Electronic supplementary information (ESI†):

Foamed lignin-silicone bio-composites by extrusion then compression molding

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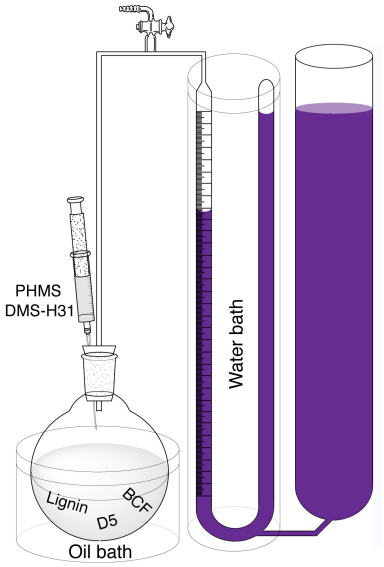


Figure S1. A gas volume meter was used to measure volume of gas produced during the reaction in solvent during preliminary optimizations and hydrolysis studies.

Table S1. Preliminarily optimizations for extrusion and molding conditions of different formulations.

No.	Lignin		PHMS		H-PDMS-H		D_5	BCF	Reaction temperature
	Mass (g)	wt% [a]	Mass (g)	wt% [b]	Type	Mass (g)	(ml)	(ppm) [a]	(°C)
O-1	0.25	50	0.02	8	H31	0.23	5	1000	30
O-2	0.25	50	0.02	8	H31	0.23	5	1000	40
O-3	0.25	50	0.02	8	H31	0.23	5	1000	50
O-4	0.25	50	0.02	8	H31	0.23	5	1000	90
O-5	0.25	51	0.01	4	H31	0.23	5	1000	50
O-6	0.25	48	0.04	16	H31	0.23	5	1000	50
O-7	0.25	50	0.02	8	H31	0.23	5	2000	30
O-8	0.25	50	0.02	8	H31	0.23	5	4000	30

[a] Weight ratios of lignin and catalyst loading are calculated without considering the solvent (D_5). [b] Weight ratios of PHMS are calculated against lignin weight only.

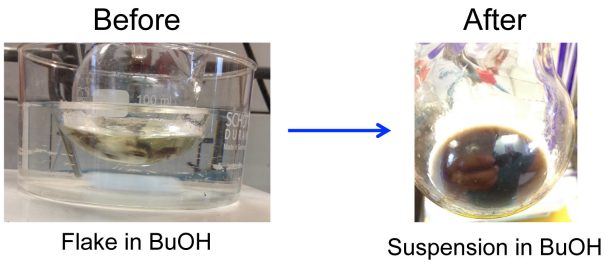


Figure S1. Testing the conversion of "Si-H" in lignin-silicone foam. Before reaction, the sliced lignin-silicone foam layers were dispersed in KOH/BuOH; the foam lost its integrity (fell apart to give a suspension) after treatment.

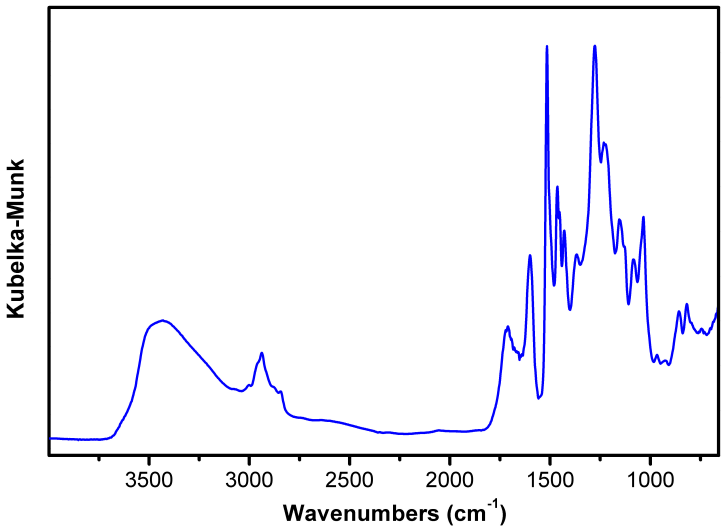


Figure S3. FT-IR for softwood kraft lignin obtained from Weyerhaeuser.

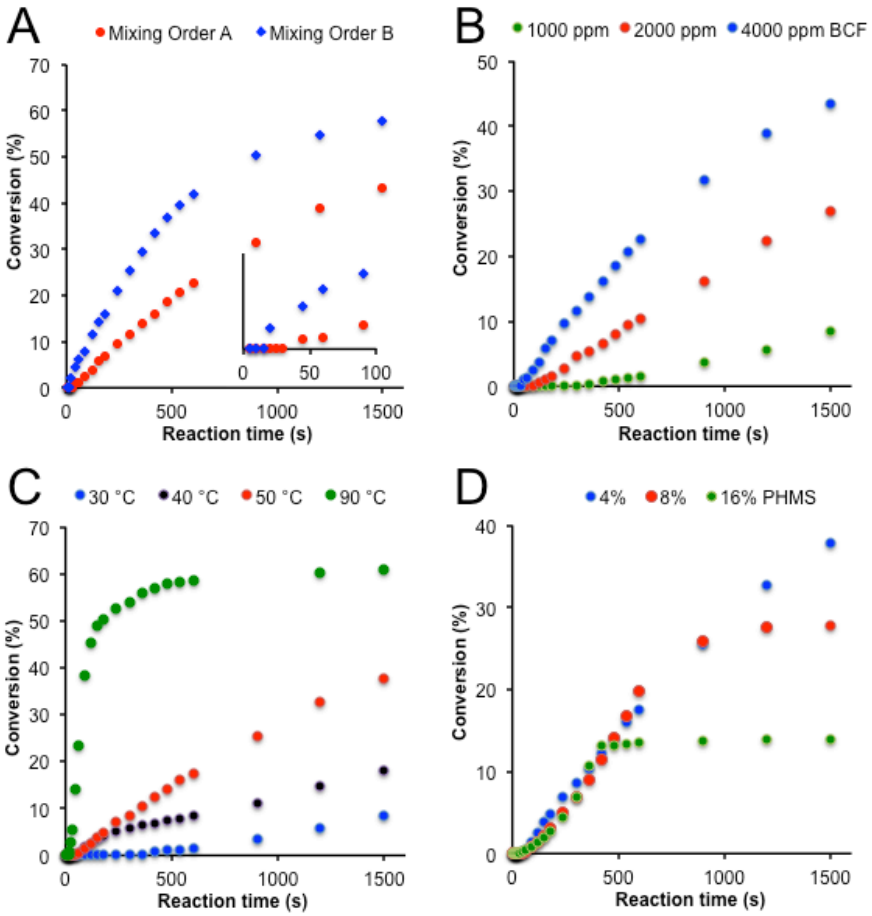


Figure S4. Kinetics studies for PHMS conversion % under different conditions and formulations: (A) mixing order: mixing order A led to longer induction times. The inset graph is the expanded range showing different induction times; (B) catalyst

loading: conversion% increased, while induction time decreased, with increased catalyst loading; (C) temperature: higher temperature led to faster reaction rates and greater conversion% of PHMS. (D) PHMS content had no impact on induction time.

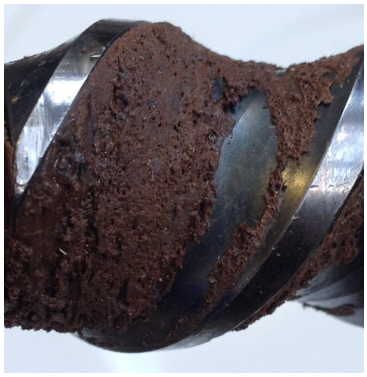


Figure S5. Gelation or crosslinking of foam precursors blocked the extruder channels.

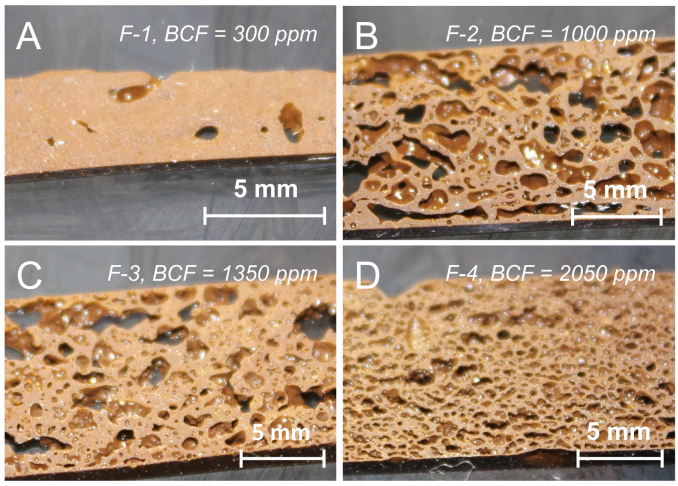


Figure S6. Cross-sectional images of lignin-silicone foams prepared from formulations with different catalyst loading: (A) F-1, BCF = 300 ppm, (B) F-2, BCF = 1000 ppm, (C) F-3, BCF = 1350 ppm, and (D) F-4, BCF = 2050 ppm.

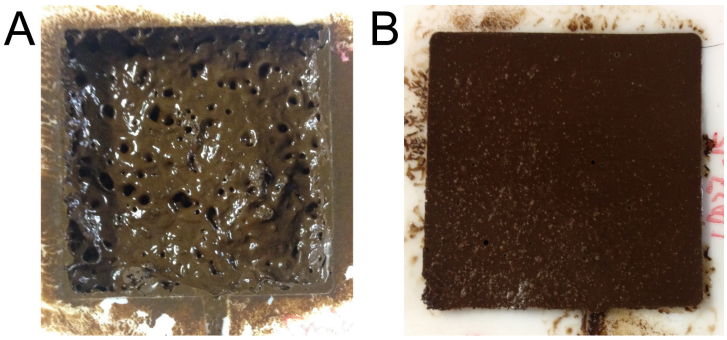


Figure S7. Images of lignin-silicone composite foams made with different catalyst content. (A) BCF = 300 ppm, curing for 180 min; (B) BCF = 1000 ppm, curing time = 5 min.

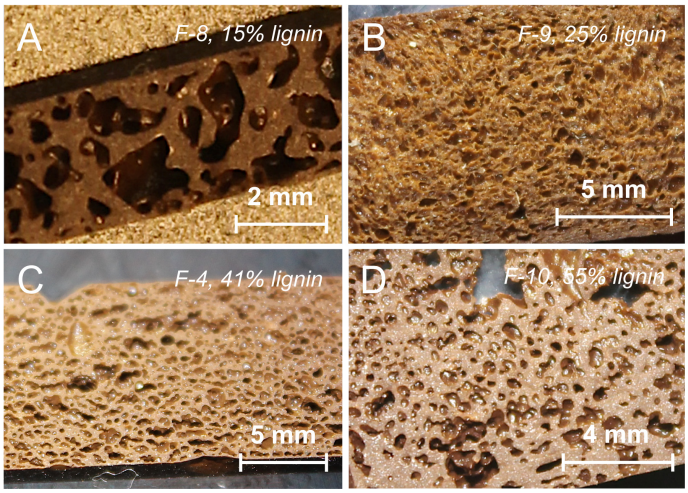


Figure S8. Cross-sectional images of lignin-silicone foam sprepared from formulations with varying lignin content: (A) F-8, 15 % of lignin, (B) F-9, 25% lignin, (C) F-4, 41% lignin, and (D) F-10, 55% lignin.

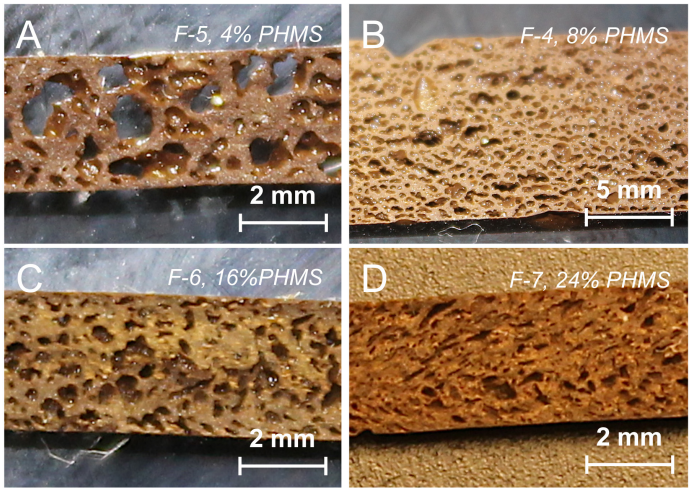


Figure S9. Cross-sectional images of lignin-silicone foams prepared from formulations with varying PHMS content: (A) F-5, 4 % of PHMS, (B) F-4, 8 % of PHMS, (C) F-6, 16 % of PHMS, and (D) F-7, 24 % of PHMS.

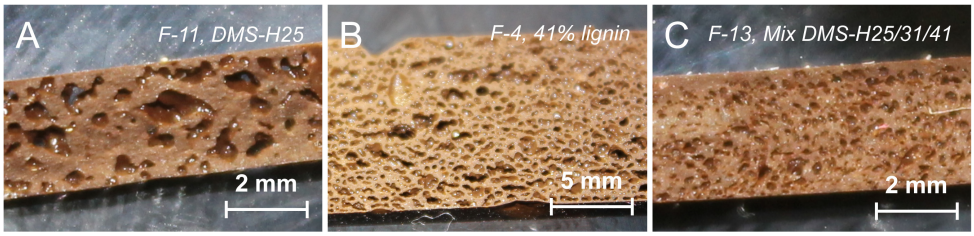


Figure S10. Cross-sectional images of lignin-silicone foams prepared from formulations with different molecular weight H-PDMS-H: (A) F-11, DMS-H25 (Mw = 6000 g/mol), (B) F-4, DMS-H31 (Mw = 28000 g/mol), and (C) F-13, Mixture of DMS-H25/31/41 (containing 26% H25, 48% H31, and 26% H41, H-41: Mw = 62700 g/mol).

Figure S11. Attempts to make lignin-silicone foams using DMS-H41 (F-12) failed due to the high intrinsic viscosity of DMS-H41. The precursor is semi-cured, and could not be processed in extruder.

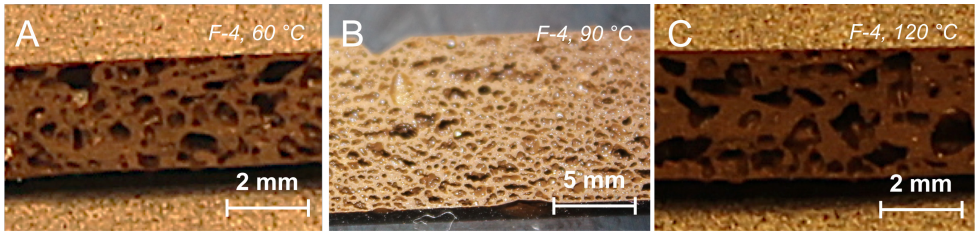


Figure S12. Cross-sectional images of lignin-silicone foams (formulation F-4) molded under different temperatures: (A) 60, (B) 90, and (C) 120°C.

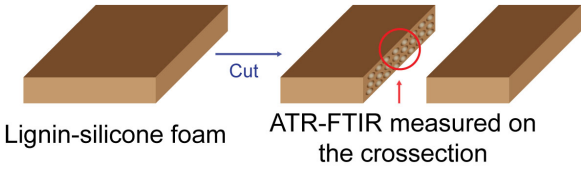


Figure S13. Preparation samples for ATR-IR characterization: how signal changes of "Si-H" groups in the foam were tracked.

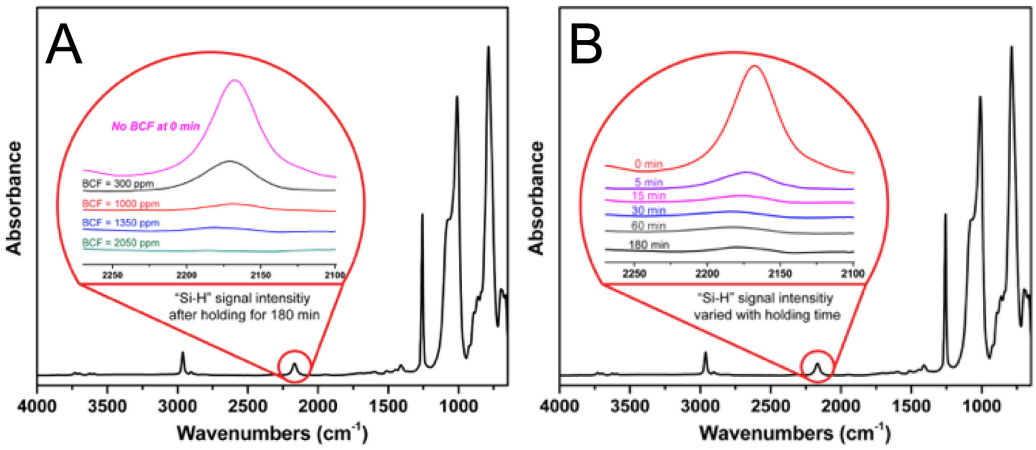
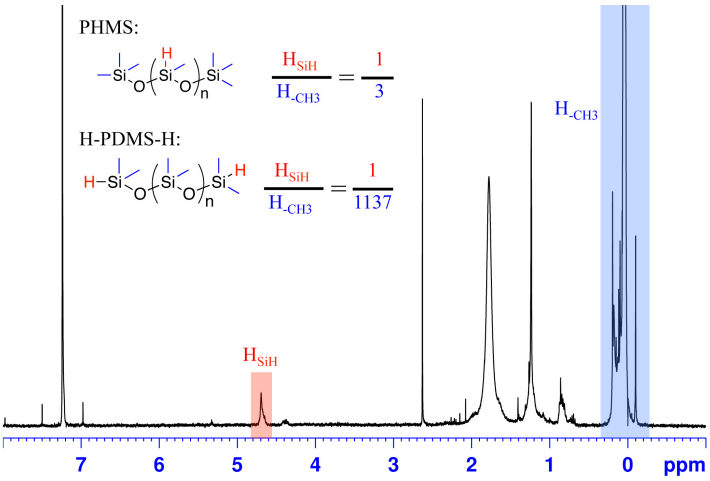


Figure S14. ATR-FTIR spectra for lignin-silicone foams made under different conditions and formulations: (A) different catalyst loading (from 300 to 2050 ppm), cured at 90 °C for 180 min, (B) foams in mold with different holding times (from 5 to 180 min), cured with 1350 ppm catalyst at 90 °C. The expanded spectra ranged from 2100 to 2250 cm^{-1} to track the intensity change of the "Si-H" signal.



BCF content (ppm)	1000			1340			2050
Curing time (min)	5	5	15	30	60	180	5
Weight loss (%)	19	12	10	9	9	8	11
$\frac{H_{SiH}}{H_{-CH3}}$	1/50	1/67		~ 0			1/142

Figure S15. NMR spectrum of the cyclohexane extract from lignin-silicone foam, and the proton ratios ("Si-H"/"Si-CH_3") of samples obtained with different conditions and formulations.

Table S1. The degree of conversion of "Si-H" in lignin-silicone foams, tested by hydrolysis in solution

Formulation	Lignin content (%)	PHMS content (%)	H-PDMS-H	Conversion of Si-H (%)[a]
F-4 B	4	8	DMS-H31	43
F-4 B, 140 °C for 12 h	4	8	DMS-H31	44
F-4 B, 220 °C for 5 h	4	8	DMS-H31	69
F-4 B, 220 °C for 12 h	4	8	DMS-H31	77
F-4 B, 300 °C for 12 h	4	8	DMS-H31	79

[a] The residual "Si-H" = 100% - Conversation of Si-H (%)

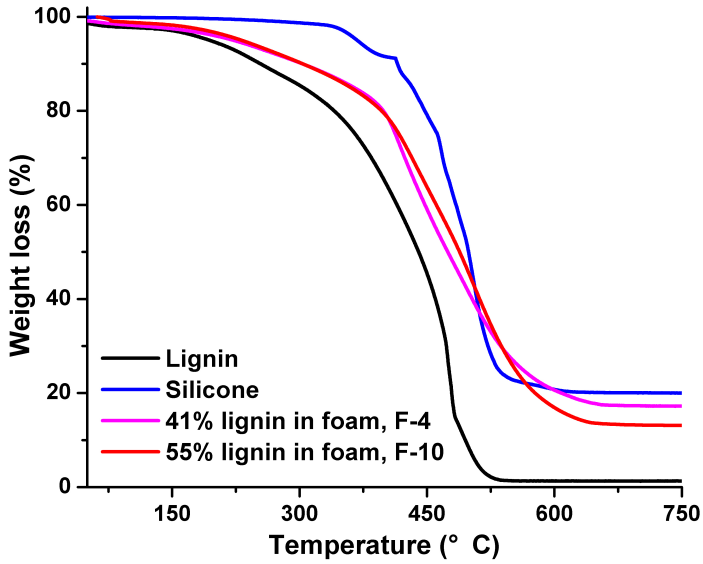


Figure S16. TGA of lignin-silicone foams (purple: 41% lignin in foam, red: 55% lignin in foam), lignin (black), and silicone (blue) under an air atmosphere.

Table S2. The mechanical properties of lignin-silicone foams characterized using DMA

		Formulation								
		F-4 B	F-5	F-6	F-7	F-8	F-9	F-10	F-11	F-13
T_g (°C)		-116	-110	-110	-111	-115	-109	-116	-111	-113
T_m (°C)		-37	-37	-37	-37	-38	-42	-36	-37	-31
Storage Modulus (E', MPa)	@ T_g - 20 °C	99	51	82	82	95	48	51	71	91
	@ T_g + 20 °C	87	43	58	85	86	46	46	59	81
	@ T_g + 50 °C	0.54	3.5	15	17	0.86	0.29	3	1.3	1.7
Loss Modulus (E", MPa)	@ T_g	7.6	2.1	3.5	7.4	4.7	11	2.5	4	3.5
	@ T_g + 50 °C	0.13	0.58	2.4	2.2	0.16	0.02	0.63	0.2	0.29
Tan Delta	@ T_g	0.09	0.05	0.05	0.09	0.06	0.55	0.05	0.07	0.04
	@ T_m	0.18	0.19	0.13	0.11	0.21	0.14	0.22	0.16	0.2