

Appendix

List of correction:

*BEA ---> Beta

Following to the revision of framework type codes for intergrowth structures in 2022

The 4th paragraph of Results:

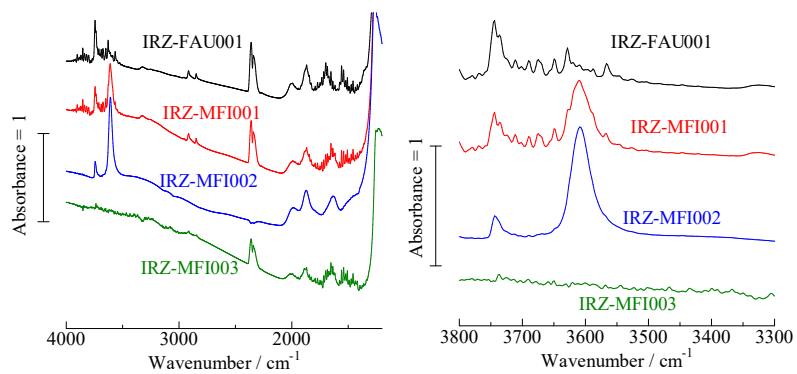
[Corrected]

The amounts of Brønsted acid sites are summarized in Table 2 as well as the chemical compositions. The NH₄-MFI sample with high Al concentration (IRZ-MFI002) had a large amount of Brønsted acid sites, corresponding to 0.982, close to 1, of [Al] - [Na] contents, indicating generation of Brønsted acid site by the isomorphous substitution of Si with Al. On the other hand, the ratio [Brønsted acid] / ([Al] - [Na]) was lower, ca. 0.8, on the H-MFI (IRZ-MFI001 and IRZ-MFI003). IRZ-MFI001 was prepared by calcination to remove NH₃ from an NH₄-MFI equivalent to IRZ-MFI002. Calcination and storage in atmosphere of H-MFI with high Al concentration was thus found to decrease the ion exchange capacity, presumably due to dealumination from Al-rich proton-form zeolite in humid atmosphere even at room temperature, while NH₄-form was stable as reported¹⁵⁻¹⁶¹⁷. The FAU sample (IRZ-FAU001) also showed low the [Brønsted acid] / ([Al] - [Na]) ratio, 0.30. In addition to the detection of Brønsted acid site enhanced by the extra-framework Al species as above, this points out the presence of considerable amount of extra-framework Al on IRZ-FAU001.

[Old]

The amounts of Brønsted acid sites are summarized in Table 2 as well as the chemical compositions. The NH₄-MFI sample with high Al concentration (IRZ-MFI002) had a large amount of Brønsted acid sites, corresponding to 0.982, close to 1, of [Al] - [Na] contents, indicating generation of Brønsted acid site by the isomorphous substitution of Si with Al. On the other hand, the ratio [Brønsted acid] / ([Al] - [Na]) was lower, 0.799, on the H-MFI (IRZ-MFI001) with Al content similar to that of IRZ-MFI002. IRZ-MFI001 was prepared by calcination to remove NH₃ from an NH₄-MFI equivalent to IRZ-MFI002. Calcination and storage in atmosphere of H-MFI with high Al concentration was thus found to decrease the ion exchange capacity, presumably due to dealumination from Al-rich proton-form zeolite in humid atmosphere even at room temperature, while NH₄-form was stable as reported¹⁵⁻¹⁶¹⁷. On the other hand, the H-MFI sample with low Al concentration showed only 0.2, far smaller than 1, of the [Brønsted acid] / ([Al] - [Na]) ratio, indicating the presence of considerable amount of extra-framework Al. In this case, the calcination and exposure of H-form to the atmosphere was not the cause of dealumination, because the Al concentration was low. The FAU sample (IRZ-FAU001) also showed low the [Brønsted acid] / ([Al] - [Na]) ratio, 0.30. In addition to the detection of Brønsted acid site enhanced by the extra-framework Al species as above, this points out the presence of considerable amount of extra-framework Al on IRZ-FAU001.

Figure 1
[Corrected]

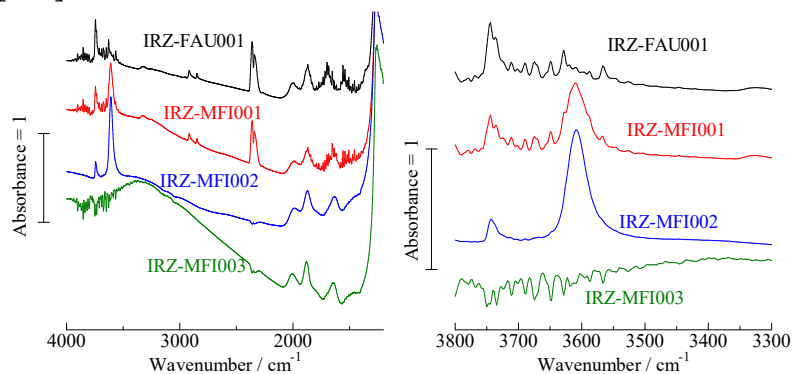


Data table:



(0.1 MB)

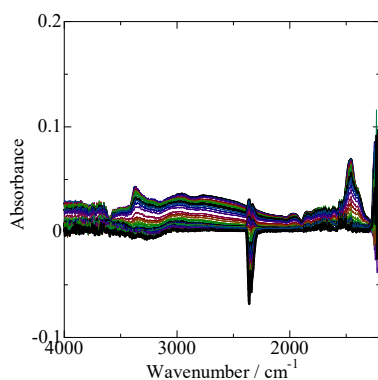
[Old]



Data table:



Figure 2, IRZ-MFI003
[Corrected]



IRZ-MFI003 Data table at 10 K interval:

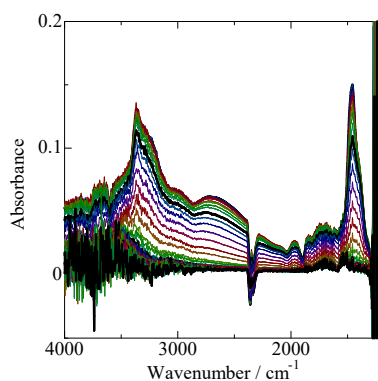


IRZ-MFI003-IRdi (2.4 MB)
f-10Kinterval.xlsx

Data table all: IRZ-MFI003-IRdi (11.0 MB)
f.xlsx



[Old]



IRZ-MFI003 Data table at 10 K interval:



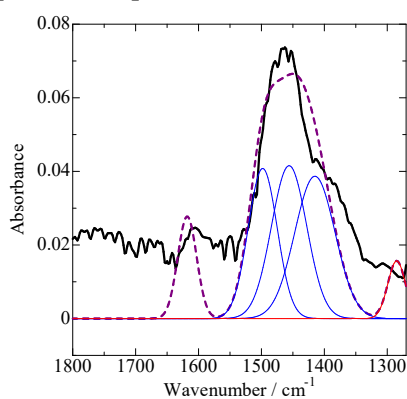
IRZ-MFI003-IRdi
f-10Kinterval.xlsx

Data table

all: IRZ-MFI003-IRdi
f.xlsx



Figure 3, IRZ-MFI003
[Corrected]

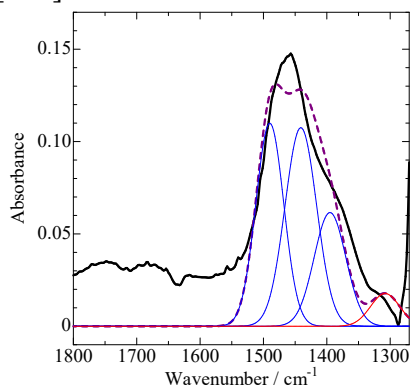


IRZ-MFI003 Data table:



IRZ-MFI003-IRd (0.07 MB)
ec373K.xlsx

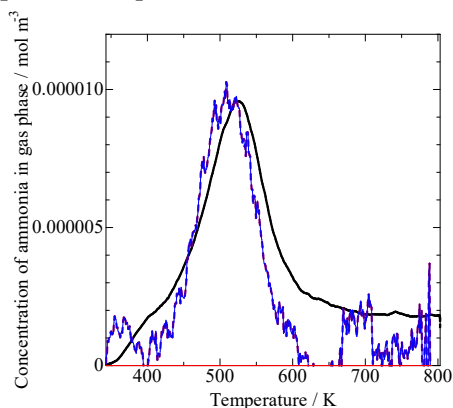
[Old]



IRZ-MFI003 Data table:  IRZ-MFI003-IRd
ec373K.xlsx

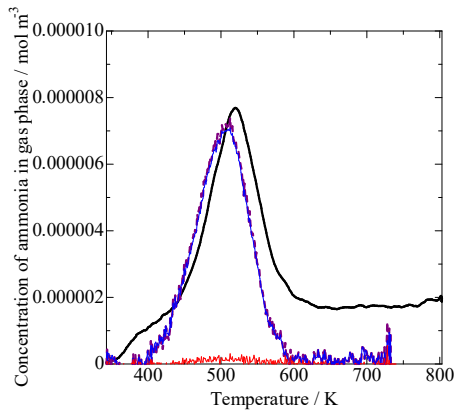
Figure 4, IRZ-MFI003

[Corrected]



IRZ-MFI003 Data table:  IRZ-MFI003-IRM (0.04 MB)
STPD.xlsx

[Old]



IRZ-MFI003 Data table:  IRZ-MFI003-IRM
STPD.xlsx

Table 1, Yellow colored parts
[Corrected]

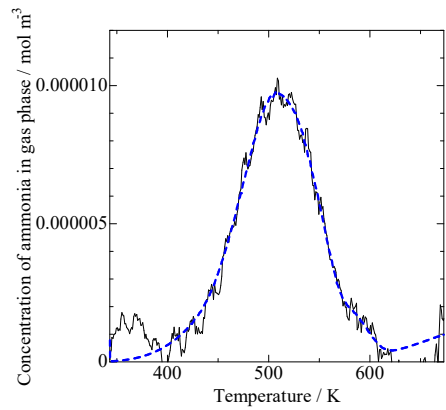
Sample	Acid amount / mol kg ⁻¹	
	Brønsted	Lewis
IRZ-FAU001	0.303354388	0.031764668
IRZ-MFI001	1.042975611	0.018365303
IRZ-MFI002	1.317670894	0.001929321
IRZ-MFI003	0.250679989	0.000000000

[Old]

Sample	Acid amount / mol kg ⁻¹	
	Brønsted	Lewis
IRZ-FAU001	0.303354388	0.031764668
IRZ-MFI001	1.042975611	0.018365303
IRZ-MFI002	1.317670894	0.001929321
IRZ-MFI003	0.074501068	0.001304755

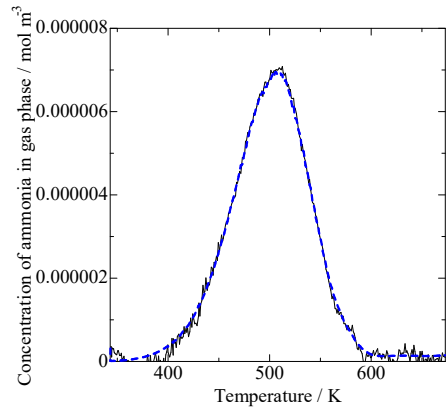
Figure 5, IRZ-MFI003

[Corrected]



 **IRZ-MFI003** Data table: IRZ-MFI003-ASD (0.04 MB)
-Efitdisp.xlsx

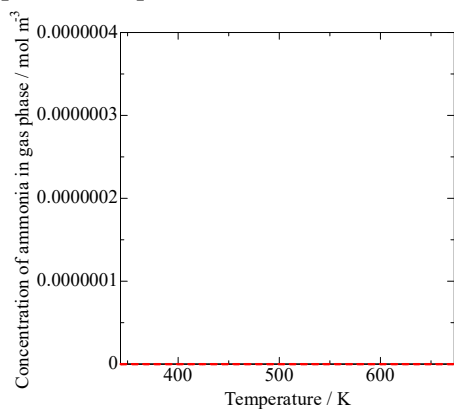
[Old]



 **IRZ-MFI003** Data table: IRZ-MFI003-ASD
-Efitdisp.xlsx

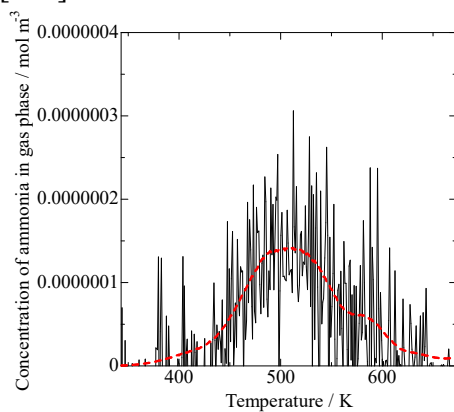
Figure 6, IRZ-MFI003

[Corrected]



IRZ-MFI003 Data table:  IRZ-MFI003-ASD (0.08 MB)
-Ffitdisp.xlsx

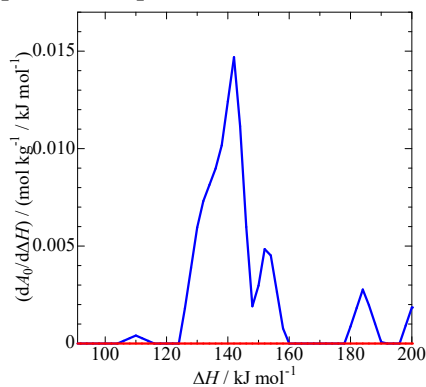
[Old]



IRZ-MFI003 Data table:  IRZ-MFI003-ASD
-Ffitdisp.xlsx

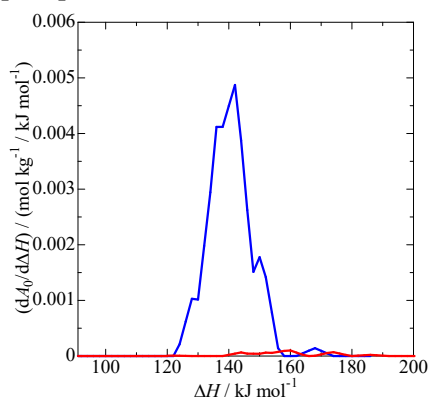
Figure 7, IRZ-MFI003

[Corrected]



IRZ-MFI003 Data table:  IRZ-MFI003-ASD (0.01 MB)
.xlsx

[Old]



IRZ-MFI003 Data table:  IRZ-MFI003-ASD
.xlsx

Table 2, Yellow-colored part
[Corrected]

Sample	SiO ₂ / Al ₂ O ₃ molar ratio	[Al] / mol kg ⁻¹	[Na] / mol kg ⁻¹	([Al] - [Na]) / mol kg ⁻¹	[B acid] / mol kg ⁻¹	[L acid] / mol kg ⁻¹	[B acid] / ([Al] - [Na])
IRZ- FAU001	30	1.05	0.032	1.02	0.30	0.03	0.30
IRZ- MFI001	23.8	1.31	<0.004	1.31	1.04	0.02	0.799
IRZ- MFI002	23.1	1.34	<0.003	1.34	1.32	0.00	0.982
IRZ- MFI003	100	0.33	unreported	0.33*	0.25	0.00	0.766

*: [Na] is assumed to be zero.

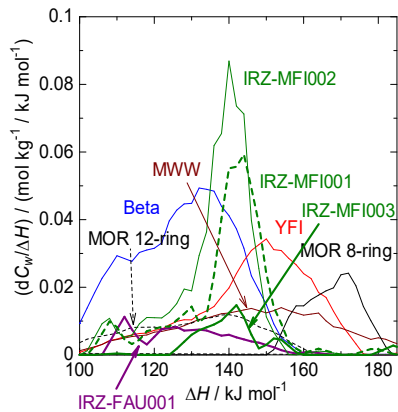
[Old]

Sample	SiO ₂ / Al ₂ O ₃ molar ratio	[Al] / mol kg ⁻¹	[Na] / mol kg ⁻¹	([Al] - [Na]) / mol kg ⁻¹	[B acid] / mol kg ⁻¹	[L acid] / mol kg ⁻¹	[B acid] / ([Al] - [Na])
IRZ- FAU001	30	1.05	0.032	1.02	0.30	0.03	0.30
IRZ- MFI001	23.8	1.31	<0.004	1.31	1.04	0.02	0.799
IRZ- MFI002	23.1	1.34	<0.003	1.34	1.32	0.00	0.982
IRZ- MFI003	100	0.33	unreported	0.33*	0.07	0.00	0.2

*: [Na] is assumed to be zero.

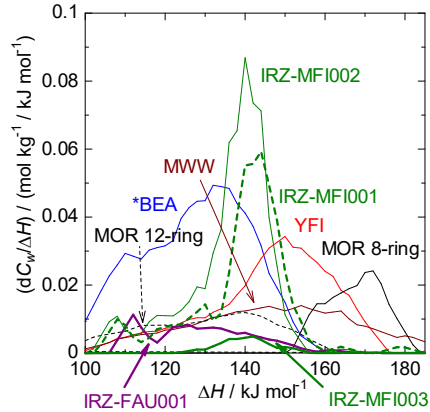
Figure 8

[Corrected]



Data table:  ASDcomparison. (0.01 MB)
xlsx

[Old]



Data table:  ASDcomparison.
xlsx