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Micromechanical compressive response of a zeolite single crystal

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In this work the mechanical properties of a widely used zeolite catalyst, ZSM-5, are measured. Single crystals of the ZSM-5, of average size 500 microns, are prepared using an organic template molecule. Testing is performed on the single crystal size scale. An in-house microdeformation tester is designed and manufactured. Loading on the crystal is provided by a high precision stepper motor. Both crystal displacement and applied load are continually monitored through a computer. The entire testing assembly is placed under an optical microscope and a CCD camera is used to obtain real time optical images of the deformation. In this way monotonic and cyclic loading compression stress strain curves are generated. The material is seen to have an average elastic modulus of 4 GPa. It is also seen to be brittle and undergo a gradual stiffness deterioration through a microcracking process. Crystal anisotropy is also investigated by performing two directional tests. However, for the [100] and [010] directions little mechanical anisotropy is observed.

1. Introduction

Aluminosilicate zeolites are widely used in many chemical applications, such as catalysis, ion exchange and gas separation. Zeolites are microporous inorganic compounds which contain small pores from about 3 Å to over 10 Å in diameter [1]. The crystal structure of zeolites comprises a three-dimensional periodic framework formed by linked silicate or aluminate tetrahedra (SiO_4 or AlO_4). The framework contains channels that may be one, two or three dimensional depending on the particular zeolite. Among zeolites, ZSM-5 is one of the most important and is used in a variety of catalytic applications in industry [2]. Fig. 1 shows the framework of ZSM-5 from two different viewpoints. From Fig. 1 it is evident that ZSM-5 has an intersecting two dimensional channel system along the [010] and [100] directions. The channel diameter in this zeolite is about 0.55 nm. ZSM-5 has an orthorhombic structure with lattice parameters $a = 2.01$ nm, $b = 1.99$ nm and $c = 1.34$ nm [3].

Typically ZSM-5 is used as a heterogeneous catalyst and it is activated and re-used many times. In many applications a continuous decrease of catalytic activity is observed with continued use, a fact which eventually leads to replacement of the catalyst at additional cost. Many factors can influence the decrease of catalytic activity observed during these industrial processes, one of them being the result of mechanical, as opposed to chemical, degradation of the crystal structure. It is therefore of interest to obtain a modeling capability that will be able to predict the mechanical degradation of ZMS-5.

The ZSM-5 material is relatively brittle and prone to microcracking. Thus, it may be possible to apply some existing microcracking models [4, 5] to understand the response of ZSM-5. As a starting point, however, detailed knowledge of the mechanical properties of the material is needed. The few studies to date that have focused on the mechanical response of zeolites are concerned either with hardness testing [6] or powder response during ball milling [7]. The objective of this work, however, is to characterize the mechanical response of ZSM-5 and to investigate the failure mechanisms that the material undergoes. An important feature of this work is that testing is conducted on *single crystals* of ZSM-5. To facilitate testing, crystals

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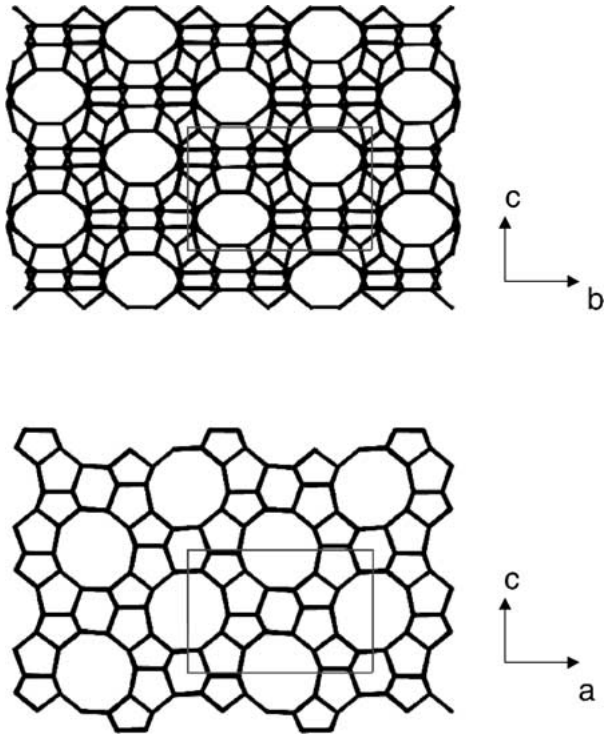


Figure 1 Framework structure of ZSM-5 along [100], top, and [010] directions.

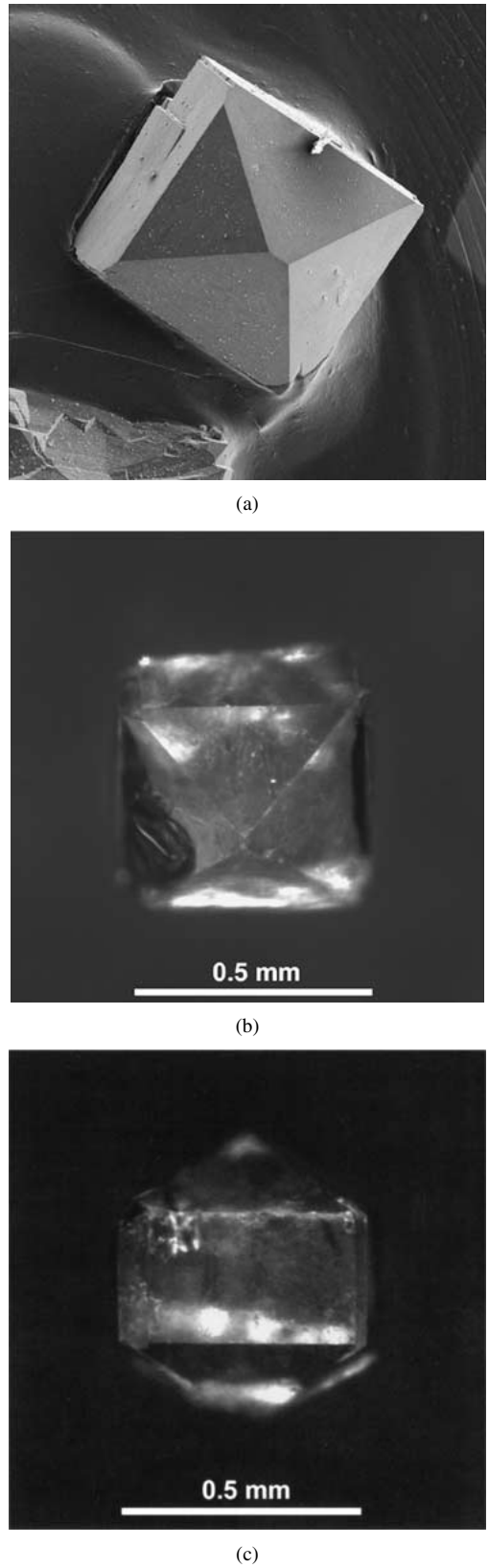
of about 0.5 mm are used. This is about 100 times larger than the commercially available catalyst. In addition to monotonic loading on the crystal, cyclic loading will be used since performance degradation is mainly seen after repeated use of the catalyst.

Mechanical testing of brittle single crystals of the size scale used here cannot be done on large scale commercially available mechanical testing equipment. Thus, to test single crystals in monotonic and cyclic loading a custom microdeformation tester was designed and constructed in-house. The device is broadly related, and is in one sense complementary, to the novel experimental technique developed in [8], which was one of the earliest studies to investigate mechanical properties of highly brittle single crystals. The details of the device constructed in the present study are presented in Section 2. Section 3 presents some typical results from compression tests of ZSM-5 single crystals. The findings of the study are then summarized in Section 4.

2. Experiments

2.1. Material preparation

The synthesis of large ZSM-5 single crystals was based on the methodology reported by [9]. The crystal framework is formed of SiO_2 only and an organic template molecule (Tetrapropylammonium fluoride) is occluded within the crystal channels. Thus, in the as-synthesized state the zeolite pores are occupied by organic guest molecules. These molecules are subsequently removed from the interior by following a two step procedure involving a treatment with a liquid oxidant (aqueous H_2O_2 or aqueous $NaClO_4$) and heating in air. More details about specimen preparation and the protocol for organic molecule removal can be found in [10]. In the subsequent experiments both the as-synthesized (non-porous) and the porous crystals were tested.



(a)

(b)

(c)

Figure 2 (a) An scanning electron micrograph and (b) and (c) optical photographs of a ZSM-5 single crystal.

The ZSM-5 single crystals used in this study have average dimensions about 500 to 800 microns. Three images of a single crystal, one from an SEM and two from an optical microscope, are shown in Fig. 2a, b and c. High quality single crystals are transparent and have smooth surfaces (Fig. 2b and c). Crystals that are not transparent contain pre existing faults that were generated during the preparation process. Such defective

crystals were not tested in this study since they had considerably lower strengths than the undamaged samples. However, it might be of interest in the future to obtain a statistical variation of crystal strength as a function of pre-existing flaws within it. Most crystals show other types of intrinsic defects, such as the ledge visible on the crystal's surface in Fig. 2a. These types of crystals were tested and any effect of the ledge was ignored in the analysis.

As can be seen in Fig. 2a the crystal consists of a rectangular parallelepiped with two square pyramidal caps. It is well know that these crystals often twin along the ⟨110⟩ directions [11]. Since the crystal has an orthorombic structure it will exhibit mechanical anisotropy. Therefore tests in all three directions should be conducted. However, note that since the zeolite framework along the a and b directions is very similar we expect the mechanical response along the [100] and [010] directions also to be similar.

2.2. Microdeformation tester: design and features

As was mentioned in the introduction, use of large scale mechanical testing equipment is not suitable for testing the ZSM-5 crystals. As shown in Fig. 2, the dimensions of these crystals are less than 1mm. One of the earliest studies to tackle the technical problems of mechanical testing of brittle crystals was by Chaudhri [8]. In that work, highly brittle and explosive crystals, of size similar to the crystals used in the present study, were compressively loaded to yield using a custom made indentation device, which also allowed for optical monitoring of the contact area between sample and loader through a transparent substrate. Loading was carefully controlled to minimize the amount of cracking, and measurements were made during loading and unloading of the crystals. As is discussed in [8], by using appropriate indentor geometry and load control, in certain instances brittle cracking was suppressed and crystal yield was possible. This was confirmed through the optical contact area information obtained during the loading-unloading.

The eventual goal of the present study is to produce a model that would account for damage occurring during in service use of the ZSM-5 catalyst. In this sense we would seek as complete a material description of the crystal as possible. In the present work a custom microdeformation tester is constructed, as discussed below, to obtain the brittle response of the crystals, i.e., investigate the elastic response of the material and its susceptibility to damage. The next step towards modeling the brittle response would be some type of statistical failure model, such as for example the Weibul type model as used in [12], that would account for the probability of pre-existing flaws. However, in instances were the deformation of the catalyst may be severe, then information close to material yield will be required. To obtain such information one would need to apply a loading technique similar to that of [8]. Thus, the present work provides a complementary testing method to that described in [8]. For a complete characterization of the material it would be optimal to undertake both types of experiments.

In more recent years considerable work has been done in developing techniques that can be used to measure mechanical properties at small size scales. For example, Yuan and Sharpe [13] and LaVan [14] have tested small scale tensile specimens of a metal weldment. In their work specimens with gage lengths as small as 1.5 mm were used. Load was applied using a piezoelectric actuator and an optical technique was used to measure specimen strain. A similar loading method was used in Shield [15] to study strain fields in the vicinity of a notch tip, but there full field optical information was obtained using a charged coupled device (CCD) camera. In addition, with the expanding interest in MEMS structures, several researchers have performed experimental testing of structures at even smaller size scales [16, 17]. In all cases the main elements that are needed are a method of applying load and precise measurements of this applied load and the resulting elongation or deflection.

The size scale of samples tested in this study is somewhere between the sizes tested in the above mentioned works. A microdeformation tester based on these principles was designed and built in-house. Loading is applied by means of a stepper motor. Load is continually monitored with a high precision load cell, and sample deformation is recorded using an Linear Variable Differential Transformer (LVDT) displacement transducer. The deformation can also be clearly observed and recorded in real time by means of a microscope and a CCD camera. All systems, i.e. motion control, optical visualization and data collection (load and deflection measurement), are controlled by a computer. Fig. 3 shows a flow chart of the entire system with the load, visualization and deformation modules shown on the left, center and right of the figure respectively.

A schematic of the microtester is shown in Fig. 4. Compressive loading is applied to the sample through a high-resolution stepper motor actuator (8.5 nm step size). The load value is transmitted from a load cell to the computer by an in-line amplifier through either the RS-232 port or a data acquisition (DAQ) board. Deformation of the sample is measured by an LVDT attached to the grips as shown in Fig. 4. The LVDT output voltage is also acquired by a DAQ board and is then converted into displacement through an experimental calibration. The entire set-up is placed under

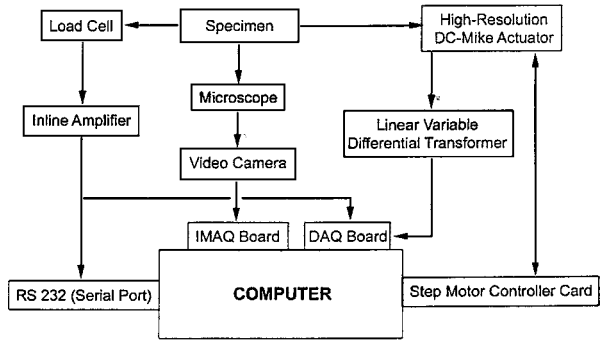


Figure 3 Flow chart of microtester modules. Displacement actuation and measurement is on the right hand side, optical imaging in the center and load recording on the left hand side. All three modules are controlled by a Personal Computer.

TABLE I Components and specifications of microdeformation tester

Component	Model/Manufacturer	Specification
Load Cell	Model 31/Sensotec	Loading capability: 220 N (compression/tension)
DC-Mike actuator and controller	M-224. 50 with C-842.20 board/Physik instrumente	Travel range: 25 mm Design resolution: 8.5 nm Maximum velocity: 1.5 mm/sec Maximum push/pull force: 100 N
LVDT	MHR 500/Schaevitz	Nominal linear range ±12.7 mm
DAQ board	PCI-MIO-16E-4/National instrument	250 kS/s sampling rate 12-bit resolution
Microscope	SZX-12/Olympus Stereo microscope	Maximum magnification: 90
Frame grabber	PCI-1408 monochrome IMAQ board/National instruments	Gray level: 256 (8-bits) Progressive scan: up to 30 frames/sec interlaced

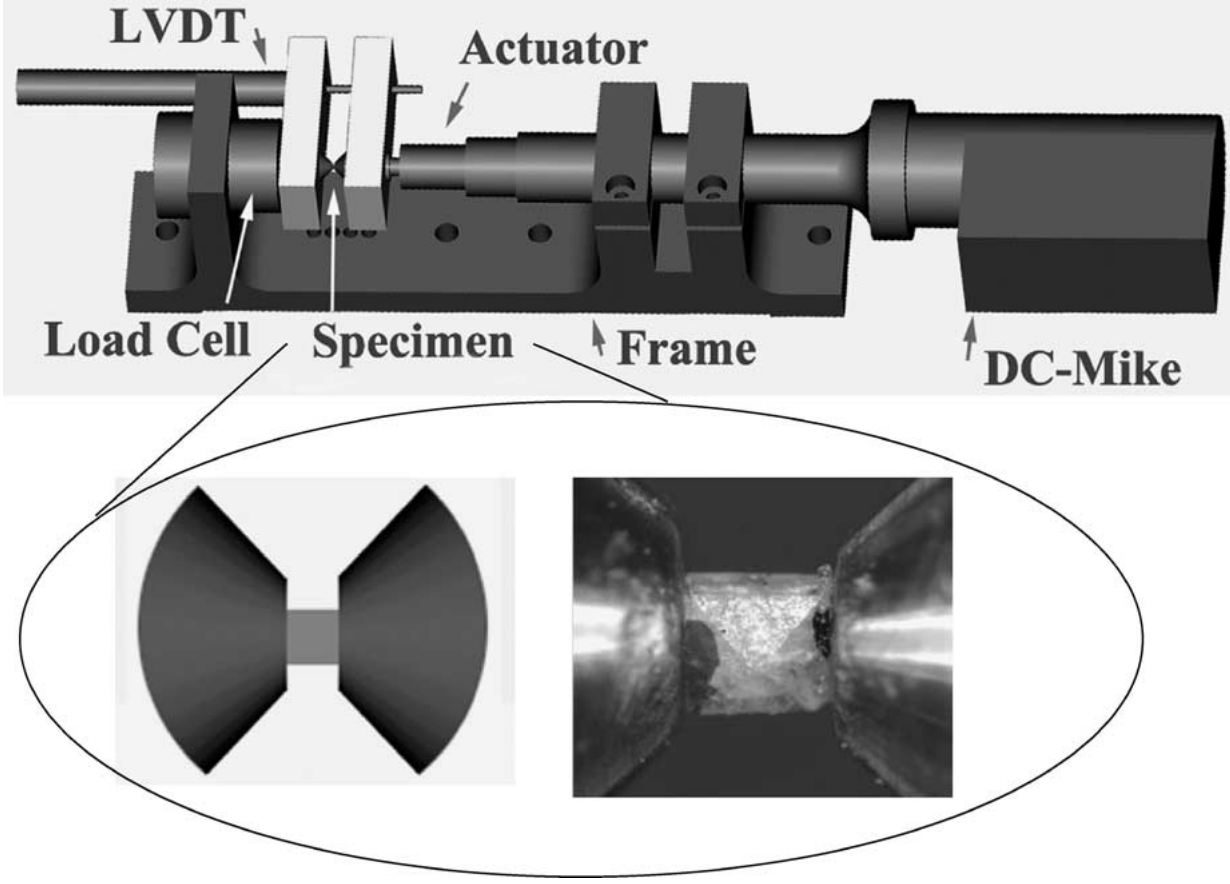


Figure 4 Schematic of microdeformation tester. Loading is provided by the DC-mike stepper motor and is recorded by the load cell. The LVDT is used to measure relative displacement of the grips. The two insets (a schematic and a photograph) show the mounted specimen.

an optical microscope. With a CCD camera attached to the microscope, deformation of the sample under loading can be recorded as a series of images captured by an image acquisition board (IMAQ). The images can then be composed into a continuous movie for reviewing the deformation process. Since load, displacement and images are all recorded on a common time base, it is possible to correlate specific images to corresponding load-displacement information. The major components used in the microdeformation tester are listed in Table I. For simplicity of machining and accuracy of assembling, a single piece steel base was designed and machined to support the set-up (Fig. 4).

To load the sample in compression a pair of cone shaped adapters were attached to the load cell and the actuator, and the sample was carefully placed between them. The blow up in Fig. 4 shows a schematic (left) and an image (right) of the mounted specimen. The specimen mounting operation was done under the microscope. No special specimen surface preparation was made, although the as-synthesized crystal surfaces were in most cases quite smooth (see Fig. 2a). Note, however, that development of frictional forces at the grip/specimen contact surface may affect the result of the mechanical test [18]. This is an important consideration in all compression testing, but especially with brittle materials where low load levels are expected. As seen in Fig. 2, the aspect ratio (length to width ratio) of these crystals is about 1.0, which according to ASTM standard E9 [18] is in the range that helps minimize effects due to friction. In addition, a small amount of lubricant was added to the grip surfaces to reduce frictional effects that would be generated from sample Poisson expansion. The lubricant can be seen on the grips in the blow up of Fig. 4. Other than aspect ratio and end face lubrication, no specific correction was made for specimen/grip friction. However, as will be seen later, both the calibration and the actual tests

gave reasonable results, thus pointing to the diminished role of friction through adequate lubrication.

A Pentium III 450 MHz computer was used to control the microdeformation tester. The interface for data acquisition and motion control was programmed with LabVIEW 5.1 (National Instruments). The resolution of captured images was 640 by 480 pixels. Every data point of load and displacement acquired during the deformation process has a corresponding image frame captured and stored. The maximum frame rate of image acquisition can be up to 30 frames per second, but the actual one depends on many factors, such as image size, hard drive speed, memory and so on.

2.3. Calibration and testing

When testing such small scale structures measurement errors introduced by machine compliance can be significant [19]. Remedies that include measuring and adjusting for machine compliance have been suggested [20], but these involve having samples of different length or precisely known properties. Alternatively displacement measurements can be made directly on the gauge length. This approach was adopted in the present study. The LVDT used to measure displacement was directly connected to the specimen grips (see Fig. 4) and therefore directly provided measurements of crystal contraction. Thus, no need for measuring machine compliance arises.

To verify that the microdeformation tester components were working correctly some calibration tests using known materials were conducted. Two different materials were used: Plexiglas (PMMA) and Polycarbonate. Rectangular samples of dimensions 2 mm by 2 mm by 3 mm were cut from each material and loaded in compression in the microdeformation tester. The LVDT and load cell readings were then used to generate partial stress strain curves for these materials. Note that the maximum applied load is limited to 100 N by the capacity of the actuator stage. Fig. 5 shows a stress strain curve for PMMA obtained using the microdeformation tester. Note that the stress levels applied to PMMA with this set-up are below the failure stress. However, elastic modulus can still be obtained from such curves. Table II shows modulus results obtained from the two polymer samples. The modulus results were in excellent agreement with known bulk values. This is implies that the lubrication used to avoid frictional effects at the specimen/grip interface was effective.

TABLE II Comparison of microdeformation tester results with bulk values

	Elastic modulus (GPa)	
Materials	Measured	Bulk
PMMA	2–5	3–4
Polycarbonate	2–3	3

3. Results and discussion

The ZSM-5 single crystals were compressed in either the [100] or [010] directions, i.e., the parallelepiped faces of the crystal. Note that because of twinning it is not possible to unambiguously distinguish between the [100] and [010] directions. In what follows these directions are identified nominally, i.e., they are perpendicular to each other. Compression tests along the [001] direction, i.e., along the pyramidal sides of the crystal, were also attempted, but the surface shape did not allow for reliable loading. Thus, results from the [001] directions tests are not reported here. The same sequence of tests were performed on the crystals as-synthesized, i.e., with the organic molecule occupying the channels of the zeolite, and after processing to remove the organic molecule was conducted. Three different tests were performed in each case:

(a) Simple compression tests: Single crystal samples were monotonically compressed until complete failure.

(b) Cyclic compression tests: Single crystals were subjected to several loading-unloading cycles. The maximum load during cycling was kept constant for three cycles and then increased by about 5 N. This cyclic loading-unloading-reloading process was repeated until complete crystal collapse occurred.

(c) Two-direction compression tests: Single crystal samples were compressed elastically (i.e., without macro-cracks having been generated in the sample) in one direction and unloaded. Then compression was applied along the second direction. This method was used to investigate anisotropy of the single crystal mechanical properties.

3.1. Simple compression tests

The load and image information collected from experiments should demonstrate the structural change of the crystal as loading is applied. Fig. 6 shows two pictures taken during compression of an as-synthesized

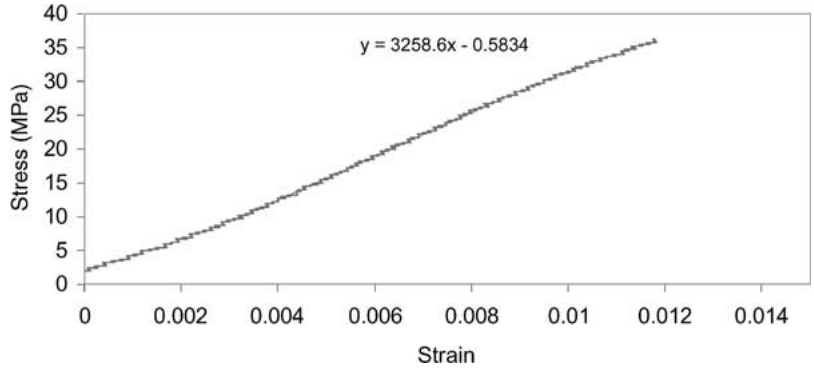
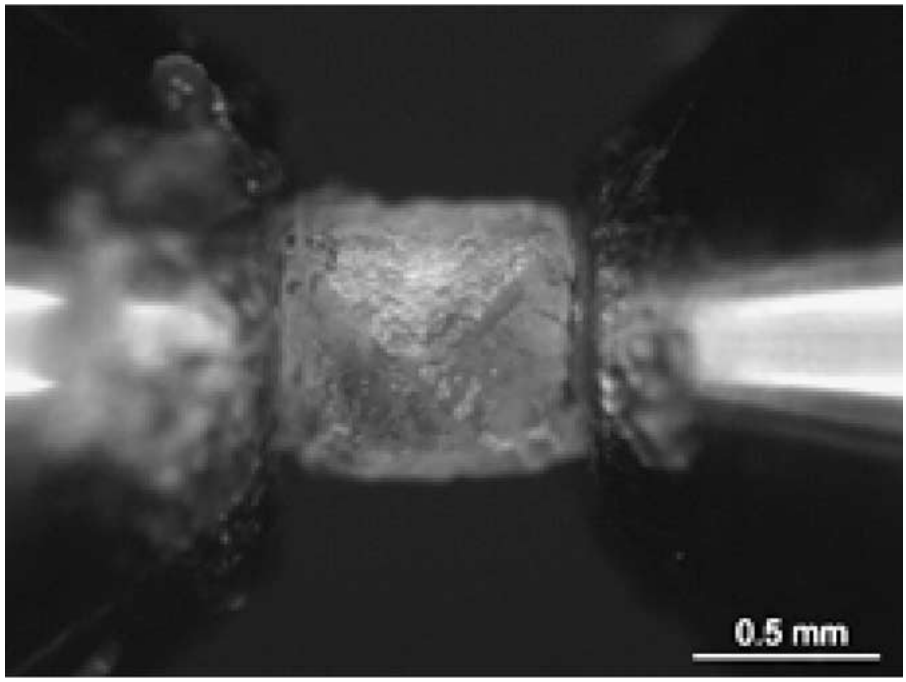
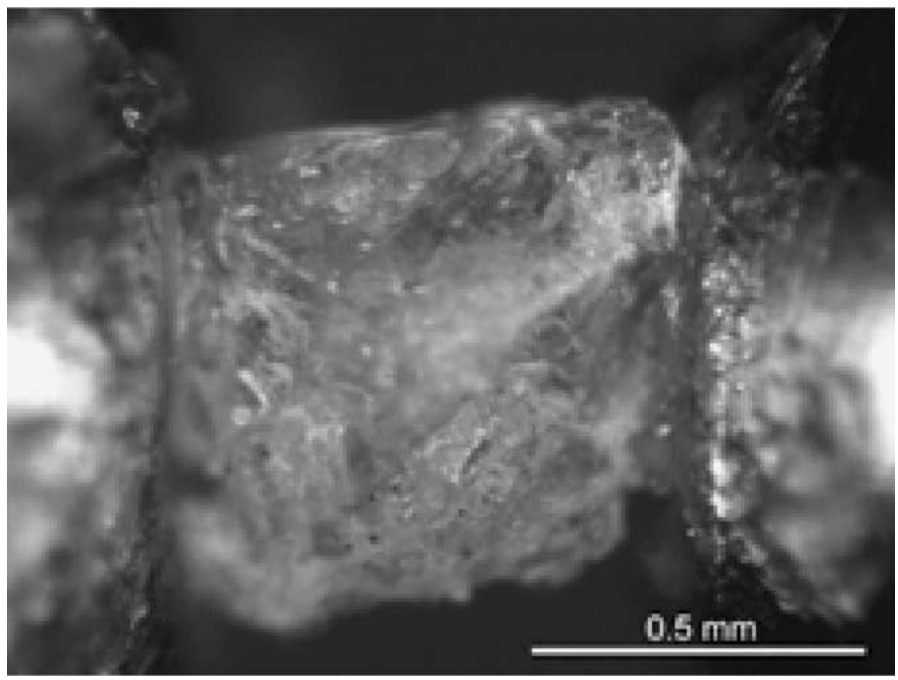


Figure 5 Stress-strain curve for PMMA used for evaluation of the microdeformation tester.



(a)



(b)

Figure 6 Optical micrographs showing two stages of ZSM-5 crystal compressive deformation.

ZSM-5 crystal. Fig. 7 presents the corresponding nominal stress strain curve for such a test. Stress is computed using the parallelepiped cross sectional area and strain is obtained by normalizing the LVDT extension results by the original crystal length measured from the microscope images. The early stages of deformation (corresponding to the first picture in Fig. 6) are purely elastic. This is also seen in the initial linear part of the stress strain curve. However, at certain time instants a sudden drop of stress occurs ($\approx$0.03 and 0.034 strain in Fig. 7). In all such instances correlation of optical images with the load-displacement data revealed the formation of an internal microcrack inside the crystal, approximately along the [110] crystal plane (second picture of Fig. 6). Upon continued loading several of these internals crack coalesced leading to global failure. By investigating the stress strain curve in Fig. 7 and the corresponding optical images we can conclude that the ZSM-5 zeolite crystal is highly brittle and subject to significant internal microcracking.

On several occasions pre-existing flaws in the as-synthesized crystals were present. Such crystals were easily identifiable when placed under the microscope as they were not transparent in the vicinity of the crack. When these crystals were tested, peak stress values were substantially less that those shown in Fig. 7, as would be expected, thus producing a distribution of lower failure loads. A similar effect of non repeatability of experimentation in the damage regime was seen in [8]. Note that in order to accurately model the effect

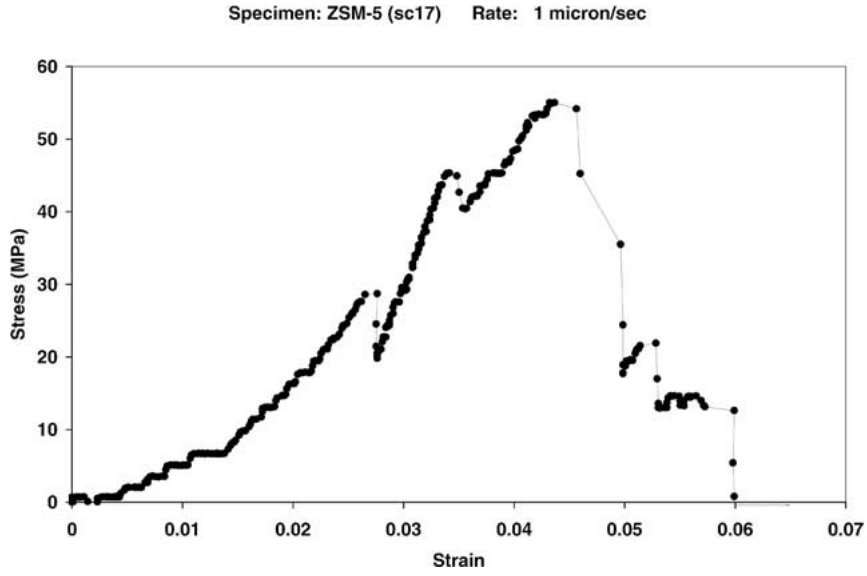


Figure 7 Typical compressive stress-strain curve for ZSM-5 single crystal.

of pre-existing flaws on failure load some type of statistical model would be required, as in [12], but this is beyond the scope of this work. Thus, Fig. 7 can be considered as the elastic stress strain curve of the "ideal" undamaged as-synthesized crystal.

From analysis of data acquired during compression testing of a group of ZSM-5 single crystals we can obtain an average value of Young's modulus for ZSM-5. These values were obtained from the slope of the stress strain curves before the first abrupt drop in stress was observed. The average value of Young's modulus from all experiments is about 4 GPa, while the actual values vary from 3 GPa to 5 GPa.

The preceding results all involved the as-synthesized crystals. A similar set of experiments was repeated using the crystals without the occluded organic molecule. Somewhat surprisingly the values of Young's modulus obtained from these experiments were similar to those of the as-synthesized crystals. However, in hindsight we know that the diameter of the organic molecule ($\approx$4 Å) is substantially smaller than the diameter of the pores ($\approx$5.5 Å). Within the range of strains achieved here, deformation of ZSM-5 can proceed unhindered. On the other hand, removal of the organic molecule did have a significant effect on peak stress recorded. Most likely this is because of generation of additional microcracks during the organic molecule removal process.

3.2. Cyclic compression tests

Motivated by the fact that in industrial applications mechanical failure of the zeolite catalyst is seen to occur after repeated use we conducted a series of compression experiments in which cyclic compressive load was applied. The maximum load value was increased by about 5 N every three cycles until specimen failure. Fig. 8 shows the stress strain variation resulting from such a load history. The first inset in Fig. 8 shows the

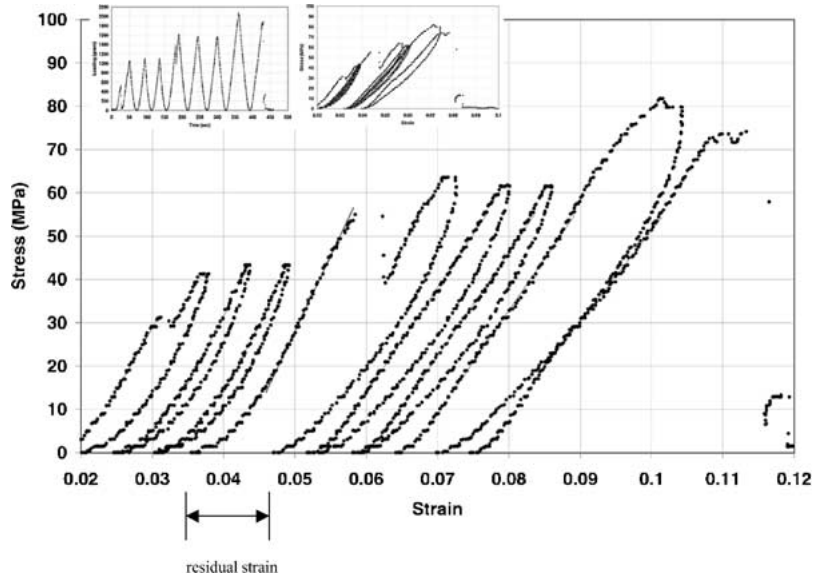


Figure 8 Stress strain history during a cyclic loading-unloading experiment. In the main figure the stress-stain curves from each loading cycle have been shifted along the strain axis so as to be separately visible. In the upper left hand corner, the first inset shows the load-time history used in the test and the second inset shows the unshifted stress-strain results.

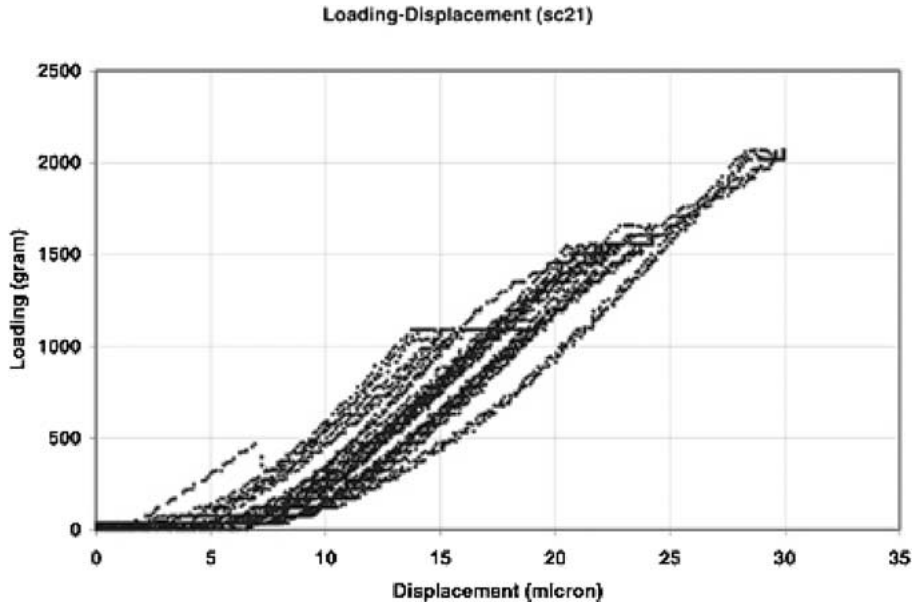


Figure 9 Unshifted stress-strain curves from cyclic loading experiments in which the material was only loaded elastically.

load history used. The second inset shows the stress strain results as recorded. In the main part of the figure each stress strain curve resulting from an individual load cycle has been arbitrarily shifted along the strain axis so that they appear separated. As can be seen the first three cycles are very similar to each other, and strain always returns to zero (see second inset). However in the fourth loading cycle, i.e., the first at an increased peak load, a sudden drop in load, corresponding to internal crack formation, appears. This results in a permanent strain appearing in the stress strain curve, i.e., a residual strain because of internal damage. Upon continued loading the crystal responds elastically, but with a decreased stiffness, until the next time that cracking occurs, which is just after 0.1 on the shifted strain axis. This again occurs at the first instant of an increased peak load. At the next cycle total catastrophic failure occurs. The original modulus computed from the first three cycles in this case was 3.5 GPa and the reduced modulus computed after the first abrupt drop in load was 2.5 GPa.

In contrast, Fig. 9 shows a purely elastic case where despite increase and decrease in the load level with time, the residual strain is zero, i.e., the stress strain curve always returns to (0, 0) albeit with considerable amounts of hysterisis.

3.3. Two-direction tests

As was mentioned in the introduction, the crystal structure of ZSM-5 is orthorhombic. This type of structure exhibits elastic anisotropy. A compression testing method was employed to qualitatively investigate the mechanical anisotropy of ZSM-5 single crystals. The first step is compressing the crystal along the nominal [100] direction, but within the elastic limit, and then repeating the compression test along the perpendicular direction [010]. Two nominal elastic constants can be obtained by estimating the slopes of two stress-strain curves from the experiments carried out in perpendicular directions. Typical values of the elastic constant obtained along [010] in this way were around 3–3.5 GPa. This value is similar to that from uniaxial compression testing along [100] (Fig. 7). So theZSM-5 single crystals do not exhibit a very strong elastic anisotropy between the [100] and [010] directions. It should be interesting to perform a test in the [001] direction since the c-axis is different than the a- or b-axis for the ZSM-5 single crystal. However, this proved unfeasible in the present set-up.

4. Conclusions

In this work ZSM-5 single crystals were tested under monotonic and cyclic compression loading. Single crystals were synthesized using an organic template molecule. Before and after molecule removal the crystals were tested in compression in order to investigate their elastic, failure and fatigue response. To test the crystals, on average 500 microns in length, a custom microdeformation tester was designed and built in-house. A high precision stepper motor provides mechanical actuation. Load and specimen contraction (as measured through an LVDT) are recorded in real time in a computer. The entire set-up is placed under a microscope and images of the deformation are also recorded throughout.

The ZSM-5 crystals tested showed a highly brittle deformation behavior with a microcracking failure mode. Elastic deformation of the crystal was reproducible before load levels at which cracks would be generated in the interior. For compression along the [100] direction an average Young's modulus of 4 GPa was measured. For loading along the [010] direction an average value of 3–3.5 GPa was determined, which within experimental scatter is similar to that of the [100] direction. A decrease in modulus was observed upon cyclic loading and by correlation of load-displacement data to optical images of the deformation this decrease was seen to be a consequence of microcracking. In the monotonic and cyclic loading cases, however, the strength of the crystal (i.e., peak load) was seen to be extremely sensitive

to the intrinsic defects, such as micro-cracks and twins produced during the synthesis procedure, necessitating an additional study that would take this effect into account.

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