	3	4	1	119(27)	0,689
65	91,5996	0,153	7,1	Sn	0	4	2	34(31)	0,692
66	91,8925	0,1533	3,55	Sn	0	4	2	17(19)	0,692
67	92,5895	0,2234	20,09	Graphite	0	2	0	57(44)	1,003
68	92,8875	0,2242	10,04	Graphite	0	2	0	28(25)	1,005
69	94,1011	0,2276	38,62	Graphite	0	2	1	107(27)	1,016
70	94,4071	0,2284	19,31	Graphite	0	2	1	53(7)	1,018
71	94,6491	0,5513	580,42	Si	1	1	5	739(134)	0,783
72	94,6491	0,5513	193,1	Si	3	3	3	246(44)	0,783
73	94,9581	0,5538	290,21	Si	1	1	5	368(79)	0,784
74	94,9581	0,5538	96,55	Si	3	3	3	122(26)	0,784
75	95,2011	0,1566	0	Sn	0	1	3	0	0,697
76	95,5131	0,1569	0	Sn	0	1	3	0	0,697
77	95,6055	0,157	16,99	Sn	3	3	2	79(24)	0,698
78	95,9197	0,1573	8,49	Sn	3	3	2	39(20)	0,698
79	96,7336	0,1582	21,38	Sn	4	4	0	98(47)	0,699
80	97,0541	0,1585	10,69	Sn	4	4	0	49(34)	0,7
81	97,458	0,159	39,65	Sn	2	5	1	181(63)	0,7
82	97,7826	0,1594	19,83	Sn	2	5	1	90(48)	0,701
83	98,6571	0,2412	27,68	Graphite	0	2	2	71(60)	1,053
84	98,9887	0,2423	13,84	Graphite	0	2	2	35(39)	1,056
85	99,6389	0,1615	14,61	Sn	2	4	2	66(43)	0,703
86	99,9763	0,1619	7,31	Sn	2	4	2	33(30)	0,704
87	101,859	0,2519	743,54	Graphite	1	1	4	1807(169)	1,08
88	102,1306	0,2529	50,84	Graphite	0	1	6	123(12)	1,082
89	102,2101	0,2532	371,77	Graphite	1	1	4	898(97)	1,083
90	102,4834	0,2541	25,42	Graphite	0	1	6	61(15)	1,085

91	103,3069	0,1661	30,76	Sn	1	2	3	134(49)	0,709
92	103,6672	0,1666	15,38	Sn	1	2	3	67(38)	0,709
93	104,8782	0,1683	11,2	Sn	0	6	0	48(28)	0,711
94	105,2488	0,1688	5,6	Sn	0	6	0	24(13)	0,712
95	106,322	0,661	683,66	Si	0	4	4	721(183)	0,8
96	106,3903	0,2688	2,4	Graphite	0	2	3	5(1)	1,118
97	106,7026	0,6651	341,83	Si	0	4	4	358(110)	0,8
98	106,7713	0,2703	1,2	Graphite	0	2	3	2,7(4)	1,121
99	111,6948	0,1791	2,77	Sn	0	3	3	11(9)	0,721
100	112,1152	0,1798	1,39	Sn	0	3	3	6(2)	0,722
101	112,1287	0,1799	40,17	Sn	1	5	2	161(50)	0,722
102	112,5525	0,1806	20,09	Sn	1	5	2	80(24)	0,722
103	113,3443	0,1821	23,96	Sn	2	6	0	95(8)	0,723
104	113,641	0,7471	1228,38	Si	1	3	5	1141(229)	0,81
105	113,7781	0,1829	11,98	Sn	2	6	0	47(3)	0,724
106	114,0773	0,7528	614,19	Si	1	3	5	566(123)	0,81
107	114,1294	0,1836	77,16	Sn	1	6	1	303(20)	0,725
108	114,5698	0,1845	38,58	Sn	1	6	1	151(21)	0,725
109	116,1671	0,781	39,29	Si	2	4	4	35(50)	0,813
110	116,6252	0,7874	19,64	Si	2	4	4	17(31)	0,814
111	117,8054	0,323	62,26	Graphite	0	2	4	110(82)	1,213
112	118,2785	0,3257	31,13	Graphite	0	2	4	55(59)	1,217
113	120,6062	0,1978	52,6	Sn	2	3	3	191(84)	0,734
114	121,1068	0,1991	26,3	Sn	2	3	3	95(64)	0,735
115	123,2479	0,2047	63,05	Sn	4	5	1	220(56)	0,738
116	123,5311	0,3588	134,42	Graphite	0	1	7	209(85)	1,26
117	123,7767	0,2062	31,52	Sn	4	5	1	109(29)	0,739
118	124,0631	0,3625	67,21	Graphite	0	1	7	103(58)	1,265
119	126,9352	0,9586	1615,52	Si	0	2	6	1160(276)	0,828
120	127,5076	0,9699	807,76	Si	0	2	6	573(168)	0,829

Figure S17. Le Bail refinement fitting for samples n-GtSi (W14): detailed views and parameters.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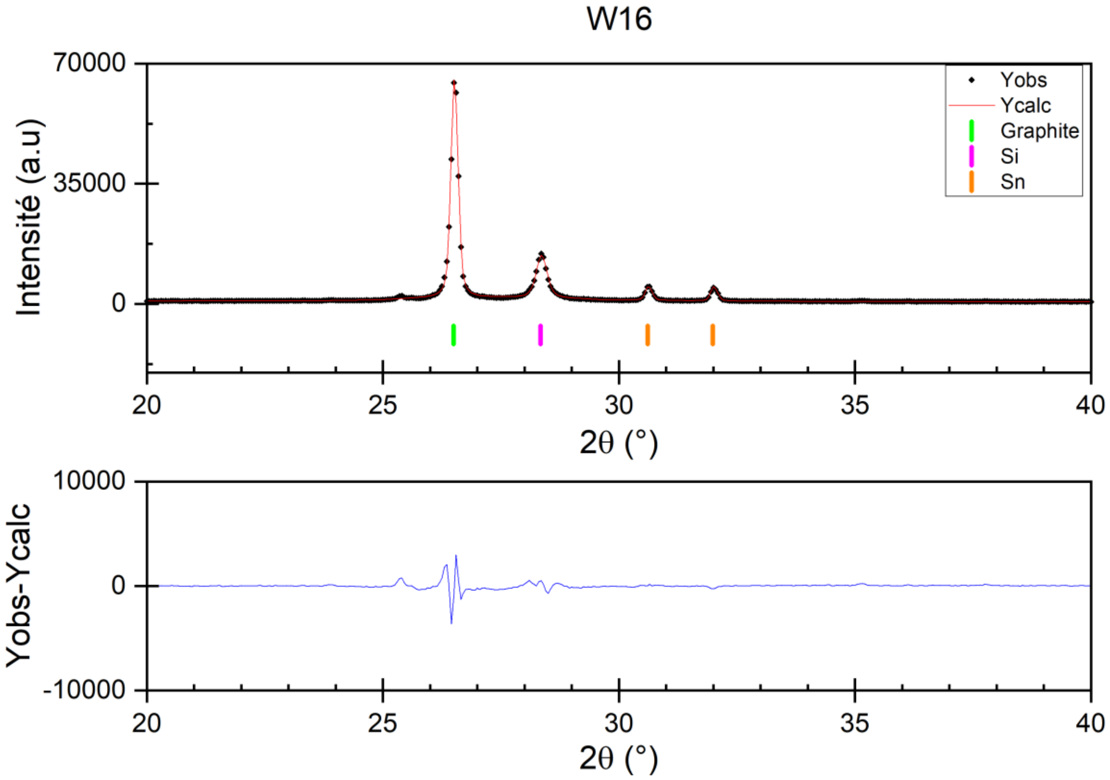
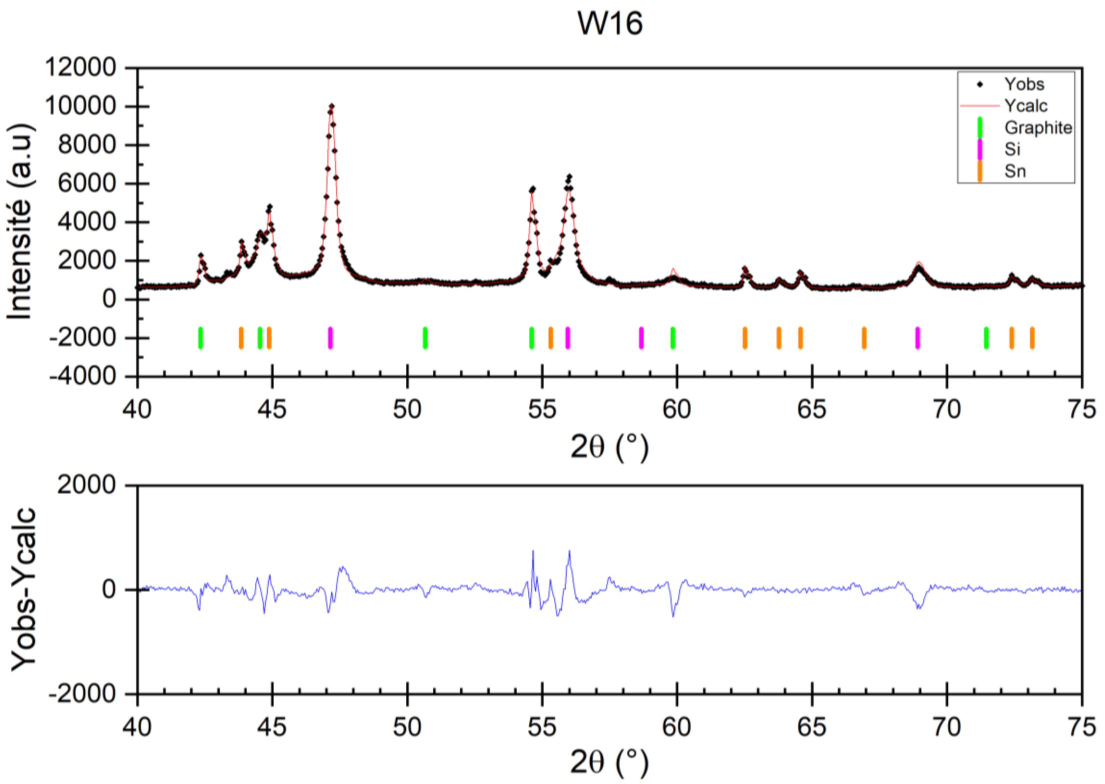
Intensité (a.u)
60000
30000
0
Yobs
Ycalc
Graphite
Si
Sn
15
30
45
60
75
90
105
120
135
2θ (°)
Yobs-Ycalc
10000
0
-10000
15
30
45
60
75
90
105
120
135
2θ (°)


W16
Intensité (a.u)
70000
35000
0
Yobs
Ycalc
Graphite
Si
Sn
20
25
30
35
40
2θ (°)
Yobs-Ycalc
10000
0
-10000
20
25
30
35
40
2θ (°)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Intensité (a.u)
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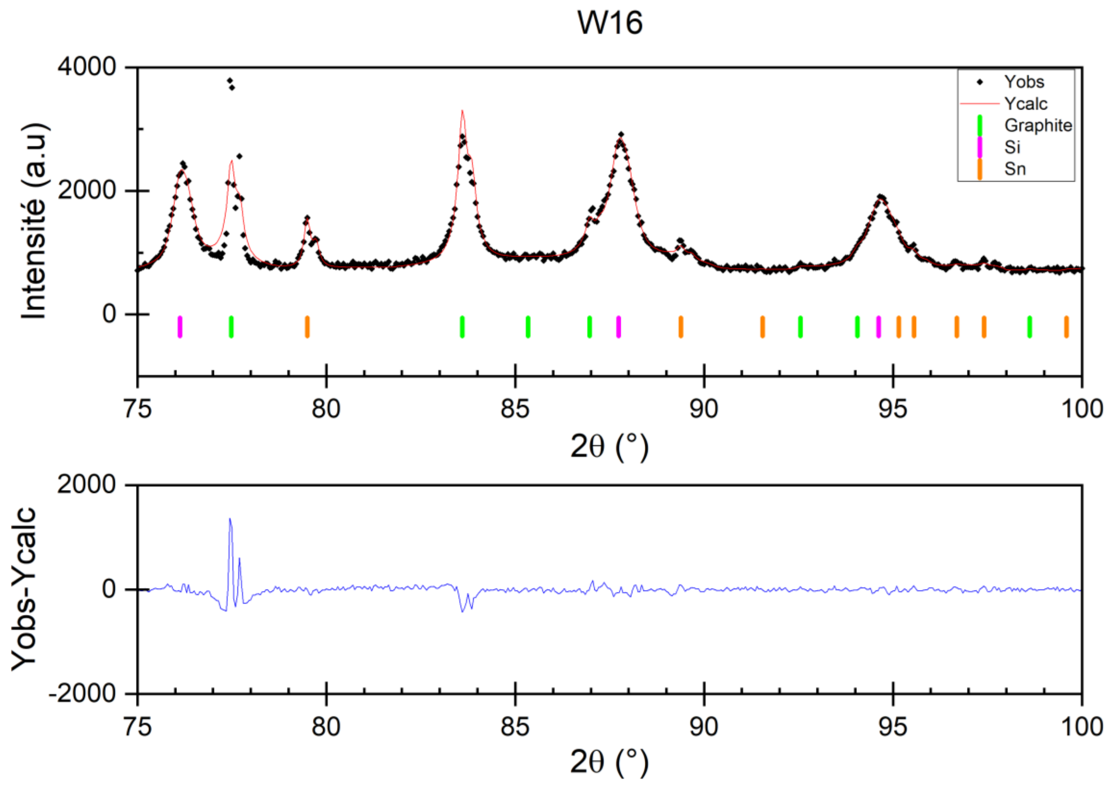
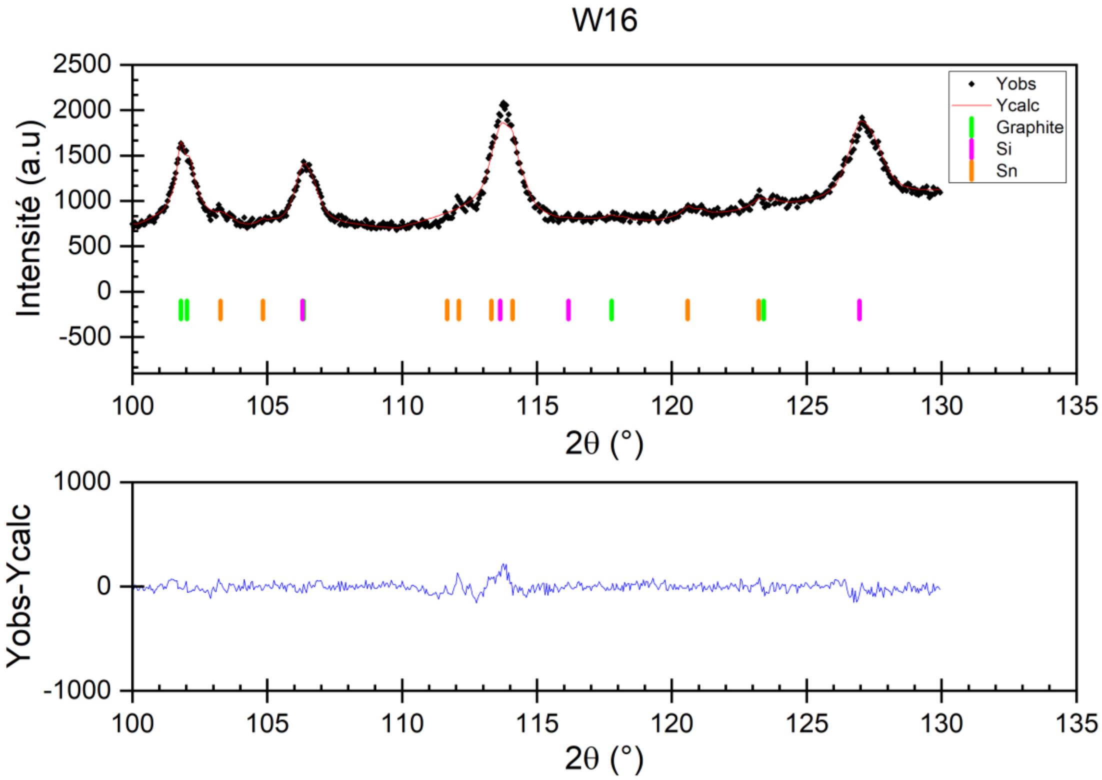
10000
8000
6000
4000
2000
0
-2000
-4000
Yobs
Ycalc
Graphite
Si
Sn
40
45
50
55
60
65
70
75
2θ (°)
Yobs-Ycalc
2000
0
-2000
40
45
50
55
60
65
70
75
2θ (°)


W16
Intensité (a.u)
4000
2000
0
Yobs
Ycalc
Graphite
Si
Sn
75
80
85
90
95
100
2θ (°)
Yobs-Ycalc
2000
0
-2000
75
80
85
90
95
100
2θ (°)



No.	2θ (°)	FWHM (°)	Area calc.	Assignment	h	k	l	Height [cts]	Shape Left
1	26,4948	0,1543	9710,83	Graphite	0	0	2	48518,58	0,557
2	26,5619	0,1542	4855,41	Graphite	0	0	2	24264,68	0,557
3	28,3384	0,2533	3031,12	Si	1	1	1	8879,45	0,652
4	28,4104	0,2536	1515,56	Si	1	1	1	4435,39	0,652
5	30,607	0,1353	587,1	Sn	0	2	0	3334,6	0,565
6	30,685	0,1352	293,55	Sn	0	2	0	1668,85	0,565
7	31,9918	0,1326	597,03	Sn	0	1	1	3446,28	0,575
8	32,0736	0,1325	298,51	Sn	0	1	1	1724,75	0,575
9	42,3422	0,1507	261,54	Graphite	0	1	0	1257,05	0,711
10	42,4526	0,1508	130,77	Graphite	0	1	0	627,89	0,712
11	43,8492	0,1172	215,93	Sn	2	2	0	1361,36	0,663
12	43,9639	0,1171	107,97	Sn	2	2	0	680,81	0,663
13	44,5344	0,1526	128,45	Graphite	0	1	1	604,02	0,732
14	44,6511	0,1528	64,23	Graphite	0	1	1	301,62	0,733
15	44,8785	0,1167	308,19	Sn	1	2	1	1945,29	0,67
16	44,9962	0,1166	154,1	Sn	1	2	1	972,69	0,671
17	47,148	0,3249	2959,58	Si	0	2	2	6662,91	0,687
18	47,2723	0,3254	1479,79	Si	0	2	2	3325,57	0,687
19	50,6613	0,1615	50,88	Graphite	0	1	2	220,41	0,792
20	50,7962	0,1618	25,44	Graphite	0	1	2	109,97	0,793
21	54,5973	0,1699	965,4	Graphite	0	0	4	3911,13	0,83
22	54,7444	0,1702	482,7	Graphite	0	0	4	1950,31	0,832

23	55,316	0,1214	6,34	Sn	0	3	1	37,28	0,748
24	55,4654	0,1216	3,17	Sn	0	3	1	18,6	0,749
25	55,9414	0,3658	1517,87	Si	1	1	3	3014,52	0,703
26	56,0928	0,3665	758,93	Si	1	1	3	1504	0,704
27	58,6668	0,3795	11,65	Si	2	2	2	22,24	0,708
28	58,8269	0,3803	5,82	Si	2	2	2	11,1	0,709
29	59,8449	0,1841	219,77	Graphite	0	1	3	803,21	0,881
30	60,0089	0,1846	109,89	Graphite	0	1	3	400,23	0,883
31	62,5115	0,1355	214,82	Sn	1	1	2	1105,08	0,801
32	62,6845	0,1359	107,41	Sn	1	1	2	550,42	0,802
33	63,766	0,1388	86,12	Sn	0	4	0	430,6	0,81
34	63,9433	0,1393	43,06	Sn	0	4	0	214,41	0,811
35	64,5658	0,1411	169,1	Sn	2	3	1	829,73	0,816
36	64,7459	0,1416	84,55	Sn	2	3	1	413,09	0,817
37	66,9348	0,1484	28,11	Sn	0	2	2	130,16	0,834
38	67,1233	0,149	14,06	Sn	0	2	2	64,77	0,835
39	68,9062	0,4361	623,37	Si	0	0	4	1027,88	0,728
40	69,1017	0,4373	311,69	Si	0	0	4	512,49	0,728
41	71,45	0,2275	27,71	Graphite	0	1	4	77,78	0,994
42	71,655	0,2284	13,85	Graphite	0	1	4	38,7	0,996
43	72,3953	0,1684	122,62	Sn	2	4	0	491,41	0,874
44	72,6039	0,1692	61,31	Sn	2	4	0	244,3	0,876
45	73,1491	0,1715	98,52	Sn	1	4	1	386,7	0,88
46	73,3607	0,1724	49,26	Sn	1	4	1	192,22	0,881
47	76,1256	0,4816	788,74	Si	1	3	3	1171,14	0,741
48	76,3488	0,483	394,37	Si	1	3	3	583,66	0,742
49	77,4799	0,2564	587,99	Graphite	1	1	0	1423,28	1,053
50	77,7087	0,2576	293,99	Graphite	1	1	0	707,59	1,055
51	79,4916	0,2006	231,45	Sn	1	3	2	760,15	0,927
52	79,7288	0,2018	115,73	Sn	1	3	2	377,52	0,928
53	83,5981	0,2905	1056,37	Graphite	1	1	2	2191,66	1,112
54	83,8531	0,292	528,18	Graphite	1	1	2	1088,74	1,115
55	85,3393	0,3011	3,91	Graphite	0	1	5	7,77	1,129
56	85,6022	0,3028	1,96	Graphite	0	1	5	3,86	1,132
57	86,9636	0,3114	240,5	Graphite	0	0	6	457,83	1,145
58	87,2341	0,3132	120,25	Graphite	0	0	6	227,33	1,148
59	87,7302	0,5669	1199,33	Si	2	2	4	1498,85	0,763
60	88,0043	0,5691	599,66	Si	2	2	4	746,32	0,763
61	89,3893	0,2572	35,04	Sn	3	4	1	86,73	1
62	89,3893	0,2572	35,04	Sn	0	5	1	86,72	1
63	89,6715	0,259	17,52	Sn	3	4	1	43,02	1,002
64	89,6715	0,259	17,52	Sn	0	5	1	43,01	1,002
65	91,5499	0,2715	5,03	Sn	0	4	2	11,71	1,016
66	91,843	0,2735	2,52	Sn	0	4	2	5,81	1,018

67	92,5488	0,3498	25,11	Graphite	0	2	0	41,38	1,199
68	92,8471	0,352	12,56	Graphite	0	2	0	20,53	1,202
69	94,0596	0,3611	103,14	Graphite	0	2	1	163,38	1,214
70	94,3659	0,3634	51,57	Graphite	0	2	1	81,04	1,217
71	94,6184	0,6267	372,42	Si	3	3	3	418,66	0,776
72	94,6184	0,6267	372,49	Si	1	1	5	418,74	0,776
73	94,9277	0,6296	186,21	Si	3	3	3	208,32	0,776
74	94,9277	0,6296	186,25	Si	1	1	5	208,36	0,776
75	95,1575	0,297	55,35	Sn	0	1	3	116,24	1,043
76	95,4697	0,2993	27,67	Sn	0	1	3	57,61	1,045
77	95,5585	0,3	55,87	Sn	3	3	2	116,02	1,046
78	95,873	0,3023	27,94	Sn	3	3	2	57,49	1,048
79	96,6844	0,3085	19,99	Sn	4	4	0	40,19	1,054
80	97,0052	0,3109	9,99	Sn	4	4	0	19,91	1,056
81	97,4101	0,3141	34,46	Sn	2	5	1	67,88	1,059
82	97,735	0,3166	17,23	Sn	2	5	1	33,63	1,062
83	98,6135	0,3975	22,58	Graphite	0	2	2	31,73	1,258
84	98,9454	0,4003	11,29	Graphite	0	2	2	15,73	1,262
85	99,5948	0,3315	10,06	Sn	2	4	2	18,62	1,076
86	99,9325	0,3343	5,03	Sn	2	4	2	9,22	1,078
87	101,7934	0,4253	528,68	Graphite	1	1	4	682,62	1,289
88	102,0233	0,4274	95,53	Graphite	0	1	6	122,59	1,291
89	102,1446	0,4285	264,34	Graphite	1	1	4	338,11	1,293
90	102,3759	0,4306	47,77	Graphite	0	1	6	60,72	1,295
91	103,2695	0,363	14,58	Sn	1	2	3	24,33	1,103
92	103,6301	0,3663	7,29	Sn	1	2	3	12,04	1,106
93	104,8352	0,3773	31,09	Sn	0	6	0	49,6	1,114
94	105,2062	0,3808	15,54	Sn	0	6	0	24,54	1,117
95	106,3049	0,7503	568,03	Si	0	4	4	528,38	0,798
96	106,3438	0,4691	10,71	Graphite	0	2	3	12,23	1,333
97	106,6859	0,7549	284,01	Si	0	4	4	262,5	0,798
98	106,7251	0,4731	5,36	Graphite	0	2	3	6,05	1,337
99	111,6651	0,447	0	Sn	0	3	3	0	1,165
100	112,086	0,4517	0	Sn	0	3	3	0	1,168
101	112,0955	0,4518	0	Sn	1	5	2	0	1,168
102	112,5198	0,4567	0	Sn	1	5	2	0	1,171
103	113,3092	0,4658	71,28	Sn	2	6	0	89,22	1,177
104	113,6343	0,8474	812,2	Si	1	3	5	664,96	0,811
105	113,7435	0,4709	35,64	Sn	2	6	0	44,05	1,18
106	114,0713	0,8538	406,1	Si	1	3	5	329,87	0,812
107	114,0959	0,4751	182,62	Sn	1	6	1	223,43	1,183
108	114,5368	0,4804	91,31	Sn	1	6	1	110,29	1,186
109	116,1646	0,8856	9,08	Si	2	4	4	7,1	0,816
110	116,6234	0,8928	4,54	Si	2	4	4	3,52	0,817

111	117,7559	0,6056	64,76	Graphite	0	2	4	53,7	1,444
112	118,2295	0,6123	32,38	Graphite	0	2	4	26,48	1,449
113	120,587	0,5606	111,15	Sn	2	3	3	112,35	1,231
114	121,0883	0,568	55,57	Sn	2	3	3	55,34	1,235
115	123,2255	0,6006	90,14	Sn	4	5	1	84,16	1,251
116	123,4077	0,6923	37,48	Graphite	0	1	7	26,28	1,499
117	123,755	0,6091	45,07	Sn	4	5	1	41,41	1,255
118	123,9392	0,7013	18,74	Graphite	0	1	7	12,93	1,505
119	126,9541	1,0861	1096,81	Si	0	2	6	692,98	0,836
120	127,5279	1,0989	548,4	Si	0	2	6	342,29	0,837

Figure S18. Le Bail refinement fitting for samples R-GtSi (W16): detailed views and parameters.