

Towards greener stone shot and stone wool materials: Binder systems based on gelatine modified with tannin or transglutaminase

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Mixing of binder compositions comprising IMAGEL® RL modified with chestnut tree tannin in the presence of sodium hydroxide

Using procedures analogous to the representative example detailed above, the following binder compositions were mixed:

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® RL	100	100	100	100	100	100	100
Chestnut tree tannin (%-wt. of gelatine)	0	3	5	10	20	30	50
Mixing of binder composition							
IMAGEL® RL (g)	12.00	12.00	12.00	12.00	12.00	11.00	9.50
Water (g)	63.95	64.92	65.42	67.47	71.33	68.71	65.3
1M NaOH (g)	5.67	5.67	5.67	5.67	5.67	5.20	4.60
pH	9.1	9.1	9.1	9.1	9.1	9.1	9.1
Chestnut tree tannin solution (g)	0	1.62	2.70	5.40	10.80	14.85	21.38
pH	-	9.1	9.1	9.0	9.1	9.1	9.1
Calculated binder composition							
Binder concentration (%-wt. in water)	15.0	15.0	15.0	15.0	15.0	15.0	14.0
IMAGEL® RL (%-wt. of gelatine/ %-wt. of all non-water components)	100.0/ 98.2	100.0/ 95.0	100.0/ 93.0	100.0/ 88.4	100.0/ 80.3	100.0/ 73.6	100.0/ 63.1
Chestnut tree tannin (%-wt. of gelatine/ %-wt. of all non-water components)	0/ 0	3.0/ 2.9	5.0/ 4.7	10.0/ 8.8	20.0/ 16.1	30.0/ 22.1	50.0/ 31.5
NaOH (%-wt. of gelatine/ %-wt. of all non-water components)	1.8/ 1.8	2.2/ 2.1	2.5/ 2.3	3.2/ 2.8	4.5/ 3.6	5.9/ 4.3	8.6/ 5.4

Mixing of binder compositions comprising IMAGEL® LA modified with chestnut tree tannin in the presence of sodium hydroxide

Using procedures analogous to the representative example detailed above, the following binder compositions were mixed:

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® LA	100	100	100	100	100	100	100
Chestnut tree tannin (%-wt. of gelatine)	0	3	5	10	20	30	50
Mixing of binder composition							
IMAGEL® LA (g)	14.00	12.00	12.00	12.00	12.00	11.00	9.50
Water (g)	75.17	65.92	66.43	68.54	71.92	69.29	65.90
1M NaOH (g)	5.16	4.65	4.67	5.00	4.75	4.29	3.97
pH	9.1	9.1	9.1	9.0	9.0	9.0	9.0
Chestnut tree tannin solution (g)	0	1.62	2.70	5.40	10.80	14.85	21.38
pH	-	9.0	9.0	9.1	9.2	9.3	9.2
Calculated binder composition							
Binder concentration (%-wt. in water)	15.1	15.0	15.0	14.9	15.0	15.0	14.9
IMAGEL® LA (%-wt. of gelatine/ %-wt. of all non-water components)	100.0/ 98.6	100.0/ 95.3	100.0/ 93.3	100.0/ 88.5	100.0/ 80.5	100.0/ 73.8	100.0/ 63.2
Chestnut tree tannin (%-wt. of gelatine/ %-wt. of all non-water components)	0/ 0	3.0/ 2.9	5.0/ 4.7	10.0/ 8.9	20.0/ 16.1	30.0/ 22.1	50.0/ 31.6
NaOH (%-wt. of gelatine/ %-wt. of all non-water components)	1.4/ 1.4	1.9/ 1.8	2.2/ 2.0	3.0/ 2.6	4.2/ 3.4	5.5/ 4.1	8.3/ 5.3

Mixing of binder compositions comprising IMAGEL® RA modified with chestnut tree tannin in the presence of sodium hydroxide

Using procedures analogous to the representative example detailed above, the following binder compositions were mixed:

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® RA	100	100	100	100	100	100	100
Chestnut tree tannin (%-wt. of gelatine)	0	3	5	10	20	30	50
Mixing of binder composition							
IMAGEL® RA (g)	12.00	12.00	12.00	12.00	12.00	11.00	9.50
Water (g)	65.42	66.43	66.95	68.54	72.51	69.88	65.90
1M NaOH (g)	3.81	3.81	3.81	3.81	3.81	3.73	3.13
pH	9.1	9.3	9.2	9.3	9.2	9.0	9.0
Chestnut tree tannin solution (g)	0	1.62	2.70	5.40	10.80	14.85	21.38
pH	-	9.3	9.2	9.3	9.3	9.2	9.1
Calculated binder composition							
Binder concentration (%-wt. in water)	15.0	15.0	15.0	15.1	15.0	15.0	15.0
IMAGEL® RA (%-wt. of gelatine/ %-wt. of all non-water components)	100.0/ 98.8	100.0/ 95.6	100.0/ 93.5	100.0/ 88.8	100.0/ 80.7	100.0/ 73.9	100.0/ 63.3
Chestnut tree tannin (%-wt. of gelatine/ %-wt. of all non-water components)	0/ 0	3.0/ 2.9	5.0/ 4.7	10.0/ 8.9	20.0/ 16.1	30.0/ 22.2	50.0/ 31.6
NaOH (%-wt. of gelatine/ %-wt. of all non-water components)	1.2/ 1.2	1.6/ 1.6	1.9/ 1.8	2.6/ 2.3	3.9/ 3.2	5.3/ 4.0	8.0/ 5.1

Mixing of binder compositions comprising IMAGEL® AP modified with chestnut tree tannin in the presence of sodium hydroxide

Using procedures analogous to the representative example detailed above, the following binder compositions were mixed:

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® AP	100	100	100	100	100	100	100
Chestnut tree tannin (%-wt. of gelatine)	0	3	5	10	20	30	50
Mixing of binder composition							
IMAGEL® AP (g)	12.00	12.00	12.00	12.00	12.00	11.00	9.50
Water (g)	65.42	66.95	67.47	69.08	73.11	70.48	66.50
1M NaOH (g)	3.14	3.14	3.14	3.14	3.14	2.87	2.49
pH	9.3	9.2	9.3	9.3	9.2	9.2	9.3
Chestnut tree tannin solution (g)	0	1.62	2.70	5.40	10.80	14.85	21.38
pH	-	9.2	9.3	9.3	9.3	9.3	9.3
Calculated binder composition							
Binder concentration (%-wt. in water)	15.0	15.0	15.0	15.0	15.0	15.0	15.0
IMAGEL® AP (%-wt. of gelatine/ %-wt. of all non-water components)	100.0/ 99.0	100.0/ 95.8	100.0/ 93.7	100.0/ 89.0	100.0/ 80.8	100.0/ 74.0	100.0/ 63.4
Chestnut tree tannin (%-wt. of gelatine/ %-wt. of all non-water components)	0/ 0	3.0/ 2.9	5.0/ 4.7	10.0/ 8.9	20.0/ 16.2	30.0/ 22.2	50.0/ 31.7
NaOH (%-wt. of gelatine/ %-wt. of all non-water components)	1.0/ 1.0	1.4/ 1.4	1.7/ 1.6	2.4/ 2.1	3.7/ 3.0	5.0/ 3.7	7.7/ 4.9

Mixing of binder compositions comprising IMAGEL® RA modified with TI transglutaminase

Using procedures analogous to the representative example detailed above, the following binder compositions were mixed:

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® RA	100	100	100	100	100	100	100
TI transglutaminase (%-wt. of gelatine)	0	3	5	10	20	30	50
Mixing of binder composition							
IMAGEL® RA (g)	12.00	14.00	13.00	12.50	12.00	11.00	9.50
Water for gelatine (g)	68.00	77.51	70.85	65.42	57.60	48.03	33.25
TI transglutaminase (g)	0	0.42	0.65	1.25	2.40	3.30	4.75
Water for transglutaminase (g)	0	4.20	6.50	12.50	24.00	33.00	47.50
pH	5.0	5.1	5.1	5.0	5.0	5.0	5.1
Calculated binder composition							
Binder concentration (%-wt. in water)	15.0	15.0	15.0	15.0	15.0	15.0	15.0
IMAGEL® RA (%-wt. of gelatine/ %-wt. of all non-water components)	100.0/ 100.0	100.0/ 97.1	100.0/ 95.2	100.0/ 90.9	100.0/ 83.3	100.0/ 76.9	100.0/ 66.7
TI transglutaminase (%-wt. of gelatine/ %-wt. of all non-water components)	0/ 0	3.0/ 2.9	5.0/ 4.8	10.0/ 9.1	20.0/ 16.7	30.0/ 23.1	50.0/ 33.3

Mixing of binder compositions comprising IMAGEL® AP modified with TI transglutaminase

Using procedures analogous to the representative example detailed above, the following binder compositions were mixed:

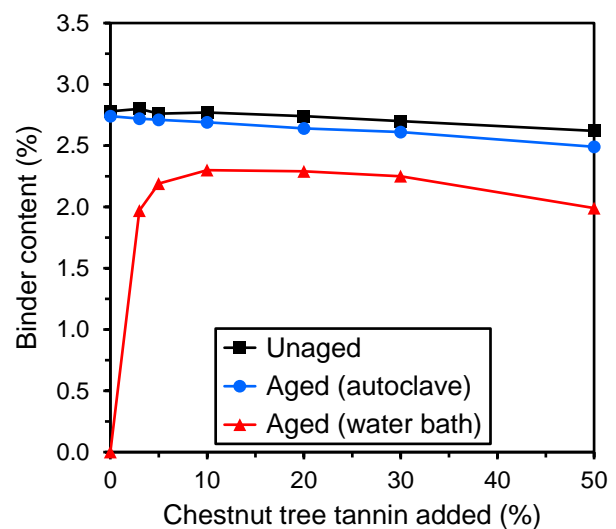
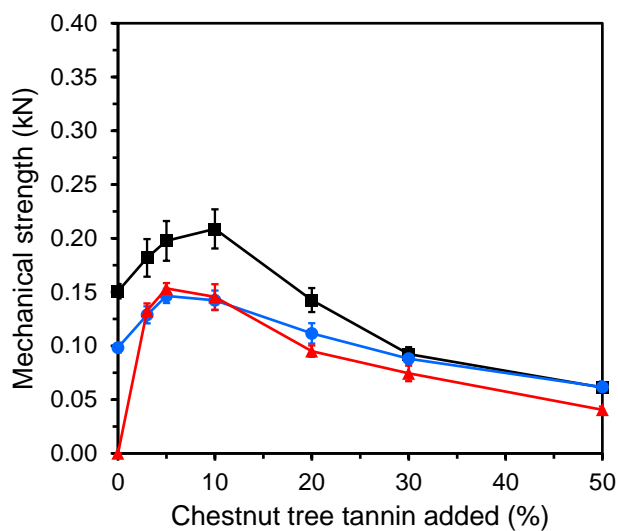
Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® AP	100	100	100	100	100	100	100
TI transglutaminase (%-wt. of gelatine)	0	3	5	10	20	30	50
Mixing of binder composition							
IMAGEL® AP (g)	12.00	14.00	13.00	12.50	12.00	11.00	9.50
Water for gelatine (g)	68.00	77.51	70.85	65.42	57.60	48.03	33.25
TI transglutaminase (g)	0	0.42	0.65	1.25	2.40	3.30	4.75
Water for transglutaminase (g)	0	4.20	6.50	12.50	24.00	33.00	47.50
pH	4.6	4.7	4.7	4.7	4.5	4.8	4.8
Calculated binder composition							
Binder concentration (%-wt. in water)	15.0	15.0	15.0	15.0	15.0	15.0	15.0
IMAGEL® AP (%-wt. of gelatine/ %-wt. of all non-water components)	100.0/ 100.0	100.0/ 97.1	100.0/ 95.2	100.0/ 90.9	100.0/ 83.3	100.0/ 76.9	100.0/ 66.7
TI transglutaminase (%-wt. of gelatine/ %-wt. of all non-water components)	0/ 0	3.0/ 2.9	5.0/ 4.8	10.0/ 9.1	20.0/ 16.7	30.0/ 23.1	50.0/ 33.3

Results for binder compositions comprising IMAGEL® RL modified with chestnut tree tannin in the presence of sodium hydroxide

The table below lists the results obtained for binder compositions comprising IMAGEL® RL modified with chestnut tree tannin in the presence of sodium hydroxide.

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® RL	100	100	100	100	100	100	100
Chestnut tree tannin (%-wt. of gelatine)	0	3	5	10	20	30	50
Composite bar results							
Avg. unaged mechanical strength (kN, n = 5)	0.151	0.182	0.198	0.209	0.143	0.092	0.061
Standard error (kN)	0.007	0.018	0.019	0.018	0.011	0.006	0.004
Binder content (%-wt of stone shots)	2.78	2.80	2.76	2.77	2.74	2.70	2.62
<hr/>							
Avg. autoclave aged mechanical strength (kN, n = 5)	0.098	0.129	0.146	0.142	0.112	0.088	0.062
Standard error (kN)	0.005	0.008	0.007	0.009	0.009	0.005	0.005
Binder content (%-wt of stone shots)	2.74	2.72	2.71	2.69	2.64	2.61	2.49
<hr/>							
Avg. water bath aged mechanical strength (kN, n = 5)	0	0.133	0.153	0.145	0.095	0.074	0.041
Standard error (kN)	-	0.007	0.005	0.012	0.005	0.007	0.003
Binder content (%-wt of stone shots)	0	1.97	2.19	2.30	2.29	2.25	1.99

These data are shown in the figures below.

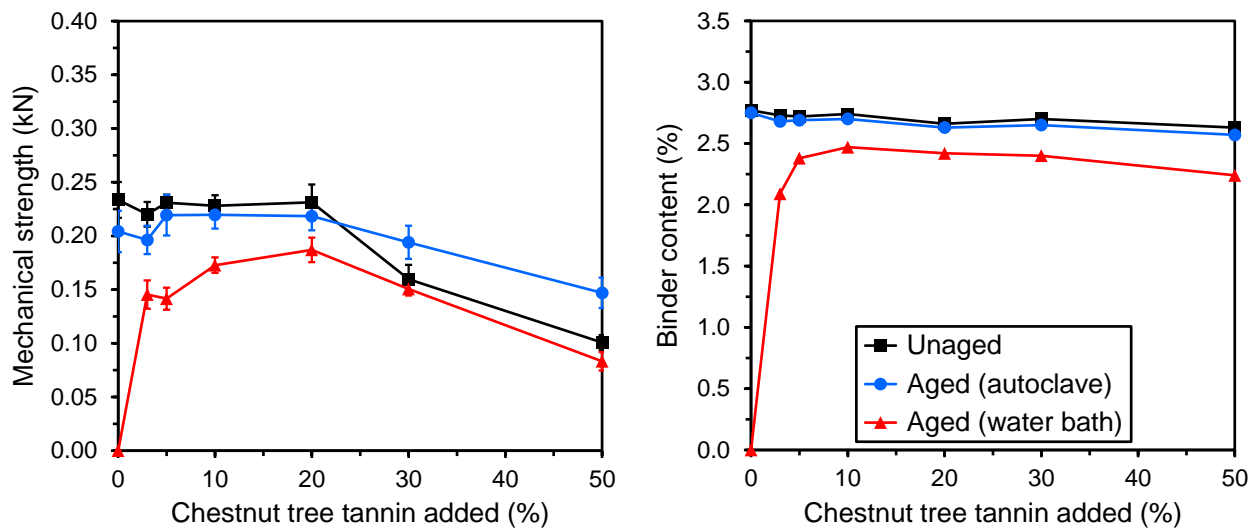


Results for binder compositions comprising IMAGEL® LA modified with chestnut tree tannin in the presence of sodium hydroxide

The table below lists the results obtained for binder compositions comprising IMAGEL® LA modified with chestnut tree tannin in the presence of sodium hydroxide.

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® LA	100	100	100	100	100	100	100
Chestnut tree tannin (%-wt. of gelatine)	0	3	5	10	20	30	50
Composite bar results							
Avg. unaged mechanical strength (kN, n = 5)	0.223	0.220	0.231	0.228	0.231	0.160	0.101
Standard error (kN)	0.017	0.011	0.007	0.010	0.016	0.014	0.007
Binder content (%-wt of stone shots)	2.77	2.73	2.72	2.74	2.66	2.70	2.63
Avg. autoclave aged mechanical strength (kN, n = 5)	0.204	0.196	0.219	0.220	0.218	0.194	0.147
Standard error (kN)	0.019	0.013	0.019	0.013	0.013	0.015	0.014
Binder content (%-wt of stone shots)	2.75	2.68	2.69	2.70	2.63	2.65	2.57
Avg. water bath aged mechanical strength (kN, n = 5)	0	0.145	0.142	0.173	0.187	0.151	0.083
Standard error (kN)	-	0.013	0.010	0.007	0.011	0.006	0.009
Binder content (%-wt of stone shots)	0	2.09	2.38	2.47	2.42	2.40	2.24

These data are shown in the figures below.

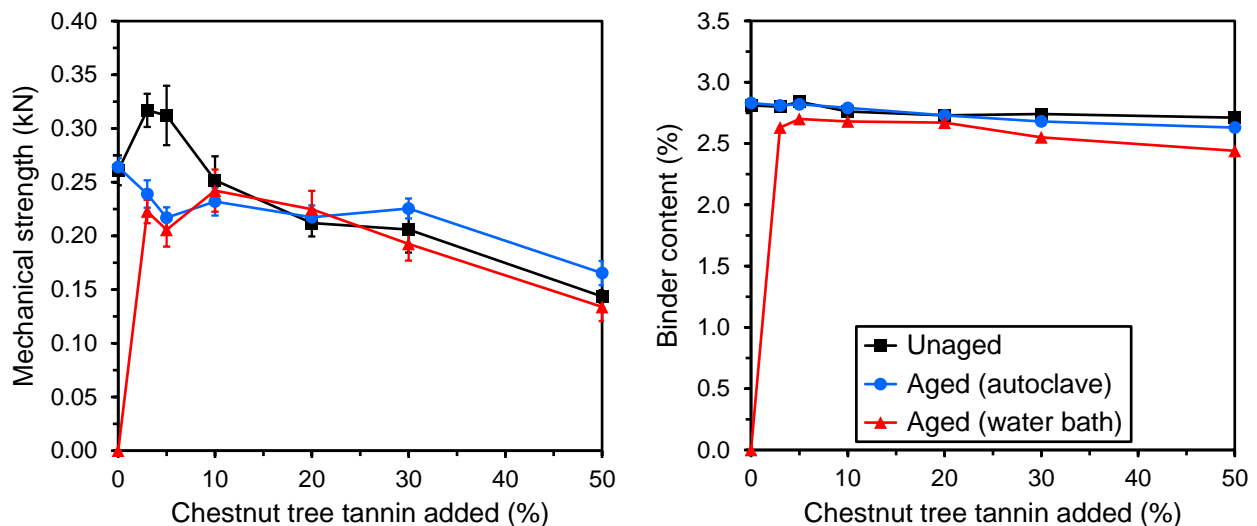


Results for binder compositions comprising IMAGEL® RA modified with chestnut tree tannin in the presence of sodium hydroxide

The table below lists the results obtained for binder compositions comprising IMAGEL® RA modified with chestnut tree tannin in the presence of sodium hydroxide.

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® RA	100	100	100	100	100	100	100
Chestnut tree tannin (%-wt. of gelatine)	0	3	5	10	20	30	50
Composite bar results							
Avg. unaged mechanical strength (kN, n = 5)	0.261	0.317	0.312	0.252	0.212	0.206	0.144
Standard error (kN)	0.014	0.015	0.028	0.022	0.013	0.021	0.006
Binder content (%-wt of stone shots)	2.81	2.80	2.84	2.76	2.73	2.74	2.71
Avg. autoclave aged mechanical strength (kN, n = 5)	0.264	0.239	0.217	0.232	0.217	0.225	0.165
Standard error (kN)	0.008	0.013	0.010	0.013	0.011	0.009	0.011
Binder content (%-wt of stone shots)	2.83	2.81	2.82	2.79	2.73	2.68	2.63
Avg. water bath aged mechanical strength (kN, n = 5)	0	0.223	0.206	0.242	0.225	0.193	0.134
Standard error (kN)	-	0.011	0.016	0.020	0.017	0.016	0.013
Binder content (%-wt of stone shots)	0	2.63	2.70	2.68	2.67	2.55	2.44

These data are shown in the figures below.

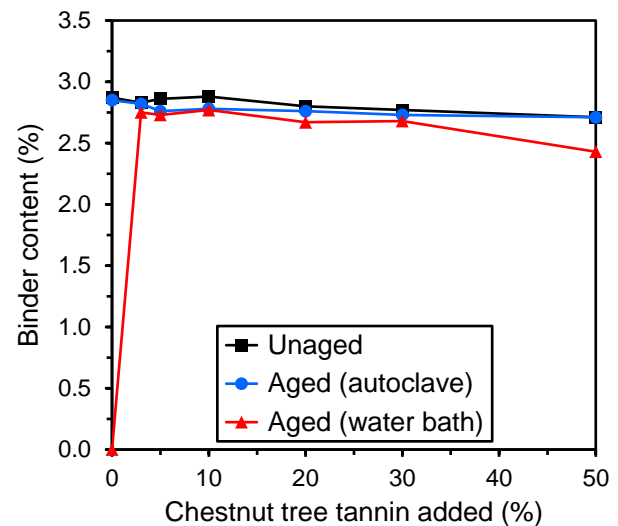
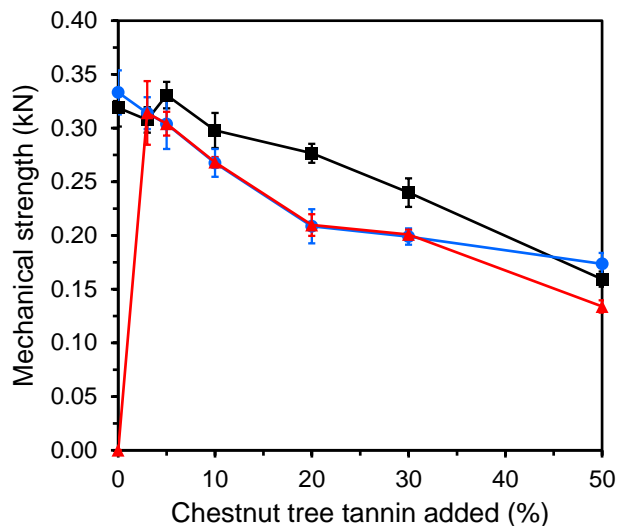


Results for binder compositions comprising IMAGEL® AP modified with chestnut tree tannin in the presence of sodium hydroxide

The table below lists the results obtained for binder compositions comprising IMAGEL® AP modified with chestnut tree tannin in the presence of sodium hydroxide.

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® AP	100	100	100	100	100	100	100
Chestnut tree tannin (%-wt. of gelatine)	0	3	5	10	20	30	50
Composite bar results							
Avg. unaged mechanical strength (kN, n = 5)	0.319	0.308	0.331	0.298	0.277	0.240	0.159
Standard error (kN)	0.018	0.012	0.012	0.016	0.009	0.013	0.007
Binder content (%-wt of stone shots)	2.87	2.83	2.86	2.88	2.80	2.77	2.71
Avg. autoclave aged mechanical strength (kN, n = 5)	0.333	0.314	0.304	0.267	0.209	0.199	0.174
Standard error (kN)	0.021	0.015	0.023	0.013	0.016	0.008	0.010
Binder content (%-wt of stone shots)	2.85	2.82	2.76	2.78	2.76	2.73	2.71
Avg. water bath aged mechanical strength (kN, n = 5)	0	0.314	0.304	0.268	0.210	0.201	0.134
Standard error (kN)	-	0.030	0.011	0.005	0.010	0.004	0.006
Binder content (%-wt of stone shots)	0	2.75	2.73	2.77	2.67	2.68	2.43

These data are shown in the figures below.

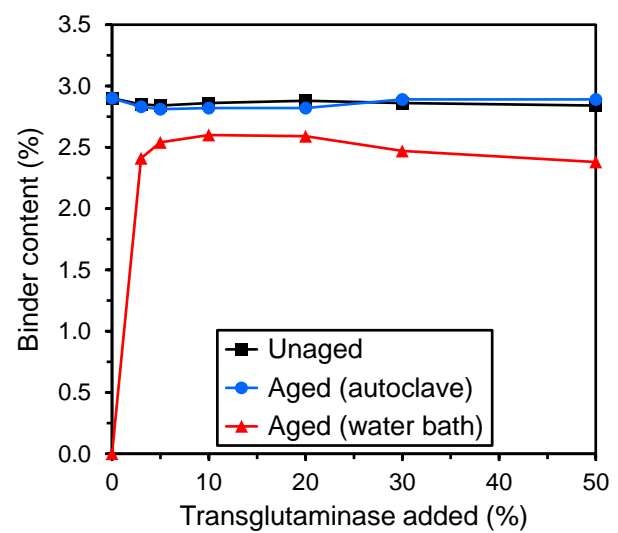
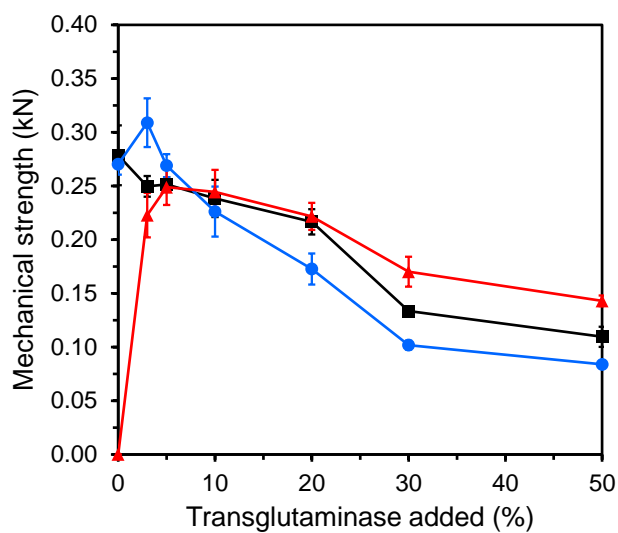


Results for binder compositions comprising IMAGEL® RA modified with transglutaminase

The table below lists the results obtained for binder compositions comprising IMAGEL® RA modified with TI transglutaminase.

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® RA	100	100	100	100	100	100	100
TI transglutaminase (%-wt. of gelatine)	0	3	5	10	20	30	50
Composite bar results							
Avg. unaged mechanical strength (kN, n = 5)	0.279	0.250	0.251	0.238	0.217	0.134	0.110
Standard error (kN)	0.028	0.010	0.007	0.017	0.012	0.003	0.009
Binder content (%-wt of stone shots)	2.90	2.85	2.84	2.86	2.88	2.86	2.84
Avg. autoclave aged mechanical strength (kN, n = 5)	0.270	0.309	0.269	0.226	0.173	0.102	0.084
Standard error (kN)	0.010	0.023	0.011	0.023	0.015	0.004	0.002
Binder content (%-wt of stone shots)	2.90	2.83	2.81	2.82	2.82	2.89	2.89
Avg. water bath aged mechanical strength (kN, n = 5)	0	0.223	0.249	0.244	0.222	0.170	0.143
Standard error (kN)	-	0.021	0.017	0.021	0.013	0.014	0.005
Binder content (%-wt of stone shots)	0	2.41	2.54	2.60	2.59	2.47	2.38

These data are shown in the figures below.

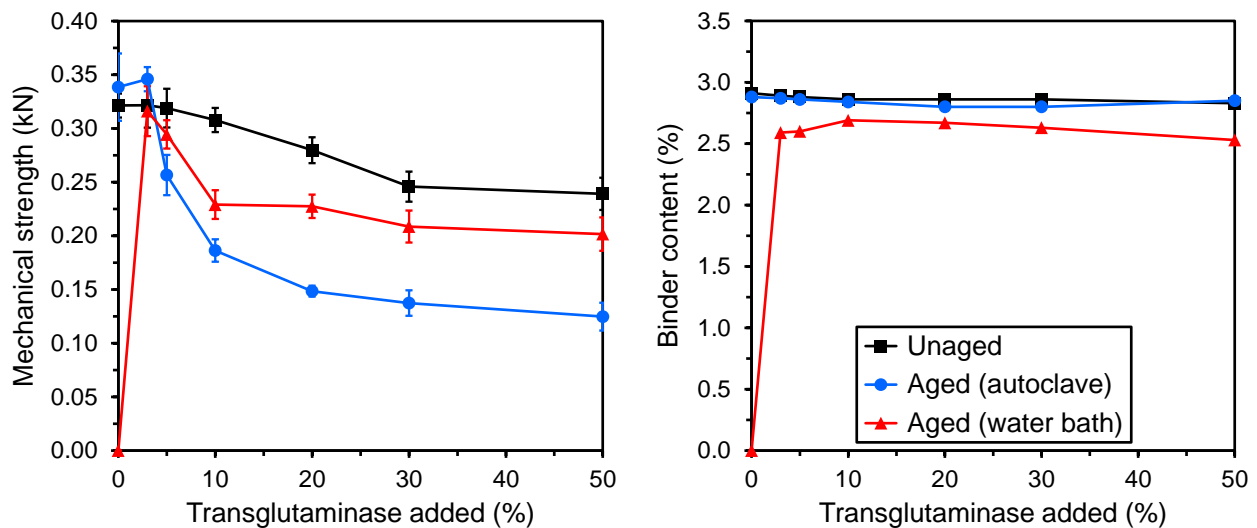


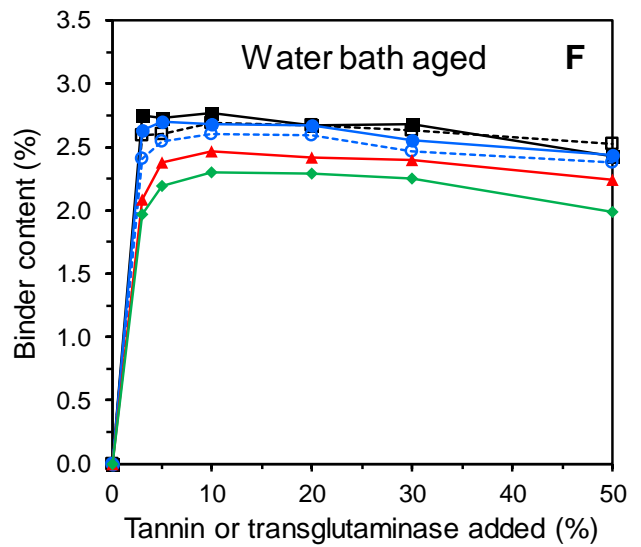
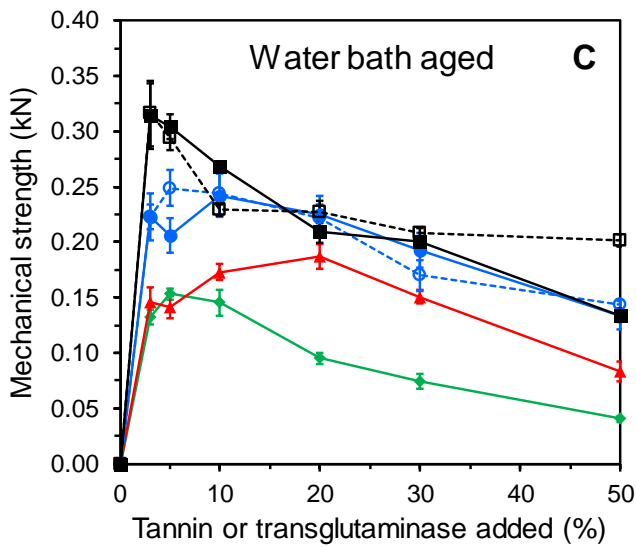
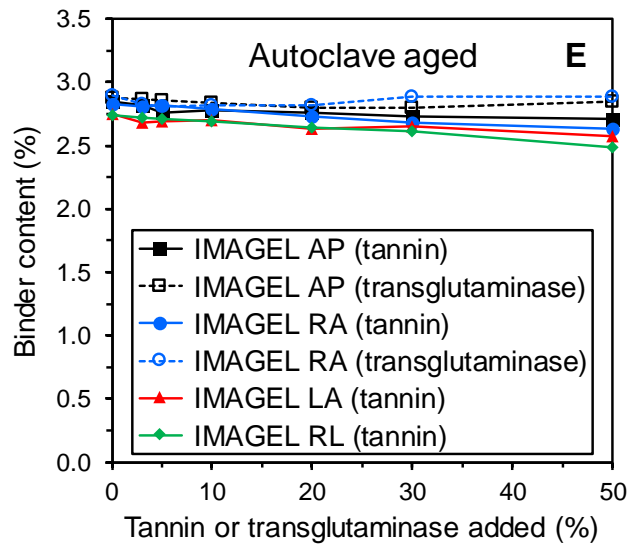
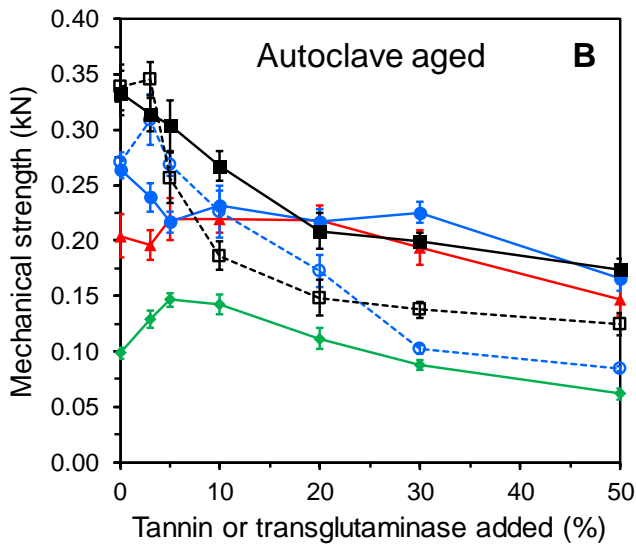
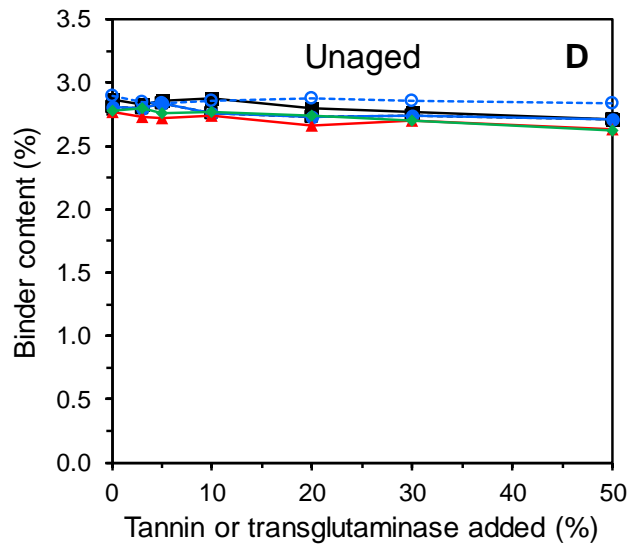
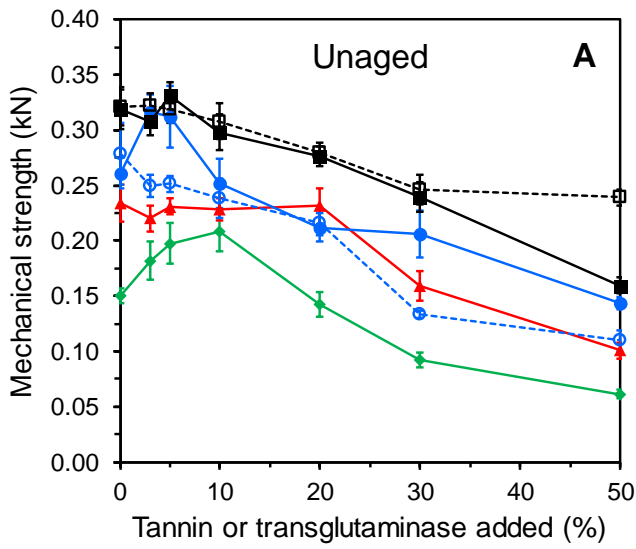
Results for binder compositions comprising IMAGEL® AP modified with transglutaminase

The table below lists the results obtained for binder compositions comprising IMAGEL® AP modified with TI transglutaminase.

Entry	1	2	3	4	5	6	7
Binder composition							
IMAGEL® AP	100	100	100	100	100	100	100
TI transglutaminase (%-wt. of gelatine)	0	3	5	10	20	30	50
Composite bar results							
Avg. unaged mechanical strength (kN, n = 5)	0.321	0.322	0.319	0.308	0.280	0.246	0.239
Standard error (kN)	0.011	0.021	0.018	0.011	0.012	0.014	0.015
Binder content (%-wt of stone shots)	2.91	2.89	2.88	2.86	2.86	2.86	2.83
Avg. autoclave aged mechanical strength (kN, n = 5)	0.338	0.346	0.257	0.186	0.148	0.137	0.125
Standard error (kN)	0.031	0.011	0.019	0.010	0.005	0.012	0.013
Binder content (%-wt of stone shots)	2.88	2.87	2.86	2.84	2.80	2.80	2.85
Avg. water bath aged mechanical strength (kN, n = 5)	0	0.316	0.294	0.229	0.227	0.209	0.202
Standard error (kN)	-	0.023	0.013	0.013	0.011	0.015	0.016
Binder content (%-wt of stone shots)	0	2.59	2.60	2.69	2.67	2.63	2.53

These data are shown in the figures below.





Large versions of Figure 3A-C

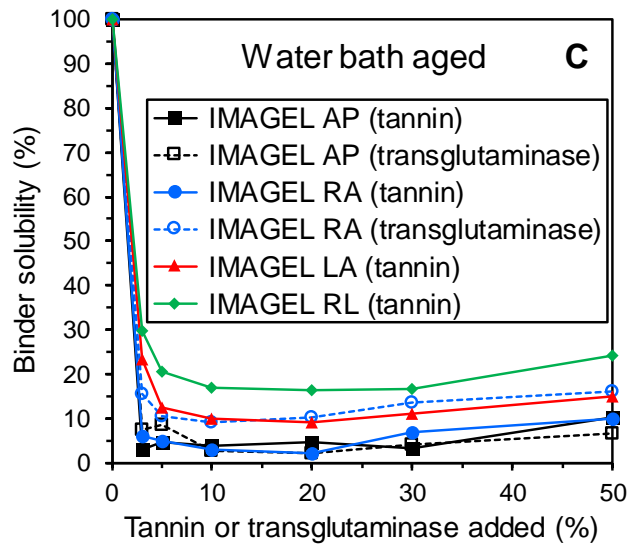
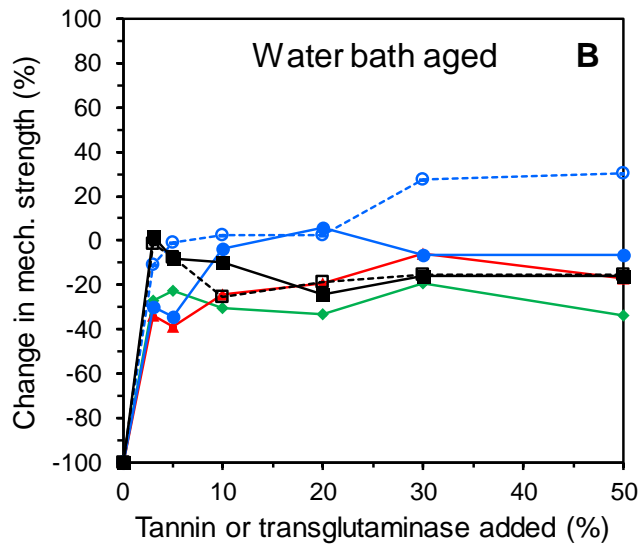
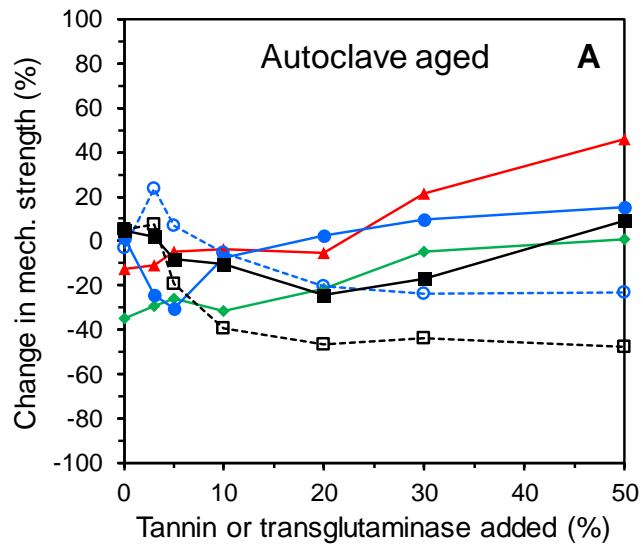


Figure 4, top left: SEM image with view field of 4.15 mm of break surface of composite bar produced using IMAGEL® LA modified with chestnut tree tannin and sodium hydroxide.

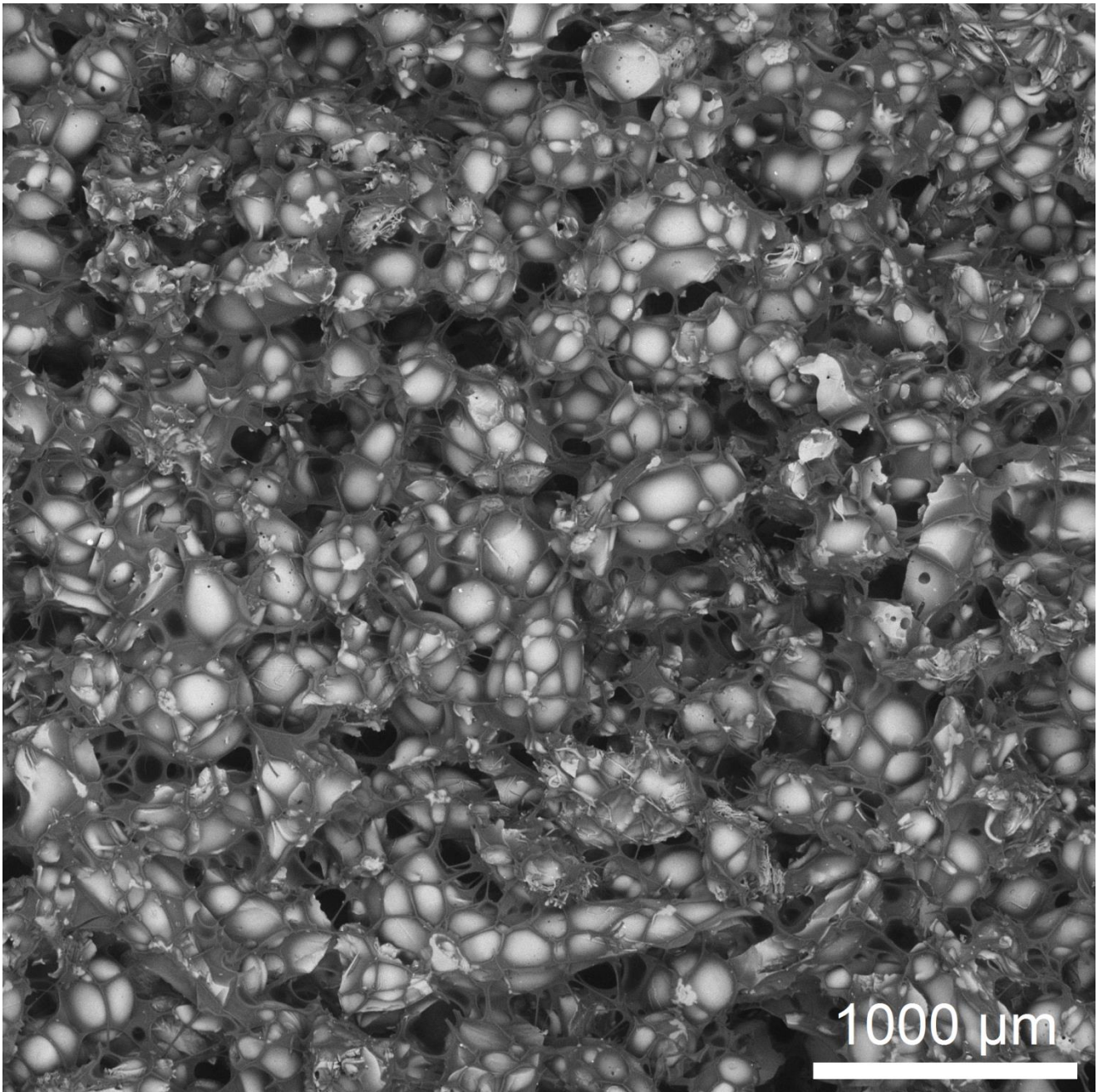


Figure 4, top right: SEM image with view field of 4.15 mm of break surface of composite bar produced using IMAGEL® RA modified with transglutaminase.

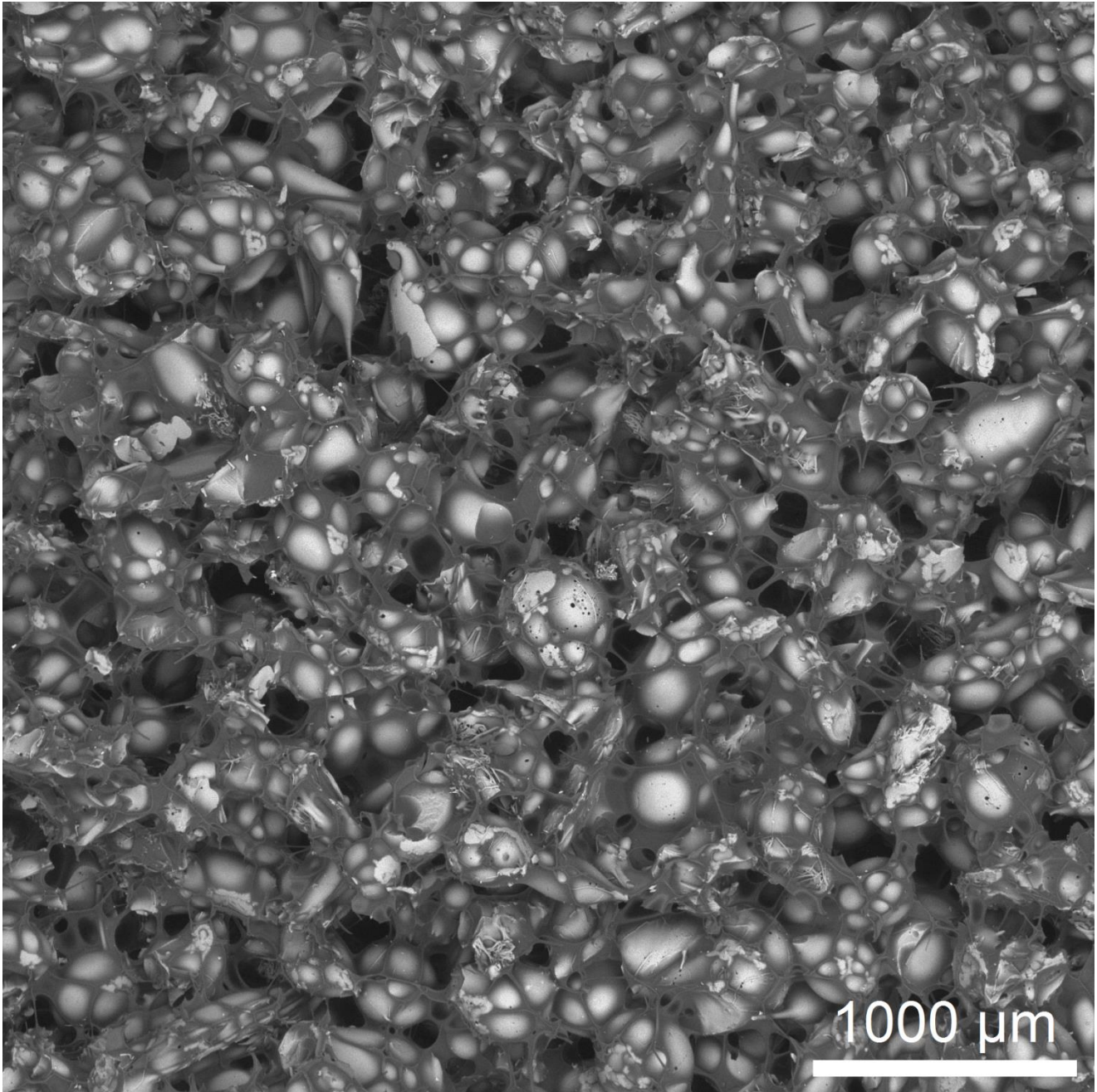


Figure 4, bottom left: SEM image with view field of 4.15 mm of break surface of composite bar produced using a conventional phenol-urea-formaldehyde binder.

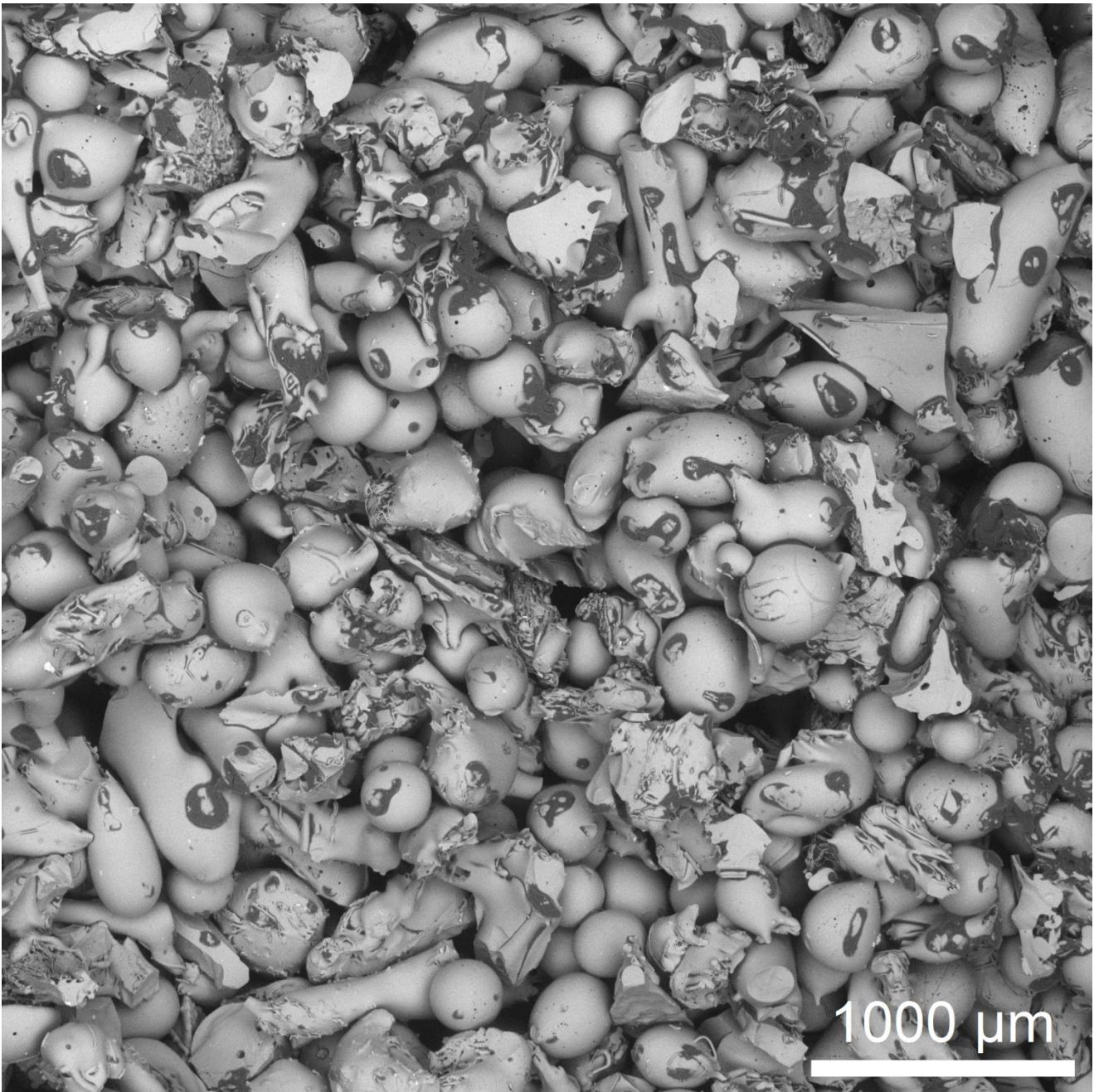


Figure 4, bottom right: SEM image with view field of 4.15 mm of binder-free stone shots.

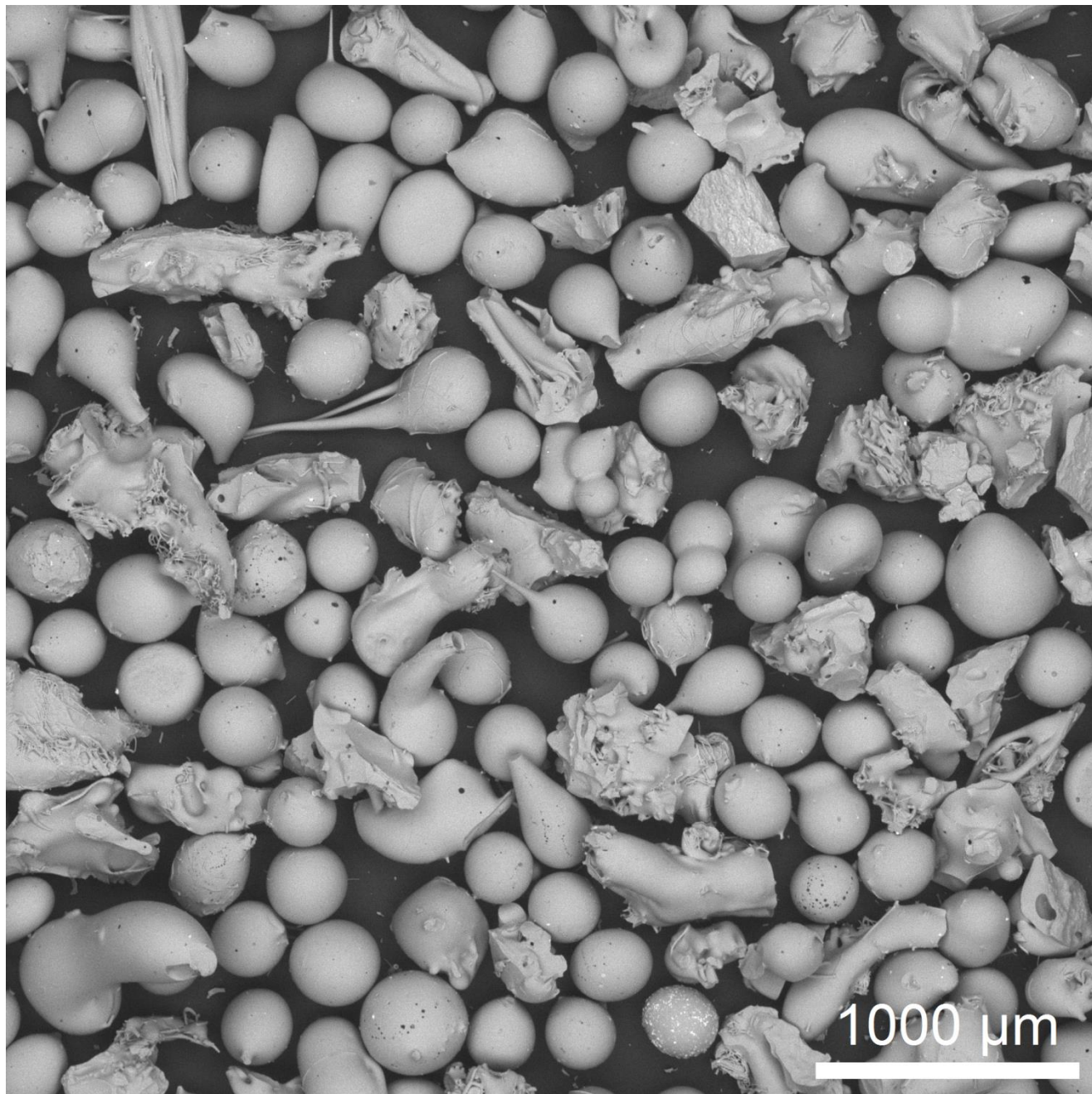


Figure 5, left: SEM image with view field of 1.38 mm of break surface of composite bar produced using IMAGEL® LA modified with chestnut tree tannin and sodium hydroxide.

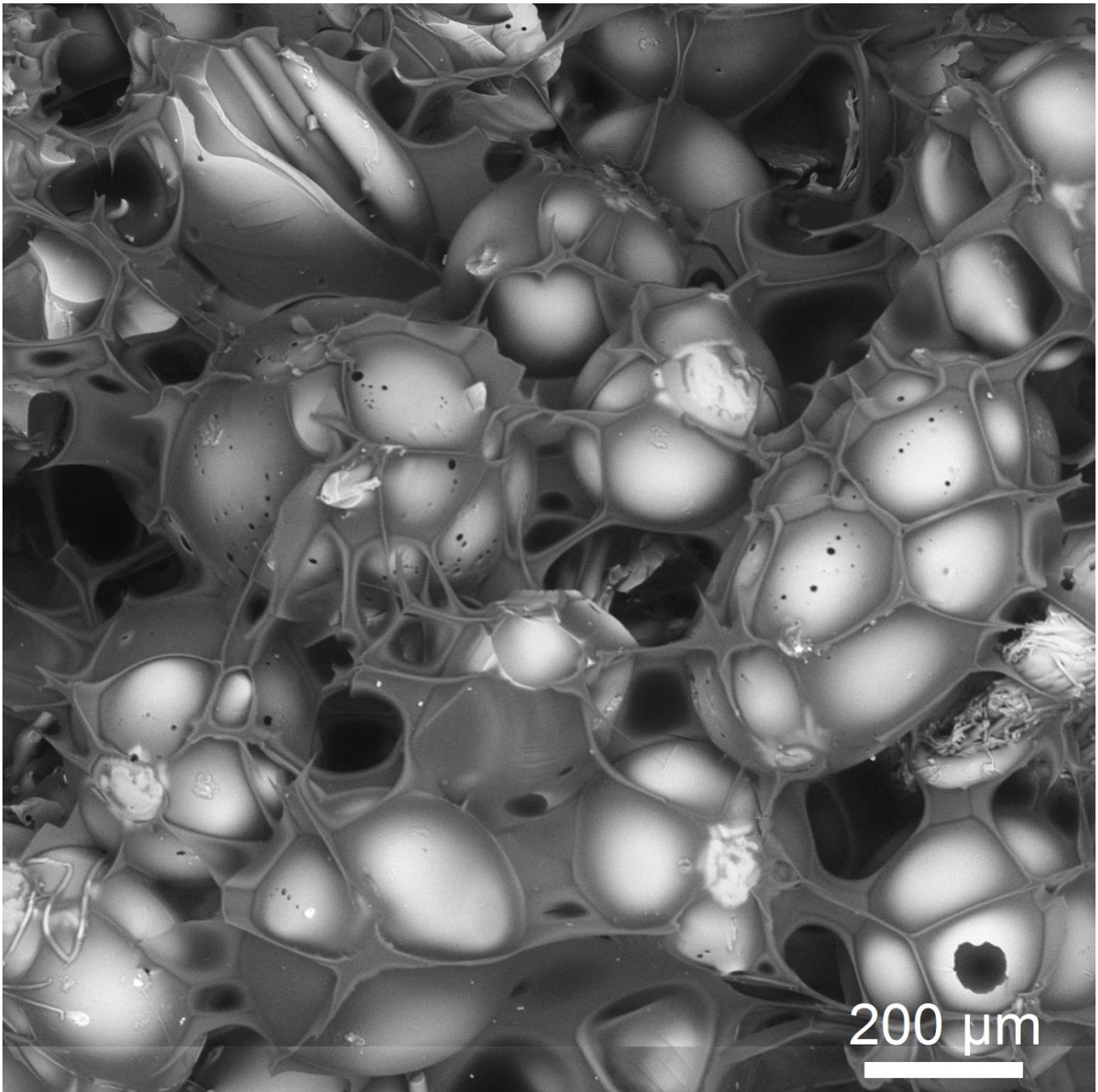


Figure 5, right: SEM image with view field of 1.38 mm of break surface of composite bar produced using IMAGEL® RA modified with transglutaminase.

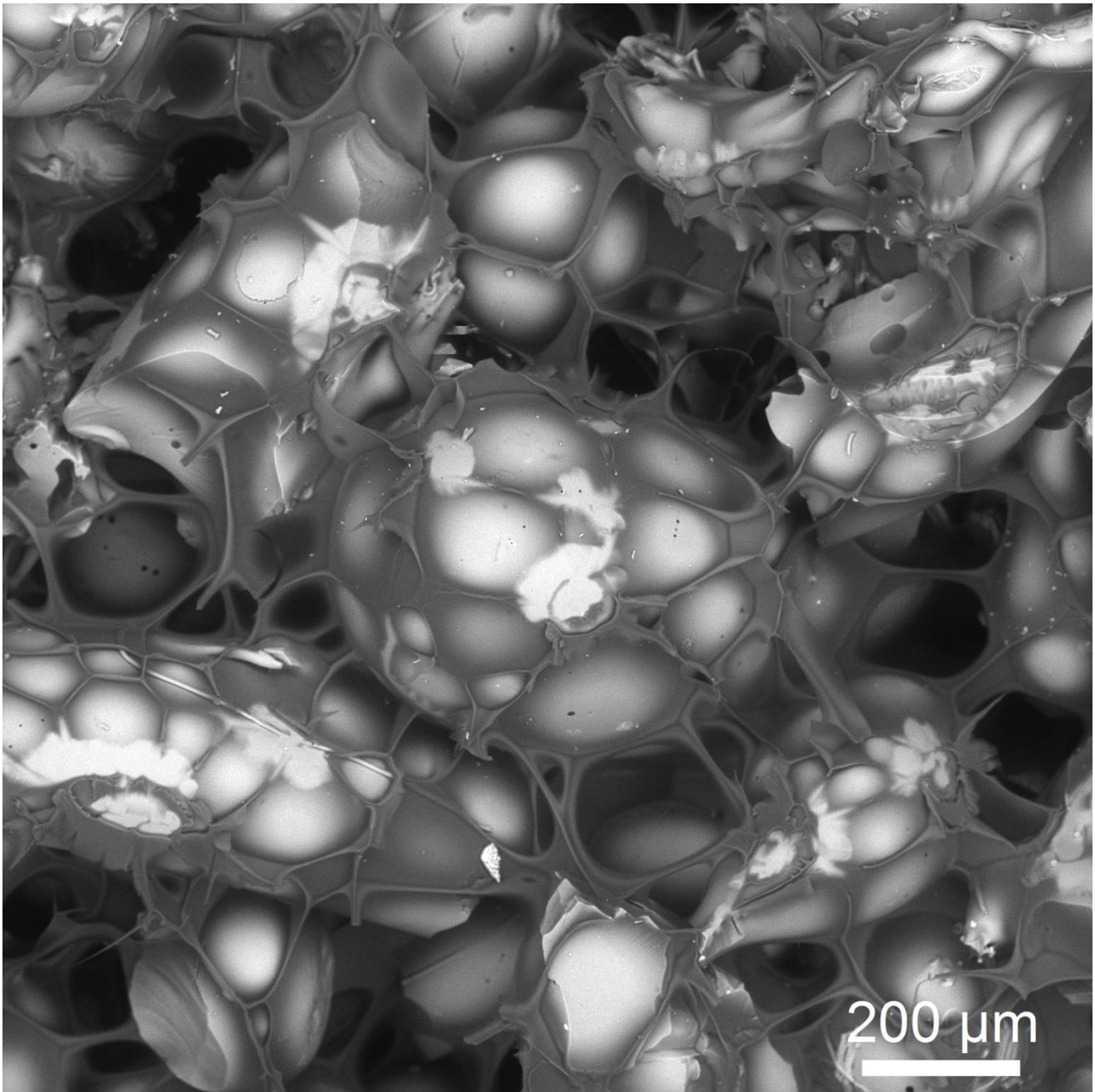


Table of contents entry, right: SEM image with view field of 0.93 mm of gold coated break surface of composite bar produced using IMAGEL® LA modified with chestnut tree tannin and sodium hydroxide.

