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Liquid phase bromination of aromatics over zeolite H-beta catalyst

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Abstract

The liquid phase bromination of chlorobenzene, toluene and xylenes (*o*-, *m*-, *p*-) is catalyzed using zeolite as catalyst and *N*-bromosuccinimide (NBS) as the brominating agent. In addition, the bromination of toluene has been investigated over various zeolites using both NBS and liquid Br_2 as brominating agents. A comparison under similar reaction conditions with H_2SO_4, in the absence of catalyst and $FeCl_3$ (in the case of toluene) is also investigated for each reaction. Zeolite H-beta is found to be selective compared to the conventional catalysts and other zeolites in the bromination of chlorobenzene and toluene whereas selectivity for 4-bromo-*o*-xylene (4-BOX/3-BOX) over H-beta and H_2SO_4 was found nearly comparable in the bromination of *o*-xylene. In the bromination of toluene, acidic H-beta favours the formation of nuclear products whereas in the absence of any catalyst, in the presence of weakly acidic H–Y and potassium exchanged zeolites K-beta and K–L, the concentration of side-chain product, œ-bromotoluene, is enhanced. The conversion of NBS, rate of NBS conversion (mmol g^{-1} h^{-1}) and selectivity for products are strongly influenced by the reaction parameters. As the reaction time, catalyst amount, reaction temperature and molar ratios of NBS/toluene are increased, an increase in the conversion of NBS is noticed. Presumably, the catalytic bromination of aromatics proceeds by an electrophile (Br^+) which is generated by the heterolytic cleavage of NBS/Br_2 by an acidic zeolite. Thus, the generated Br^+ attacks the aromatic ring resulting in the formation of brominated nuclear products. © 1999 Elsevier Science B.V. All rights reserved.

Keywords: Bromination of aromatics; Zeolite H-beta; Bromination of chlorobenzene; Toluene; Xylenes

1. Introduction

Selective bromination of aromatic compounds is investigated in view of the importance of the brominated compounds in organic synthesis [1]. Conventionally, the ring bromination of aromatics is catalyzed by Lewis acids such as the chlorides of aluminum, zinc and iron [2]. Lambert et al. [3] reported on the bromination of benzene and toluene using the NBS–H_2SO_4–H_2O system. This method is unsuitable for the industrial bromination of aromatic compounds due to the need for stoichiometric amount of catalyst, formation of polybrominated products and difficulty in separation of catalyst from the final product. *N*-bromosuccinimide (NBS) is a radical type reagent used in allylic bromination and in aromatic side-chain bromination, also bromine when applied in daylight to, e.g., toluene, will lead to side-chain bromination.

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Now sulphuric acid (in the case of NBS) and metal halides (in the case of bromine) are able to turn the reaction to the ionic mechanism, leading to nuclear substitution. Further, the use of zeolite catalysts inhibit the radical reaction and favours the formation of nuclear substituted products by ionic reaction mechanism.

Mechanistically, it is believed [4–8] that bromination of aromatics proceeds by the generation of an electrophile (Br^+) which is produced by the heterolytic cleavage of NBS/Br_2 by an acidic catalyst. Thus, the generated Br^+ attacks the aromatic ring resulting in the formation of brominated nuclear products [4–8]. The side-chain or α-bromination of aromatics takes place by a radical mechanism [6,9,10].

During the last decade, solid acidic catalysts and in particular, cation-exchange resins in their H^+-form have attracted considerable attention due to important advantages over soluble mineral acids [11,12]. In addition, the bromination of aromatics mediated by silica gel has been reported [1,13–15]. Zeolites are widely used in the petrochemical industry [16,17]. More recently, zeolite catalyzed chlorination of aromatic compounds has attracted much interest owing to the simplicity in the use of zeolites and their shape-selective properties [9,18–24]. However, their use in the bromination of aromatic compounds has been limited [25–34]. In addition, the potential of large pore zeolites like H-beta has not yet been exploited in the bromination of various aromatics. Hence, studies on the catalytic activity of beta zeolites in the bromination of chlorobenzene, toluene and xylenes (*o*-, *m*-, *p*-) with NBS were carried out. The influence of various catalyst modifications, reaction time, brominating agent (NBS/Br_2), catalyst concentration, reaction temperature and molar ratios of NBS/toluene was examined on the conversion of NBS, rate of NBS conversion (mmol g^{-1} h^{-1}) and selectivity for 4-bromotoluene using zeolite H-beta in the bromination of toluene. The results obtained over H-beta were compared with those obtained over conventional catalysts like H_2SO_4 or $FeCl_3$.

2. Experimental

2.1. Materials

Zeolites ZSM-5, beta and K–L were synthesized according to the literature procedures [35–37]. Organic templates were eliminated from the zeolite channels by calcination in dry air at 823 K for 16 h. To convert ZSM-5 and beta into their protonic forms, the zeolites were exchanged thrice with 1 M NH_4NO_3 solution at 353 K for 8 h. Zeolites H-mordenite and H–Y were supplied by Laporte Inorganics, Cheshire, UK.

2.2. Characterization

The SiO_2/Al_2O_3 molar ratios and degree of ion-exchange of the zeolites were determined by a combination of wet atomic absorption methods (Hitachi 800). The crystallinity and phase purity of the zeolites were characterized by X-ray diffraction (XRD) using CuKα radiation. The average particle size of the zeolites were determined using Shimadzu (model UV-2101 PC) scanning electron microscope. The surface area of the zeolites was measured by nitrogen BET method using an Omnisorb 100CX apparatus. The properties of the catalysts are listed in Table 1. Before the reaction, the zeolite samples were dehydrated at 473 K under N_2 for 6 h.

2.3. Catalytic reaction experiments

The bromination reactions were carried out, in a glass reactor, batchwise. An appropriate amount of catalyst, aromatic substrate (0.021), NBS (0.004 mol) and 1,2-dichloroethane (10 g) were stirred at the required temperature. Product samples were analyzed by a gas-chromatograph (Blue star model 421) fitted with flame ionization detector and a capillary column (50 m × 0.2 mm) with methyl silicone gum. Products were identified by GC-MS (Shimadzu-MS QP-2000A) with reference to standard samples.

Table 1
Physico-chemical properties of zeolites

Catalyst	SiO_2/Al_2O_3 (molar ratio)	Degree of H^+-exchange (%)	Crystal size (μm)	Surface[a] area (m^2 g^{-1})
H-beta	26.0	98.7	0.5	745
H-ZSM-5	41.0	98.1	0.5	413
H-mordenite	22.0	98.0	1.0	552
HY	4.1	98.2	1.0	615
K–L	6.8	–	0.2	215
K-beta	26.0	9.8	0.5	743

[a] Measured by N_2 adsorption.

Conversion (and bromine consumption) is reported as the wt.% of NBS consumed. The rate of NBS/Br_2 conversion (mmol g^{-1} h^{-1}) indicates moles of NBS/Br_2 converted per gram of catalyst in an hour.

3. Results and discussion

Table 1 lists the SiO_2/Al_2O_3 ratio, degree of H^+-exchange (%), crystal size and surface areas of the various zeolite catalysts used in the bromination reactions. XRD studies indicated high crystallinity and phase purity in all the samples. XRD examination also gave no evidence of structure change of the zeolite sample after various cation exchange. The surface area and scanning electron micrographs revealed well defined materials without any occluded amorphous matter in the zeolites.

3.1. Bromination of toluene

3.1.1. Comparison of various catalysts: bromination with NBS

The bromination of toluene with NBS over H-ZSM-5, H-mordenite, H-beta, H–Y, conventional catalyst H_2SO_4 and in the absence of catalyst yielded 2- and 4-bromotoluenes (2-BT and 4-BT,respectively) as the main products and the side-chain brominated α-bromotoluene (α-BT) as the side product. The absence of consecutive products even in the presence of H_2SO_4 may be attributed to the lower reaction temperature. In addition, strong acid catalyst (H_2SO_4) does not favour the formation of side-chain halogenated products [18,20,31]. The formation of 2- and 4-monobromotoluenes takes place by parallel reactions [21,31]. Table 2 compares the activities of catalysts under identical reaction conditions. Zeolite H-beta and H_2SO_4, produced predominantly nuclear brominated products. The blank experiment (no catalyst), zeolites H-ZSM-5, H-mordenite and H–Y produced mainly the side-chain brominated product α-bromotoluene in the bromination of toluene. It is reported that NBS is a reagent which selectively brominates allylic and benzylic positions [10]. However, a number of exceptions have been reported where NBS was shown to brominate nuclear rather than benzylic positions [4,5]. In addition, the side-chain bromination of toluene in the absence of any catalyst may be attributed to a free radical mechanism [6,9].

The results in Table 2 indicate that zeolite H-beta is more active and selective (4-BT/2-BT = 5.5) for nuclear bromination of toluene compared to other zeolite catalysts. The acidic H–Y gave a higher rate of NBS conversion (3.62 mmol g^{-1} h^{-1}) for the side-chain product. The conventional catalyst, H_2SO_4 was found to be more active (rate of NBS conversion = 2.68 mmol g^{-1} h^{-1}) but less selective (4-BT/2-BT = 1.3) than H-beta. Among the zeolites H–Y and H-beta, the largest yield of side-chain product (α-BT) was observed over H–Y zeolite. It is reported [9] that silyloxy

Table 2
Bromination of toluene[a]

Catalyst	Brominating[b] agent	Conversion of NBS/Br_2 (wt.%)	Rate of NBS[c] or Br_2 conversion (mmol g^{-1} h^{-1})	Product distribution (wt.%)[d]			4-BT/2-BT[e]
				2-BT	4-BT	α-BT	
H_2SO_4	NBS	49.5	2.68	42.4	53.5	4.0	1.3
No catalyst	NBS	4.0	–	–	–	100.0	–
H-ZSM-5	NBS	4.5	0.24	11.1	22.2	66.7	2.0
H-mordenite	NBS	3.5	0.19	–	–	100.0	–
H-beta	NBS	41.5	2.24	14.5	79.5	6.0	5.5
H–Y	NBS	67.0	3.62	2.3	3.0	94.7	1.3
$FeCl_3$	Br_2	100.0	5.38	47.0	53.0	–	1.1
No catalyst	Br_2	100.0	5.38	7.0	11.5	81.5	1.6
H-beta	Br_2	76.5	4.11	15.7	63.4	20.9	4.0
K-beta	Br_2	85.2	4.57	7.6	17.0	75.4	2.2
K–L	Br_2	40.0	1.93	15.0	31.3	53.8	2.1

[a] Reaction conditions: catalyst = 18.4 g mol^{-1} of toluene; reaction temperature = 353 K; toluene = 0.02 mol; NBS = 0.0043 mol; Br_2 = 0.0043 mol; reaction time = 2 h.

[b] NBS = *N*-bromosuccinimide; Br_2 = liquid bromine.

[c] Rate of NBS or Br_2 conversion is defined as the amount (mmol) of NBS or Br_2 converted per gram per hour of catalyst.

[d] 2-BT = 2-bromotoluene; 4-BT = 4-bromotoluene; α-BT = α-bromotoluene (benzyl bromide).

[e] Ratio of 4-BT/2-BT.

radicals are produced by the homolytic opening of $O_3SiOAlO_3$ structure of zeolites into $O_3SiO^{\cdot} + O_3Al^{\cdot}$. These radicals initiate the radical chain reactions. The highest concentration of such centres is expected with aluminum rich zeolites (in H–Y compared to the H-beta) and hence the higher amount of α-BT is observed over H–Y than that of H-beta.

3.1.2. *Comparison of various catalysts: bromination of toluene with Br_2*

Liquid bromine was also employed as the brominating agent instead of NBS in the bromination of toluene. The results of the conversion of bromine, rate of bromine conversion (mmol g^{-1} h^{-1}), product distribution and isomer ratio (4-BT/2-BT) obtained during bromination of toluene over zeolite H-beta, K-beta and K–L are presented in Table 2. The reaction was also carried out using conventional catalyst, $FeCl_3$, and in the absence of any catalyst for comparison. Zeolite H-beta was found to be the most active (4.11 mmol g^{-1} h^{-1}) and selective (4-BT/2-BT = 4.0) among zeolite catalysts, in the nuclear bromination of toluene. Zeolites K-beta, K–L and the blank experiment (no catalyst) favoured the formation of the side-chain product, α-BT. The higher amount of nuclear product over H-beta compared to K-beta and K–L may be attributed to its acidic nature. The Lewis acid catalyst, $FeCl_3$, was found to be slightly more active (5.38 mmol g^{-1} h^{-1}) but less selective (4-BT/2-BT = 1.1) than H-beta.

Zeolite H-beta performed better than the other zeolites and conventional catalysts, H_2SO_4 and $FeCl_3$, in the bromination of toluene with NBS. The influence of various process parameters on this reaction are discussed, using H-beta, in the following sections.

3.1.3. *Influence of reaction time: bromination of toluene with NBS*

Fig. 1 shows the time-dependence of the conversion of NBS (wt.%) on H-ZSM-5, H-mordenite, H-beta, H–Y, H_2SO_4 and in the absence of catalyst under similar reaction conditions. H-beta showed a lower activity compared to H_2SO_4. Zeolites H-ZSM-5 and H-mordenite were considerably less active than H-beta in the bromination of toluene.

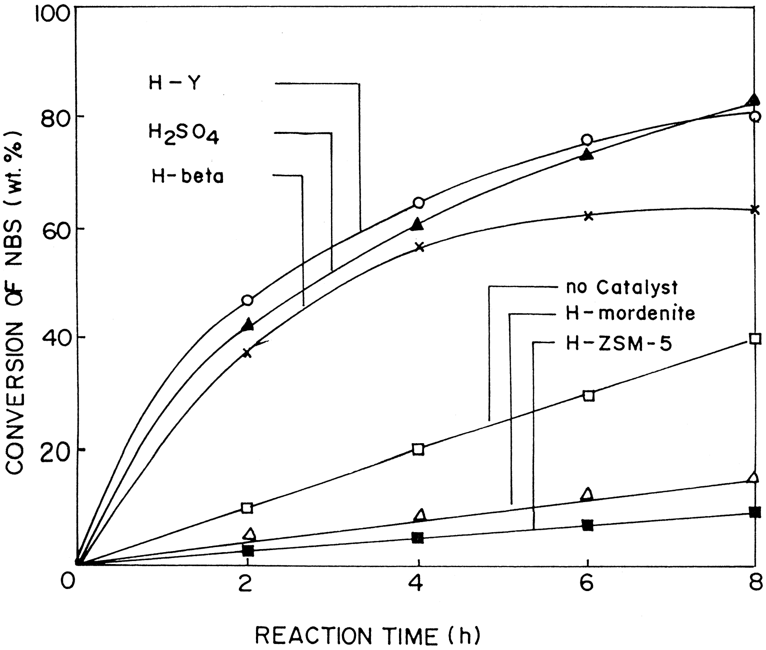

Fig. 1. Conversion of NBS over various catalysts as a function of reaction time (h). Reaction conditions: catalyst (g mol^{-1} toluene) = 19.04; reaction temperature (K) = 353; toluene (mol) = 0.021; NBS (mol) = 0.004, 1,2-dichloroethane = 10 g.

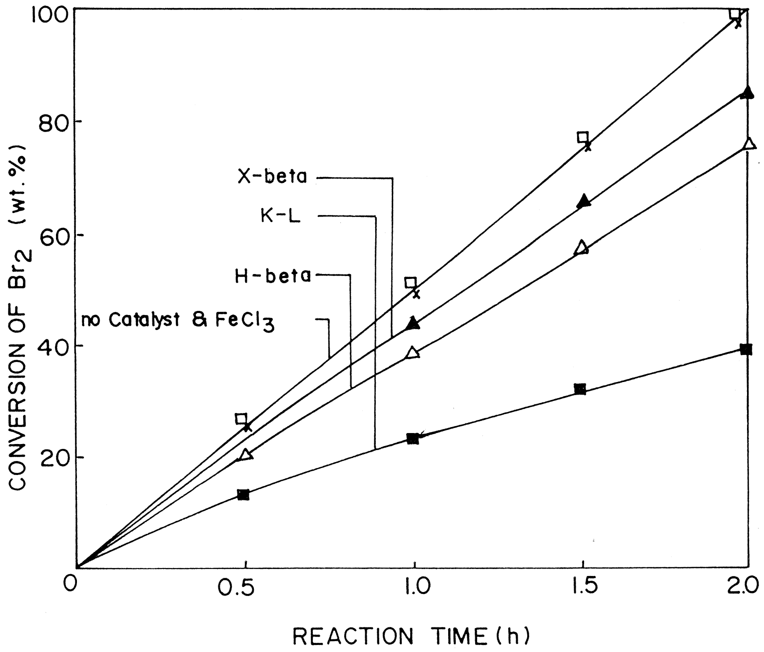

Fig. 2. Conversion of Br_2 over various catalysts as a function of reaction time (h). Reaction conditions: catalyst (g mol^{-1} toluene) = 19.04; reaction temperature (K) = 353; toluene (mol) = 0.021; Br_2 (mol) = 0.004, 1,2-dichloroethane = 10 g.

3.1.4. Influence of reaction time: bromination of toluene with Br_2

The results are illustrated in Fig. 2. The conventional catalyst, $FeCl_3$, was found to be more active than the zeolite catalysts.

3.1.5. Influence of catalyst concentration

Fig. 3 shows the effect of concentration of zeolite H-beta on the conversion of NBS, product selectivity and 4-BT/2-BT ratio in the bromination of toluene. The catalyst to NBS ratio was changed by keeping the concentration of NBS constant in the reaction mixture. When the catalyst to NBS ratio was increased from 23.3 to 69.8 g mol^{-1} of NBS, the conversion of NBS increased from 7.0 to 38.0 wt.%, respectively. Above this ratio the conversion of NBS leveled off. However, the ratio of 4-BT/2-BT is not influenced to a significant extent by the change in catalyst to NBS ratio. œ-Bromotoluene was the main side-chain product in the absence of any catalyst. These results confirm that H-beta catalyzed mainly the ring bromination of toluene.

3.1.6. Influence of reaction temperature

The temperature dependence of NBS conversion and product yields in the bromination of toluene is shown in Fig. 4. The conversion of NBS over H-beta increased steadily from 8.5 to 41.5 wt.% as the temperature was raised from 333 to 353 K. In addition, an increase in the reaction temperature increased the formation of 4-BT and decreased the concentration of 2-BT and side-chain product, α-BT, in the reaction mixture. The isomer ratio (4-BT/2-BT) increased steadily from 1.1 to 5.5 with the increase in reaction temperature from 333 to 353

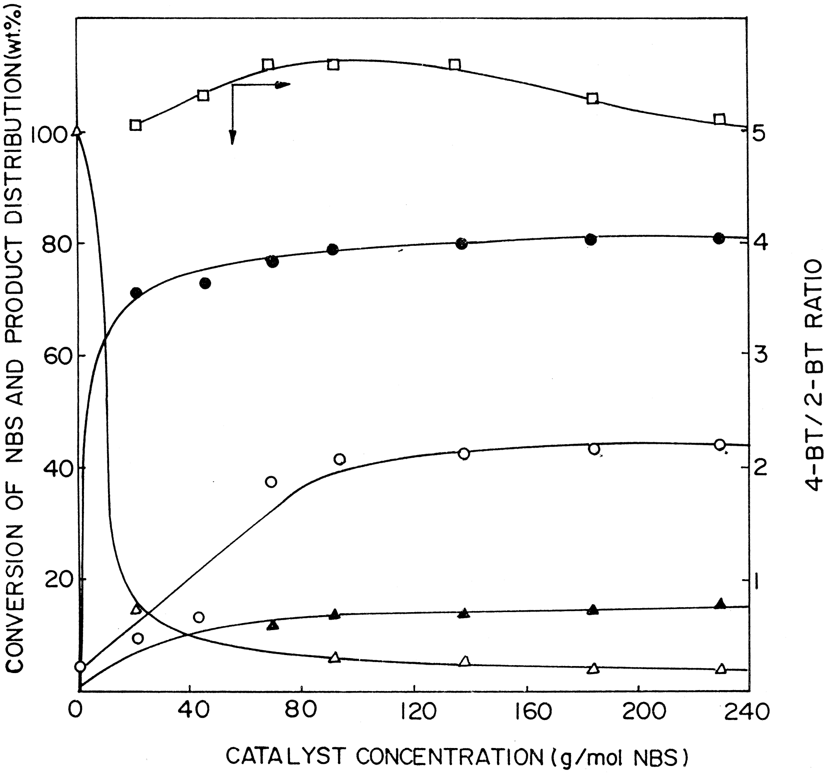

Fig. 3. Effect of catalyst H-beta concentration on the NBS conversion (○), and product yields, 4-BT (●), 2-BT (▲), œ-BT (△), 4-BT/2-BT (□). Reaction conditions: reaction temperature (K) = 353; toluene (mol) = 0.021; NBS (mol) = 0.004; 1,2-dichloroethane = 10 g.

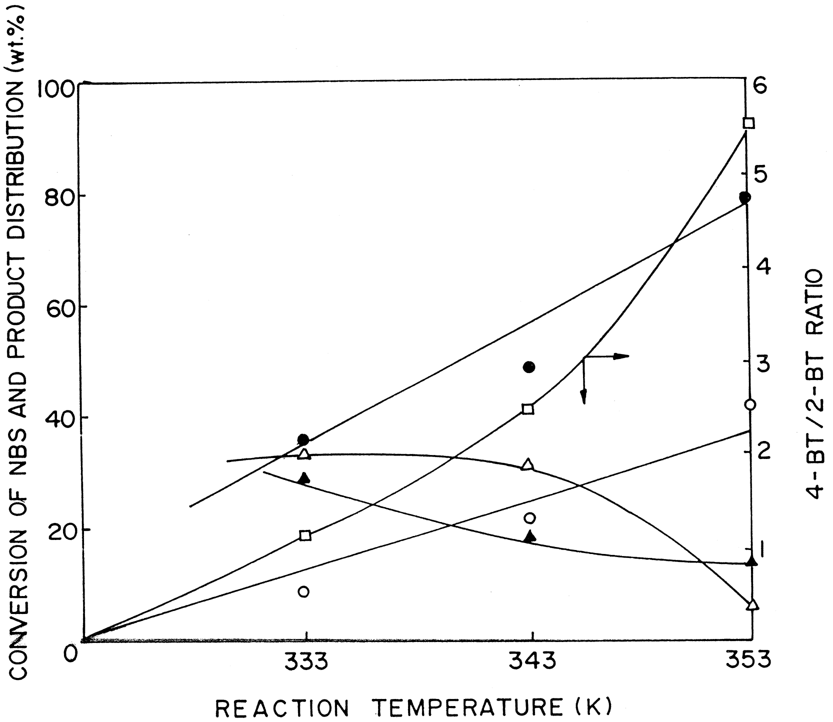

Fig. 4. Effect of reaction temperature on the NBS conversion (○), And product yields, 4-BT (●), 2-BT (▲), œ-BT (△), 4-BT/2-BT (□). Reaction conditions: toluene (mol) = 0.021; NBS (mol) = 0.004; 1,2-dichloroethane = 10 g; zeolite H-beta (g mol^{-1} toluene) = 19.04.

K, respectively. These results are consistent with the earlier reports [22].

3.1.7. Influence of toluene / NBS molar ratio

The (toluene/NBS) ratios were changed by keeping the amount of toluene constant. The increase in toluene/NBS ratio up to 8 gave a linear rise in the NBS conversion (Fig. 5). The conversion of NBS increased from 8.5 to 53.1 wt.% as the toluene/NBS molar ratio increased from 2 to 8. The selectivity for the products was also influenced by the toluene/NBS molar ratio (Fig. 5).

3.2. Bromination of chlorobenzene

H-beta was catalytically active also in the bromination of chlorobenzene. The results were compared with those of the conventional catalyst H_2SO_4. The reaction conditions and the results are summarized in Table 3. The main products of the reaction were 4-bromo-chlorobenzene (4-BCB) and 2-bromo-chlorobenzene (2-BCB). H-beta is less active than H_2SO_4. A blank run (without any catalyst) failed to give any conversion of chlorobenzene.

3.3. Bromination of xylenes (o-, m-, p-) over H-beta

The bromination of xylenes (*o*-, *m*- and *p*-) was investigated in the presence of 1,2-dichloroethane using zeolite H-beta at 353 K. The conversion of NBS, rate of NBS conversion, product distribution and isomer ratio obtained over H-beta, H_2SO_4, and in the absence of any catalyst, for all xylenes are included in Table 4. The bromination of xylenes with NBS over H-beta leads to nuclear as well as side-chain products whereas H_2SO_4 gave mostly nuclear brominated products. Higher yields of secondary products was also obtained over H_2SO_4.

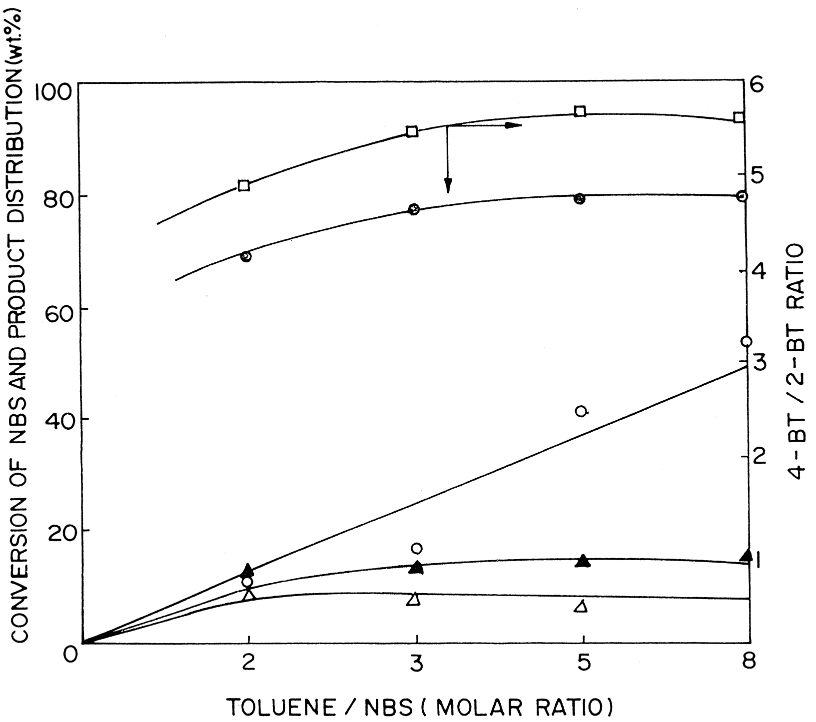

Fig. 5. Effect of toluene/NBS molar ratio on the NBS conversion (○), and product yields, 4-BT (●), 2-BT (▲), œ-BT (△), 4-BT/2-BT (□). Reaction conditions: reaction temperature (K) = 353; 1,2-dichloroethane = 10 g; zeolite H-beta (g mo^{-1} toluene) = 19.04.

In the absence of any catalyst, only the side-chain brominated products from the corresponding xylenes were obtained. The bromination of *o*-xylene over zeolite H-beta leads to the formation of 3-bromo-*o*-xylene (3-BOX), 4-bromo-*o*-xylene (4-BOX) and α-bromo-*o*-xylene. With m-xylene, the reaction gives 5-bromo-*m*-xylene (5-BMX), 2-bromo-*m*-xylene (2-BMX), 4-bromo-*m*-xylene (4-BMX) and α-bromo-*m*-xylene (α-BMX). Bromination of *p*-xylene gave monobrominated nuclear product, 2-bromo-*p*-xylene (2-BPX) and side-chain product, α-bromo-*p*-xylene. The product ratios of 4-BOX/3-BOX and 4-BMX/2-BMX over H-beta were found to be 5.14 and 2.73, respectively.

4. Conclusions

In the liquid phase bromination of toluene using NBS as the brominating agent, zeolite H-beta is more active in the formation of nu-

Table 3
Bromination of chlorobenzene[a]

Catalyst	Conversion of NBS (wt.%)[b]	Rate of NBS[c] conversion (mmol g^{-1} h^{-1})	Product distribution[d] (wt.%)		4-BCB/2-BCB[e]
			4-BCB	2-BCB	
H_2SO_4	34.5	1.86	62.3	37.7	1.65
H-beta	11.2	0.61	87.5	12.5	7.0

[a] Reaction conditions: catalyst = 0.4 g; NBS = 0.77 g; chlorobenzene = 0.021 mol; solvent (1,2-dichloroethane) = 10 g; reaction temperature = 353 K; reaction time = 2 h.

[b] NBS = *N*-bromosuccinimide.

[c] Rate of NBS conversion (mmol g^{-1} h^{-1}) = mass of NBS converted/mass of catalyst used × reaction time (h^{-1}).

[d] 4-BCB = 4-bromochlorobenzene; 2-BCB = 2-bromochlorobenzene.

[e] 4-BCB/2-BCB = isomer ratio.

Table 4
Bromination of xylenes[a]

Xylenes	Catalysts	Conversion[b] of NBS (wt.%)	Rate of NBS[c] conversion (mmol g^{-1} h^{-1})	Product distribution (wt.%)[d]										4-BOX/3-BOX ratio	4-BMX/2-BMX ratio
				3-BOX	4-BOX	œ-BOX	5-BMX	2-BMX	4-MBX	œ-BMX	2-BPX	œ-BPX	Others		
o-Xylene	None	71.5	–	–	–	100	–	–	–	–	–	–	–	–	–
	H_2SO_4	61.5	3.32	12.2	74.0	3.2	–	–	–	–	–	–	10.6	6.06	–
	H-beta	51.5	2.78	4.9	25.2	69.9	–	–	–	–	–	–	–	5.14	–
m-Xylene	None	12.5	–	–	–	–	–	–	–	100	–	–	–	–	–
	H_2SO_4	45.0	2.43	–	–	–	2.2	4.4	90.0	–	–	–	3.4	–	20.45
	H-beta	63.0	3.40	–	–	–	9.5	8.7	23.8	57.9	–	–	–	–	2.73
p-Xylene	None	9.0	–	–	–	–	–	–	–	–	–	100	–	–	–
	H_2SO_4	52.0	2.81	–	–	–	–	–	–	–	81.7	–	18.3	–	–
	H-beta	16.5	0.89	–	–	–	–	–	–	–	60.6	39.4	–	–	–

[a] Reaction conditions: catalyst = 0.4 g; NBS = 0.004 mol; xylene = 0.021 mol; solvent (1,2-dichloroethane) = 10 g; reaction temperature = 353 K; reaction time = 2 h.

[b] NBS = *N*-bromosuccinimide.

[c] Rate of NBS conversion (mmol g^{-1} h^{-1}) = mass of NBS converted/mass of catalyst used × reaction time (h^{-1}).

[d] 3-BOX = 3-bromo-*o*-xylene; 4-BOX = 4-bromo-*o*-xylene; œ-BOX = œ-bromo-*o*-xylene; 5-BMX = 5-bromo-*m*-xylene; 2-BMX = 2-bromo-*m*-xylene; 4-BMX = 4-bromo-*m*-xylene; œ-BMX = œ-bromo-*m*-xylene; 2-BPX = 2-bromo-*p*-xylene; œ-BPX = œ-bromo-*p*-xylene; others = consecutive products.

clear and para-selective brominated products compared to the other zeolites studied. The higher activity and selectivity may be due to its higher acidity, three dimensional channel structure and large pore openings. The conventional catalyst H_2SO_4 is more active but poorly para-selective compared to H-beta. Similarly, H-beta is a more para-selective but less active catalyst for the bromination of toluene compared to the conventional catalyst $FeCl_3$ when liquid bromine is used as the brominating agent. In the liquid phase bromination of chlorobenzene, using NBS as the brominating agent, zeolite H-beta is more selective in the production of 4-bromochlorobenzene compared to the conventional catalyst H_2SO_4. Bromination of xylenes over H-beta gave both nuclear and side-chain brominated products whereas H_2SO_4 gave mostly the nuclear products.

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