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Tellurium-Loaded Zeolites. III. The Structure of Te/NaX

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A single-crystal X-ray study of a hydrogen-treated Te/NaX catalyst has revealed scattering matter, assigned to telluride ion on the basis of chemical environment, in both the sodalite cage and the supercage of the X-zeolite structure. Both telluride ion sites have relatively low occupancy factors. The telluride ion in the center of the sodalite cage is coordinated to four site I′ and possibly to four site II′ sodium ions. The supercage telluride ion is coordinated to a site II sodium ion and, probably, to at least one additional sodium ion (not detected) located near the conventional site III. The chemical involvement of hydrogen, necessary for the reduction of tellurium, is supported by diffuse reflectance spectra and hydrogen sorption data. Probable hydrogen locations on the zeolitic framework are assigned.

INTRODUCTION

The selective aromatization capability of tellurium-loaded zeolites has been rationalized (*1*) on the basis of an aromatization site consisting of a telluride ion coordinated to zeolitic cations. Cracking activity has been presumed to arise from protonic hydrogen generated by the reduction of the tellurium atoms. This explanation was based primarily, but not exclusively, on a crystallographic study of a single crystal of a Te/NaX catalyst, the details of which are presented here. In addition, positive evidence of the chemical involvement of hydrogen in the production of the catalyst is offered.

EXPERIMENTAL

A 100 mg batch of nominal 11% Te/NaX was prepared by heating a mixture of finely divided tellurium and dehydrated NaX-zeolite crystals (*2*) for 116 hr at 540°C in an evacuated and sealed U-tube following a 1-hr pretreatment in flowing helium at 540°C. From this preparation, a well-formed octahedral crystal, ~80 μ in length, was selected and wedged in a thin-walled glass capillary which was then attached to a glass manifold. The specimen was heated at 475°C in flowing hydrogen for 16.5 hr. Before cooling to room temperature, the open end of the capillary was sealed and the sample evacuated with a mechanical pump. The portion of the capillary containing the crystal was then sealed off and placed in an aluminum pin for mounting on the X-ray apparatus.

The batch of crystals after the helium treatment appeared brown to the eye, but had a metallic luster when viewed under magnification. Slicing some of the crystals showed that the coloration was uniform throughout and not a surface effect. After the hydrogen treatment, the selected crystal was a translucent brown under the microscope.

The intensity data were collected with a General Electric Eulerian cradle mounted on a Siemens goniometer using the moving-crystal/moving-counter technique and nickel-filtered copper radiation. One-minute background counts were taken at each end of a 2.5°, 10-min scan. The standard deviations of the structure factors were computed as described earlier (*3*). Of the 260 structure factors obtained, only those (173) having magnitudes greater than their standard deviations were used in the solution and refinement of the structure.

The scattering factors computed by Hanson and Pohler (*4*), and Hanson, Herman, Lea, and Skillman (*5*), selected and modified as described earlier (*3*), were used in the structure factor calculations. The crystallographic computer programs ORFLS (*6*), ORFFE (*7*), FOUR (*8*), and ORTEP (*9*) were used in this study.

Results and Discussion

Determination and Refinement of Structure

The X-ray diffraction patterns were much less intense than expected for a crystal of this size—this is attributed primarily to a loss in crystal quality since the absorption of X-rays by tellurium can account for only about a 10% decrease in intensity relative to a NaX crystal of the same size. The lattice parameter, $a_0 = 24.76 \pm 0.02$ Å, was contracted by 1% with respect to NaX, which is also consistent with some loss in crystal quality.

TABLE 1

Final Fractional Coordinates and E.S.D's[b]

Atom	Set	P	X	Y	Z	T[a]
T	i	1.00	−0.0537(4)	0.0351(4)	0.1260(5)	2.2
01	h	1.00	−0.1085(10)	0.1085(10)	0.0000	3.6
02	g	1.00	−0.0046(11)	−0.0046(11)	0.1429(13)	3.6
03	g	1.00	0.1746(10)	0.1746(10)	−0.0312(14)	3.6
04	g	1.00	0.1800(10)	0.1800(10)	0.3233(15)	3.6
Te(LC)	g	0.039(7)	0.336(3)	0.336(3)	0.240(4)	3.6
Te(U)	a	0.16(3)	0.125	0.125	0.125	3.6
Na(I′)	e	1.00(8)	0.0646(10)	0.0646(10)	0.0646(10)	4.2
Na(II′)	e	0.28(8)	0.202(5)	0.202(5)	0.202(5)	4.2
Na(II)	e	0.73(7)	0.2447(14)	0.2447(14)	0.2447(14)	4.2

[a] The E.S.D. in the overall temperature factor is 0.3.

[b] The estimated standard deviations (E.S.D.) are in parentheses.

Diffraction photographs exhibited symmetry and extinctions consistent with the previously assigned space group, *Fd*3*m* (*10*).

Estimates of initial cation and framework positional parameters and cation occupancy factors were made on the basis of previous structural studies (*11*). In addition to these expected atom sites, three scattering sites were revealed on difference maps. On the basis of interatomic distances and chemical environment, one of these was assigned to sodium ion and two to tellurium species; all three sites have relatively low occupancy factors.

The site assigned to the sodium ion is in the site II′ position, 1.8 Å from the sodium ion of site II.* Scattering matter in site U, the center of the sodalite cage, was assigned to telluride ion of partial occupancy on the basis of its tetrahedral coordination to four sodium ions at about 2.6 Å. A scattering site in the supercage was also assigned to tellurium ion due to its distances from the Na(II) ion and from the 01, 02, and 04 oxygen ions.†

Since the accuracy of the data did not appear to warrant the use of individually refined temperature factors, the T (Si and

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* The site designations are defined as follows: I, within the hexagonal prism; II, in the supercage and on a hexagonal face of a sodalite unit; I′ and II′, *within* the sodalite unit, opposed to sites I and II, respectively; III, within the supercage, and conventionally centered on the square faces of sodalite units (*12*); and U, at the center of the sodalite cage (*13*).

† The framework oxygen ions are: 01, the bridging oxygen ion of the hexagonal prism; 02, the oxygen ion common to the hexagonal-prism six-ring and the supercage six-ring; 03, the second type of oxygen ion in the hexagonal-prism six-ring, alternating in the ring with type 02; 04, the second type of oxygen ion in the supercage six-ring, alternating in the ring with type 02.

TABLE 2

Observed and Calculated Structure Factors

H	K	FOBS	FCAL	H	K	FOBS	FCAL	H	K	FOBS	FCAL	H	K	FOBS	FCAL
***L	=	0****		17	11	952	-795	7	7	873	631	13	9	1429	-1066
12	0	1906	-2158	13	13	1092	466	9	7	1076	-1079	15	9	1087	-1100
16	0	3893	4278	***L	=	2****		11	7	784	594	11	11	2852	-3211
20	0	963	-775	4	2	692	572	13	7	612	419	15	13	1218	1086
2	2	1176	2409	6	2	1557	-975	15	7	1036	1110	***L	=	6****	
6	2	920	873	8	2	2508	1806	19	7	851	-895	6	6	2685	2861
14	2	1305	-1009	10	2	1322	-1676	9	9	682	780	8	6	727	681
4	4	4143	-3491	12	2	920	196	11	9	2630	-2630	10	6	907	-897
8	4	2075	2455	14	2	690	-175	13	9	824	-674	12	6	451	-732
16	4	1034	1023	16	2	845	-692	17	9	1993	-1847	16	6	4010	4066
6	6	1998	2500	20	2	997	-1108	15	11	1554	1706	8	8	1140	1440
18	6	1372	-1237	4	4	1343	-1241	13	13	1499	1626	14	8	2165	-2518
8	8	5026	4623	6	4	3162	-2886	***L	=	4****		10	10	1005	-1036
12	8	521	796	8	4	1257	877	4	4	910	-329	12	10	1836	-2059
16	8	876	-560	10	4	540	691	6	4	974	401	12	12	749	876
10	10	4567	-4489	12	4	1384	1313	8	4	2256	2150	***L	=	7****	
14	10	1062	-847	14	4	697	-1001	10	4	909	989	7	7	1067	1150
12	12	5722	5108	16	4	548	-721	12	4	1065	775	9	7	1238	1207
16	12	785	-681	18	4	715	791	16	4	763	138	11	7	1150	-1016
***L	=	1****		6	6	563	-394	18	4	1047	-235	13	7	753	-798
3	1	1366	2013	8	6	1282	1456	6	6	3159	-3200	17	7	555	-250
5	1	1772	1919	10	6	749	-1086	8	6	681	-909	11	9	1042	962
7	1	1037	-792	12	6	910	-950	12	6	889	-771	15	9	654	902
9	1	785	676	14	6	956	726	14	6	1236	1215	17	9	1011	1139
11	1	1065	-1754	16	6	646	230	16	6	453	62	15	11	808	-757
17	1	1735	-1761	10	8	1618	1890	8	8	1316	-1759	13	13	433	603
3	3	2187	-3082	12	8	1004	-973	10	8	444	-833	***L	=	8****	
5	3	1146	358	16	8	1157	826	12	8	1289	-993	8	8	1929	1564
9	3	1446	1951	18	8	610	-782	16	8	658	-1057	12	8	2157	2477
11	3	807	-761	10	10	715	-971	12	10	508	-515	16	8	2933	2972
13	3	672	-349	14	10	949	-1041	14	12	862	654	10	10	1695	1751
15	3	696	729	16	10	917	-672	14	14	1059	963	14	10	1033	-734
19	3	757	-821	14	12	1294	1512	***L	=	5****		12	12	619	-368
5	5	1108	830	***L	=	3****		5	5	7999	-7439	***L	=	9****	
7	5	1309	723	5	3	4678	-5145	7	5	1214	-786	9	9	1726	1528
9	5	637	1059	7	3	2056	-1355	9	5	909	-713	11	9	1110	1354
13	5	1288	-940	11	3	2786	2422	11	5	629	-406	13	9	1418	-1233
17	5	670	-585	13	3	1119	1021	13	5	535	-730	11	11	640	499
9	7	869	1309	15	3	1415	-1282	15	5	1554	-1228	13	11	891	-777
15	7	697	-1030	19	3	683	846	17	5	3145	2786	***L	=	10****	
19	7	734	812	5	5	916	-80	7	7	1307	1463	10	10	867	-752
9	9	1423	-1284	7	5	1321	1421	9	7	749	-769	12	10	1730	1449
11	9	542	-626	11	5	648	337	11	7	1204	-1522	14	10	562	-651
13	9	1244	-896	13	5	536	-584	13	7	1483	1231	12	12	1080	426
15	9	882	-620	15	5	1057	1189	17	7	1842	1878	***L	=	11****	
11	11	3617	3630	19	5	1831	-2204	9	9	1808	2107	13	11	670	-1200
13	11	923	670												

Al), oxygen, tellurium and sodium ions were assigned initial isotropic temperature factors of 1.0, 2.4, 2.4, and 3.0 Å^2, respectively, and subsequently only the overall temperature factor was allowed to vary. The final R value was 0.19.* Final atomic parameters, structure factor listing, and interatomic distances and angles are given in Tables 1–4.

Structural Character

The framework interatomic distances and angles are characteristic of the X-zeolite structure and do not warrant further comment. The dispositions of the sodium and tellurium ions, on the other hand.

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* $R = \dfrac{\Sigma \| F_o| - |F_c\|}{\Sigma |F_o|}$

TABLE 3
FRAMEWORK INTERATOMIC DISTANCES AND ANGLES IN Te/NaX[a]

Distances(Å)		Angles (Deg.)	
T–01	1.67(2)	T–01–T	134(2)
T–02	1.62(2)	T–02–T	148(3)
T–03	1.70(2)	T–03–T	140(2)
T–04	1.67(2)	T–04–T	139(2)
Mean value	1.66(1)		140(1)
01–02	2.71(3)	01–T–02	111(2)
01–03	2.80(4)	01–T–03	112(2)
01–04	2.63(2)	01–T–04	104(2)
02–03	2.67(3)	02–T–03	107(2)
02–04	2.68(3)	02–T–04	109(2)
03–04	2.79(4)	03–T–04	112(2)
Mean value	2.71(1)		109(1)

[a] Estimated standard deviations are in parentheses.

TABLE 4
INTERATOMIC DISTANCES AND ANGLES IN Te/NaX INVOLVING NONFRAMEWORK ATOMS[a]

Distances (Å)	
Na(I′)–03	2.41(4)
Na(I′)–02	3.10(4)
Na(I′)–Te(U)	2.59(4)
Na(I′)–Na(I′)′	4.22(7)
Na(II′)–02	2.35(10)
Na(II′)–04	3.10(10)
Na(II′)–Te(U)	3.30(10)
Te(U)–02	4.56(4)
Te(U)–03	4.24(4)
Te(U)–04	5.25(4)
Na(II)–02	2.54(4)
Na(II)–04	2.99(3)
Na(II)–Te(LC)	3.20(12)
Te(LC)–01	3.98(11)
Te(LC)–02	3.76(12)
Te(LC)–04	4.17(10)
Angles (Deg.)	
03–Na(I′)–03′	125.9(2)
Na(I′)–Te(U)–Na(I′)′	109.5(0)
02–Na(II′)–02′	112(3)
02–Na(II)–02′	101(2)
02–Na(II)–Te(LC)	80(2)
02′–Na(II)–Te(LC)	129(2)
04–Na(II)–Te(LC)	85(2)
04–Na(II)–Te(LC)	140(3)
01–Te(LC)–01′	57(2)

[a] A prime indicates an atom related to the unprimed by symmetry. Estimated standard deviations are in parentheses.

are of great significance for the catalytic properties of the system and are summarized in Table 5. A total of sixty-four sodium ions per unit cell was observed in sites I′, II, and II′—leaving site I unoccupied. The twenty-one ions (85–64) unaccounted for and necessary for electrical neutrality are listed as occupying sites III. The tellurium is distributed between the large cavity and the center of the sodalite cage in a 3:1 ratio. The tellurium population corresponds to 4.5 wt %—a value consistent with the tellurium contents of active catalysts (3.5–6 wt %) previously discussed (*1*, *14*).

The full occupancy of site I′ by sodium ion, i.e., four sodium ions/sodalite cage or 32/unit cell, was unexpected, since electrostatic arguments appear to favor a distribution between sites I and I′ (*15*) and only about ⅙ of the sodalite units contain a tellurium species. More recently, however, an X-ray study (*16*) of *dehydrated* NaX has revealed very high occupancy of site I′ by sodium ion and may indicate that the result observed here is not abnormal, An alternative argument recognizes that with sodium ions filling the sites I′ of about ⅙ of the sodalite cages (in coordination with tellurium), it is possible that the resulting

TABLE 5
DISTRIBUTION OF FRAMEWORK SPECIES

Species	Location	Number per unit cell
Na(I)	Site I	0
Na(I′)	Site I′	32(3)
Na(II)	Site II	23(2)
Na(II′)	Site II′	9(3)
Na(III)	Site III (undetected)	21
Te(LC)	Large Cavity	3.7(7)
Te(U)	Center of Sodalite Cage	1.3(3)

tetrahedral Na_4Te arrangement establishes a pattern that totally excludes site I occupancy because of the energetically unfavorable situation of simultaneous occupancy of site I and I′. Because not all of the sodium ions of site I′ have the same chemical environment (only $\frac{1}{6}$ of them are bonded to tellurium), there is probably positional disorder in the Na(I′) scattering site. As a result, the estimated standard deviations in the bond lengths involving Na(I′) may be too low so that these bond lengths should be regarded with caution.

The occupancy factor of the Na(II′) ion is only three times its estimated standard deviation and hence the presence of this ion in site II′ should not be accepted without reservation. The close proximity of sites II and II′ rules out their simultaneous occupancy by sodium ion, and suggests a complementarity such that the combined occupancy of these sites by sodium ion should be less than or equal to 32. The observed total of 32 listed in Table 5 thus provides justification for treating the observed occupancy of Na(II′) as real.

The undetected sodium ions listed as occupying sites III are important for the inferred coordination (to be discussed) between them and the tellurium present in the supercage scattering site. In the past, these ions have not been detected by X-ray diffraction and, conventionally, they have been placed at the center of the square faces of the sodalite unit. The study of dehydrated NaX, referred to above (*16*), has shown these ions to be near the edges of the square faces; because their site occupancies are low, these ions were not detected in the present study.

The coordination of the telluride ion by sodium ions of site I′ is presented in Fig. 1. In this respect, the unit is isostructural with the sodalite cages of the nosean sodalite felspathoids where anions (Cl^-, Br^-, OH^-), occluded during crystallization, are also tetrahedrally coordinated to sodium ions. However, the presence of sodium ions in site II′—Na(II′)-Te(U) = 3.3 Å—admits the possibility of an eight-fold coordination of the sodalite-cage telluride ion with the sodium ions of both sites I′ and II′. The site occupancies necessary for this arrangement are within the estimated experimental precision. Finally, passage of the large tellurium species—Te^0 of 1.6 Å radius or Te^{2-} of 2.2 Å radius—through the site II six-ring (0.9 Å radius) would require bond breakage at some point in the ring, and would result in the crystal deterioration mentioned above.

Coordination of the large-cage tellurium is of particular interest because its accessibility to reactant molecules has led to its selection as the probable aromatization site. As Fig. 2 shows, the tellurium is co-

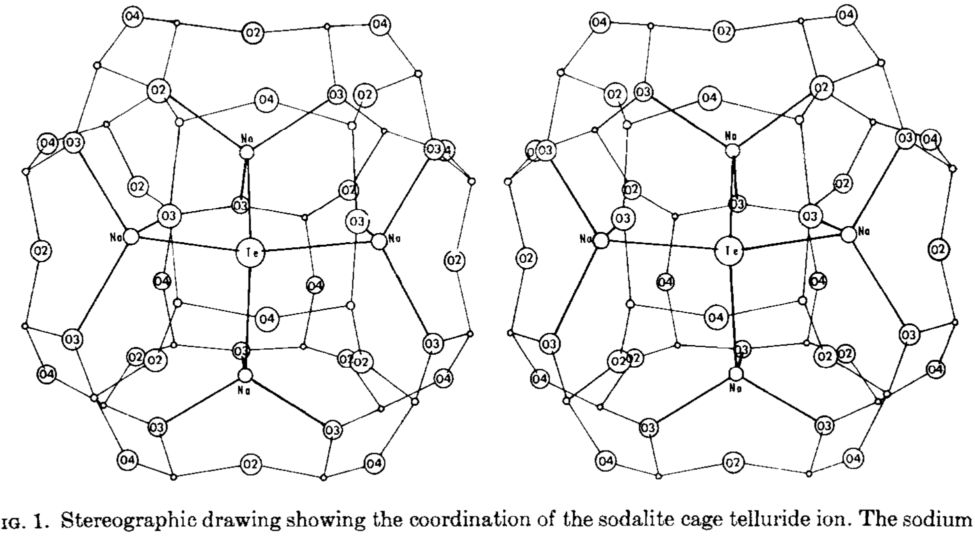

FIG. 1. Stereographic drawing showing the coordination of the sodalite cage telluride ion. The sodium ions shown bonded to Te and 03 are in sites I′.

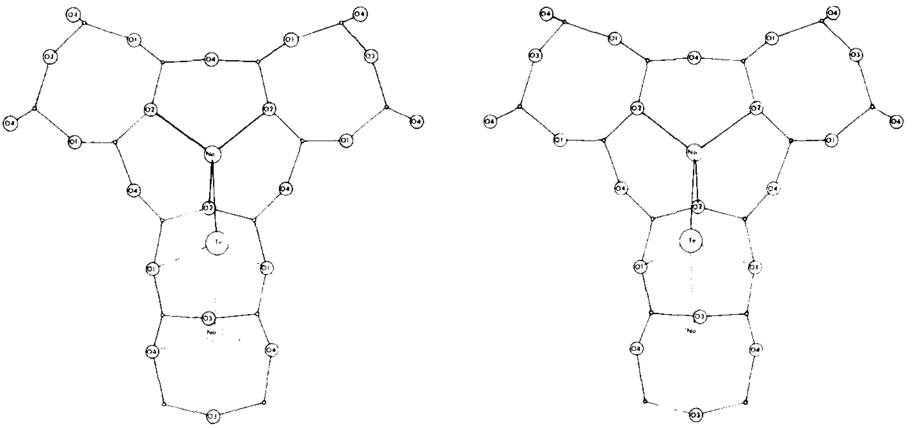

FIG. 2. Stereographic drawing showing the position of the supercage tellurium ion. The supercage and sodalite cage extend above and below the plane of the paper, respectively. Also shown are the sodium ion observed in site II and the sodium ion placed in site III (dotted lines).

ordinated to a Na(II) ion at a distance of 3.2 Å (the sum of the Pauling ionic radii is 3.16 Å). The fact that the tellurium species is considerably displaced from the three-fold symmetry axis strongly suggests that it is also coordinated to at least one additional sodium ion.

An appropriate position for a single additional sodium ion is the *conventional* site III, shown by the dotted circle in Fig. 2 (fractional coordinates; X = Y = 0.375, Z = 0.128). A sodium ion in this site would be 2.4 Å from each of the two 04 oxygen ions and 3.1 Å from the telluride ion, Te(LC); the Na(II)-Te(LC)-Na(III) angle would be 118°. It appears that the formation of the active catalyst stabilizes sodium ions in the conventional site III positions. The source of these additional ions would be those unobserved ions already listed as occupying sites III.

Catalytic Considerations

The salient results of this crystal study have been used previously (*1*) to rationalize the selective aromatization capability of this type of catalyst. However, the results, by establishing a coordination between tellurium and zeolitic cations and hence, most probably, a negative oxidation state for tellurium, strongly suggest the production of protons to retain electrical neutrality and raise questions regarding their generation and accommodation by the crystal lattice.

Coordination of the tellurium in the large cavity to a site II sodium ion and its inferred coordination to a site III sodium ion involve bond lengths equal to the sum of the Na^+ and Te^{2-} ionic radii. The qualifications noted above on the Na(I′) position prevent us from identifying the tellurium within the sodalite cage as telluride ion on the basis of bond length; however, its coordination to four sodium ions makes the assumption of a negative oxidation state reasonable. A negative oxidation state for tellurium was inferred earlier (*1, 14*) from: (1) the observation of the best catalysts being formed in the presence of hydrogen, (2) the establishment, under hydrogen elutant, of a stationary value of the tellurium content that does not exist with helium elutant, and (3) oxidized catalysts undergoing an activation by product hydrogen under reaction conditions.

Olson and Dempsey have deduced (*17*) from a crystallographic study of hydrogen faujasite that cationic hydrogen is bonded to the 01 and 03 oxygen ions. In the Te/NaX system all of the 03 oxygen ions are coordinated to sodium ions in sites I′ which leaves the 01 oxygen ions as the most probable sites for protonic hydrogen produced by the reduction of the tellurium. The 01–H hydroxyl group formed would be expected to produce an absorption band at 3650 cm^{-1} in the ir region (*17–19*). The assignment of cracking activity to this hydroxyl group is based on its accessibility to reactant molecules.

In Fig. 3, we offer a positive graphical indication that hydrogen is chemically involved in forming a Te/NaX catalyst and that it produces an adsorption band at 3650 cm^{-1}. The figure shows the pertinent region of diffuse reflectance spectra of the base sodium X–zeolite and a nominal 6% Te/NaX catalyst. Details of the experiment are given in Appendix A. As can be seen, the presence of tellurium and an *in-situ* hydrogen treatment (17 hr at 540°C in flowing hydrogen) results in an approximately ten-fold increase of the area of the 3650 cm^{-1} absorption band over that observed with the base zeolite.

Although difficult to do definitively, hydrogen sorption experiments have also shown hydrogen uptake by this catalytic system at elevated temperatures. In Appendix B are given the details of an experiment designed to measure the adsorption of hydrogen by a Te/NaX mixture at 510°C with a minimum loss of tellurium from the system. The linear form of the resultant isotherm is presented as Fig. 4. The predicted saturation coverage (reciprocal slope) amounts to 0.19 mmoles hydrogen per g. Chemical analysis of the Te/NaX at the conclusion of the experiment gave a tellurium content of 0.14 milliatoms/g. The formation of telluride ion would require an equality of these two concentrations which differ by ~25%.

The model developed above is also consistent with our knowledge of tellurium retention. The main factor controlling tellurium retention should be the cationic shielding of the tellurium ion from the anionic oxide framework—an effect which would increase with increasing cation size and number of cations. Observations on the

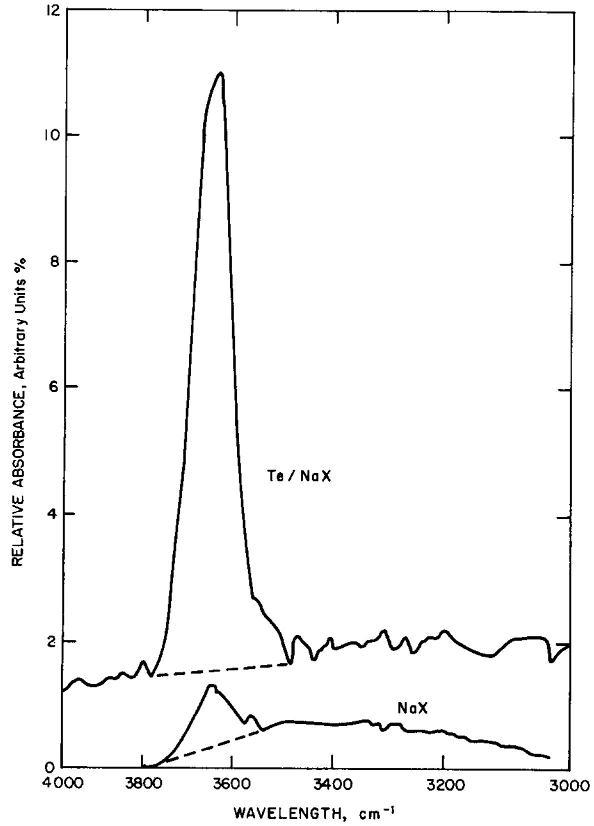

Fig. 3. Diffuse-reflectance spectra for Te/NaX and NaX.

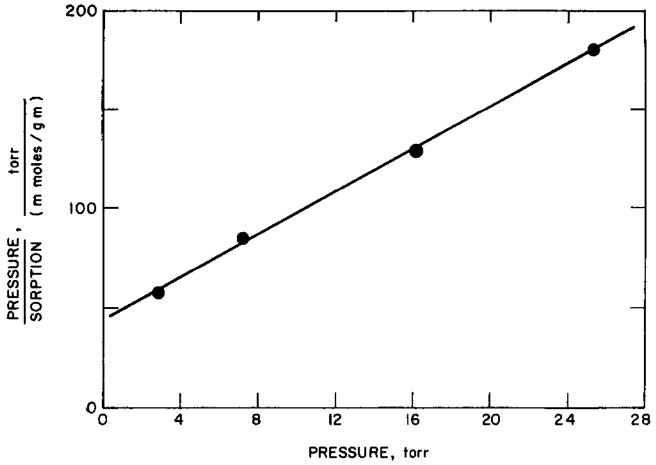

Fig. 4. Hydrogen sorption on Te/NaX.

elution of tellurium from alkali-metal faujasites (*1*, *20*) confirm this expectancy.

The chemical interaction of *both* tellurium and hydrogen with the zeolite and the siting of the resultant species thus generates a satisfactory model for the catalytically active species (*1*). Although there is good qualitative agreement between the model and our knowledge of this system, to date, more quantitative aspects await further work.

Appendix A: Diffuse Reflectance of Te/NaX

Diffuse-reflectance measurements were made with a Perkin–Elmer Spectrophotometer (Model 13U). This dual-beam instrument was equipped with a diffuse-reflectance attachment, also built by Perkin–Elmer Corporation, that directed the incident beam upon a horizontally positioned sample, and collected the diffusely reflected beam so that its intensity could be measured with a cesium bromide thermocouple.

The sample cell was of Vycor glass and was constructed so that the sample container (~10 mm × 3 cm diam) could be reproducibly positioned either in a heated zone or in the ir beam. The sample within the cell could be subjected to high vacuum or gas flow at any time during the experiment.

The sodium X–zeolite was a Linde preparation (13X, L137518, 15.1% Na_2O, 37.8% Al_2O_3, 47.4% SiO_2) and the tellurium-containing catalyst was a nominal 6% tellurium mixture with NaX, that had been ball-milled for 4 hr.

Procedurally, the sample was placed in the heated zone of the cell and subject to hydrogen flow for 17 hr at 540°C. The sample was then moved into the ir beam, allowed to cool and the spectrum recorded while the hydrogen flow continued. The reflectance spectra were referenced to powdered sodium chloride. The reflectance measurements were then converted to absorbance (*21*) and the results plotted as Fig. 3.

Appendix B: Hydrogen Sorption on Te/NaX

The manometric sorption apparatus has been described previously (*22*). The amount of hydrogen adsorbed was measured by monitoring the change in pressure of a constant volume of hydrogen in contact with the catalyst. Pressure measurements were referenced to a constant-pressure vessel by a capacitance manometer and were displayed on a recorder chart.

The catalyst was a nominal 6 wt % Te/NaX mixture that had been ball-milled for 4 hr. The material was treated for 1 hr at 538°C in flowing helium and cooled to room temperature under the helium flow. About 2.4 g were transferred to the sorption cell, which was then evacuated. The temperature of the cell, during evacuation, was raised to 350°C and maintained for 1 hr to produce an ultimate vacuum of 5×10^{-5} Torr. Over the next 45 min the temperature of the cell was raised to 510°C at which point the evacuation was stopped, hydrogen admitted to the cell and pressure monitoring begun. Incremental amounts of hydrogen were subsequently added and the pressure changes recorded over 20–60 min after each addition to ensure reaching equilibrium. After the pressure measurements were made, the catalyst was cooled, removed from the cell, weighed and analyzed for its tellurium content.

The linear form of the Langmuir isotherm was used with the data to produce Fig. 4. Saturation coverage was established as 0.19 mmoles H_2 per g and the tellurium content was found to be 1.8 wt % which corresponds to 0.14 milliatoms/g.

During the evacuation and temperature elevation, tellurium distilled from the sample up onto the cool area of the sorption cell. Previous experience (*1*), however, leads us to expect that contact of the catalyst with hydrogen reduced this loss to essentially zero during the course of the sorption experiment. A control experiment showed that no hydrogen sorption was occurring on the tellurium mirror formed by the distillation.

We, furthermore, assumed that essentially no chemisorption of hydrogen would occur on the NaX base without the involvement of tellurium. This was based on the diffuse-reflectance experiments, described in Appendix A, where the hydroxyl content of the NaX-zeolite was found to be less than 10% of that of the tellurium-containing catalyst under conditions comparable to those of the sorption experiments.

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