Supplementary information

S1 Synthesis of three primary colored amphoteric polymers

(1) Synthesis of red amphoteric polymer (AAD-R)

Firstly, 20 g of AAD was dissolved in 80 mL of deionized water. To activate the reaction system, 2.3 g of EDC (12 mmol) and 0.7 g of NHS (6 mmol) were added with slow stirring for 30 minutes. Then, 2 g of activated Red 180 (5.4 mmol) was introduced, and the reaction mixture was heated to 70℃ for 24 hours. The reaction mixture was recrystallized in acetone three times, and the resulting precipitate was collected and dried in a vacuum oven at 60℃ for 24 hours. The final product, a red powder named AAD-R, was obtained with a yield of 92.6%.

(2) Synthesis of blue amphoteric polymer (AAD-B)

In a similar procedure, 20 g of AAD was dissolved in 80 mL of deionized water. Then, 0.95 g of EDC (8 mmol) and 0.46 g of NHS (4 mmol) were added and stirred slowly for 30 minutes to activate the reaction system. Subsequently, 2 g of activated Blue 19 (3.3 mmol) was added. The reaction conditions and purification methods were identical to those used for AAD-R. The final product, a blue powder named AAD-B, was obtained with a yield of 90.3%.

(3) Synthesis of yellow amphoteric polymer (AAD-Y)

For the yellow polymer, 20 g of AAD was dissolved in 80 mL of anhydrous ethanol. To activate the reaction system, 2.6 g of EDC (22 mmol) and 1.3 g of NHS (11 mmol) were added with slow stirring for 30 minutes. Then, 3 g of naphthylimide derivatives (DMENA, 10.6 mmol) was added. The reaction conditions and purification process followed the same protocol as used for AAD-R. The final product, a yellow powder named AAD-Y, was obtained with a yield of 91.5%.

Based on the three primary colored amphoteric polymers (AAD-R, AAD-B, and AAD-Y), integrated amphoteric polymers with various colors were prepared: Orange

(AAD-O): Mixing AAD-R and AAD-Y in a ratio of 1:3 yielded the orange amphoteric material named AAD-O; Green (AAD-G): Mixing AAD-Y and AAD-B in a ratio of 1:3 yielded the green amphoteric material named AAD-G. Cyan (AAD-C): Mixing AAD-Y and AAD-B in a ratio of 3:1 yielded the cyan amphoteric material named AAD-C. Purple (AAD-P): Mixing AAD-R and AAD-B in a ratio of 1:1 yielded the purple amphoteric material named AAD-P.

AAD, AAD-R, AAD-O, AAD-Y, AAD-G, AAD-C, AAD-B, AAD-P are collectively referred to as AADs.

(4) **The application of AADs in leather manufacturing**

The AADs were utilized in the tanning-wet finishing processes of pickled sheepskins (Tab. S1). These were compared with samples of leathers tanned and wet finished using commercia chrome-free tanning agents, specifically AT, F-90, and TWS (Tab. S2-S5). To minimize errors due to variations in different parts of the leather, all samples were taken symmetrically along the dorsal spine of the same pickled sheepskin. Additionally, waste liquids from each process were collected during the experiment for subsequent environmental data analysis.

(5) Production of anti-counterfeiting patterns

To produce anti-counterfeiting patterns, the integrated material is mixed with either a temperature-sensitive dye or a light-sensitive dye in a 1:1 ratio. The anti-counterfeiting patterns, shaped like “ginkgo biloba leaves” and “clovers”, are created using a vacuum filtration device. The leather specimen was placed with the flesh side upwards, and the mould was placed on the top of the leather specimen, the mixed dyes are added slowly into the mould, and the vacuum is drawn for 1 minute, then the leather specimen is placed in the oven for 10 minutes at 60°C. Additional patterns such as “SUST”, barcodes, and QR codes were created by brushing the mixed dyes onto the leather surface.

Tab. S1 The tanning-wet finishing process of integration amphoteric polymers

Process	Chemicals	Offer/%	T/°C	t/min	Remarks
Rewetting	Water	150	25	20	Drain
	Sodium chloride	7			
Tanning-Wet finishing	Water	50	25	300	Adjust the pH of the bath liquid to 6.5 ±
	Sodium chloride	3			
	Triethylamine	0.5			
	"Integration" amphoteric polymers	12			
	Sodium bicarbonate	5	40	180	Slowly adjust pH of the bath liquid to 8.5 ± and stop mechanical action overnight

Tab. S2 Tanning process of AT tanning agents

Process	Chemicals	Offer/%	T/°C	t/min	Remarks
Basification	Water	200	30	120	Sodium carbonate was added twice at an interval of 15 min. The pH of the system was stable at 8.5 ~ 9.0. Drain.
	Sodium chloride	10			
	Sodium carbonate	0.2			
Tanning	Water	100	25	360	pH=8.5~9.0
	AT	6			
	Water	100	45	120	Stop the drum overnight, turn 60 min the next day, remove the drum and stand still.

Tab. S3 Tanning process of F-90 tanning agents

Process	Chemicals	Offer/%	T/°C	t/min	Remarks
Depickling	Water	150	25	3×15+20	Adjust pH to 7.0
	Sodium bicarbonate	0.2			
	Sodium chloride	10			
Tanning	F-90	10	35	90	Drain water.
	Water	40			

	Water	30	40	150	
	Water	30	45	180	

Tab. S4 Tanning process of TWS tanning agents

Process	Chemicals	Offer/%	T/℃	t/min	Remarks
Depickling	Water	150	25	3×15+20	Adjust pH to 5.5.
	Sodium bicarbonate	0.2			
	Sodium chloride	10			
Tanning	Water	200	25	300	
	TWS	10			
	Sodium bicarbonate	1.0	40	180	Adjust pH to 8.0. Drain water.

Tab. S5 Wet finishing process of AT, F-90 and TWS tanned leather

Process	Chemicals	Offer/%	T/℃	t/min	Remarks
Rewetting	Water	150	40	60	Drain water
	Degreasing agent	1			
Washing	Water	250	37	15	
Neutralizing	Water	100	40	40	Adjust pH to 5.5 or 7.5.
	Sodium bicarbonate	1		3×15+30	
	Formic acid	0.2			
Fatliquoring	Water	150	45	60	
	Anionic fatliquor	18			
Dyeing	Water	150	45	60	
	Anionic dyes (TRUPOCOR YELLOW BB)	2			
	Formic acid	0.2		3×15+30	Adjust pH to 3.5. Drain water.

S2 LCA impacts of the AADs compare to SACG and EHBPs

The life cycle inventory (LCI) data of the upstream producytion of chemicals, electricity, steam, and water were collected from the Ecoinvent database and literature. The LCI data for SACG and EHBPs were estimated based on the literature *J. Clean. Prod., 2020, 270: 122351* and *Green Chem., 2021, 23, 5924*.

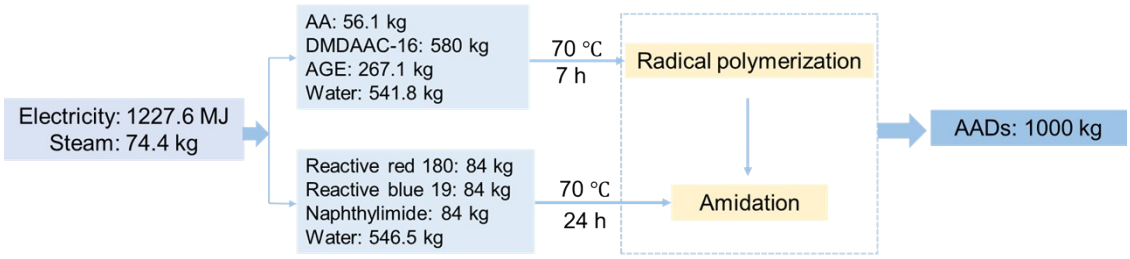


Fig. S1 Flow sheet of AADs preparation process.

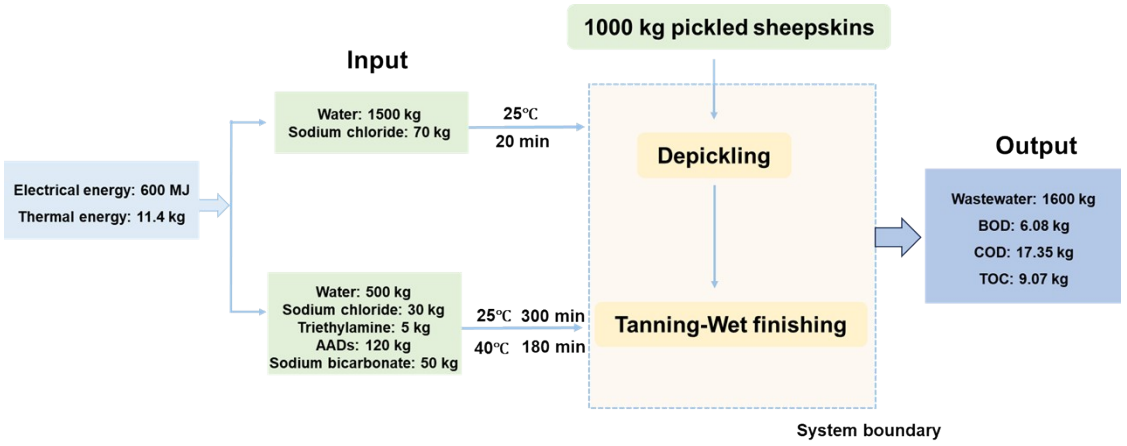


Fig. S2 Flow sheet of AADs tanning-wet finishing system.

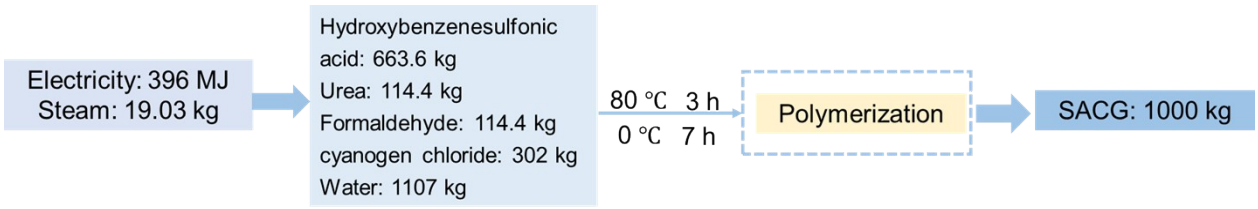


Fig. S3 Flow sheet of SACG reparation process [1].

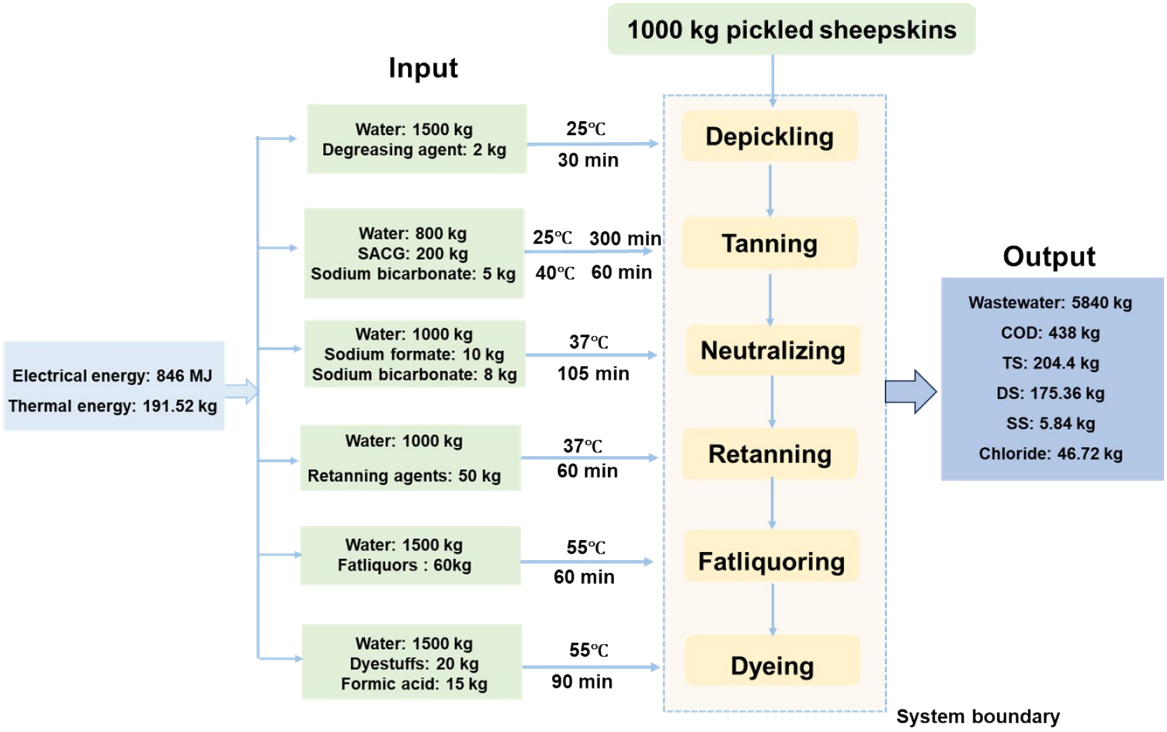


Fig. S4 Flow sheet of SACG tanning and wet-finishing system [1].

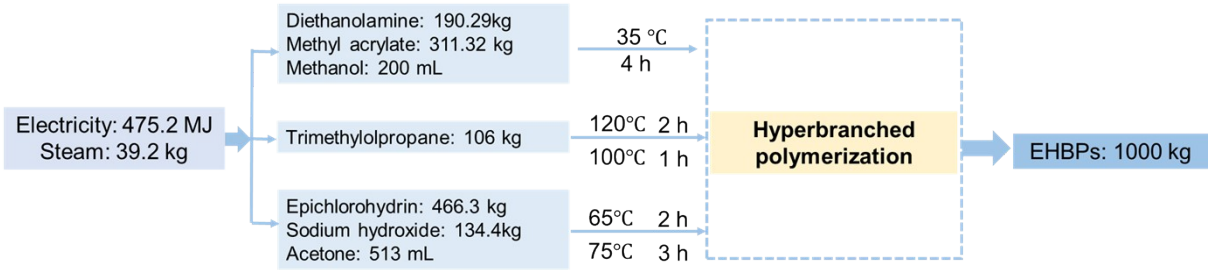


Fig. S5 Flow sheet of EHBPs preparation process [2].

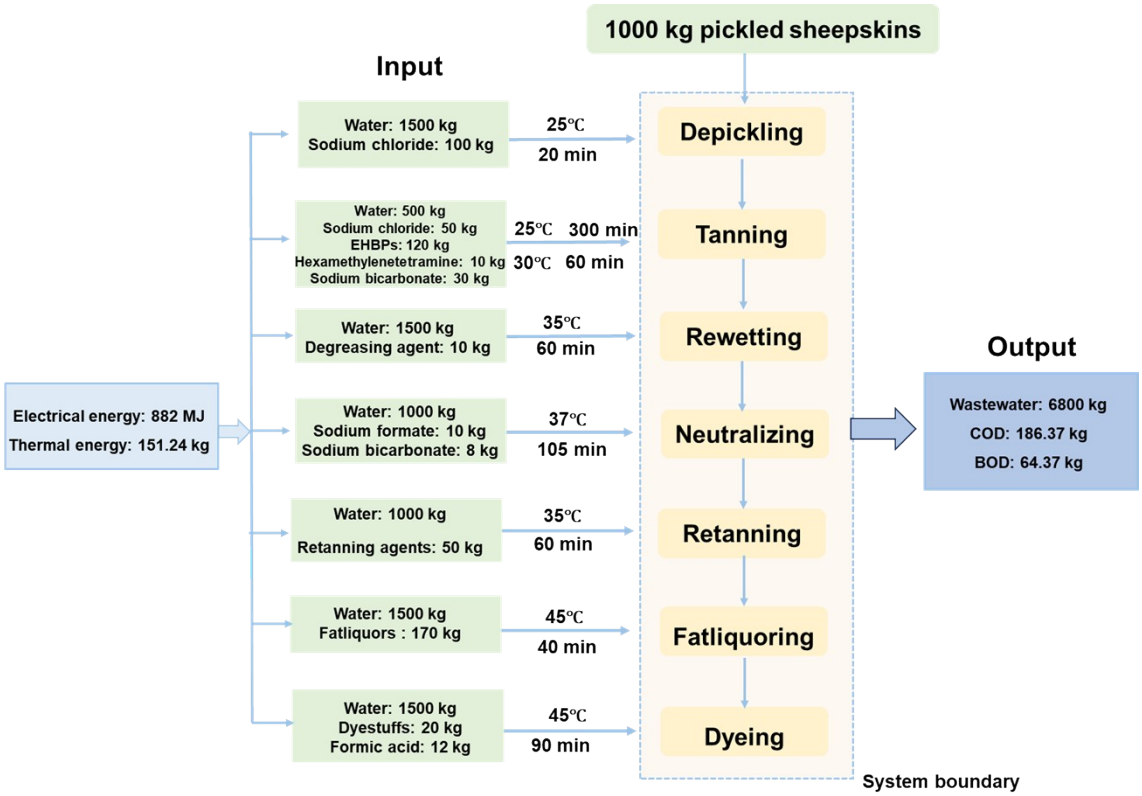


Fig. S6 Flow sheet of EHBPs tanning and wet-finishing system [2].

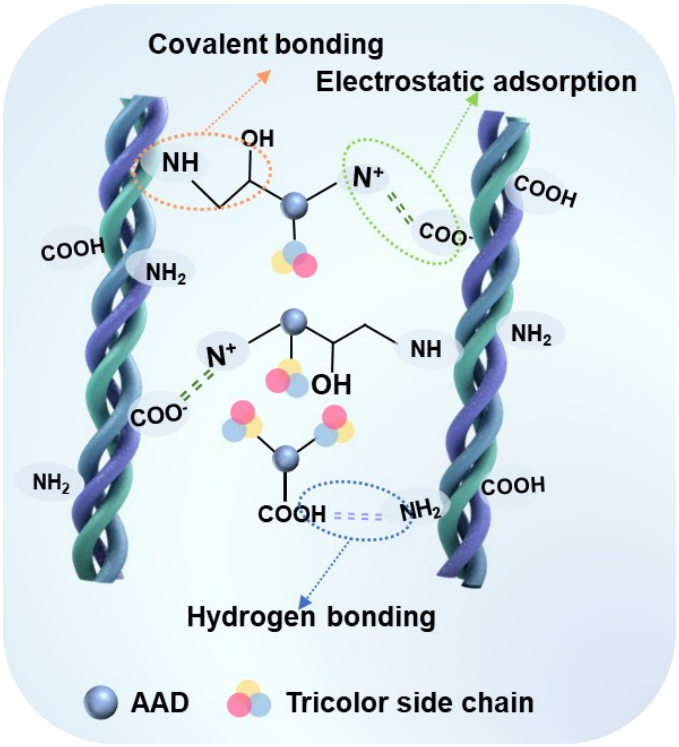


Fig. S7 Schematic diagram of AADs tanning mechanism.

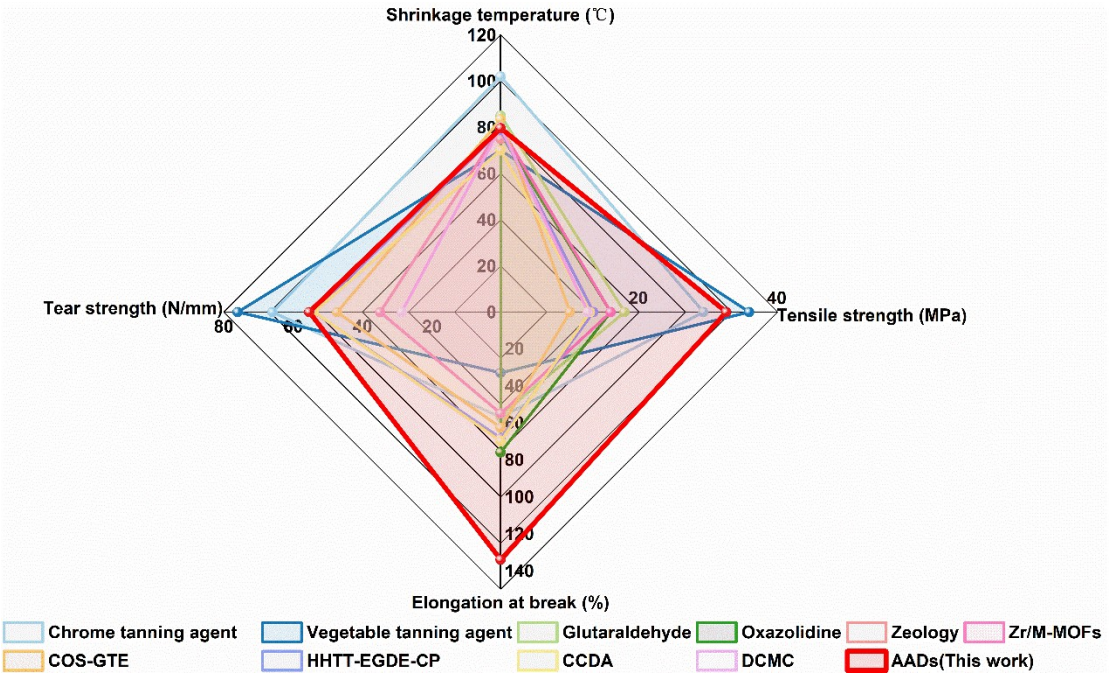


Fig. S8 Comparison of thermal stability and mechanical properties between integrated polymer treated leather and typical tanning agent tanned leather

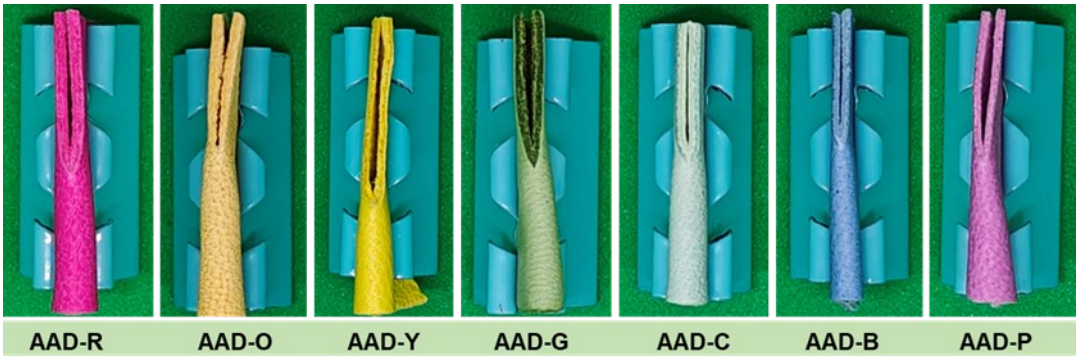
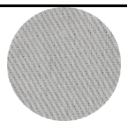
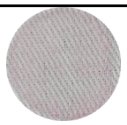
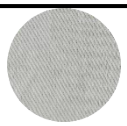
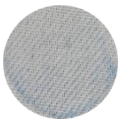


Fig. S9 Penetration of polymeric dyes AADs in leather

Tab. S6 Surface color of cotton cloth after dry and wet rubbing in AADs dyed leather.

Samples	Dry-rubbing	Wet-rubbing
AAD-R		
AAD-O		

AAD-Y		
AAD-G		
AAD-C		
AAD-B		
AAD-P		

Tab. S7 Surface color of cotton cloth after dry and wet rubbing in anionic dyed leather.

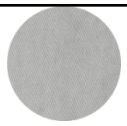
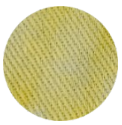
Samples	**Dry-rubbing**	**Wet-rubbing**
TWS		
F-90		
AT		

Fig. S10 Color Changes on the surface of AAD-dyed leather after 1 to 5 wash cycles.

Tab. S8 Evaluation of wash fastness ratings for AAD-dyed leather

Samples	Wash 1 time	Wash 2 times	Wash 3 times	Wash 4 times	Wash 5 times
AAD-Y	Grade 5	Grade 5	Grade 5	Grade 5	Grade 5
AAD-R	Grade 5	Grade 4~5	Grade 5	Grade 5	Grade 4~5
AAD-B	Grade 5	Grade 5	Grade 5	Grade 4~5	Grade 5

Tab. S9 Chemical usage, water consumption and processing time of AADs and traditional tanning-wet finishing processes

	Chemicals usage (%)	**Water consumption (%)**	**Processing time (min)**	**Ref**
Zirconium-aluminium-titanium combination tanning agent	88.9	1400	1330	[3]
Chrome tanning agent	45.9	1150	2360	[4]
Glutaraldehyde tanning agent	42.7	2000	1095	[5]
Polymer chrome-free tanning agent	64.8	1330	1235	[6]
Integration tanning agent (This work)	22.4	200	500	-

Tab. S10 TOC content of baths before and after tanning-wet finishing for different tanning processes.

Samples	TOC (mg/L)		Absorption rate (%)
	Initial	Residual	
AAD	25424	3125	87.71
AAD-R	27574	4055	85.29
AAD-Y	29234	4796	83.59
AAD-B	31254	5028	83.91
TWS	54231	18865	65.21
F-90	51259	24452	52.29
AT	60257	16528	72.57

Tab. S11 BOD_5, COD_{Cr}, and BOD/COD content of baths tanning-wet finishing for different tanning processes.

Samples	BOD_5 (mg/L)	COD_{Cr} (mg/L)	BOD_5/COD
AAD	2058	5415	0.38
AAD-R	4190	12326	0.33
AAD-Y	2660	7714	0.34
AAD-B	2510	7076	0.35
TWS	6480	23258	0.28
F-90	7580	32849	0.23
AT	6870	23404	0.29

Tab. S12 Characteristic Values of Various Impact Categories of AADs

Category	Unit	Depickling	Tanning	Rewetting	Neutralizing	Retanning	Fatliquoring	Dyeing	Total
PED	MJ	3.64E+02	1.53E+04	0	0	0	0	0	1.56E+04
ADP	kg Sb eq.	5.90E-04	5.38E-03	0	0	0	0	0	5.97E-03
WU	kg	1.14E+01	5.80E+02	0	0	0	0	0	5.92E+02
GWP	kg CO_2 eq.	2.70E+01	8.55E+02	0	0	0	0	0	8.82E+02
AP	kg SO_2 eq.	1.47E-01	4.43E+00	0	0	0	0	0	4.58E+00
EP	kg PO_4^{3-} eq.	5.54E-02	1.74E+00	0	0	0	0	0	1.80E+00
ET	CTUe	2.25E+02	1.23E+04	0	0	0	0	0	1.25E+04
HTC	CTUh	2.39E-08	4.58E-07	0	0	0	0	0	4.82E-07
HTNC	CTUh	7.09E-07	1.28E-05	0	0	0	0	0	1.35E-05

Tab. S13 Characteristic Values of Various Impact Categories of SACG

Category	Unit	Depickling	Tanning	Rewetting	Neutralizing	Retanning	Fatliquoring	Dyeing	Total
PED	MJ	3.35E+02	1.66E+04	0.00E+00	1.13E+03	2.50E+03	3.71E+03	4.69E+03	2.90E+04
ADP	kg Sb eq.	6.52E-05	5.32E-03	0.00E+00	4.96E-04	1.04E-03	1.55E-03	1.78E-03	1.03E-02
WU	kg	1.80E+01	4.73E+02	0.00E+00	3.91E+01	1.22E+02	2.02E+02	2.12E+02	1.07E+03
GWP	kg CO_2 eq.	1.97E+01	7.70E+02	0.00E+00	7.97E+01	1.32E+02	1.41E+02	2.82E+02	1.43E+03
AP	kg SO_2 eq.	8.02E-02	3.71E+00	0.00E+00	3.96E-01	5.60E-01	1.50E+00	1.36E+00	7.61E+00
EP	kg PO_4^{3-} eq.	5.46E-02	1.19E+00	0.00E+00	1.92E-01	3.28E-01	1.20E+00	5.03E-01	3.47E+00
ET	CTUe	2.43E+02	1.65E+04	0.00E+00	9.54E+02	5.05E+02	3.75E+03	4.95E+03	2.69E+04
HTC	CTUh	1.95E-08	5.96E-07	0.00E+00	3.14E-08	1.41E-06	1.25E-07	1.10E-07	2.29E-06
HTNC	CTUh	2.58E-07	1.42E-05	0.00E+00	1.04E-06	1.51E-06	1.00E-05	3.33E-06	3.03E-05

Tab. S14 Characteristic Values of Various Impact Categories of EHBPs

Category	Unit	Depickling	Tanning	Rewetting	Neutralizing	Retanning	Fatliquoring	Dyeing	Total
PED	MJ	4.82E+02	1.81E+04	1.39E+03	1.13E+03	2.49E+03	9.32E+03	4.38E+03	3.73E+04
ADP	kg Sb eq.	8.38E-04	8.62E-03	2.84E-04	4.96E-04	1.04E-03	4.33E-03	1.74E-03	1.73E-02
WU	kg	1.59E+01	5.25E+02	8.55E+01	3.91E+01	1.22E+02	5.66E+02	1.91E+02	1.55E+03
GWP	kg CO_2 eq.	3.51E+01	9.70E+02	7.03E+01	7.97E+01	1.31E+02	3.02E+02	2.63E+02	1.85E+03
AP	kg SO_2 eq.	1.94E-01	6.93E+00	2.63E-01	3.96E-01	5.57E-01	3.89E+00	1.29E+00	1.35E+01
EP	kg PO_4^{3-} eq.	7.52E-02	2.66E+00	2.38E-01	1.92E-01	3.27E-01	3.33E+00	4.82E-01	7.31E+00

ET	CTUe	3.10E+02	1.30E+04	1.13E+03	9.54E+02	5.02E+02	1.04E+04	4.88E+03	3.12E+04
HTC	CTUh	3.21E-08	7.75E-07	7.91E-08	3.14E-08	1.41E-06	3.27E-07	1.04E-07	2.76E-06
HTNC	CTUh	9.63E-07	1.15E-05	8.30E-07	1.04E-06	1.51E-06	2.76E-05	3.19E-06	4.67E-05

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