

SUPPLEMENTARY INFORMATION

Interactions of Hydrophilic Birch Wood (*Betula pendula* ROTH) Extractives with Adhesives for Load-bearing Timber Structures

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Authors: Max ENGELHARDT^{1,*}
orcid.org/0000-0002-9781-7476 (engelhardt@hfm.tum.de)

Thomas BÖGER¹
orcid.org/0000-0001-8747-8506 (boeger@hfm.tum.de)

Michael GIGL²
orcid.org/0000-0002-5617-9236 (michael.gigl@tum.de)

Chen MENG²
orcid.org/0000-0002-5968-6719 (chen.meng@tum.de)

Viktor SOPRUNYUK³
orcid.org/0000-0001-7073-6900 (viktor.soprnyuk@univie.ac.at)

Wilfried SCHRANZ³
orcid.org/0000-0002-9842-3532 (wilfried.schranz@univie.ac.at)

Klaus RICHTER¹
orcid.org/0000-0002-6583-0254 (richter@hfm.tum.de)

Antoni SÁNCHEZ-FERRER^{1,*}
orcid.org/0000-0002-1041-0324 (sanchez@hfm.tum.de)

¹ Chair of Wood Science, TUM School of Life Sciences, Technical University of Munich

² Bavarian Center for Biomolecular Mass Spectrometry, TUM School of Life Sciences, Technical University Munich

³ Faculty of Physics, University of Vienna

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Experimental Section – Tensile Shear Strength Experiments

Table SI–1 lists the relevant bonding processing parameters (conducted at 20 °C/65%-RH).

Table SI–1: Bonding parameters for the applied adhesive systems

adhesive type	PUR	MUF
quantity [g/m ²]	175 – 180	375 – 385
open waiting time [min]	1 – 2	2 – 4
closed waiting time [min]	20 ± 4	40 ± 5
pressure [MPa]	0.8	1.2
pressing time [h]	1.5	10
application	toothed spatula, single-sided	toothed spatula, both-sided

Table SI–2 lists the treatments applied to the specimens after bonding.

Table SI–2: Applied treatment procedures of tensile shear specimens as defined in EN 302–1

designation	treatment description	measurement conditions
A1	equilibrate at standard climate (20 °C/65%-RH)	standard climate (20 °C/65%-RH)
A2	A1 + 4 days soaking in cold water at 20 °C	wet state
A4	A1 + 6 h soaking in boiling water, 2 h soaking in cold water (20 °C)	wet state
A5	A1 + 6 h soaking in boiling water, 2 h soaking in cold water (20 °C)	recondition in standard climate

The tensile shear specimen geometry and loading schematic and the cutting pattern of specimens prepared from the glued boards are shown in Figure SI–1.

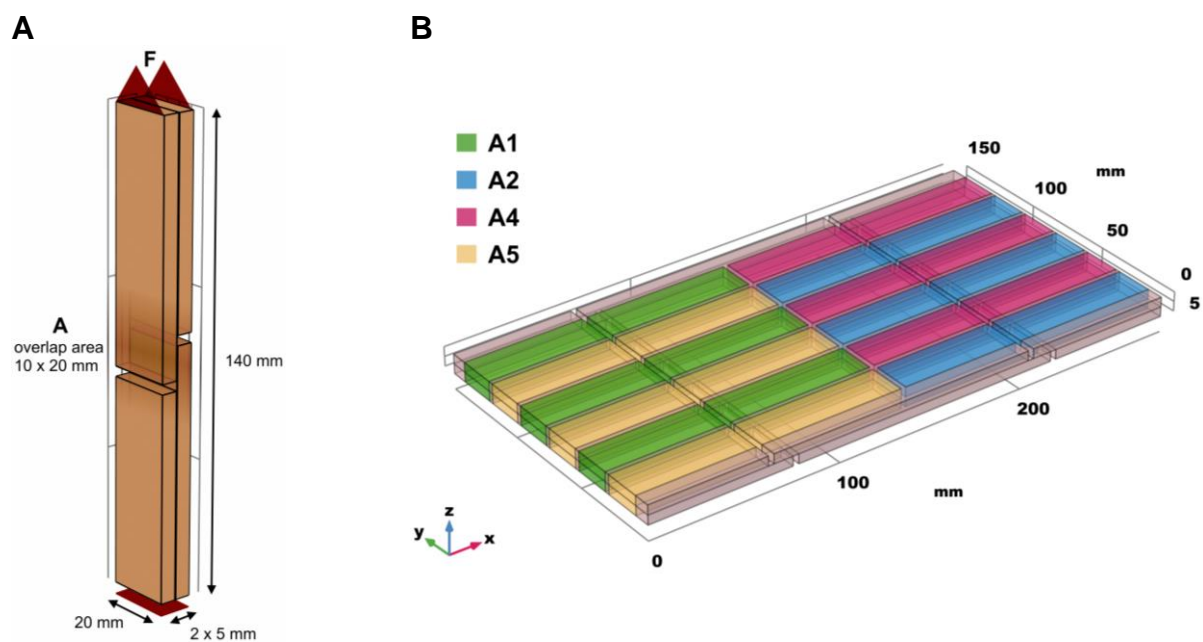


Figure SI–1: A) Tensile shear strength specimen acc. to EN 302–1. B) Cutting pattern of tensile shear specimens from glued boards. Color indication of treatment type (designation code acc. to EN 302–1).

Experimental Section – Rheological Experiments of Isothermal Curing

The determination of gelation *via* multi-frequency time-sweep in a small-angle-oscillatory-shear (SAOS) experiment is visualized in Figure SI–2 below. Figure SI–2A shows the progression of the loss factor $\tan \delta$ for different oscillation frequencies f . In Figure SI–2B, for selected times / frequency sweep cycles, the loss factors $\tan \delta$ (detrended) are plotted as a function of f . While regression analysis shows positive slopes prior to gelation and negative slopes after gelation ($\tan \delta \propto f^n$), the gel point is identified as the time when the slope is zero ($n = 0$) by calculating the variance of $\tan \delta$ and finding the local minimum (Figure SI–2C).

The gel point can be identified in (A) by the temporary convergence of the loss factors, in (B) by constant loss factor values and resulting horizontal regression line of zero slope value and in (C) by the local minimum of loss factor variance s^2 , each shown using pink color. The crossover, when $\tan \delta = 1$, can be seen occurring at considerably later times and depending on the frequency considered (dashed lines in (A)).

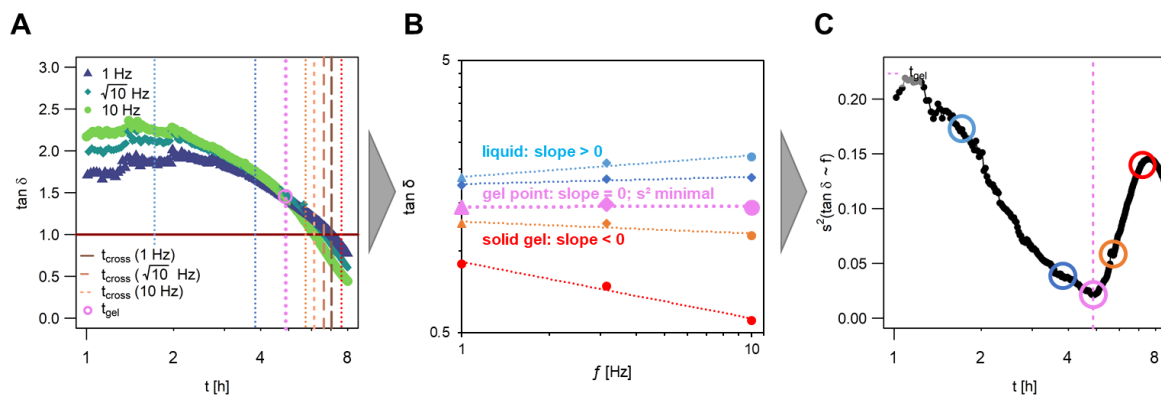


Figure SI–2: Schematic of gel time evaluation using multi-frequency SAOS experiments. A: progression of $\tan \delta$ for the frequencies 1 Hz, $\sqrt{10}$ Hz and 10 Hz. Dark and light blue, pink, orange and red dotted lines in (A) correspond to the respective colored frequency cycle data in (B) and loss factor variance s^2 in (C).

Results and Discussion – Chemical Characteristics of Extractives

The concentrations of saccharides as measured by LC sugar analyzer are listed in Table SI–3. The chemical compounds in the birch wood extractive (cold-water extraction), classified as compound types per GC-MS analysis and estimated concentrations via the internal standard of the birch extractive sample used in this study, are listed in Table SI–4. The compounds, as classified into compound types per UHPLC-ESI-TOF-MS analysis with abundance (peak area), are listed in Table SI–5, whereby high-precision MS data is given as median m/z of the parent ion with 4 digits to 0.0001 Da.

Table SI–3: Free and hydrolyzed saccharide concentrations in birch wood cold-water extractive as identified per LC sugar analyzer using different methods of hydrolysis

saccharide	concentration [mg/g]	
	hydrolysis method I	hydrolysis method II
Oligosaccharides [#]	3.59	2.59
cellobiose	1.2	< 1
Rhamnose	2.62	2.74
peak at 16 min [°]	< 1	< 1
Mannose	3.38	3.83
peak at 24 min ⁺	2.59	---
Arabinose	4.98	4.07
Galactose	4.25	4.21
Xylose	15.08	11.71
Glucose	39.75	45.04
total	77.44	74.19

[#]Oligo calculated as Cellobiose

[°]calculated as Rhamnose

⁺calculated as Arabinose

In the study, the higher results of the hydrolysis method per saccharide are considered, giving a total of 81.1 mg/g.

Table SI-4: Gas-chromatography coupled with mass spectrometry (GC-MS) on silylated water extract sample of silver birch (*Betula pendula* Roth) – Most abundant compounds. Qual: NIST MS Search reported quality. Concentrations and molar mass calculated via peak area (derivatized) using an internal standard (heneicosanoic acid) were corrected for silylation weight gain.

t_R [min]	compound type	silylated concentration [µg/mL]	concentration [µg/mL]	concentration [mg/g]	concentration in wood [µg/g]	molar mass [Da]
12.62	Saccharide (mono)	300.2	99.9	25.2	353.3	180.2
10.29	Saccharide (mono)	114.4	39.2	9.9	138.4	150.1
10.90	Fatty Acid and Conjugates (Carboxylic acid)	48.3	27.3	6.9	96.6	188.2
10.84	Saccharide (mono)	80.1	26.7	6.7	94.1	180.2
10.80	Saccharide (mono)	53.3	17.8	4.5	62.8	180.2
11.68	Saccharide (mono)	31.8	10.6	2.7	37.4	180.2
10.71	Saccharide (mono)	20.9	7.2	1.8	25.3	150.1
4.73	Polyol	30.7	9.2	2.3	32.3	92.1
5.87	Fatty Acid and Conjugates (Carboxylic acid)	11.8	7.9	2.0	27.9	146.2
8.70	Fatty Acid and Conjugates (Carboxylic acid)	15.6	6.9	1.7	24.5	116.1
10.95	Fatty Acid and Conjugates (Dicarboxylic acid)	13.2	6.1	1.5	21.5	187.2
9.86	Fatty Acid and Conjugates (Dicarboxylic acid)	9.4	5.2	1.3	18.2	174.2
19.69	Fatty Acid and Conjugates (Carboxylic acid)	9.1	3.4	0.9	11.9	170.1
8.93	Fatty Acid and Conjugates (Carboxylic acid)	12.1	4.4	1.1	15.5	74.1
8.57	Saccharide (mono)	17.0	5.4	1.4	19.2	136.1
11.76	Saccharide (mono)	14.7	5.1	1.3	18.2	180.2
12.41	Polyol	5.6	3.1	0.8	11.1	180.2
13.50	Polyol	8.6	2.5	0.6	9.0	180.2
18.60	Fatty Acid and Conjugates (Carboxylic acid)	5.2	2.0	0.5	7.0	89.1
5.17	Fatty Acid and Conjugates (Carboxylic acid)	6.1	2.4	0.6	8.3	100.1
8.82	Fatty Acid and Conjugates (Dicarboxylic acid)	3.2	1.7	0.4	6.0	160.2
11.59	Saccharide (mono)	3.3	1.1	0.3	4.0	150.1
17.34	Saccharide (mono)	3.0	1.0	0.2	3.4	150.1
7.20	Fatty Acid and Conjugates (Dicarboxylic acid)	2.2	1.0	0.3	3.6	190.1
12.51	Saccharide (mono)	1.9	0.7	0.2	2.3	150.1

Table SI-5: List of compound features detected via UHPLC-ESI-TOF-MS analysis of birch wood amphiphilic and hydrophilic extractives with retention time, mass-to-charge ratio, compound type annotation and TIC abundance values.

tr [s]	m/z [Da]	compound type	abundance (peak area)
41.01	1080.7848	Saccharide	74620
41.01	944.8092	Saccharide	97804
41.08	808.8334	Saccharide	132950
41.08	876.8216	Saccharide	116307
41.16	740.8462	Saccharide	135039
41.16	834.8401	Saccharide	58090
41.16	1004.8091	Saccharide	47667
41.24	902.8279	Saccharide	55629
41.24	800.8458	Saccharide	48529
41.24	766.8540	Saccharide	39251
41.26	868.8343	Saccharide	57409
41.27	672.8582	Saccharide	128712
41.29	604.8706	Saccharide	135104
41.29	536.8827	Phenyl glycoside	109451
47.40	211.0817	Saccharide	7072
47.76	179.0559	Saccharide	44881
48.98	195.0513	Saccharide	2663623
49.99	711.2203	Saccharide	13227
50.10	665.2148	Saccharide	10351
50.38	341.1088	Saccharide	15580
50.47	503.1607	Saccharide	22223
52.36	379.0831	Saccharide	1996
52.37	377.0857	Saccharide	5902
54.78	96.9606	Inorganic Acid	21601
82.17	96.9607	Inorganic Acid	50403
197.22	451.1445	Phenyl glycoside	106263
197.48	737.2363	Phenyl glycoside	133362
203.64	161.0815	Fatty Acid and Conjugates	89200
208.71	169.0141	Phenol	1471
209.70	391.1238	Phenyl glycoside	40403
210.51	460.1504	Phenyl glycoside	38749
210.82	203.0922	Fatty Acid and Conjugates	31509
210.86	459.1490	Phenyl glycoside	171298
211.90	175.0957	Fatty Acid and Conjugates	235182
212.09	176.1004	Fatty Acid and Conjugates	19615
212.34	451.1456	Phenyl glycoside	2092
212.69	189.0771	Fatty Acid and Conjugates	88321
213.54	359.0985	Phenyl glycoside	200188
215.72	215.0919	Fatty Acid and Conjugates	18326
215.94	195.0663	Phenol	1445
216.65	173.0816	Fatty Acid and Conjugates	29607
217.01	445.1757	Phenyl glycoside	203704
218.01	447.1168	Phenyl glycoside	19064
218.43	405.1395	Phenyl glycoside	74974
218.43	217.1077	Fatty Acid and Conjugates	289911
218.86	199.0969	Fatty Acid and Conjugates	45198
218.94	201.0765	Fatty Acid and Conjugates	32258
220.78	461.1285	Phenyl glycoside	15071

Table SI-5: List of compound features detected via UHPLC-ESI-TOF-MS analysis of birch wood amphiphilic and hydrophilic extractives with retention time, mass-to-charge ratio, compound type annotation and TIC abundance values. (CONTINUED)

t_R [s]	m/z [Da]	compound type	abundance (peak area)
221.01	444.1935	Phenyl glycoside	259914
221.05	443.1914	Phenyl glycoside	1197706
221.62	551.1607	Phenyl glycoside	11064
224.94	521.1006	Phenyl glycoside	7433
225.36	241.0718	Phenol	31482
225.36	489.1652	Phenyl glycoside	35886
226.31	203.0929	Fatty Acid and Conjugates	312996
226.69	527.1751	Phenyl glycoside	123993
226.95	557.1849	Phenyl glycoside	47988
229.31	161.0813	Fatty Acid and Conjugates	22639
229.99	541.1585	Phenyl glycoside	31160
230.76	523.1666	Phenyl glycoside	842357
230.76	524.1700	Phenyl glycoside	189947
230.83	525.1722	Phenyl glycoside	68070
230.83	577.0860	Phenyl glycoside	13784
230.98	513.1374	Phenyl glycoside	40377
230.99	575.1285	Phenyl glycoside	40968
231.01	540.1559	Phenyl glycoside	23962
231.01	515.1350	Phenyl glycoside	12266
231.01	576.0819	Phenyl glycoside	11727
231.05	591.1529	Phenyl glycoside	22552
231.26	478.1636	Phenyl glycoside	25070
231.33	477.1611	Phenyl glycoside	112230
231.37	693.2607	Phenyl glycoside	39947
231.42	479.1652	Phenyl glycoside	13471
231.86	215.0925	Fatty Acid and Conjugates	102624
233.45	505.1906	Phenyl glycoside	154065
233.98	427.1583	Phenyl glycoside	183186
234.44	507.1862	Phenyl glycoside	546173
234.45	391.1245	Phenyl glycoside	392164
234.86	503.1754	Phenyl glycoside	44530
235.68	509.1892	Phenyl glycoside	39956
235.68	625.1236	Phenyl glycoside	15021
236.08	585.1818	Phenyl glycoside	331447
236.14	586.1844	Phenyl glycoside	98557
237.59	583.2013	Phenyl glycoside	68822
237.80	201.0785	Fatty Acid and Conjugates	63634
238.62	401.1443	Phenyl glycoside	39028
238.86	447.1511	Phenyl glycoside	54938
240.08	319.1193	Polyphenol	51753
240.39	555.1711	Phenyl glycoside	330303
240.49	271.0963	Polyphenol	42911
240.84	287.1503	Fatty Acid and Conjugates	9787
241.21	301.1113	Polyphenol	25079
241.43	245.1354	Fatty Acid and Conjugates	33948
241.82	627.2230	Phenyl glycoside	12252
242.23	443.1918	Phenyl glycoside	1032781
242.23	444.1953	Phenyl glycoside	224539
242.64	541.1584	Phenyl glycoside	26346

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t_R [s]	m/z [Da]	compound type	abundance (peak area)
242.68	432.1948	Glycolipid	184873
242.68	431.1916	Glycolipid	884015
242.69	159.0664	Fatty Acid and Conjugates	86052
242.84	161.0816	Fatty Acid and Conjugates	57655
243.28	567.1768	Phenyl glycoside	45929
243.98	423.1795	Glycolipid	160300
244.15	539.1417	Phenyl glycoside	29736
244.96	517.2580	Glycolipid	72309
246.12	195.0660	Phenol	21554
246.14	491.0856	Phenyl glycoside	3063
247.27	557.1568	Phenyl glycoside	80844
247.39	573.1758	Phenyl glycoside	63371
247.39	527.1716	Phenyl glycoside	57573
247.43	459.1871	Phenyl glycoside	4800610
247.52	460.1898	Phenyl glycoside	1020254
247.67	495.1623	Phenyl glycoside	87130
247.77	461.1834	Phenyl glycoside	217071
248.24	787.2676	Phenyl glycoside	59673
248.25	788.2701	Phenyl glycoside	22178
248.46	505.1912	Phenyl glycoside	129033
249.81	579.2054	Phenyl glycoside	89178
250.08	465.1033	Phenyl glycoside	2677
250.19	569.1855	Phenyl glycoside	234351
252.65	580.2104	Phenyl glycoside	39570
252.80	489.1366	Phenyl glycoside	11491
252.96	203.0927	Fatty Acid and Conjugates	75790
253.07	391.1372	Phenyl glycoside	6520
253.29	553.1795	Phenyl glycoside	101700
254.30	515.1897	Polyphenol	71312
254.75	585.2184	Phenyl glycoside	182709
254.75	586.2212	Phenyl glycoside	52905
255.27	549.1918	Phenyl glycoside	111582
256.06	581.2240	Phenyl glycoside	2219749
256.06	627.2291	Phenyl glycoside	914613
256.06	582.2264	Phenyl glycoside	631063
256.13	628.2322	Phenyl glycoside	283610
256.25	521.2018	Phenyl glycoside	555452
256.43	419.1154	Phenyl glycoside	48415
256.47	583.2223	Phenyl glycoside	196023
256.55	261.1343	Glycerolipid	65138
256.67	447.1022	Phenyl glycoside	8795
258.46	541.1495	Phenyl glycoside	69859
259.01	539.1433	Phenyl glycoside	486430
259.05	540.1451	Phenyl glycoside	120723
259.06	517.1921	Phenyl glycoside	18415
259.13	525.1607	Phenyl glycoside	190894
261.21	491.1913	Phenyl glycoside	424277
262.09	509.1307	Phenyl glycoside	391926

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t_R [s]	m/z [Da]	compound type	abundance (peak area)
262.11	567.2067	Phenyl glycoside	141812
262.38	247.1189	Glycerolipid	337389
263.98	185.0820	Fatty Acid and Conjugates	49254
264.87	713.2662	Phenyl glycoside	36484
266.82	577.1926	Phenyl glycoside	45353
267.18	595.2032	Phenyl glycoside	711661
267.31	596.2058	Phenyl glycoside	212146
267.43	551.2137	Phenyl glycoside	770497
267.43	552.2160	Phenyl glycoside	218667
267.49	597.2171	Phenyl glycoside	463304
267.56	598.2204	Phenyl glycoside	132193
268.50	261.1344	Glycerolipid	115304
268.74	449.2010	Phenyl glycoside	322027
269.09	475.1972	Phenyl glycoside	266713
269.26	451.1264	Phenyl glycoside	109013
269.87	489.1746	Phenyl glycoside	692178
271.37	481.1344	Phenyl glycoside	283517
271.38	459.1474	Phenyl glycoside	21988
271.65	173.0820	Fatty Acid and Conjugates	470543
271.82	195.0639	Fatty Acid and Conjugates	11408
272.04	523.1387	Phenyl glycoside	2552
272.06	513.1757	Phenyl glycoside	6857
274.10	423.1299	Phenyl glycoside	51011
275.24	205.1081	Fatty Acid and Conjugates	361501
275.67	159.1020	Fatty Acid and Conjugates	203168
275.77	569.2213	Phenyl glycoside	84031
275.84	521.2013	Phenyl glycoside	99493
276.05	431.1365	Phenyl glycoside	16760
276.09	553.1899	Phenyl glycoside	160960
276.64	611.1965	Phenyl glycoside	27509
276.95	495.1496	Phenyl glycoside	268226
278.44	573.2032	Phenyl glycoside	22126
279.15	549.2529	Phenyl glycoside	81965
279.29	245.1392	Fatty Acid and Conjugates	8479
279.73	551.2133	Phenyl glycoside	53286
279.73	552.2162	Phenyl glycoside	13937
280.18	217.1083	Fatty Acid and Conjugates	194732
280.71	593.2051	Phenyl glycoside	18190
281.36	461.0739	Phenyl glycoside	1614
282.18	579.2077	Phenyl glycoside	809362
282.32	580.2106	Phenyl glycoside	241517
282.38	427.1584	Phenyl glycoside	4485
283.03	449.1449	Phenyl glycoside	2721
283.21	505.1335	Phenyl glycoside	2807
283.31	625.2132	Phenyl glycoside	166867
283.63	515.1765	Phenyl glycoside	41472
283.74	175.0971	Fatty Acid and Conjugates	1134999
283.74	176.1006	Fatty Acid and Conjugates	93180
284.26	496.1174	Phenyl glycoside	67627

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t_R [s]	m/z [Da]	compound type	abundance (peak area)
284.32	325.1092	Polyphenol	1327
286.13	261.1345	Glycerolipid	1149636
286.78	713.2650	Phenyl glycoside	16222
286.78	169.0870	Fatty Acid and Conjugates	10076
287.48	201.1131	Fatty Acid and Conjugates	29020
288.37	477.2117	Phenyl glycoside	89158
289.40	417.2083	Glycolipid	515144
290.29	287.1500	Fatty Acid and Conjugates	142356
291.03	171.0991	Fatty Acid and Conjugates	29738
291.21	523.1855	Phenyl glycoside	241697
291.21	524.1882	Phenyl glycoside	64695
291.66	327.1233	Polyphenol	438358
291.69	489.1955	Phenyl glycoside	14071
292.01	189.1132	Fatty Acid and Conjugates	311231
292.72	345.2285	Glycolipid	12480
293.01	241.1075	Phenol	57114
294.69	325.1085	Polyphenol	194216
295.71	195.0304	Phenol	1861
295.79	505.1331	Phenyl glycoside	18476
296.57	507.1846	Phenyl glycoside	238919
298.50	583.2065	Phenyl glycoside	51378
298.75	525.1611	Phenyl glycoside	2580155
298.76	526.1639	Phenyl glycoside	655334
298.79	475.2034	Phenyl glycoside	1486170
298.79	527.1673	Phenyl glycoside	143984
298.96	713.2658	Phenyl glycoside	239783
299.36	401.1236	Polyphenol	2122
299.50	431.1231	Polyphenol	17192
299.58	188.1008	Fatty Acid and Conjugates	214550
299.60	187.0981	Fatty Acid and Conjugates	2388556
299.71	169.0865	Fatty Acid and Conjugates	37009
300.01	209.0791	Fatty Acid and Conjugates	18964
300.30	359.1547	Polyphenol	22013
300.40	549.2063	Phenyl glycoside	12584
302.53	496.1522	Phenyl glycoside	63271
302.68	495.1496	Phenyl glycoside	252238
302.78	343.2113	Fatty Acid and Conjugates	20459
310.03	203.0917	Fatty Acid and Conjugates	38487
310.58	145.0871	Fatty Acid and Conjugates	799226
311.44	261.1331	Glycerolipid	71128
311.73	159.1027	Fatty Acid and Conjugates	1040
314.04	201.1136	Fatty Acid and Conjugates	7577
315.03	593.2604	Phenyl glycoside	44422
315.09	594.2638	Phenyl glycoside	15108
315.11	639.2631	Phenyl glycoside	35786
317.00	199.0972	Fatty Acid and Conjugates	30184
317.68	169.0868	Fatty Acid and Conjugates	45778
320.32	325.1100	Polyphenol	31468
322.18	553.2180	Phenyl glycoside	6508

Table SI–5: List of compound features detected via UHPLC-ESI-TOF-MS analysis of birch wood amphiphilic and hydrophilic extractives with retention time, mass-to-charge ratio, compound type annotation and TIC abundance values. (CONTINUED)

t_R [s]	m/z [Da]	compound type	abundance (peak area)
322.76	569.2238	Phenyl glycoside	65186
323.89	287.1485	Fatty Acid and Conjugates	28267
323.94	345.2270	Glycolipid	129213
324.47	159.1026	Fatty Acid and Conjugates	105229
324.75	273.1707	Fatty Acid and Conjugates	75948
326.86	201.1135	Fatty Acid and Conjugates	259524
330.11	433.2055	Glycolipid	27952
335.87	583.2153	Phenyl glycoside	8839
336.31	171.1028	Fatty Acid and Conjugates	470984
343.27	327.2177	Fatty Acid and Conjugates	29317
343.81	245.1386	Fatty Acid and Conjugates	32307
345.12	301.2021	Fatty Acid and Conjugates	17220
350.27	159.1020	Fatty Acid and Conjugates	23332
357.74	171.1025	Fatty Acid and Conjugates	307768
358.77	273.1698	Fatty Acid and Conjugates	5382
360.39	169.0866	Fatty Acid and Conjugates	13367
361.77	201.1134	Fatty Acid and Conjugates	97369
362.86	331.2382	Fatty Acid and Conjugates	47866
364.73	225.1492	Fatty Acid and Conjugates	14888
365.26	343.2112	Fatty Acid and Conjugates	66047
366.16	329.2329	Fatty Acid and Conjugates	2160958
371.00	345.2272	Glycolipid	125420
371.14	301.1651	Fatty Acid and Conjugates	12121
380.04	331.2474	Fatty Acid and Conjugates	82937
385.14	169.0846	Fatty Acid and Conjugates	44562
389.80	329.2331	Fatty Acid and Conjugates	691429
394.42	301.2007	Fatty Acid and Conjugates	7506
406.27	331.2468	Fatty Acid and Conjugates	30040
408.75	297.1498	Polyphenol	13702
415.63	293.1193	Polyphenol	12522
417.85	331.2494	Fatty Acid and Conjugates	209142
419.96	207.1380	Phenol	2835
428.08	516.3530	Phospholipid	17135
428.99	293.1757	Polyphenol	15056
433.64	207.1390	Phenol	37118
442.05	293.1750	Polyphenol	3511
458.20	293.1760	Polyphenol	37926
474.16	293.2098	Fatty Acid and Conjugates	3713
517.22	293.2100	Fatty Acid and Conjugates	6204

Results and Discussion – Chemical Characteristics of Cured Adhesive-Extractives Mixtures

In Figure SI–3, the fingerprint region of FTIR spectra for MUF components and cured adhesive as well as cured and uncured PUR adhesive are shown.

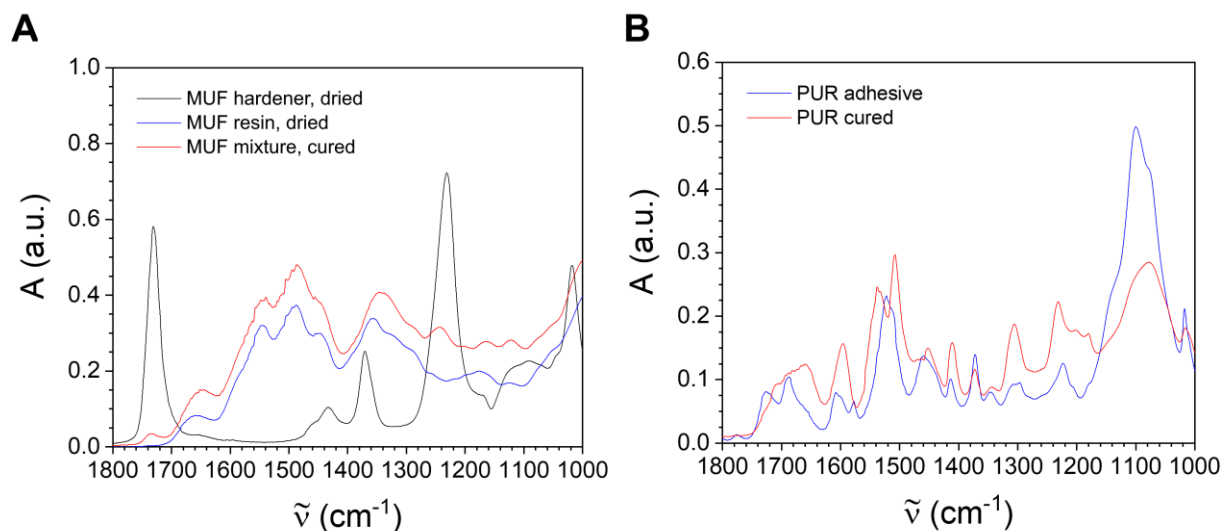


Figure SI–3: FTIR spectra of A) MUF components (dried hardener – black; dried resin – blue) and cured MUF mixture (red) and B) PUR (uncured – blue; cured – red)

Results and Discussion – Rheological Experiments – Adhesive Curing Behavior

Experimental data for MUF and PUR curing rheological experiments are shown in the following figures (from Figure SI-4 to Figure SI-19): The progression of the storage modulus (G') and loss modulus (G'') at 1 Hz together with the loss modulus peak, indicating glass formation (t_{glass} , blue dashed line) and the crossover time when of G' and G'' at 1 Hz (t_{cross} , brown dashed line), are shown in A. The dynamic viscosity (η^*) together with the gel point t_{η} – indicated by reaching 100 Pa·s – are shown in B. The loss factor ($\tan \delta$) is shown for the lowest frequency (1 Hz), highest frequency (10 Hz) and the geometric mean ($\sqrt{10}$ Hz) in C, together with the time of crossover at the respective frequencies (dashed lines). In D, the progression of the frequency-dependent variance of the loss factor ($\tan \delta$) and its local minimum (t_{gel} , violet dashed line), indicating percolation and, thus, the occurrence of the gelation, is shown, which is also marked in C (empty violet circle).

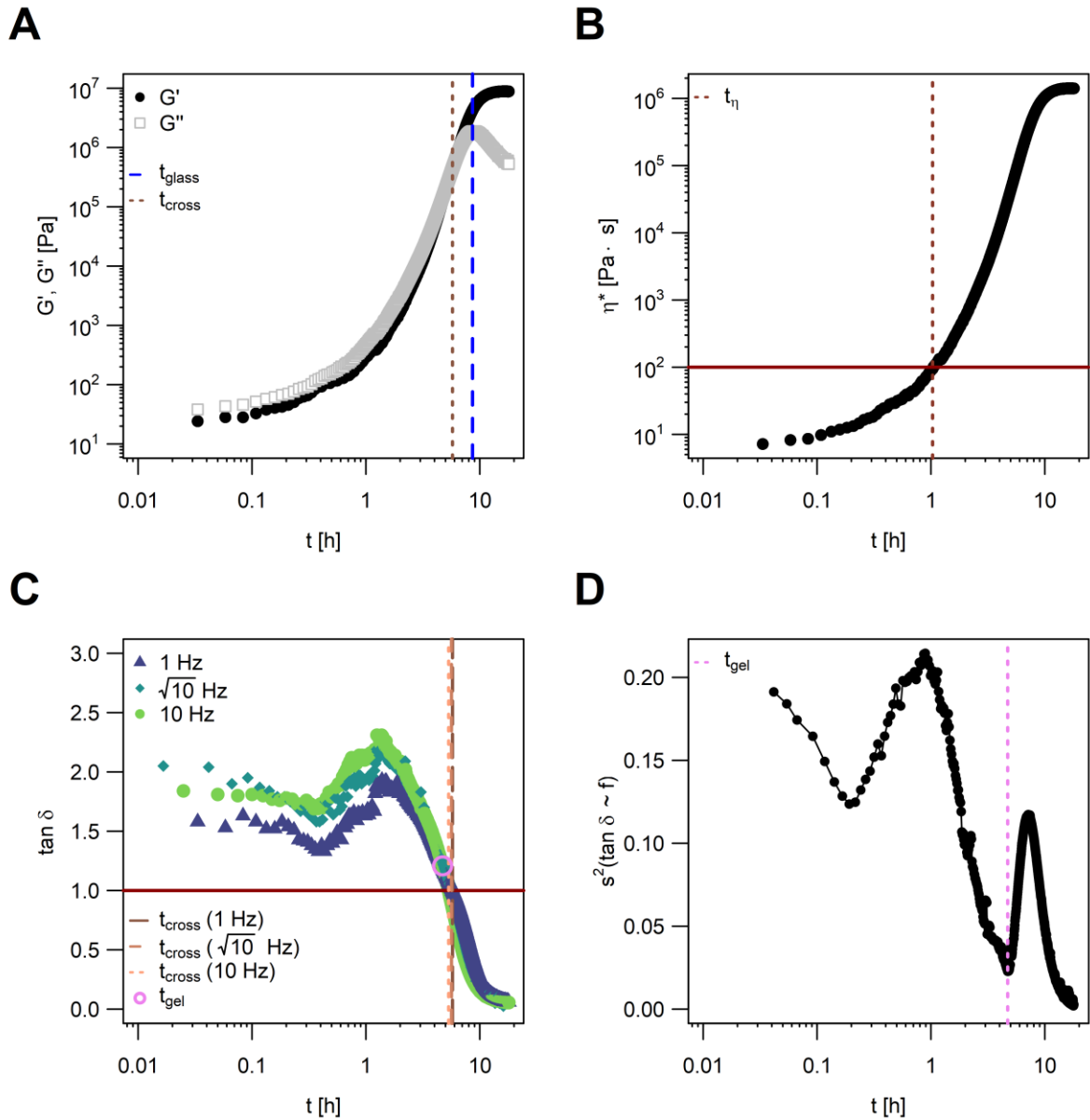


Figure SI-4: Isothermal time-sweep experiment at 20 °C/65%-RH during the curing process of MUF adhesive without extractives addition on pristine birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$) -; B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} -; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} -; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

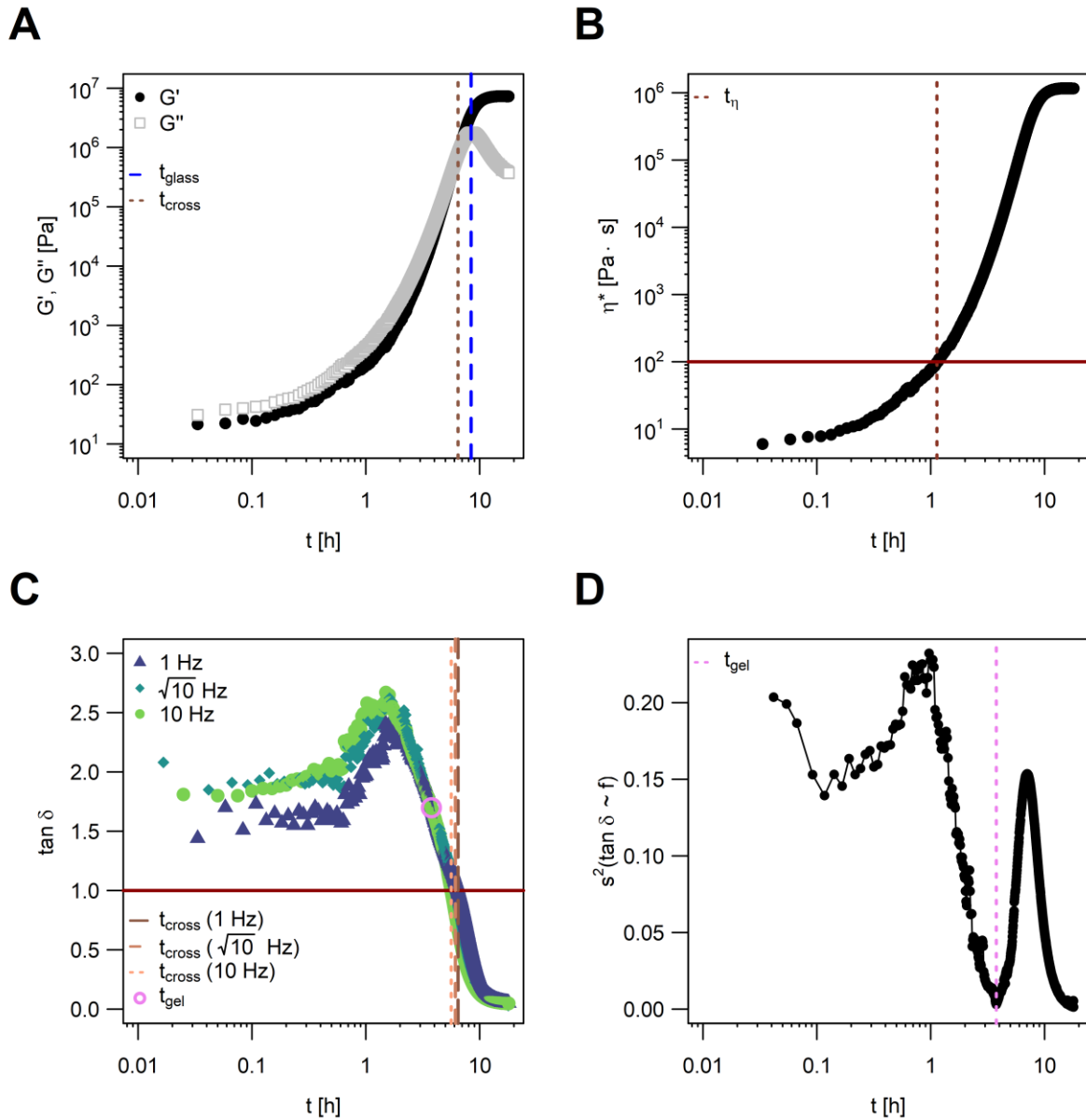


Figure SI-5: Isothermal time-sweep experiment at 20 °C/65%-RH during the curing process of MUF adhesive without extractives addition on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$) -; B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} -; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} -; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

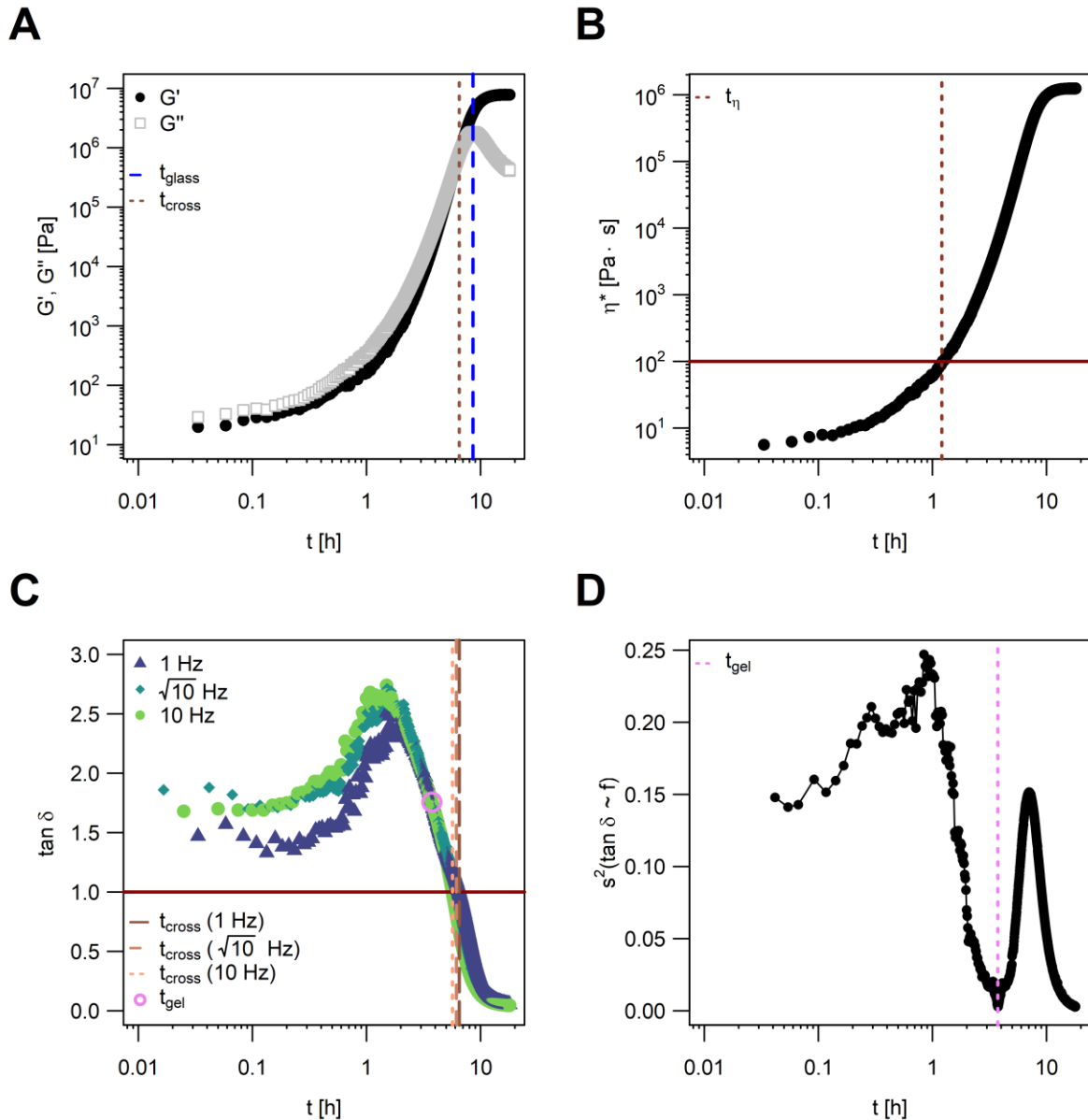


Figure SI-6: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of MUF adhesive with added extractive concentration of 0.2% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

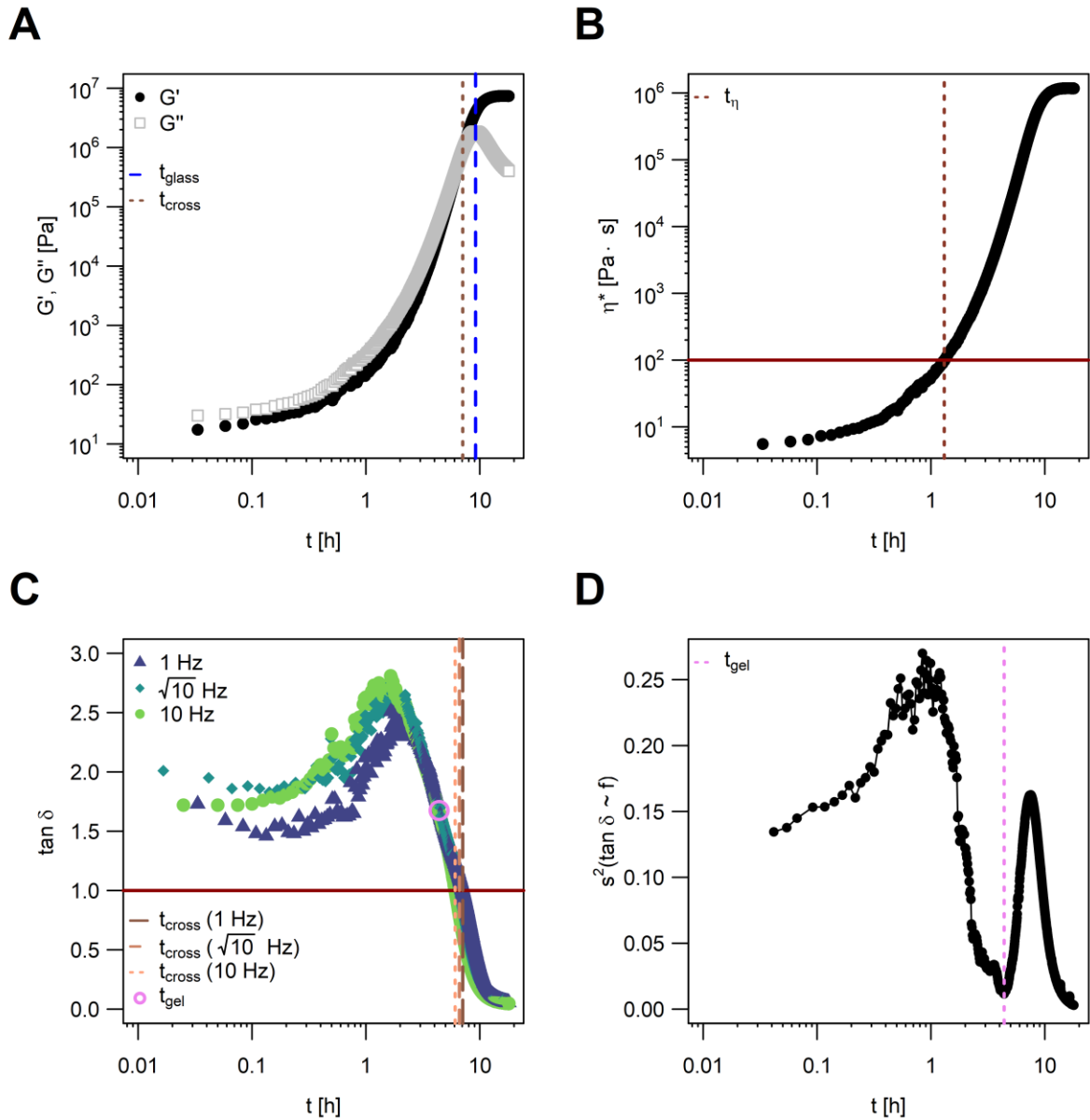


Figure SI-7: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of MUF adhesive with added extractive concentration of 0.4% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

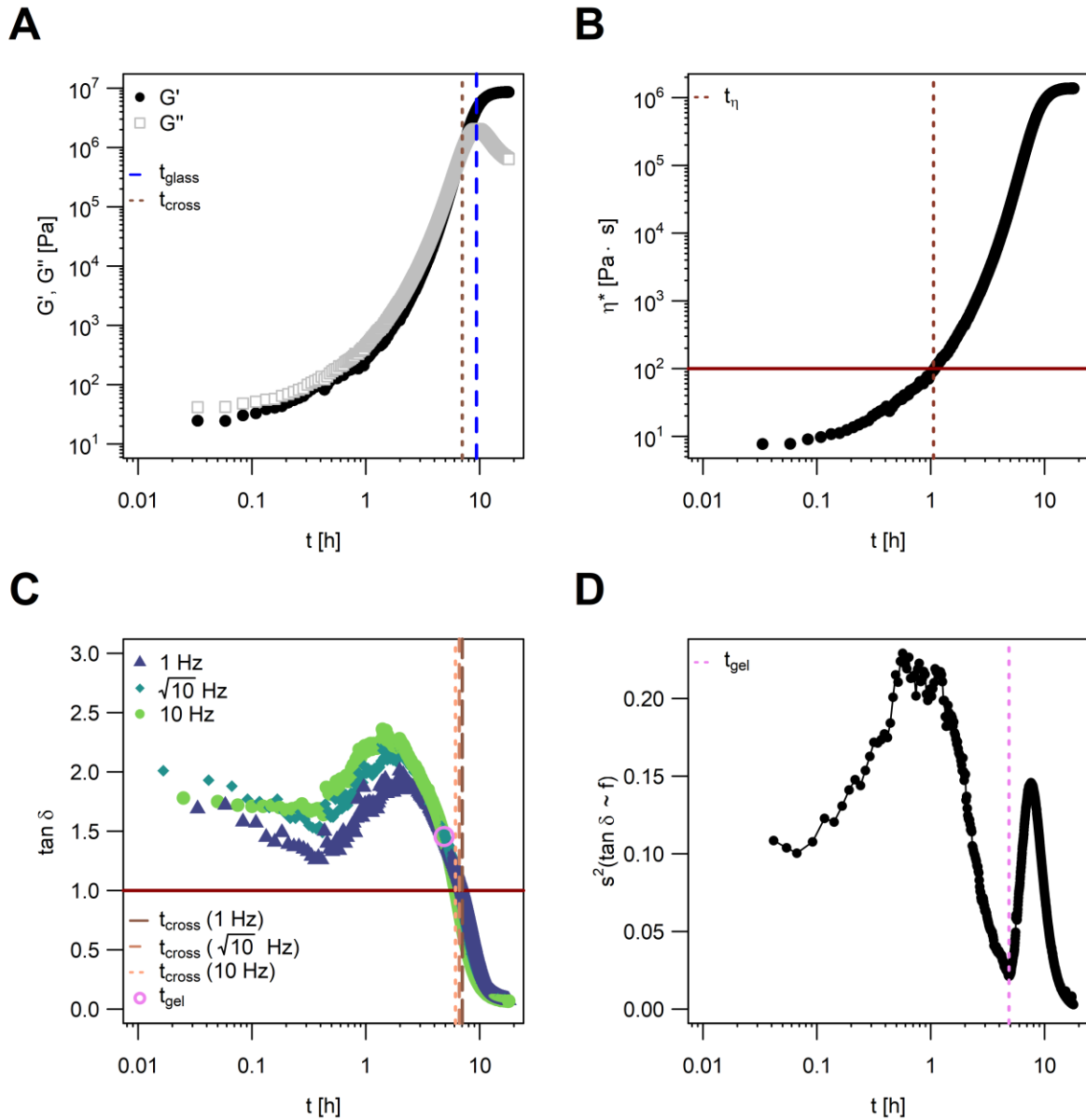


Figure SI-8: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of MUF adhesive with added extractive concentration of 0.6% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

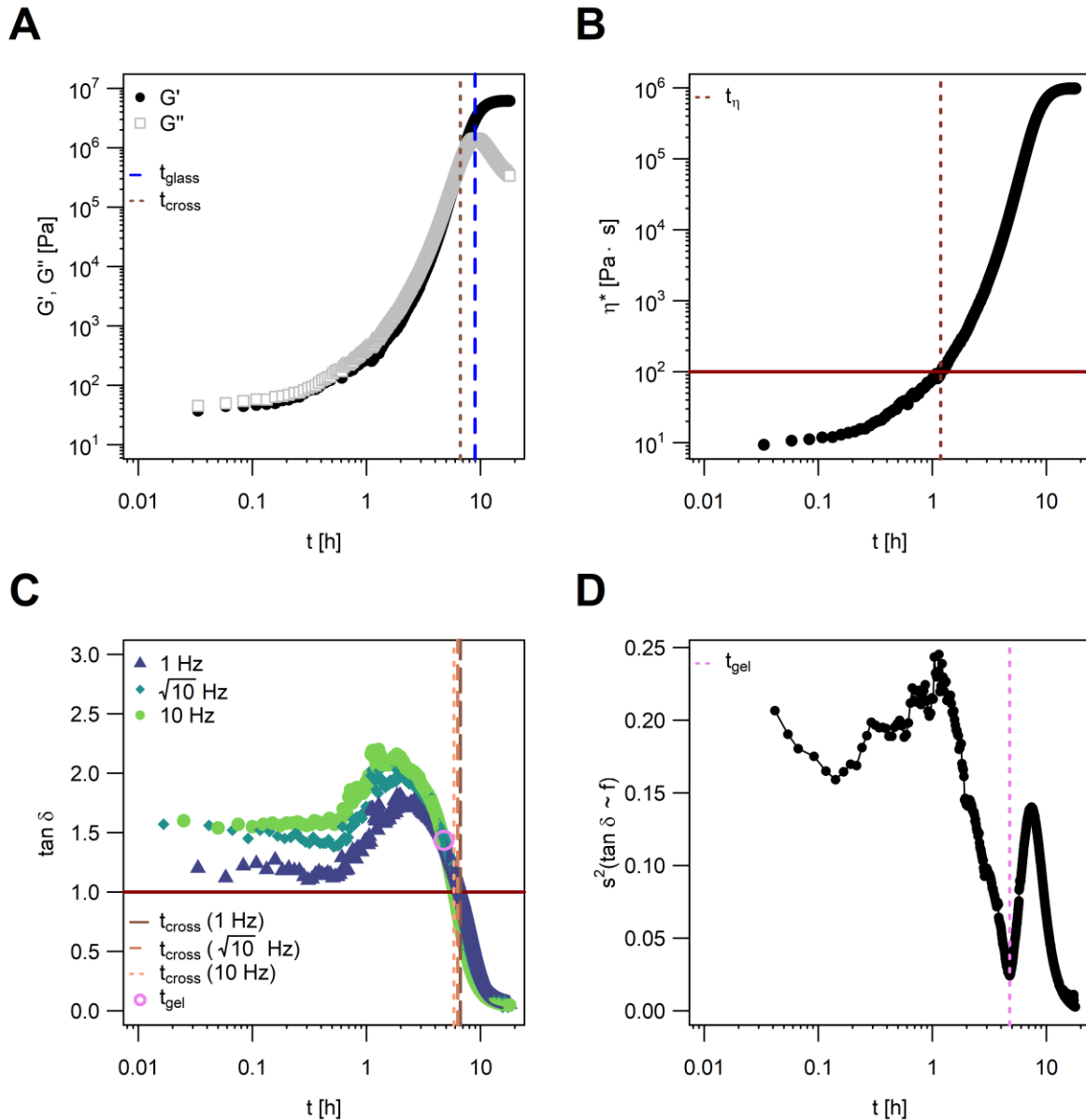


Figure SI-9: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of MUF adhesive with added extractive concentration of 0.8% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

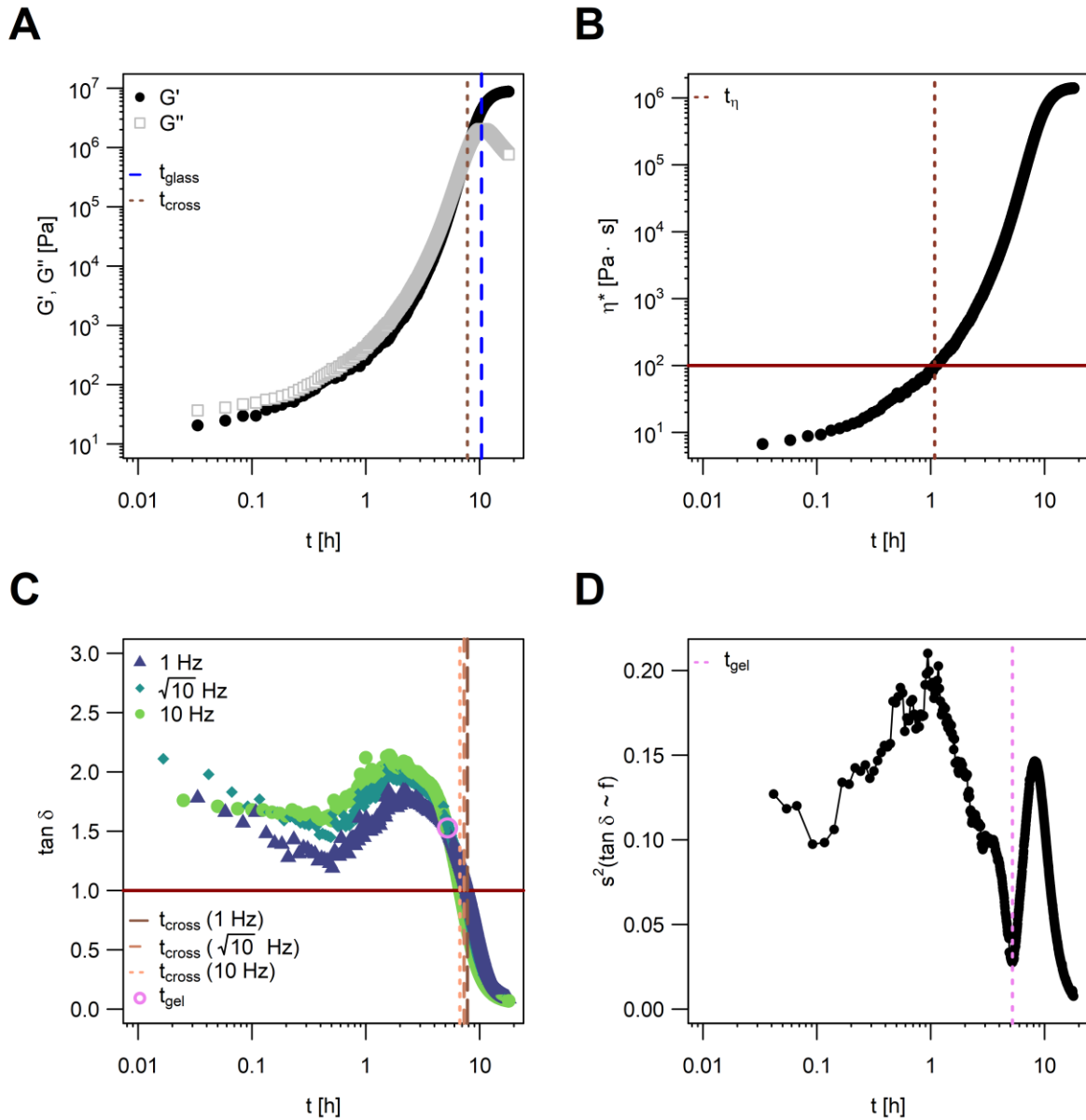


Figure SI-10: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of MUF adhesive with added extractive concentration of 1.0% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

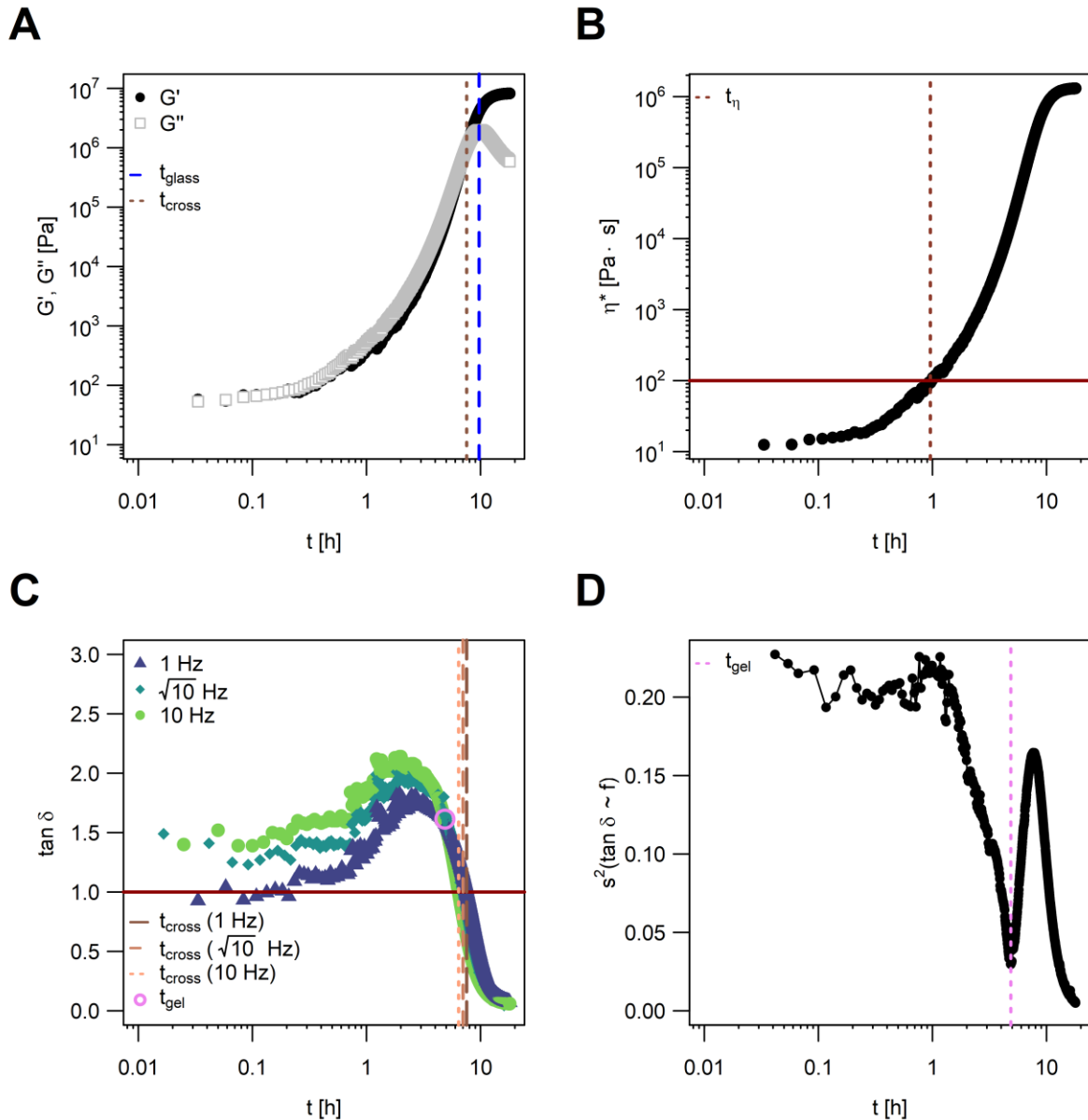


Figure SI-11: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of MUF adhesive with added extractive concentration of 1.2% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

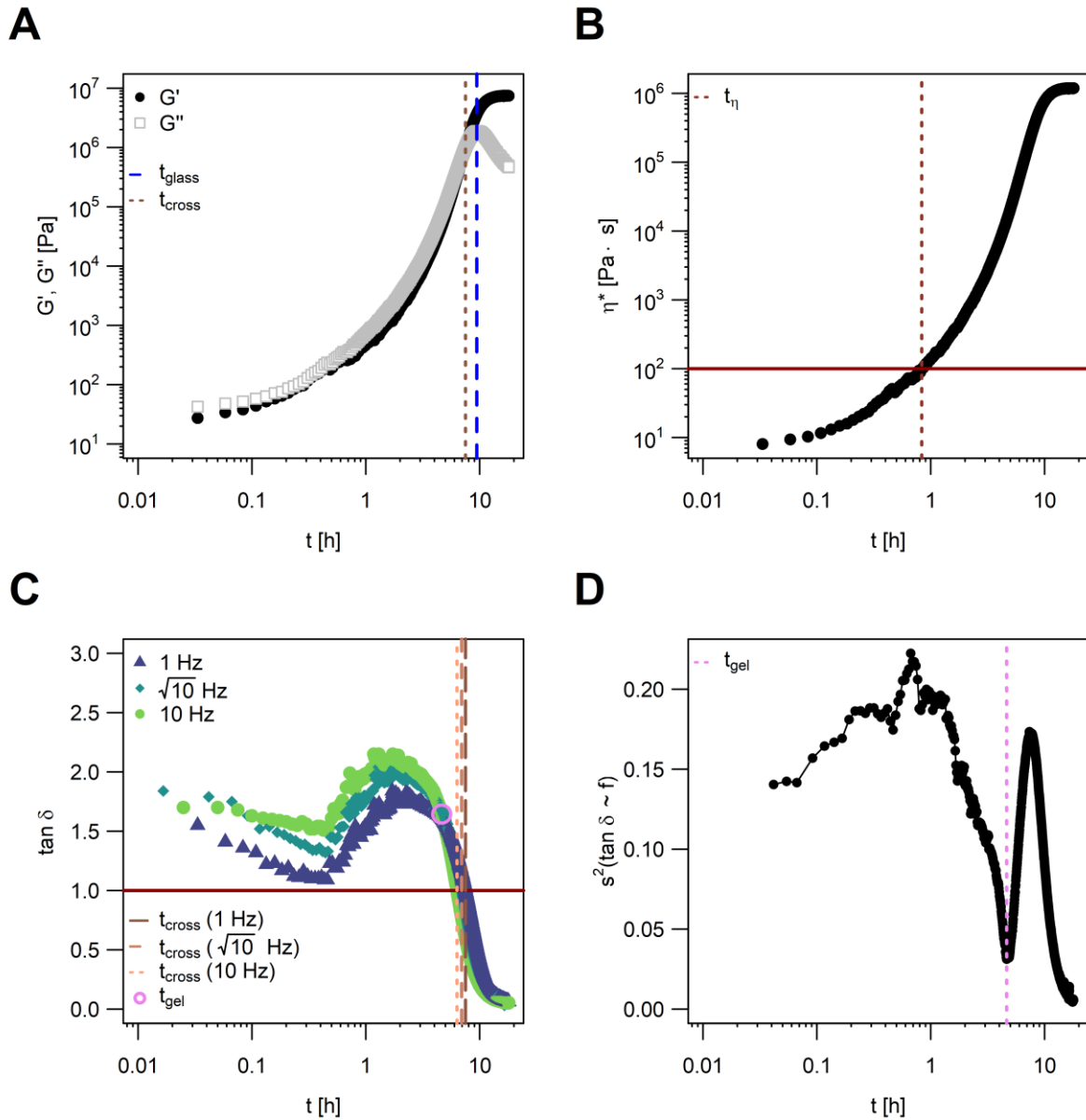


Figure SI-12: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of MUF adhesive with added extractive concentration of 1.4% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

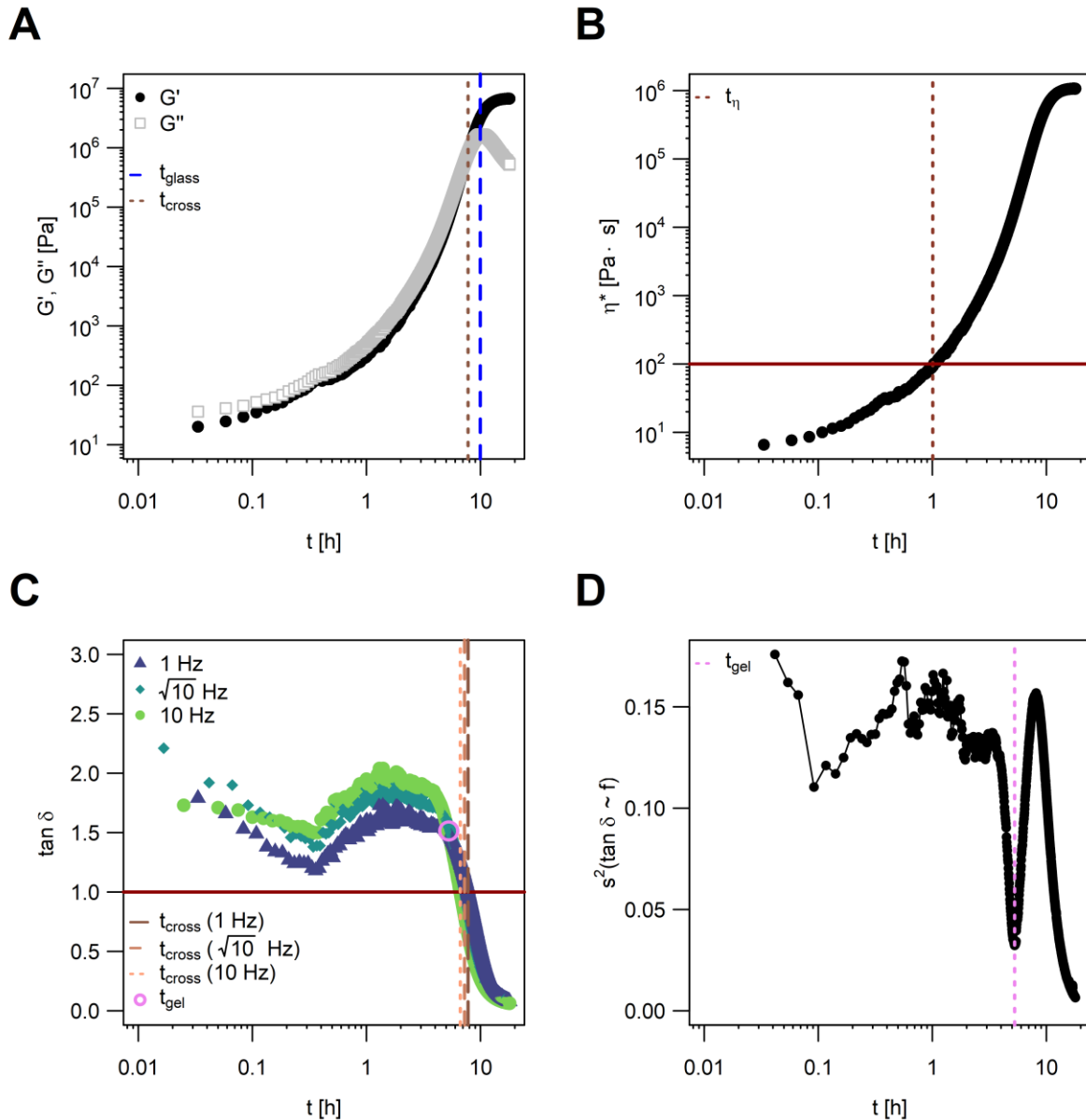


Figure SI-13: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of MUF adhesive with added extractive concentration of 1.6% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

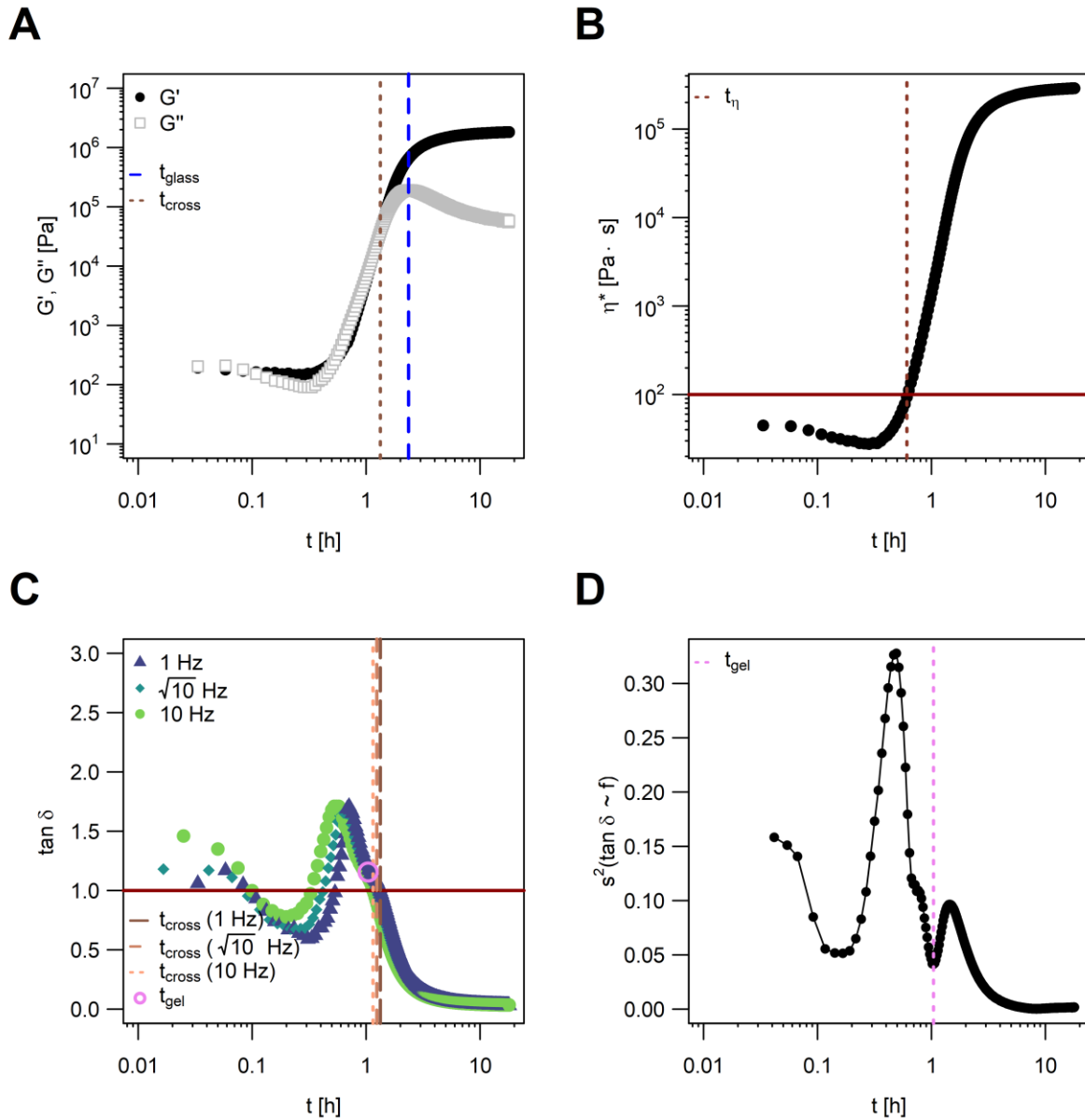


Figure SI-14: Isothermal time-sweep experiment at 20 °C/65%-RH during the curing process of PUR adhesive without extractives addition on pristine birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

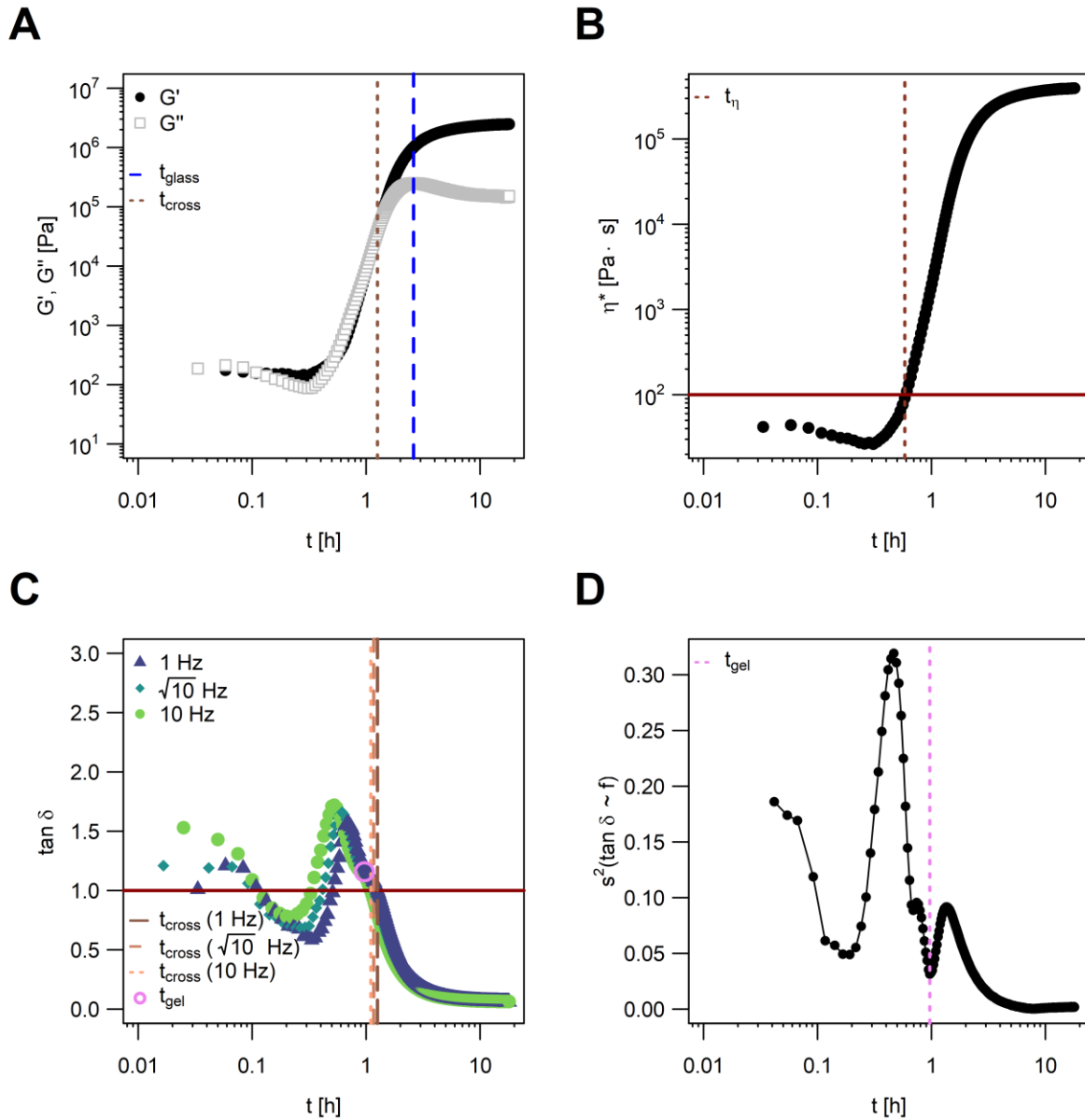


Figure SI-15: Isothermal time-sweep experiment at 20 °C/65% -RH during the curing process of PUR adhesive without extractives addition on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

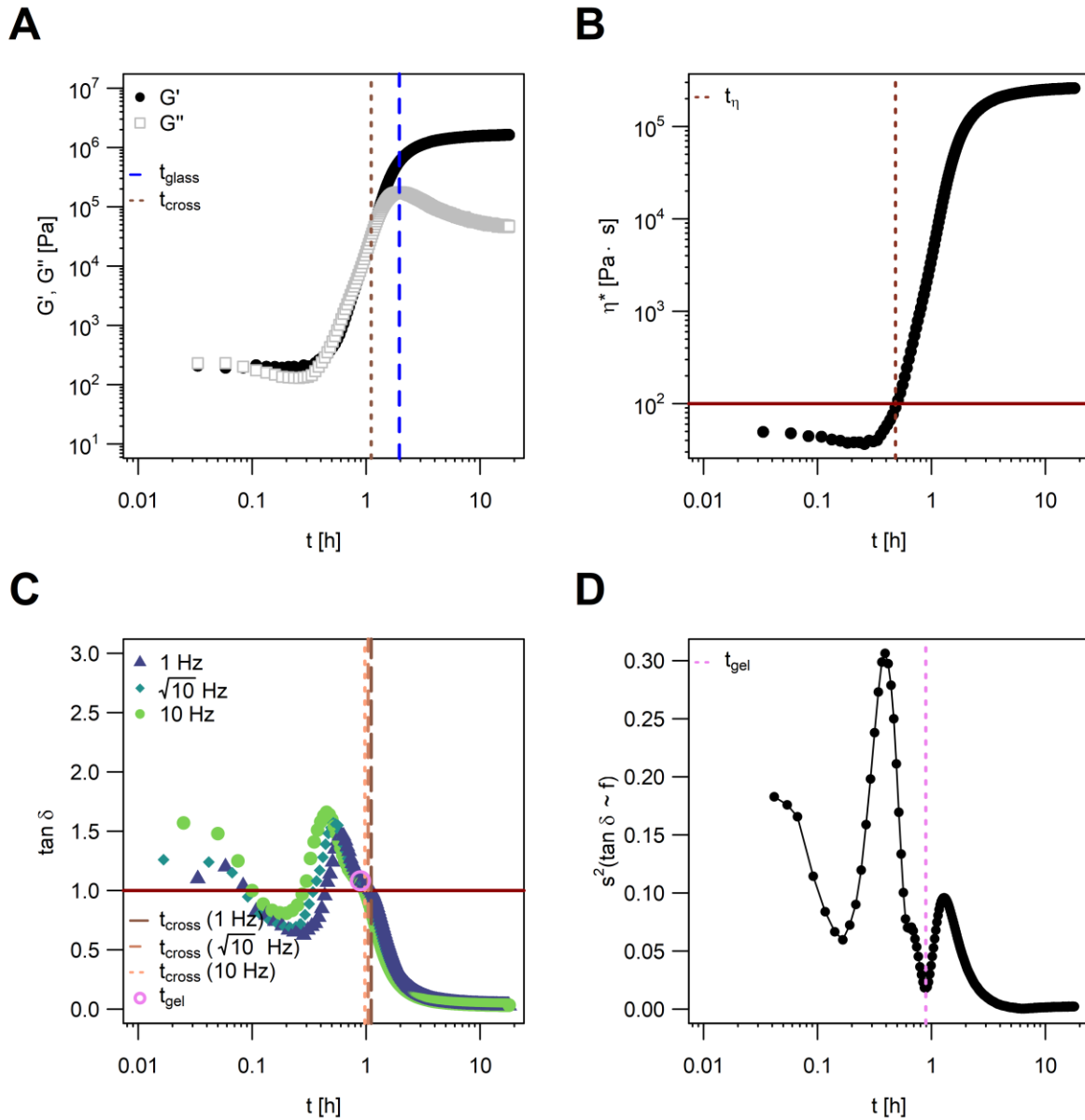


Figure SI-16: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of PUR adhesive with added extractive concentration of 0.05% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

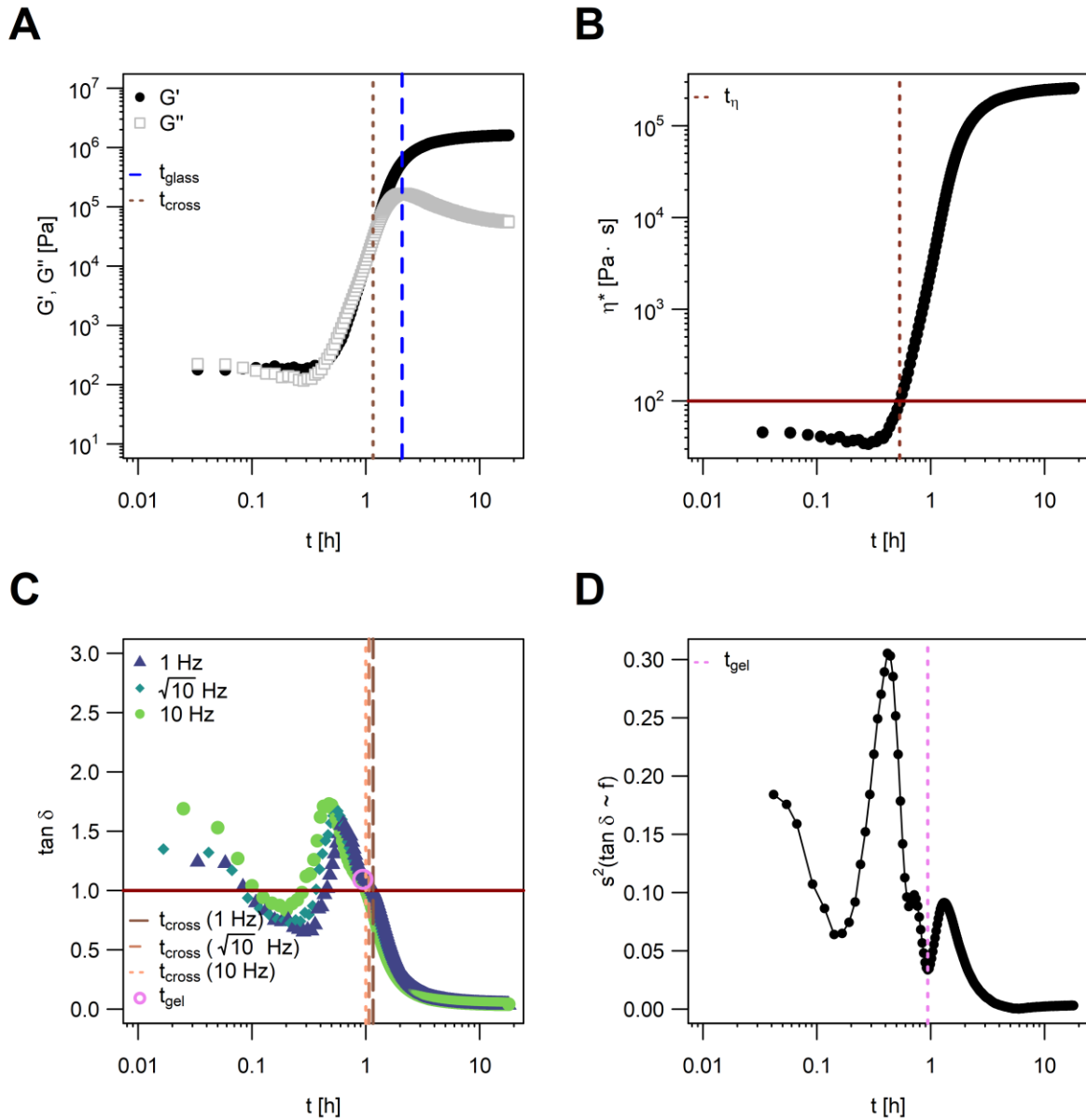


Figure SI-17: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of PUR adhesive with added extractive concentration of 0.1% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

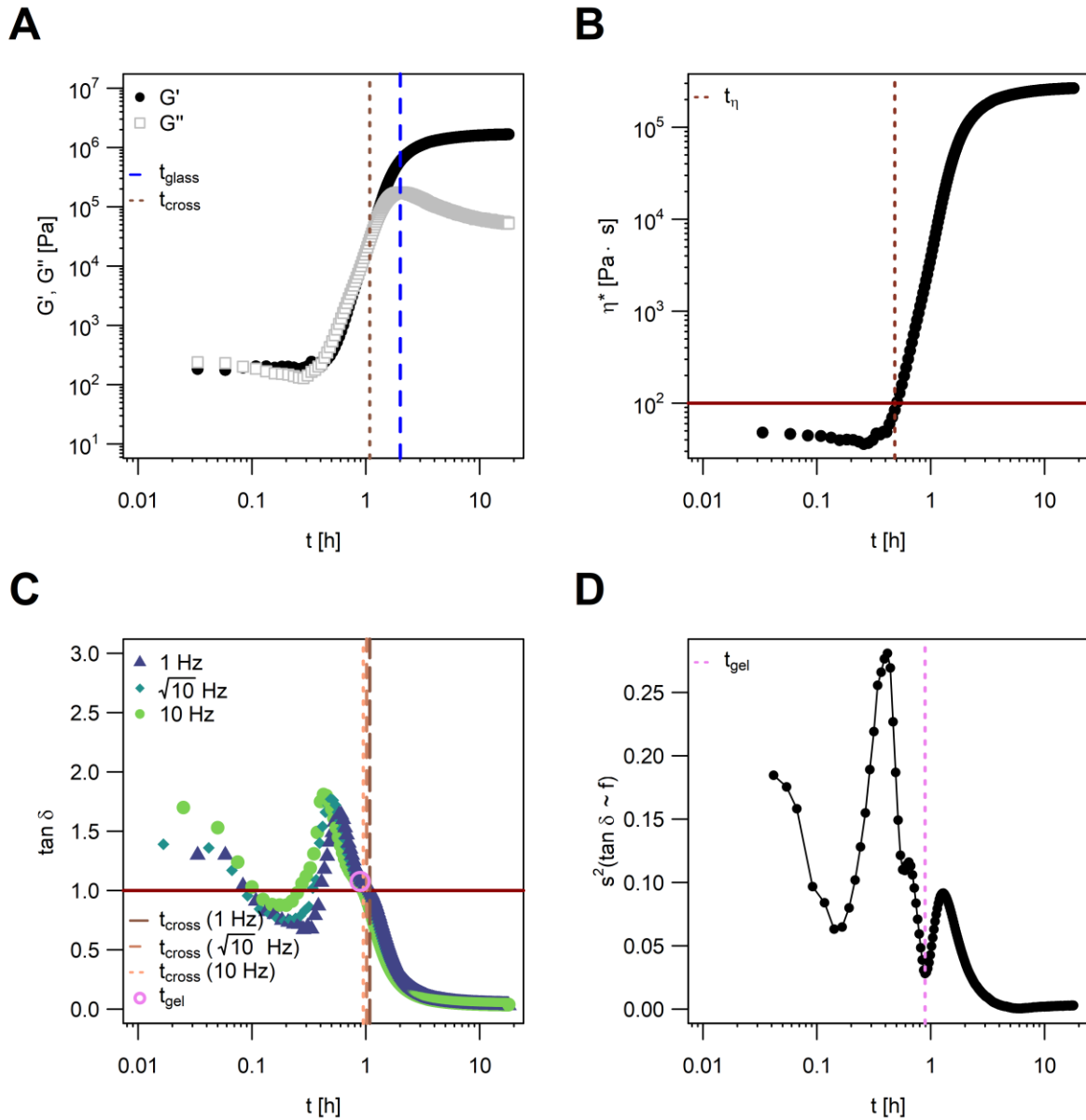


Figure SI-18: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of PUR adhesive with added extractive concentration of 0.2% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

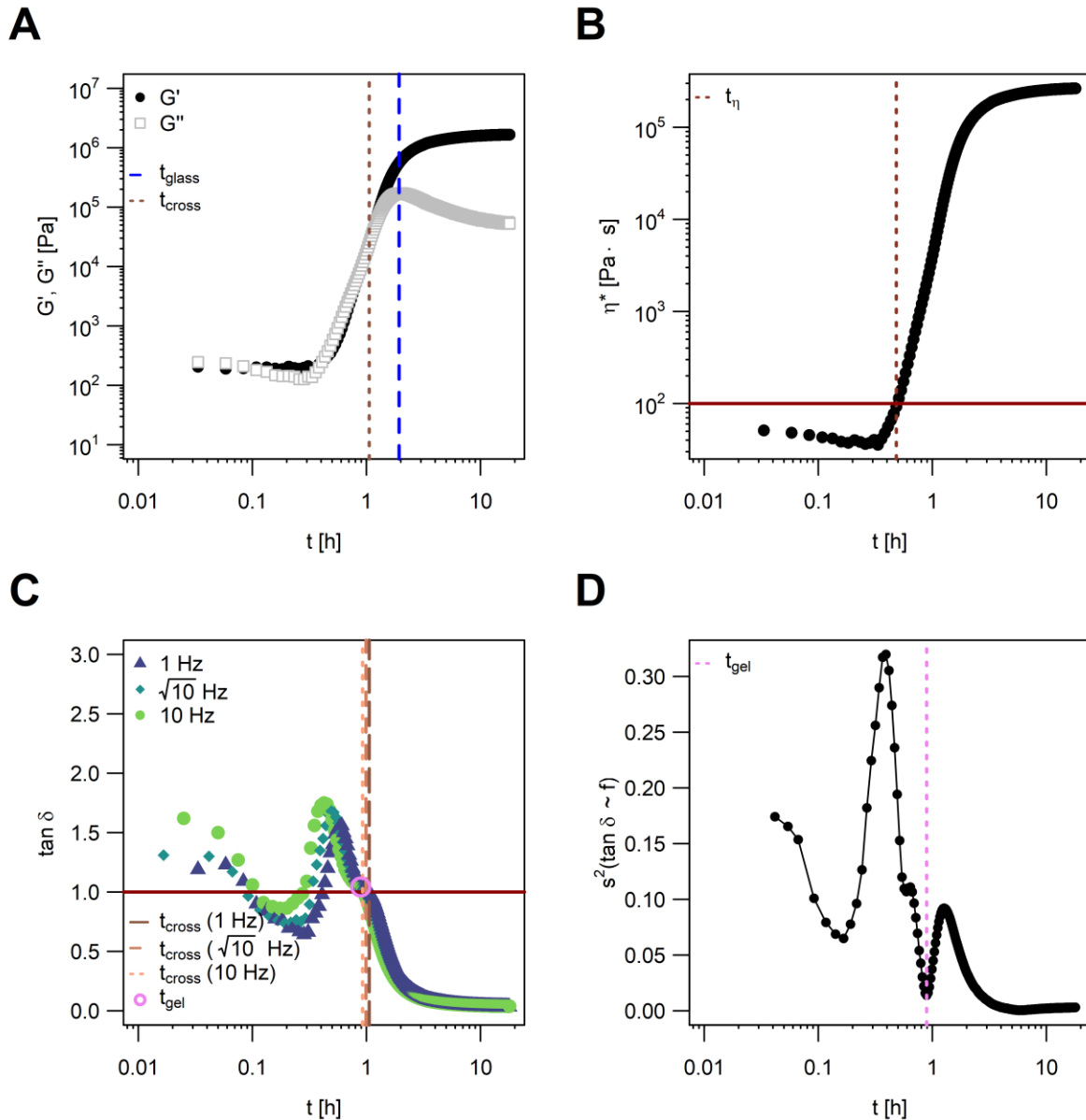


Figure SI-19: Isothermal time-sweep experiment at 20 °C/65% RH during the curing process of PUR adhesive with added extractive concentration of 0.4% (W/W) on extracted birch wood: A) storage modulus G' and loss modulus G'' at 1 Hz – the blue dashed line corresponds to the vitrification time t_{glass} and the brown dashed to the crossover time t_{cross} ($\tan \delta = 1$); B) dynamic viscosity η^* at 1 Hz – the dashed line corresponds to the gel point t_{η} ; C) loss factor $\tan \delta$ with at different frequency values – the dashed lines correspond to the crossover time t_{cross} ($\tan \delta = 1$) and violet circle to the gel time t_{gel} ; and D) loss factor variance – the dashed line corresponds to the local minimum variance indicating gel time t_{gel} .

In Figure SI–20A, the progression of the rheometer gap size (adhesive layer thickness, d) throughout the MUF curing experiments on pristine and extracted wood is shown. The thick lines are average curves, and the light curves show the original experimental data. In Figure SI–20B, the progression of the axial pressure in the initial phase of PUR curing experiments due to CO_2 development on pristine and extracted wood is shown.

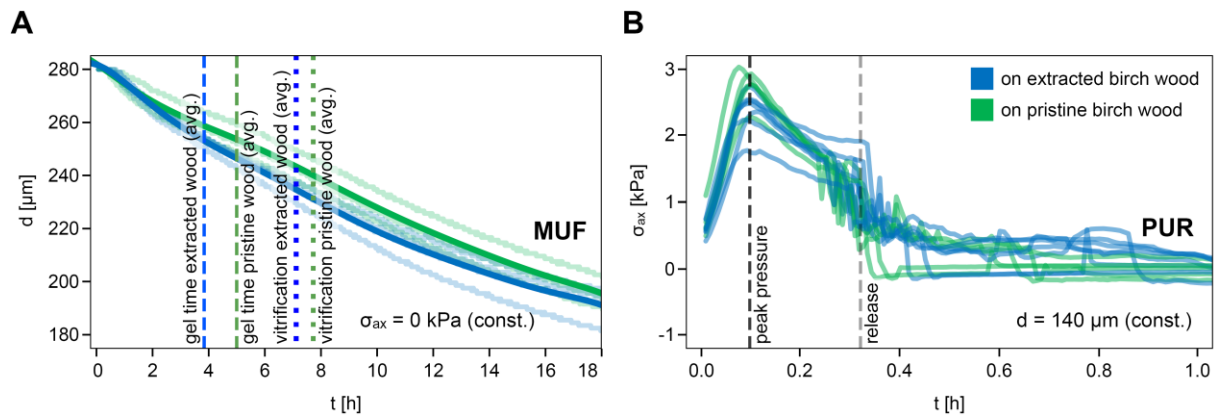


Figure SI–20: A: Progression of the rheometer gap size (adhesive layer thickness, d) throughout the MUF curing experiments on pristine wood (in green, $n = 4$) and extracted wood (blue, $n = 4$). The thick lines are average curves, and the light curves show the original experimental data. B: Progression of the axial pressure in the initial phase of PUR curing experiments due to CO_2 development on pristine wood (in green, $n = 5$) and extracted wood (blue, $n = 7$).

Results and Discussion – Mechanical Characteristics of Cured Adhesive Films

The stress-strain curves of the performed uniaxial tensile stress-strain (UTSS) measurements on MUF (A and C) and PUR (B and D) adhesive curves with varying concentrations of extractives are shown in Figure SI–21. The color of the respective curves is mapped to extractives concentration as shown as scales in (A) for MUF (0 to 1.0% (W/W)) and in (B) for PUR (0 to 0.2% (W/W)).

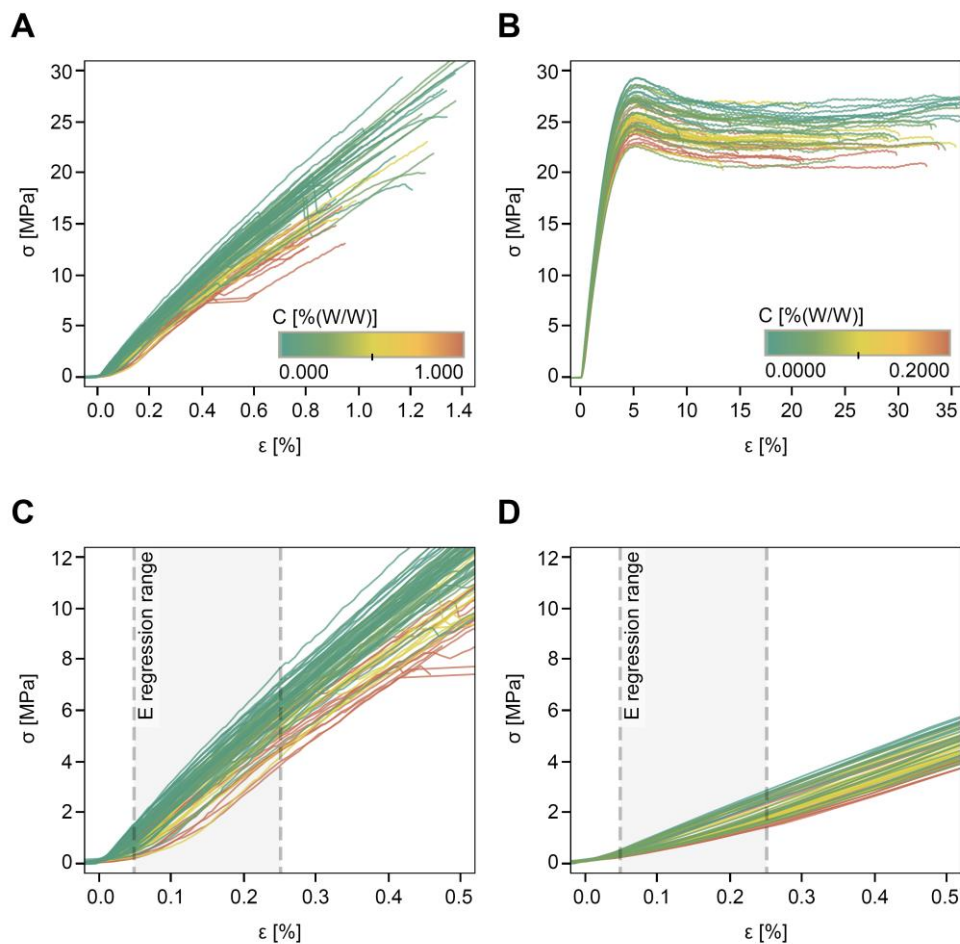


Figure SI–21: Uniaxial tensile stress-strain (UTSS) curves for MUF (A and C) and PUR (B and D) adhesive films. A and B show the complete curves, while C and D show low strain range with the moduli of elasticity regression interval ($0.05 < \epsilon < 0.25$, as defined in ISO 527–1) in grey. The color of the respective curves is mapped to extractives concentration as shown as scales for (A) MUF and (B) PUR.

The glass transition temperatures T_g as given by TMA measurement and the mid-point evaluation method are shown in Figure SI–22. A polynomial fit with 95% confidence band is shown as a guide to the eyes.

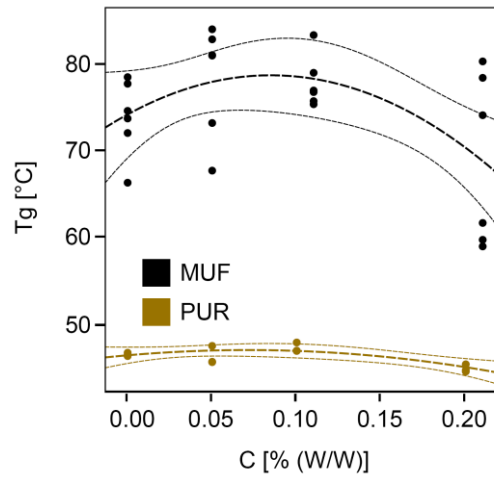


Figure SI–22: Glass transition temperature T_g determined by the mid-point method for adhesive films with increased extractives concentration. Dashed lines show the 2nd-degree polynomial fit with 95% confidence band as a guide to the eyes.

The temperature- and frequency-sweep performed via Dynamic Mechanical Analysis (DMA) on PUR and MUF are shown in Figure SI–23 below. The frequency-sweep was performed at equilibrium climate conditions of 0%-RH, 65%-RH and 90%-RH. Although the MUF is expected to have a higher sorptive moisture content at the respective RH, the effects on the storage modulus E' are smaller than in the case of PUR, since the highly cross-linked it less plasticizable.

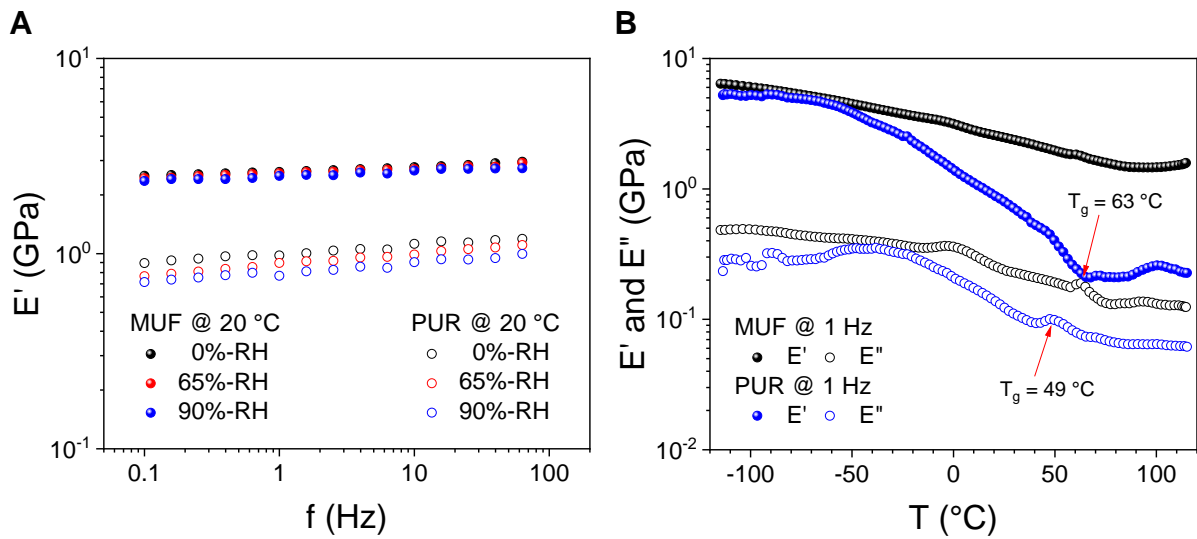


Figure SI–23: Dynamic Mechanical Analysis (DMA) experiments on MUF and PUR adhesive films: A) frequency-sweep experiments at 20 °C and different RH-values; and B) temperature-sweep experiments at 1 Hz and 0%-RH. The glass transition temperature (T_g) is indicated by the peak in the E'' profile.

Results and Discussion – Tensile Shear Strength Experiments

The statistics of the TSS results, the average change in TSS comparing extracted specimens to pristine wood specimens, and the p-value of ANOVA are shown in Table SI–6.

Table SI–6: Tensile shear strength (TSS) measurements using pristine and extracted birch wood: result statistics.

specimen group: adhesive and treatment		tensile shear strength (TSS) average \pm standard deviation [MPa]		rel. change in avg. TSS [%]	ANOVA: p-value
		pristine wood	extracted wood		
PUR	A1	12.5 \pm 2.8	11.3 \pm 1.7	+11%	0.13
	A2	7 \pm 2.4	6.8 \pm 1.7	+3%	0.80
	A4	6.2 \pm 2.1	6.3 \pm 1.0	-2%	0.81
	A5	11.7 \pm 2.1	11.4 \pm 2.0	+2%	0.72
MUF	A1	10.7 \pm 1.7	9.6 \pm 1.0	+11%	0.026
	A2	8.2 \pm 1.5	6.7 \pm 1.3	+22%	0.003
	A4	7.4 \pm 1.7	6.7 \pm 1.0	+11%	0.13
	A5	12 \pm 2.2	10.7 \pm 1.3	+12%	0.049