Supplementary figures

Figure S1

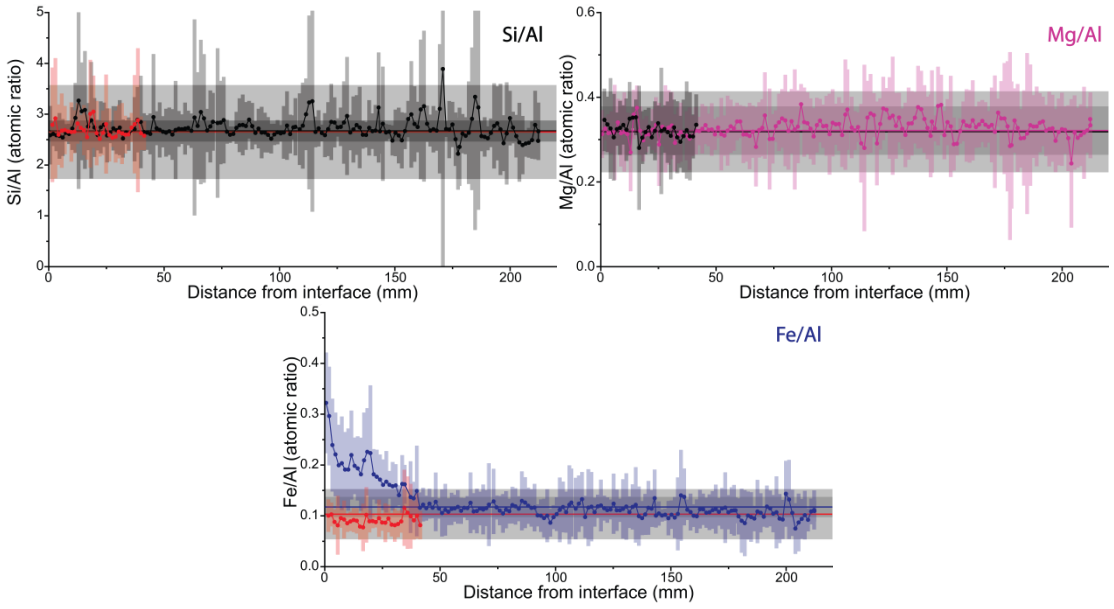

Figure S1: "Al-normalized" chemical profiles of major elements Si, Mg, and Fe in block BM-B-41-1 (long profile, ~220 mm) and block BM-B-41-2 (short profile, ~40 mm) from section 62. Horizontal lines and gray areas represent same data as in Figure 4 (*i.e.* reference and bulk values).

d 2

Figure S2

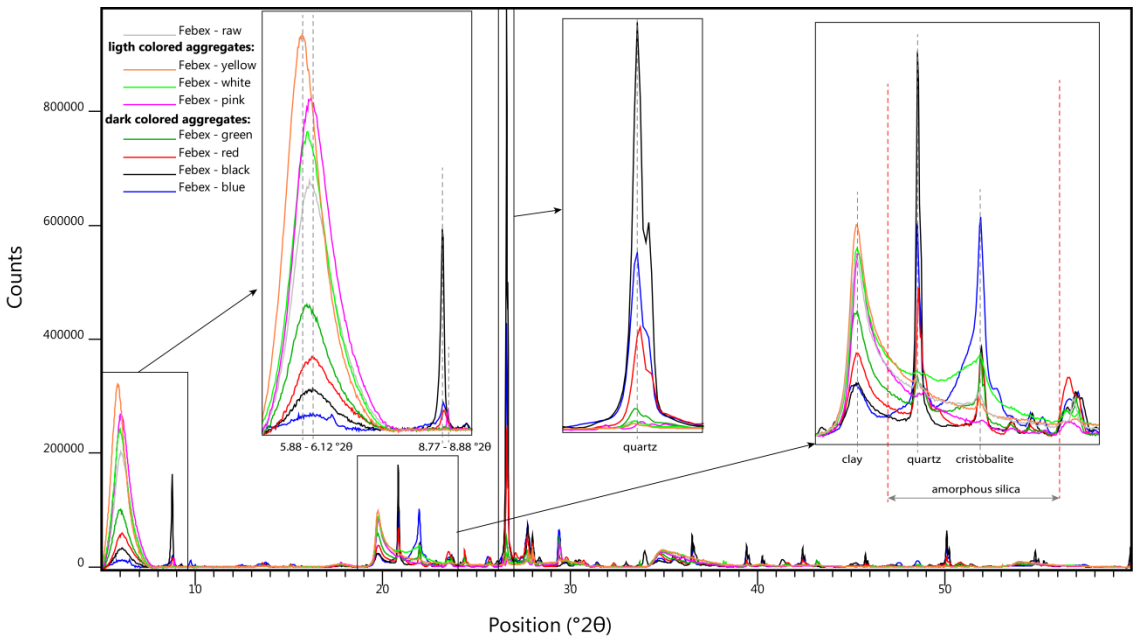

Figure S2: Diffractograms of the various colored aggregates isolated from FEBEX raw bentonite and the bulk raw material.

d

3

Figure S3

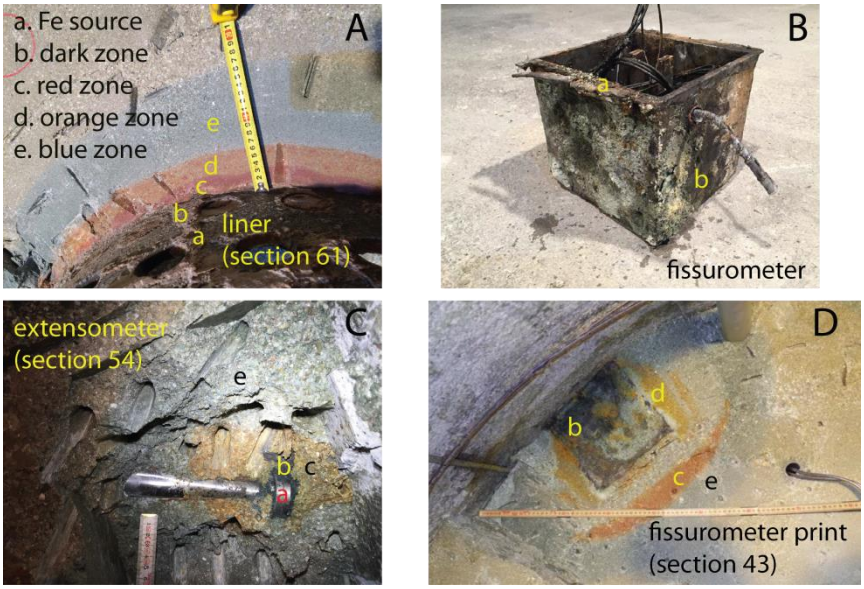

Figure S3: Colored corrosion halos observed around various steel components retrieved upon dismantling of the FEBEX experiment.

Figure S4

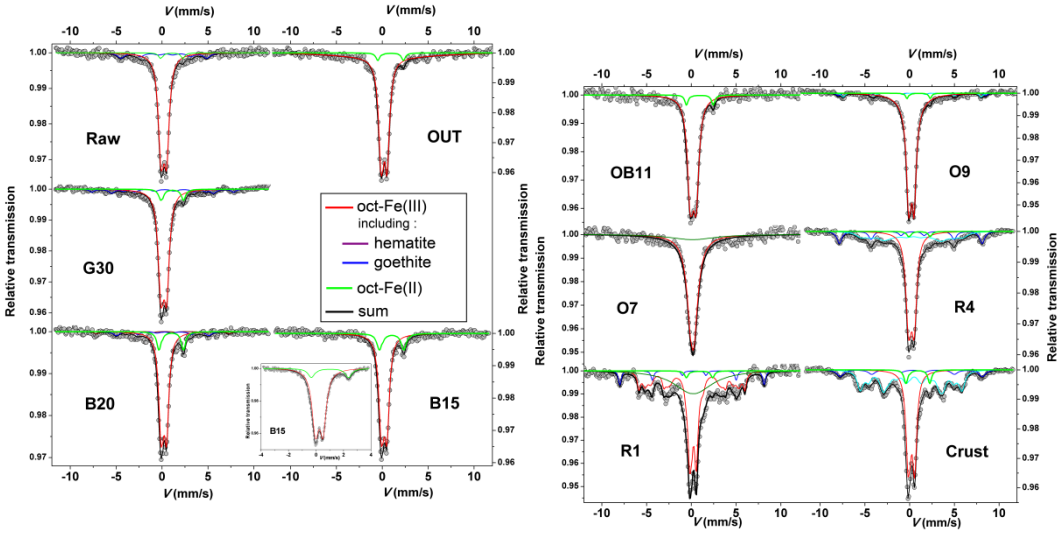

Figure S4: 300 K Mössbauer spectra of the raw FEBEX material and of 11 samples from block BM-B-41-1. The refined values of the hyperfine parameters are listed in Table S3 and Table S4.

Supplementary tables

Table S1 and S2

Table S1: Chemical composition of eighteen powdered FEBEX samples from block BM-B-41-1 determined by XRF (and ATG). Relative uncertainty on major elements is 5%.

Sample	R1	R2	R3	R4	R5	R6	O7	O8	O9	OB10	OB11	B12	B13	B14	B15	B16	B17	B18
d (mm)	2.5	8.2	13.9	19.6	25.3	31.0	36.6	42.3	48.0	53.7	59.4	65.1	70.8	76.5	82.2	87.9	93.5	99.2
%H_2O	5.1	4.2	4	3.1	3.7	4.7	3.7	4.7	4.1	4.9	4.1	3.9	3.8	4.4	4.1	4.8	3.2	5.3
LOI	8.1	7.9	8.1	8.1	8.1	8.1	8.2	8	8	8.1	8.1	8	8.2	8.3	8.3	8.2	8.2	8.2
Σ	99.17	99.37	98.60	99.06	99.26	99.35	99.09	99.02	98.66	98.76	99.07	99.68	99.04	99.03	99.19	99.06	98.16	98.46
majors elements (in oxide wt%)																		
SiO_2	62.52	62.11	63.64	63.73	64.43	64.47	64.58	64.22	64.23	64.55	64.64	65.16	64.62	64.50	64.77	64.57	63.85	64.11
TiO_2	19.41	19.35	19.47	19.95	19.70	20.02	20.11	20.16	19.75	20.14	20.21	20.35	20.13	20.19	20.12	20.15	19.91	19.90
Al_2O_3	7.71	8.55	6.36	5.90	5.70	5.35	5.15	4.93	5.38	4.27	4.27	4.36	4.38	4.51	4.31	4.32	4.40	4.31
Fe_2O_3	4.51	4.52	4.57	4.66	4.57	4.67	4.69	4.69	4.70	4.75	4.90	4.89	4.85	4.85	4.87	4.89	4.84	4.83
MnO	1.67	1.67	1.68	1.69	1.66	1.72	1.69	1.72	1.72	1.96	2.01	1.92	1.95	2.01	2.03	2.06	2.09	2.09
MgO	1.73	1.60	1.35	1.58	1.61	1.52	1.29	1.56	1.27	1.46	1.47	1.38	1.43	1.35	1.45	1.44	1.43	1.57
CaO	1.19	1.16	1.09	1.13	1.17	1.17	1.16	1.29	1.18	1.20	1.14	1.18	1.22	1.17	1.20	1.17	1.19	1.20
Na_2O	0.27	0.25	0.27	0.26	0.26	0.26	0.26	0.28	0.25	0.26	0.26	0.26	0.28	0.27	0.26	0.26	0.27	0.27
TiO_2	0.05	0.04	0.05	0.04	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.04	0.05	0.05	0.05	0.05	0.05	0.05
P_2O_5	0.04	0.04	0.03	0.03	0.04	0.03	0.03	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.04	0.05	0.04	0.04
MnO	62.52	62.11	63.64	63.73	64.43	64.47	64.58	64.22	64.23	64.55	64.64	65.16	64.62	64.50	64.77	64.57	63.85	64.11
Trace elements (in ppm)																		
Ba	130	182	188	146	117	178	123	309	113	172	133	244	145	167	178	228	177	130
Cr	10	18	17	15	14	8	14	17	12	20	13	9	17	10	10	10	13	14
Cu	6	6	8	7	6	8	7	7	8	8	9	12	9	10	11	10	9	9
Nb	12	15	12	15	13	13	13	13	14	14	13	14	14	14	13	15	15	14
Ni	19	25	22	21	22	22	20	21	23	22	33	49	36	34	35	32	25	28
Pb	30	30	26	28	30	31	32	33	36	36	38	40	40	39	39	38	36	39
Rb	47	47	45	46	49	47	46	50	50	49	48	50	47	48	49	48	47	49
Sr	180	183	184	188	187	191	189	199	191	202	199	208	199	204	204	211	213	212
V	23	18	16	13	77	76	71	24	45	11	13	35	12	21	10	28	9	28
Y	18	21	18	34	25	26	26	25	28	34	31	35	35	36	36	35	32	32
Zn	70	70	68	71	70	74	72	73	74	74	77	77	77	77	76	77	75	75
Zr	215	217	209	226	225	218	209	209	236	210	213	211	218	227	211	218	216	215

Sample identity: letters corresponds to the color of the bentonite sampled area (R = red, O = orange, B = blue G = green, OB = transition from orange to blue, BG = transition from blue to green, out = outer layer of the block), and number to the order of sampling, (starting from the from the interface)

d: approximate distance between the sampled layer and the interface

LOI: Loss On Ignition

30 $\%H_2O$: water content measured at 105°C
31 Σ: sum of elements (excludes LOI and $\%H_2O$)

Table S2: Chemical composition of fourteen powdered FEBEX samples determined by XRF (and ATG), including the raw material. Relative uncertainty on major elements is 5%.

Sample	B19	B20	B21	B22	BG23	BG24	BG25	BG26	BG27	G28	G29	G30	out	raw
d (mm)	104.9	110.6	116.3	122.0	127.7	133.4	139.1	144.8	150.4	156.1	161.8	167.5	217.5	-
%H_2O	4.8	3.2	4	3.1	3.2	4.2	4.7	4.4	4.3	7.6	5.3	3	3.3	11
LOI	8.4	8.1	8.2	8.6	8.2	8.5	8.4	8.5	8.2	8.5	8.4	8.3	8.5	8.8
Σ	98.67	98.75	99.35	98.49	99.28	98.61	98.74	99.15	99.25	99.20	99.82	101.29	98.93	98.67
majors elements (in oxide wt%)														
SiO_2	64.38	64.47	64.78	63.64	64.42	64.24	64.62	64.81	64.89	64.99	65.42	66.06	64.61	64.38
TiO_2	20.04	20.04	20.13	19.66	19.73	19.71	19.85	20.15	20.17	20.01	20.17	20.80	20.04	20.04
Al_2O_3	4.23	4.22	4.19	4.13	4.09	3.97	3.98	3.82	3.87	3.86	3.87	3.93	3.91	4.23
Fe_2O_3	4.85	4.84	4.87	4.80	4.84	4.77	4.84	4.93	4.94	4.87	5.00	5.01	4.98	4.85
MnO	2.13	2.12	2.17	3.18	2.22	2.71	2.42	2.47	2.32	2.33	2.35	2.38	2.32	2.13
MgO	1.40	1.35	1.51	1.40	2.34	1.54	1.36	1.34	1.44	1.48	1.37	1.42	1.39	1.40
CaO	1.21	1.25	1.24	1.21	1.18	1.21	1.22	1.18	1.16	1.21	1.18	1.24	1.22	1.21
Na_2O	0.26	0.27	0.27	0.26	0.27	0.26	0.26	0.27	0.27	0.26	0.26	0.27	0.26	0.26
TiO_2	0.05	0.05	0.05	0.07	0.05	0.04	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.05
P_2O_5	0.04	0.04	0.04	0.06	0.05	0.05	0.05	0.05	0.05	0.05	0.05	0.06	0.05	0.04
MnO	64.38	64.47	64.78	63.64	64.42	64.24	64.62	64.81	64.89	64.99	65.42	66.06	64.61	64.38
Trace elements (in ppm)														
Ba	179	195	217	188	179	188	190	193	141	174	254	155	179	195
Cr	15	16	13	18	15	12	16	14	10	19	18	6	15	16
Cu	7	9	8	8	10	9	11	11	9	9	10	11	7	9
Nb	13	14	12	13	13	12	14	15	13	13	13	14	13	14
Ni	25	25	24	22	24	21	22	21	23	21	22	22	25	25
Pb	34	32	37	35	38	34	35	39	33	35	37	36	34	32
Rb	50	51	50	50	48	49	48	47	48	48	48	47	50	51
Sr	218	223	228	237	229	235	238	236	239	239	238	239	218	223
V	26	34	29	16	16	16	19	29	25	32	28	31	26	34
Y	32	31	30	33	31	35	31	31	28	32	32	29	32	31
Zn	75	73	74	74	75	73	75	74	75	76	75	75	75	73
Zr	212	209	216	213	221	212	213	214	221	211	220	215	212	209

Sample identity: letters corresponds to the color of the bentonite sampled area (R = red, O = orange, B = blue G = green, OB = transition from orange to blue, BG = transition from blue to green, out = outer layer of the block), and number to the order of sampling, (starting from the from the interface)

d: average distance between the sampled layer and the interface

LOI: Loss On Ignition

38 $\%H_2O$: water content measured at 105°C
39 Σ: sum of elements (excludes LOI and $\%H_2O$)

Table S3 and S4

Table S3: refined values of Mössbauer parameters and doublet structural attributions for the raw FEBEX material and fives samples collected in block BM-B-41-1 (spectra shown in Figure 9 and S4). Uncertainties are 0.02 mm·s^{-1} for I.S., Q.S., 2ε, and F.W.H.M.; 0.5 T for B_{hf}, and 2% for the area.

300K						77K					
Hyperfine parameters					Attribution	Hyperfine parameters					Attribution
I.S.	F.W. H.M.	Q.S. /2ε	B_{hf}	Area (%)		I.S.	F.W. H.M.	Q.S. /2ε	B_{hf}	Area (%)	
raw											
1.20	0.50	2.35		4	para-Fe(II)	1.35	0.50	2.54		5	para-Fe(II)
0.35	0.67	0.54		83	para-Fe(III)	0.46	0.71	0.56		75	para-Fe(III)
0.45	0.90	-0.20	28.8	13	goethite	0.47	1.07	-0.25	48.6	11	goethite
						0.53	0.69	0.06	55.0	9	hematite
out (220 mm)											
1.26	0.56	2.37		3	para-Fe(II)	1.26	0.5	2.76		5	para-Fe(II)
0.34	2.00	0.55		46	para-Fe(III)	0.46	1.50	0.21		33	para-Fe(III)
0.34	0.62	0.58		51	para-Fe(III)	0.46	0.56	0.59		51	para-Fe(III)
						0.49	0.44	-0.23	48.9	7	goethite
						0.45	0.44	0.12	54.5	4	hematite
G30 (168 mm)											
1.26	0.56	2.37		8	para-Fe(II)	1.29	0.72	2.71		9	para-Fe(II)
0.34	0.67	0.55		80	para-Fe(III)	0.46	0.73	0.57		81	para-Fe(III)
0.32	0.70	-0.20	34.2	7	goethite	0.47	0.46	-0.20	48.9	4	goethite
0.34	0.56	-0.20	47.7	5	hematite	0.44	0.46	0.00	53.7	6	hematite
BG25 (139 mm)											
						1.27	0.4	2.85		4	para-Fe(II)
						0.55	0.72	0.00		20	para-Fe(III)
						0.47	0.62	0.58		62	para-Fe(III)
						0.55	0.67	-0.4	48.6	4	goethite
						0.55	1.10	0.06	55.2	10	hematite
B20 (111 mm)											
1.32	0.50	2.27		14	para-Fe(II)	1.26	0.60	2.92		15	para-Fe(II)
0.32	0.62	0.59		86	para-Fe(III)	0.46	0.64	0.56		75	para-Fe(III)
						0.47	0.46	-0.23	49.8	6	goethite
						0.57	0.46	0.25	55.6	4	hematite
B15 (82 mm)											
1.17	0.67	2.57		16	para-Fe(II)	1.28	0.66	2.88		16	para-Fe(II)
0.34	0.62	0.53		84	para-Fe(III)	0.47	0.66	0.56		74	para-Fe(III)
						0.53	0.70	-0.20	48.0	4	goethite
						0.41	0.56	0.22	53.5	6	hematite

I.S. = Isomer shift value relative to that of the α-Fe at 300 K. (mm·s^{-1})
F.W.H.M. = Full width of line at half of its maximum intensity. (mm·s^{-1})
Q.S./2ε = Quadrupolar splitting/quadrupolar shift
B_{hf} = Magnetic hyperfine field (T)

Table S4: refined values of Mössbauer parameters and doublet structural attributions for six samples collected in block BM-B-41-1 (spectra shown in Figures 9 and S4). Uncertainties are 0.02 mm·s^{-1} for I.S., Q.S., 2ε, and F.W.H.M.; 0.5 T for B_{hf}, and 2% for the area.

300K						77K					
Hyperfine parameters					Attribution	Hyperfine parameters					Attribution
I.S.	F.W. H.M.	Q.S. /2ε	B_{hf}	Area (%)		I.S.	F.W. H.M.	Q.S. /2ε	B_{hf}	Area (%)	
OB11 (59 mm)											
1.10	0.44	2.89		6	para-Fe(II)	1.20	0.50	3.05		6	para-Fe(II)
0.36	0.73	0.57		94	para-Fe(III)	0.45	0.65	0.58		81	para-Fe(III)
						0.40	0.50	-0.21	42.7	3	goethite
						0.44	0.50	-0.26	49.3	4	goethite
						0.34	0.50	-0.05	54.0	6	hematite
O9 (48 mm)											
0.37	2.00	0.00		33	para-Fe(III)	1.26	0.36	2.80		4	para-Fe(II)
0.35	0.52	0.52		57	para-Fe(III)	0.46	0.32	0.57		71	para-Fe(III)
1.16	0.36	2.51		3	para-Fe(II)	0.50	0.71	-0.23	45.0	4	goethite
0.54	0.44	-0.21	47.6	4	goethite	0.50	0.51	-0.23	49.5	17	goethite
0.51	0.44	-0.20	50.8	3	hematite	0.46	0.50	-0.04	55.0	4	hematite
O7 (37 mm)											
						1.26	0.40	2.81		3	para-Fe(II)
0.31	3.00	0.00		19	para-Fe(III)	0.47	0.64	0.58		69	para-Fe(III)
0.31	1.20	0.00		81	para-Fe(III)						
						0.49	0.47	-0.22	49.2	12	goethite
						0.49	0.47	-0.22	46.0	4	goethite
						0.47	0.49	-0.15	52.8	12	hematite
R4 (20 mm)											
1.25	0.50	2.32		2	para-Fe(II)	1.32	0.50	2.76		4	para-Fe(II)
0.34	0.66	0.56		53	para-Fe(III)	0.47	0.70	0.54		55	para-Fe(III)
<0.43>		<-0.25>	<31.6>	31	goethite	0.48	0.65	-0.23	49.1	29	goethite
0.38	0.59	-0.20	49.3	14	hematite	0.51	0.49	-0.11	52.9	12	hematite
R1 (3 mm)											
						1.45	0.40	2.55		2	para-Fe(II)
						0.47	0.71	0.59		43	para-Fe(III)
						0.49	0.52	-0.24	49.3	48	goethite
						0.47	0.37	-0.11	53.0	7	hematite
crust (first few hundreds of μm)											
1.17	0.50	2.57		5	para-Fe(II)	1.24	0.71	2.74		8	para-Fe(II)
0.36	0.58	0.63		41	para-Fe(III)	0.46	0.56	0.63		37	para-Fe(III)
0.44	0.89	-0.20	49.1	9	goethite	0.50	0.42	-0.22	49.8	37	goethite
<0.46>		<-0.30>	<30.2>	45	goethite	0.50	0.64	-0.23	48.1	18	goethite

I.S. = Isomer shift value relative to that of the α-Fe at 300 K. (mm·s^{-1})
F.W.H.M = Full width of line at half of its maximum intensity. (mm·s^{-1})
Q.S./2ε = Quadrupolar splitting/quadrupolar shift
B_{hf} = Magnetic hyperfine field (T)
a. the shape of this spectra accounts for the presence of very fast relaxation phenomena.

Figure S1: “Al-normalized” chemical profiles of major elements Si, Mg, and Fe in block BM-B-41-1 (long profile, ~220 mm) and block BM-B-41-2 (short profile, ~40 mm) from section 62. Horizontal lines and grey areas represent same data as in Figure 4 (i.e. reference and bulk values).

Figure S2: Diffractograms of the various colored aggregates isolated from FEBEX raw bentonite and the bulk raw material.

Figure S3: Colored corrosion halos observed around various steel components retrieved upon dismantling of the FEBEX experiment.

Figure S4: 300 K Mössbauer spectra of the raw FEBEX material and of 11 samples from block BM-B-41-1. The refined values of the hyperfine parameters are listed in Table S3 and Table S4.

66 Table S1: Chemical composition of eighteen powdered FEBEX samples from block BM-B-41-1
67 determined by XRF (and ATG). Relative uncertainty on major elements is 5%.

68 Table S2: Chemical composition of fourteen powdered FEBEX samples determined by XRF
69 (and ATG), including the raw material. Relative uncertainty on major elements is 5%.

70 Table S3: refined values of Mössbauer parameters and doublet structural attributions for the raw
71 FEBEX material and fives samples collected in block BM-B-41-1 (spectra shown in Figure 9 and S4).
72 Uncertainties are 0.02 mm·s^{-1} for I.S., Q.S., 2ε, and F.W.H.M.; 0.5 T for B_{hf}, and 2% for the area.

73 Table S4: refined values of Mössbauer parameters and doublet structural attributions for six samples
74 collected in block BM-B-41-1 (spectra shown in Figure 9 and S4). Uncertainties are 0.02 mm·s^{-1} for
75 I.S., Q.S., 2ε, and F.W.H.M.; 0.5 T for B_{hf}, and 2% for the area.

76