Assign to Mauper Due 7/29 DNW DN

South Carolina Department of Health and Environmental Control

2600 Bull Street
Columbia, S.C. 29201

Commissioner
Michael D. Jarrett

June 29, 1988

Donald A. Nussbaumer
Assistant Director for
State Agreements Program
US Nuclear Regulatory Commission
Washington, DC 20555

Dear Mr. Nussbaumer:

In regard to the report "Review of the Structural Design of Polyethylene High Integrity Containers" by S.A. Silling, the Department has several comments and questions.

1) Please verify the use of secant modulus instead of Young's Modulus. Secant modulus is usually used for metals.

2) Assumption's that are based purely on tensile data cannot be considered adequate. Although there is little data on HDPE, most of the loads experienced by the containers are compressive.

3) The tests performed by BNL were done to establish a baseline stress value. These tests would be more adequate than the data given by Silling since Silling's data are for linear HDPE in pure tensile loads.

4) Silling states that HDPE cannot be designed to overcome creep buckling. The statement totally neglects the idea that HDPE may have a threshold stress below which containers could be designed using HDPE. Soo, in his Brookhaven report, also believes that a threshold value exists as it does with most all structural materials.

5) The report totally neglects real world situations by taking into account backfill and percent filling of the containers.

6) We do agree that data for these specific applications is limited. Perhaps BNL should continue its research in the structural analysis of HDPE and take into account compressive loads, radiation, and the crosslinking of Marlex CL 100 specifically.

Although many questions have been raised concerning the adequacy of HDPE for HIC's, the Department does not feel that Silling's report confirms this inadequacy. We will continue to research this area of concern in the future. Please provide responses to the above mentioned concerns.

Should there be any questions, please contact Mr. Virgil R. Autry at (803) 734-4633.

Very truly yours,

Heyward G. Shealy

Heyward G. Shealy, Chief
Bureau of Radiological Health

OTP/ac

REVIEW OF THE STRUCTURAL DESIGNS OF POLYETHYLENE HIGH INTEGRITY CONTAINERS

Stewart A. Silling

Assistant Professor of Engineering
Brown University

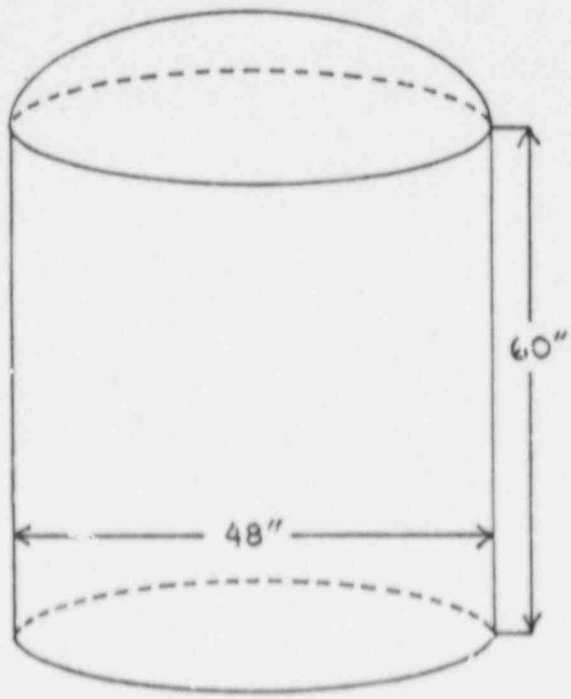
presented to
U.S. Nuclear Regulatory Commission
Advisory Committee on Nuclear Waste
June 28, 1988

b|26

GENERIC HIC

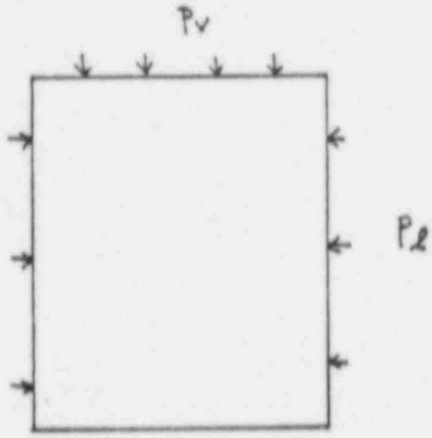
Typical geometry:

- Cylindrical shell
- Torospherical dome
- Rotationally molded Marlex CL-100
  - –Cross-linked high-density polyethylene (HDPE)
- 0.5 inch thickness



MAGNITUDE OF LOADS

- Loads are from overburden
- Affected by
  - –Depth
  - –Soil conditions
  - –Burial configuration
  - –Arching (Unequal stiffnesses of structure and soil influence load)
- Orders of magnitude (25 ft depth):
  - –Vertical pressure $p_v \approx 21$ psi
  - –Lateral pressure $p_\ell \approx 7$ psi
- Total load on generic HIC is 19 tons



CREEP: Definition

Uniaxial test at constant load – responses:

- *Elastic*

  –Instantaneous

  –Fully recoverable strain

- *Plastic*

  -Instantaneous

  –Nonrecoverable strain

  –Only important above the yield stress

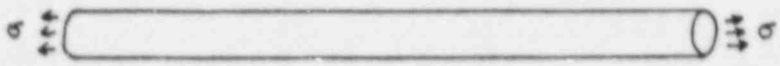
- *Creep*

  –Time-dependent

  –Sometimes recoverable

  –No threshold stress in general

  –Time scales vary widely
  (Microseconds to centuries, e.g. glaciers)

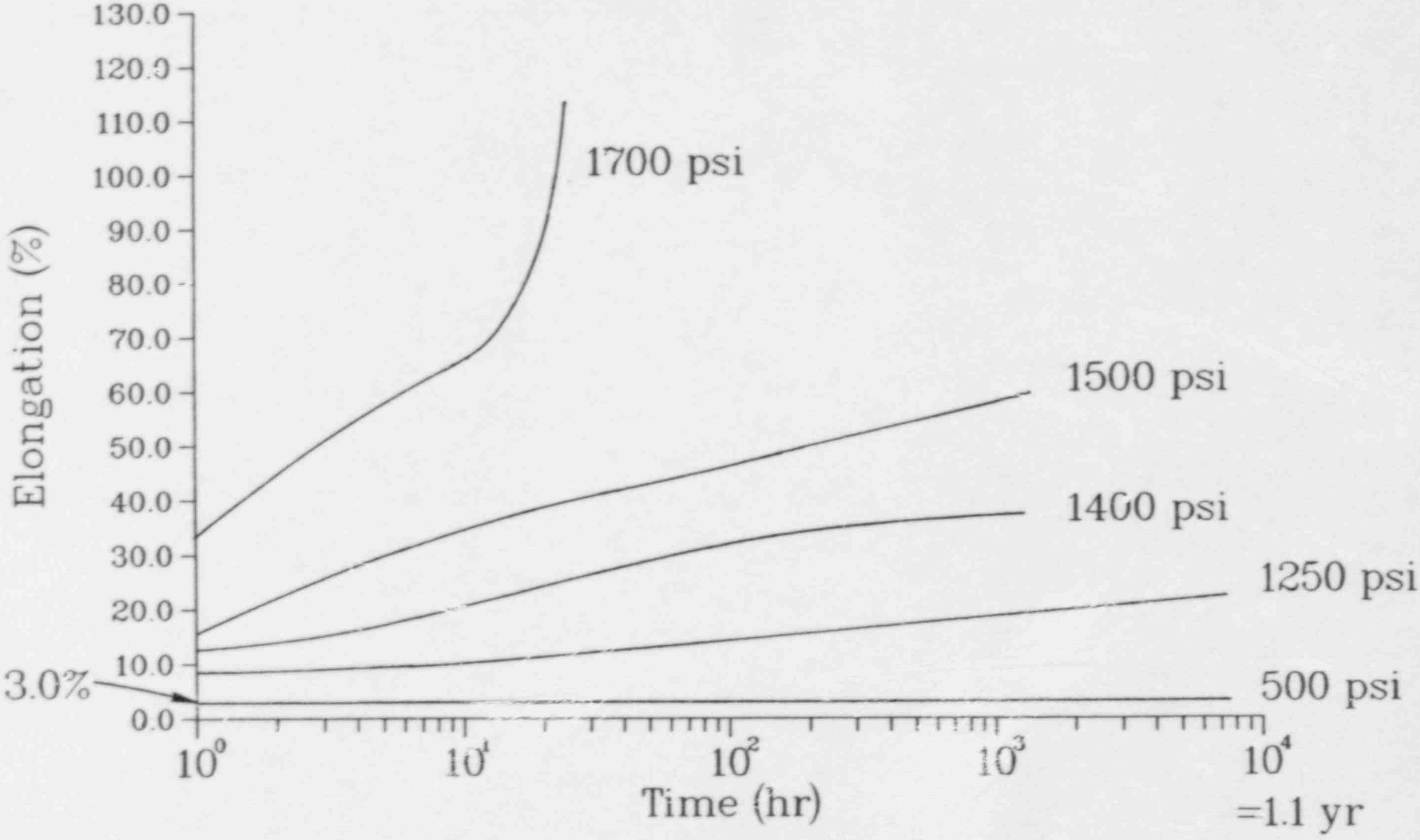


CREEP IN POLYMERS

- Elastic/plastic in short term
- Creep is the main long-term mode of deformation
  - –Marlex CL-100 low-stress creep properties are largely unknown
  - –Next slide shows Phillips data
  - –Result: at 500 psi (low stress),
    total strain $\approx$ 6 times elastic strain

Marlex CL–100 creep test (25 degC, air)

Phillips data



DESIGN UNDER CONDITIONS OF CREEP

If the loads are constant:

- Define *secant modulus* (effective Young's modulus at time t)

$$E_s(t) = \sigma/\varepsilon(t)$$

where σ =stress, $\varepsilon(t)$ =strain in uniaxial creep test.

- Marlex CL-100 after 1 hour (Phillips):

$$E_s = 16,700\ psi$$

- Vendors generally ignored creep, used

$$E = 100,000\ psi$$

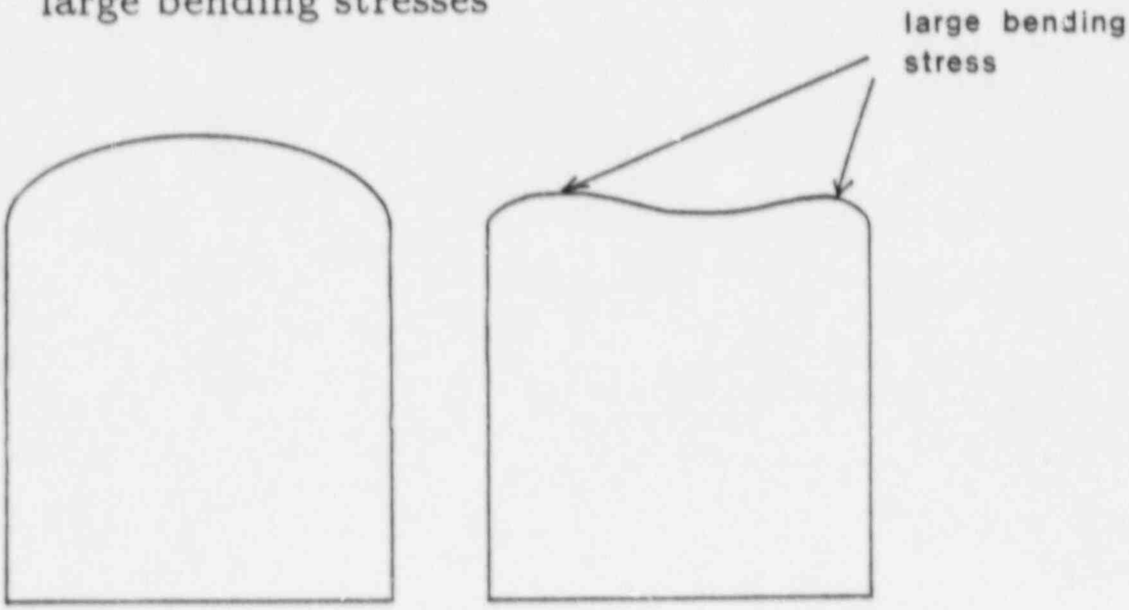
–which is the (elastic) Young's modulus

- Effect of creep: reduced stiffness, increased deflections, different failure modes.

SIGNIFICANCE OF LARGE DEFORMATION EFFECTS IN HICs

Likely effects of creep:

- Large shape changes will alter the stress distribution
  - –Small-deflection analysis probably invalid
  - –This is why flexible structures are hard to design
- Example: Torospherical dome under load
  - –Small-deflection analysis: load is supported by small compressive membrane stresses ("egg stresses")
  - –Large-deflection analysis: shape change leads to large bending stresses



FAILURE OF HDPE

Strongly affected by:

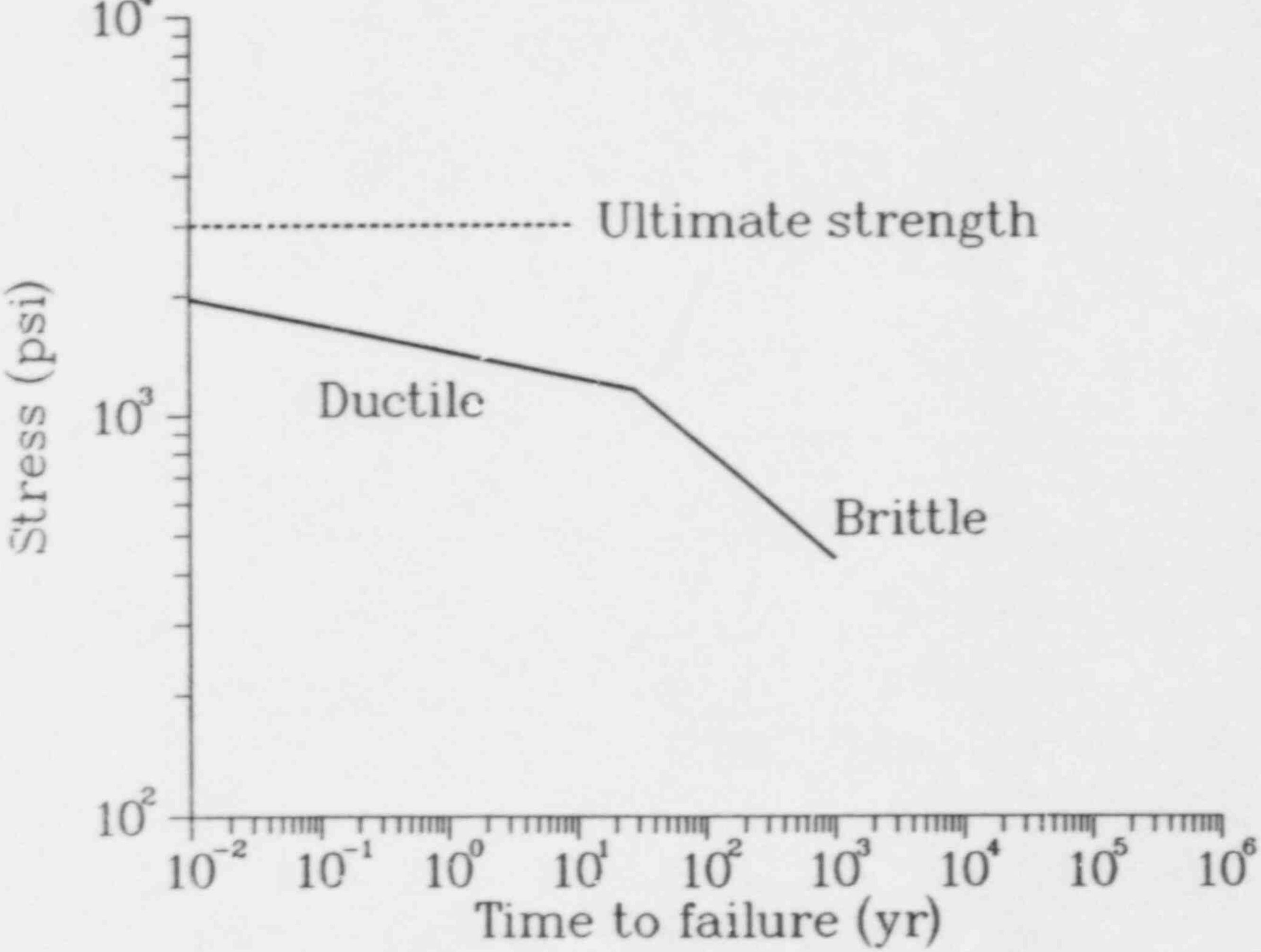
- Strain rate (higher strength at higher rates)
- Radiation
- Age (ductile/brittle transition)
- Chemical environment
- Temperature
- Exact material composition
- Molecular weight and cross-linking
- Microscopic defects
- Fabrication and processing methods

SHORT-TERM/LONG-TERM FAILURE OF HDPE

Time to failure depends on stress

- High stress (above ≈2500 psi):
  - –Rupture at "ultimate strength"
  - –Failure due to excessive plastic strain
  - –Time scale: seconds to minutes
  - –Only failure mode considered by vendors
- Moderate stress (1000 to 2500 psi):
  - –Ductile creep rupture
  - –Time scale: hours to weeks
- Low stress (below ≈1000 psi):
  - –Brittle failure
  - –Time scale: months to years
- 20-year test results (Graube) on next slide
  - –Hostalen GM 5010 – Linear HDPE, unirradiated
  - –Extruded pressurized pipe
  - –20 degC, extrapolated from high temperature data
  - –Marlex CL-100 may behave differently

Failure of an unirradiated HDPE



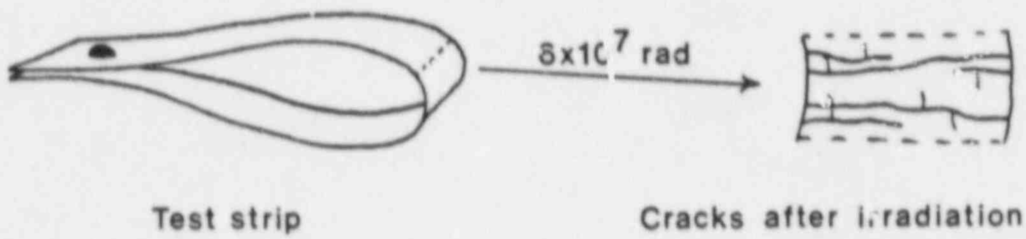
RADIATION EFFECTS ON POLYMER MATERIAL STRENGTH

General effects, moderate dose

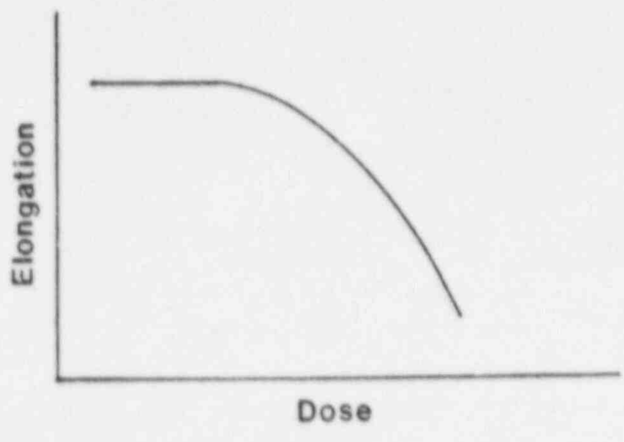
- Increases hardness and plastic strength
  - Not relevant to HICs
- Creep rate may increase or decrease
  - Depends on dose rate
  - Scission may increase creep
  - Cross-linking decreases creep
- Embrittlement
  - Failure by crack propagation

RADIATION EMBRITTLEMENT OF MARLEX CL-100

- U-bend tests (Soo)
  - –Radiation-induced cracks at constant strain
  - –Cracking is in spite of stress relaxation
  - –Strains comparable to buckled HICs



- Uniaxial test: decreased elongation at break (Soo)
  - –Effect is sensitive to dose rate



CREEP BUCKLING

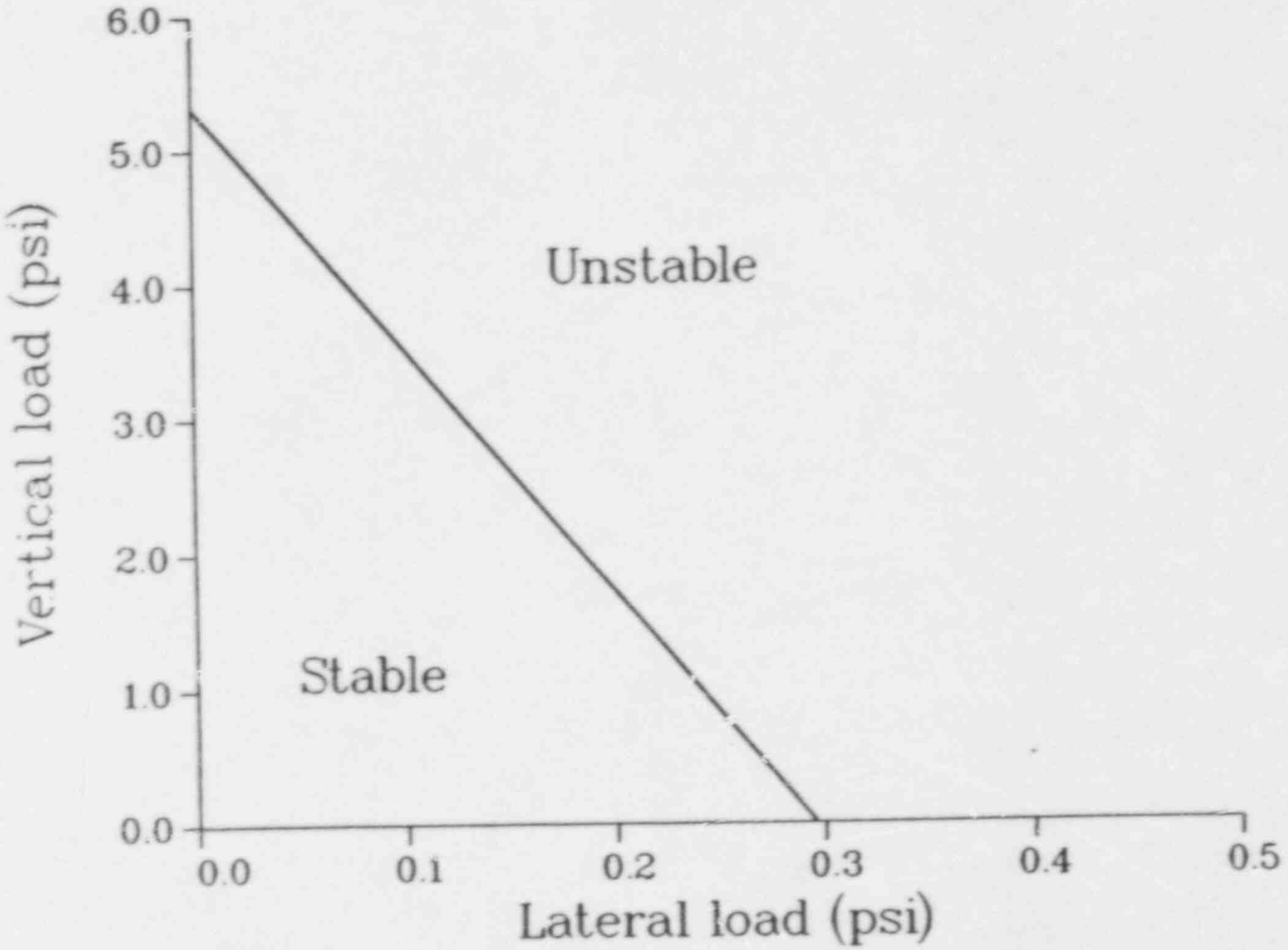
- *Buckling:* Large deflection occurring at a critical compressive load

- Creep strongly affects critical load
  - –Approximation: critical load proportional to material stiffness (secant modulus $E_s(t)$) for a given geometry
  - –Thus the critical load is a function of time
  - –Not considered by vendors

- Both vertical and lateral loads are important

- Soil and waste will have an unknown but helpful effect

Generic HIC buckling loads



TESTS SO FAR MEANINGLESS
in regard to buckling

- Typical "compression test" by vendor:
  - –Container filled with water, sand, etc.
  - –Load is applied at ends only (no lateral loads)
  - –Test is run for a short time (up to ≈ 1 day)
- Meaningless because:
  - –Contents will prevent buckling
  - –Small lateral loads can cause buckling
  - –Creep buckling may be slow to develop

IMPORTANCE OF BUCKLING

- Buckling means collapse
  - –Waste supports load
- Excessive strains in kinks
  - –Radiation causes cracks in strained HDPE in spite of stress relaxation
- Integrity of seals cannot be assured
- Little is known about post-buckling in shells

SUMMARY
Problems identified

- Uncertainty in long-term creep properties
- Creep ignored in the designs
  - –No design limits on deformation
- Brittle failure modes, including radiation embrittlement, were not considered
- Creep buckling appears unavoidable

PROSPECTS FOR REDESIGN

- HDPE is a poor material from a structural mechanics point of view
  - –Problems with HDPE are of a fundamental nature
- None of the following is well understood:
  - –Creep buckling
  - –Post-buckling behavior
  - –Long-term properties of polymers
  - –Design of flexible structures
- Composite materials a possibility

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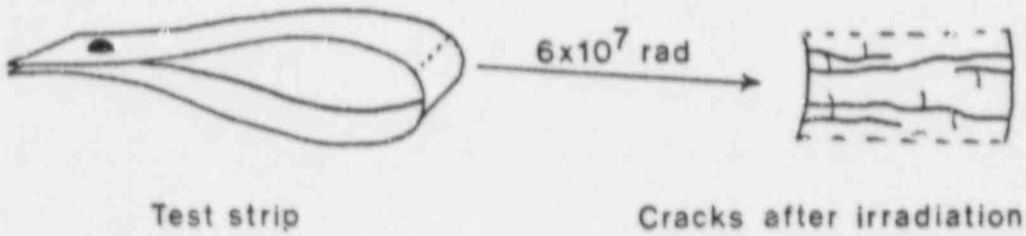

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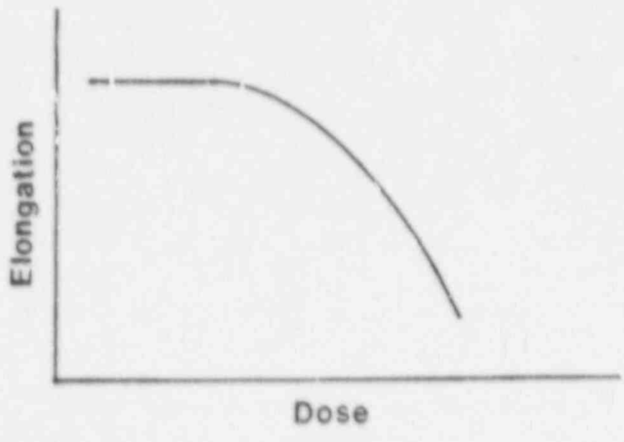

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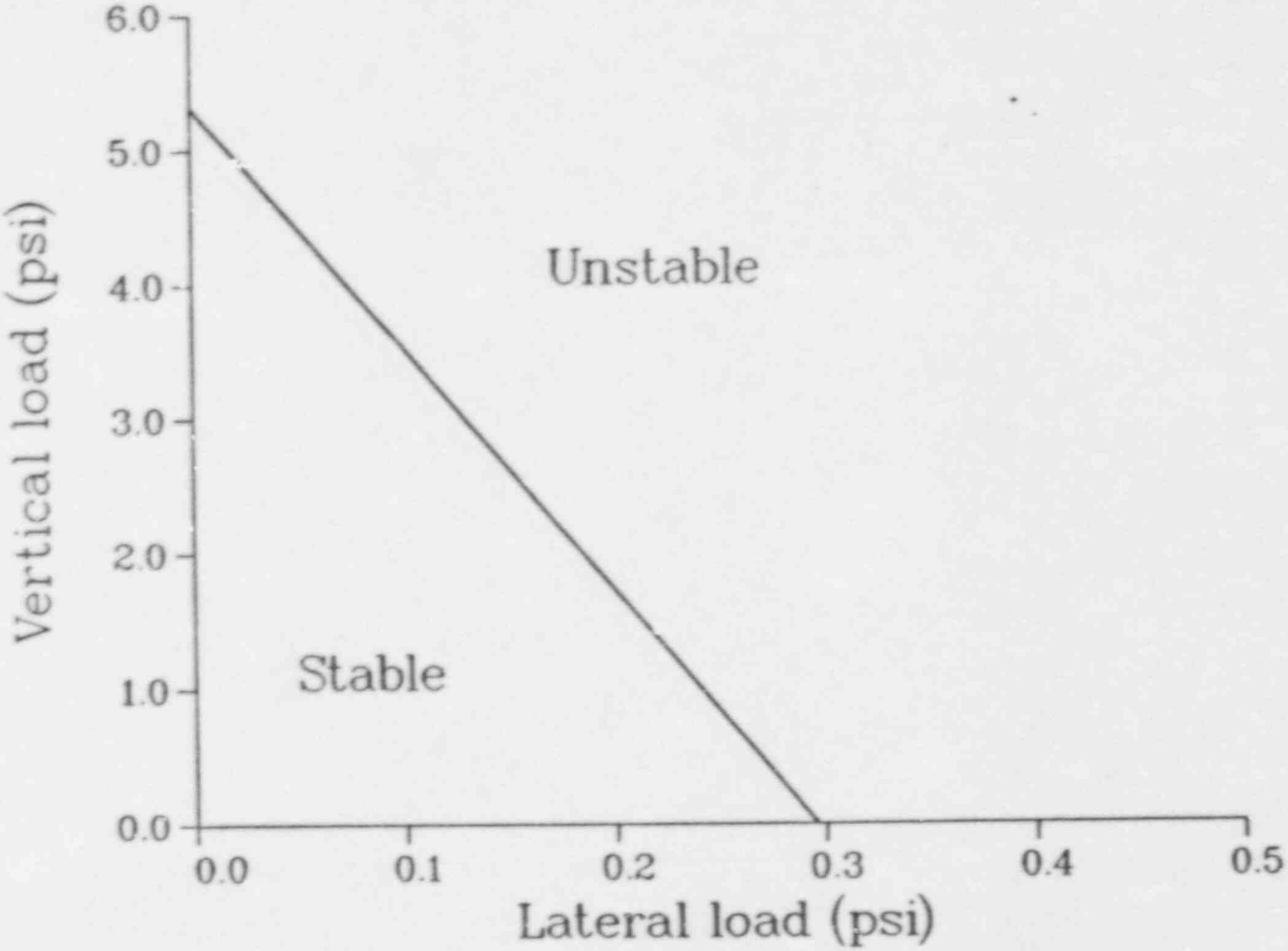

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(8)

NRC STAFF PRESENTATION TO THE ACNW

SUBJECT: UPDATE ON STATUS OF CEMENT WASTE FORM SOLIDIFICATION ISSUES.

DATE: JUNE 28,1988

PRESENTER: DR. MICHAEL TOKAR

PRESENTER'S TITLE/BRANCH DIV.:

SECTION LEADER / TECHNICAL BRANCH / LOW-LEVEL WASTE MANAGEMENT DIVISION

PRESENTER'S NRC TEL. NO.: (301) 492-0590

SUBCOMMITTEE: ACNW

TO BE USED ALL PRESENTATIONS TO THE ACNW BY NRC EMPLOYEES

UPDATE ON

STATUS OF CEMENT

WASTE FORM SOLIDIFICATION

ISSUES

Dr. Michael Tokar
ACNW Meeting
June 28, 1988

LAST DISCUSSION (On March 17, 1988)

* GENERIC ISSUES
* TMI—2 WASTE LINER EVENT
* STATUS OF (4) VENDOR TOPICAL REPORT REVIEWS
* MISCELLANEA

SUBJECTS FOR DISCUSSION TODAY

* WEST VALLEY CEMENT SOLIDIFICATION SYSTEM (CSS)

* STATUS OF 4 CEMENT VENDOR TR REVIEWS

WEST VALLEY LLW ACTIVITIES

- SOLIDIFICATION OF 39 WEIGHT PERCENT SUPERNATANT
- EXTRACTION OF CS−137 FROM HIGH−LEVEL SUPERNATANT
- 600,000 GALLONS OF WASTE
- 15,000 DRUMS OF WASTE

CEMENT FORMULATION – WV

- INITIAL FORMULATION

  -- FOAMING
  -- LOW COMPRESSIVE STRENGTHS
  CAUSE -- HIGH SHEAR MIXING

- MODIFIED FORMULATION

  -- SLOW SETTING
  -- BLEED WATER
  CAUSE -- ORGANICS

- FINAL FORMULATION

  -- INCLUDES ADDITIVES – CALCIUM NITRATE, ANTIFOAM AGENT & SODIUM SILICATE

WV CEMENT WASTE FORMS – TESTING

- NRC – ADDITIONAL QUALIFICATION TESTING – APRIL 1988

- DOE – HOT CHECKOUT TESTING

  300 DRUMS PRODUCED

  CONSIDERING FULL-SCALE TESTING

  DEVELOPING LONG-TERM (5 Year) TEST PROGRAM

CHEM-NUCLEAR RESPONSE

- WITHDREW 2 EXISTING CEMENT TOPICAL REPORTS.
- SUBMITTED 3 NEW TOPICAL REPORTS (GROUPED BY SOLIDIFICATION BINDER TYPE.)
    1) PMC BINDER
    2) POZZOLANIC BINDER
    3) CEMENT BINDER
- REVIEW SCHEDULES FOR THE NEW TOPICAL REPORTS ARE BEING PREPARED.

LN TECHNOLOGY RESPONSE

COMPRESSIVE STRENGTH AFTER IMMERSION VS. CURE TIME

* LN IS PERFORMING TESTING ON BEAD RESIN WASTE FORMS TO DETERMINE IF THE WASTE FORMS RETAIN STRENGTH WITH EXTENDED CURE TIME.
* THE POST-IMMERSION COMPRESSIVE STRENGTH OF WASTE, CURED FOR VARIOUS TIMES, IS BEING MEASURED.
* WASTE FORMS: CATION BEAD RESIN
MIXED BED BEAD RESIN
* CURE TIME (days): 14, 28, 42, 56, 70, 84
* IMMERSION MEDIA: DEMINERALIZED WATER
SYNTHETIC SEA WATER
* IMMERSION TIME: 90 DAYS
* REDUCED WASTE LOADINGS

RESPONSES TO NRC LETTER – STOCK

- JANUARY 8, 1988 – STOCK REQUESTS INFO ON STATUS OF BTP & BNL REPORT.
- MARCH 1, 1988 – NRC RESPONDS TO STOCK LETTER & REPEATS REQUEST FOR INFO.
- MARCH 31,1988 – STOCK DISCUSSES NUMARC REPORT.
- JUNE 6, 1988 – LAST RESPONSE RECEIVED
- RESPONSE INADEQUATE
- SIGNIFICANT DEFICIENCIES WITH CERTAIN WASTE STREAMS
  - DRAMATIC LOSS IN COMPRESSIVE STRENGTHS
- NRC NOTIFIES STOCK OF PLANS TO DISCONTINUE – JUNE 1988

RESPONSES TO NRC LETTER – HITTMAN

- MEETING – MARCH 28, 1988
- AGREEMENT ON SCOPE OF ADDITIONAL TESTING – APRIL, 1988
- TESTING DURATION – JULY 1988 TO JANUARY 1989
- MONTHLY SUMMARY LETTER REPORTS
- RESUBMITTING REVISED TOPICAL REPORT

TOPICAL REPORT REVIEW STATUS SUMMARY

SOLIDIFIED WASTE FORM and HIGH INTEGRITY CONTAINERS (HICs)

June 30, 1988

Vendor	Docket No.	Type	Disposition
Waste Chem	WM-90***	Solidification (bitumen)	Approved.
General Electric	WM-88	Solidification (polymer)	Approved.
U.S. Gypsum	WM-51***	Solidification (gypsum)	Approved *.
Chichibu	WM-81	HIC (poly impreg/concrete)	Approved.
Nuclear Packaging	WM-45	HIC (ferralium/FL-50)	Approved.
Nuclear Packaging	WM-85***	HIC (ferralium/family)	Approved.
DOW	WM-82***	Solidification (polymer)	Approved**.
ATI	WM-91***	Solidification (bitumen)	Discontinued.
VIKEM	WM-13	Solidification/oil (cement)	Discontinued.
Stock	WM-92***	Solidification (cement)	Discontinued.
Nuclear Packaging	WM-71	Solid/Encap (cement/gypsum)	Withdrawn.
LN Technologies	WM-57	HIC (polyethylene)	Withdrawn.
Chem-Nuclear	WM-47	HIC (fiberglass/poly)	Withdrawn.
Chem-Nuclear	WM-19***	Solidification (cement)	Withdrawn.
Chem-Nuclear	WM-96***	Solidification (cement)	Withdrawn.
Hittman	WM-79***	Solidification (SG-95)	Withdrawn.
Chem-Nuclear	TBD	Solidification (cement #1)	Under review.
Chem-Nuclear	TBD	Solidification (cement #2)	Under review.
Chem-Nuclear	TBD	Solidification (cement #3)	Under review.
LN Technologies	WM-20	Solidification (cement)	Under review.
Hittman	WM-46	Solidification (cement)	Under review.
Chem-Nuclear	WM-18	HIC (polyethylene)	Under review.
Hittman	WM-80	HIC (polyethylene)	Under review.
TFC	WM-76	HIC (polyethylene)	Under review.
Nuclear Packaging	WM-83	HIC (316-stainless)	Under review.
LN Technologies	WM-93	HIC (stainless/poly)	Under review.
Bondico	WM-94	HIC (fiberglass/poly)	Under review.
Babcock & Wilcox	WM-95	HIC (coated carbon steel)	Under review.

* Approved for single waste stream for one year.

** Approved pending satisfactory completion of thermal cycling tests.

*** Actions completed in Calendar Year 1988.

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(6)

NRC STAFF PRESENTATION TO THE ACNW

SUBJECT: HIGH DENSITY POLYETHYLENE (HDPE) HIGH INTEGRITY CONTAINER (HIC) REGULATORY ISSUES.

DATE: JUNE 28,1988

PRESENTER: DR. MICHAEL TOKAR

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8/26

HIGH DENSITY POLYETHYLENE (HDPE)

HIGH INTEGRITY CONTAINER (HIC)

REGULATORY ISSUES

Low-Level Technical Branch
June / 1988

CURRENT SITUATION

* HDPE HICS HAVE BEEN ACCEPTED FOR SEVERAL YEARS AT THE BARNWELL LLW DISPOSAL FACILITY (See list of certificates of compliance).

* NRC IS REVIEWING 3 TOPICAL REPORTS (from CNSI, TFC-Nuclear, & W-Hittman) ON HDPE HIC DESIGNS.

* NRC CONSULTANTS AT BNL AND BROWN UNIVERSITY HAVE RAISED QUESTIONS CONCERNING ABILITY OF HDPE HICS TO PROVIDE LONG-TERM (300 yr.) STRUCTURAL STABILITY AS REQUIRED BY 10 CFR PART 61.

Certificates of Compliance

State of South Carolina

HIC Certificates of Compliance

Issued to:	Issued what:	Issued when:
Adwin Equipment Company	55-gallon HIC	5/29/84
Chem-Nuclear	HDPE HICs (x 14)	5/26/81
Chem-Nuclear	FRP HIC	2/23/82
Chem-Nuclear	Overpack HICs (x3)	4/8/83
Philadelphia Electric Comp.	PECO-HIC-1	9/28/81
Hittman	Radlok-55 HIC	6/17/82
Hittman	Radlok-100 HIC	6/17/82
Hittman	Radlok-200 HIC	5/5/83
Hittman	Radlok-500 HIC	9/31/85
LN Technologies	Barrier-55 HIC	9/1/83
TFC	NUHIC-120 HIC	11/1/83
NUPAC	HDPE 142 HIC	8/20/84
NUPAC	FL-50 HIC	9/26/85
Chichibu	Concrete HICs (x2)	8/12/86
Vermont Yankee	HDPE HIC	10/10/83

SUMMARY

* HDPE USE BEGAN IN EARLY 80s IN S.C.
* NRC IMPLEMENTS PART 61 STABILITY REQUIREMENTS – 1983.
* VENDORS SUBMIT TR's FOR HDPE – 1984.
* TECHNICAL PAPER CRITICAL OF HDPE – 1986.
* BNL/NRC DEVELOPS METHODOLOGY & CRITERIA – 1987.
* VENDORS HAVE TROUBLE MEETING CRITERIA.
* INDEPENDENT REVIEW BY S. SILLING.
* STAFF POSITION TO BE DEVELOPED BY LATE SUMMER 1988.

SCHEDULE FOR HDPE HIC REPORT ACTIONS

ACTION	COMPLETED BY
• NRC requests study on HDPE HICs from Consultant Silling	3-11-88
• Draft HDPE HIC Report received from Silling	5-1?-88
• Peer review of Draft HDPE HIC Report completed	6- 6-88
• Final HDPE HIC Report received from Silling	6-13-88
• Letters sent transmitting HDPE HIC Report HDPE HIC vendors - Hittman Nuclear - Chem-Nuclear Systems, Inc. - TFC Nuclear Associates, Inc. Advisory Committee on Nuclear Waste (ACNW) South Carolina DHEC	6-15-88
• Meeting with South Carolina DHEC	6-20-88
• ACNW Meeting that includes discussion on HDPE HIC Report	6-28-88
• Comments on HDPE HIC Report received from vendors	7-15-88
• Meetings with Vendors on HDPE HIC Report	Summer 88
• Final Decisions on HDPE HIC issues	Summer 88

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Assistant Professor of Engineering
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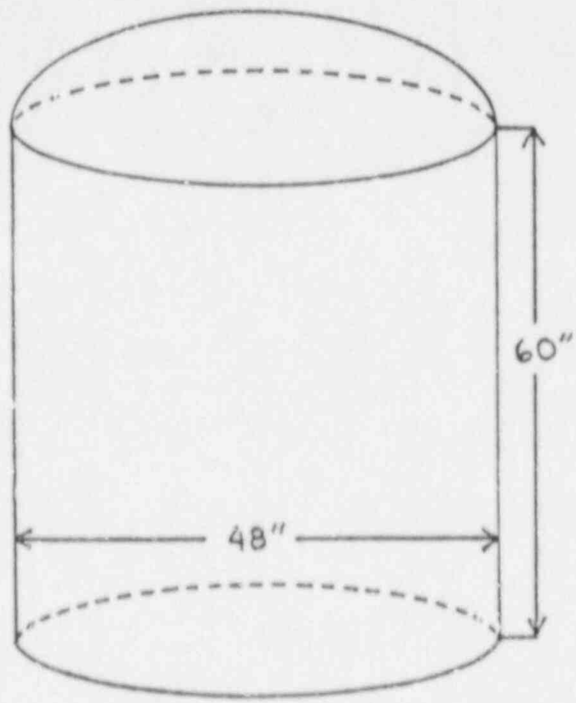

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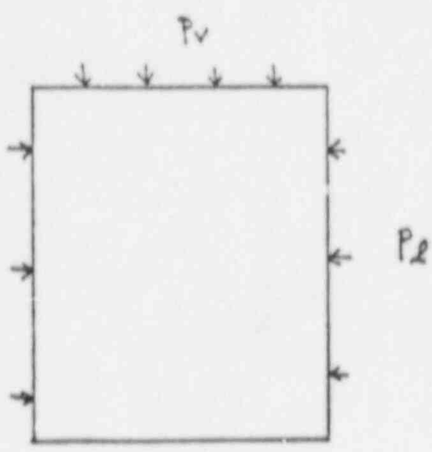

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  –No threshold stress in general

  –Time scales vary widely
  (Microseconds to centuries, e.g. glaciers)

CREEP IN POLYMERS

- Elastic/plastic in short term
- Creep is the main long-term mode of deformation

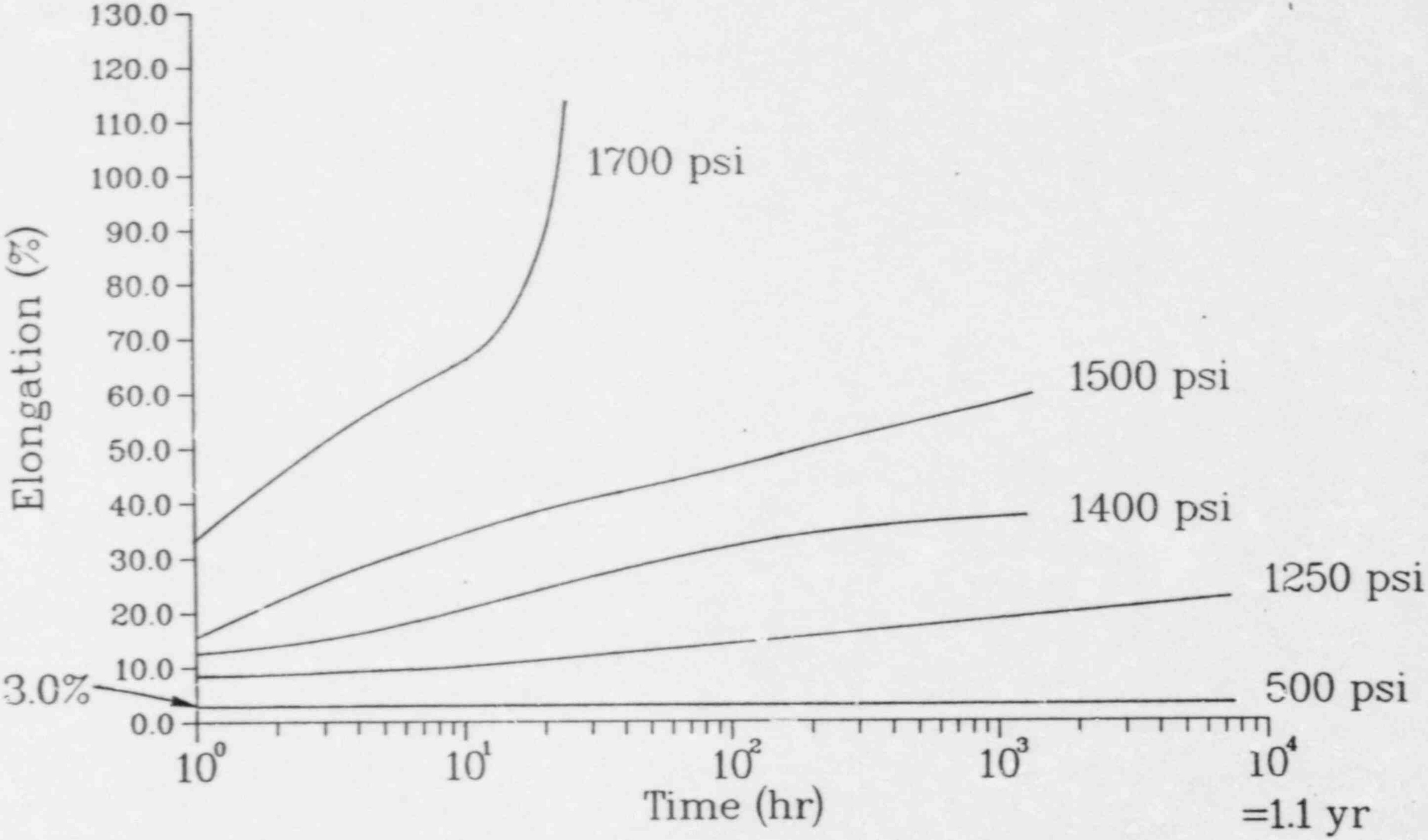
  –Marlex CL-100 low-stress creep properties are largely unknown

  –Next slide shows Phillips data

  –Result: at 500 psi (low stress),
  total strain $\approx$ 6 times elastic strain

Marlex CL-100 creep test (25 degC, air)

Phillips data



DESIGN UNDER CONDITIONS OF CREEP

If the loads are constant:

- Define *secant modulus* (effective Young's modulus at time t)

$$E_s(t) = \sigma/\varepsilon(t)$$

where σ =stress, $\varepsilon(t)$ =strain in uniaxial creep test.

- Marlex CL-100 after 1 hour (Phillips):

$$E_s = 16,700\, psi$$

- Vendors generally ignored creep, used

$$E = 100,000\, psi$$

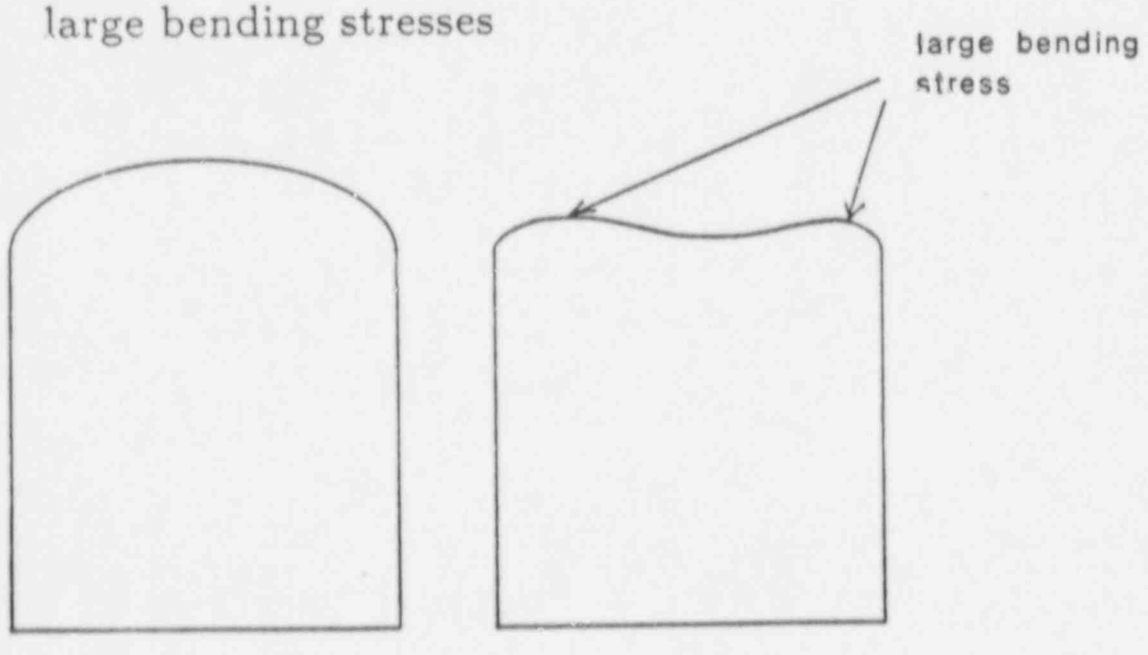
–which is the (elastic) Young's modulus

- Effect of creep: reduced stiffness, increased deflections, different failure modes.

SIGNIFICANCE OF LARGE DEFORMATION EFFECTS IN HICs

Likely effects of creep:

- Large shape changes will alter the stress distribution
  - –Small-deflection analysis probably invalid
  - –This is why flexible structures are hard to design
- Example: Torospherical dome under load
  - –Small-deflection analysis: load is supported by small compressive membrane stresses ("egg stresses")
  - –Large-deflection analysis: shape change leads to large bending stresses



FAILURE OF HDPE

Strongly affected by:

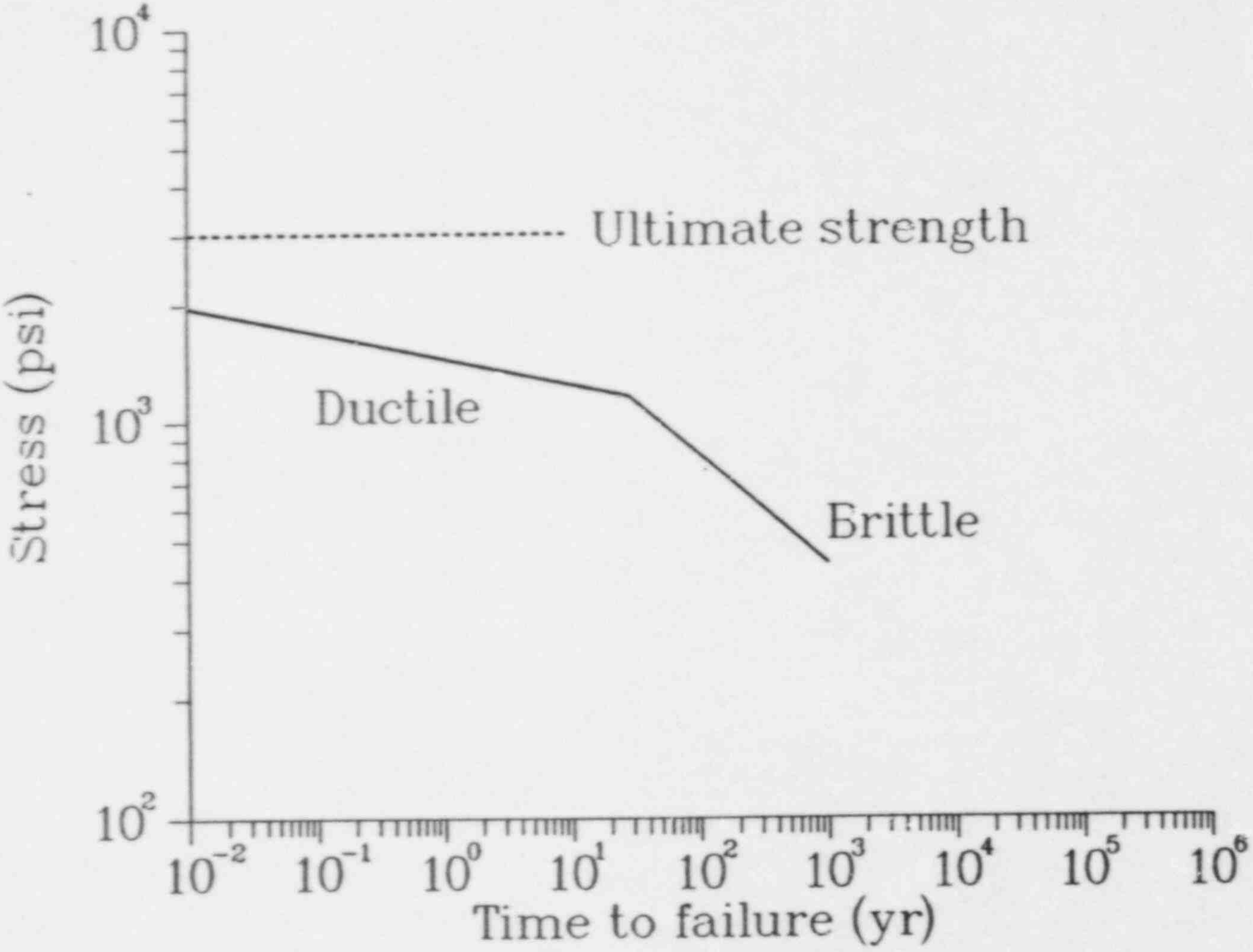
- Strain rate (higher strength at higher rates)
- Radiation
- Age (ductile/brittle transition)
- Chemical environment
- Temperature
- Exact material composition
- Molecular weight and cross-linking
- Microscopic defects
- Fabrication and processing methods

SHORT-TERM/LONG-TERM FAILURE OF HDPE

Time to failure depends on stress

- High stress (above ≈2500 psi):
  - –Rupture at "ultimate strength"
  - –Failure due to excessive plastic strain
  - –Time scale: seconds to minutes
  - –Only failure mode considered by vendors
- Moderate stress (1000 to 2500 psi):
  - –Ductile creep rupture
  - –Time scale: hours to weeks
- Low stress (below ≈1000 psi):
  - –Brittle failure
  - –Time scale: months to years
- 20-year test results (Graube) on next slide
  - –Hostalen GM 5010 – Linear HDPE, unirradiated
  - –Extruded pressurized pipe
  - –20 degC, extrapolated from high temperature data
  - –Marlex CL-100 may behave differently

Failure of an unirradiated HDPE



RADIATION EFFECTS ON POLYMER MATERIAL STRENGTH

General effects, moderate dose

- Increases hardness and plastic strength
  - Not relevant to HICs
- Creep rate may increase or decrease
  - Depends on dose rate
  - Scission may increase creep
  - Cross-linking decreases creep
- Embrittlement
  - Failure by crack propagation

Widmayer

TECHNICAL REPORT

WM-3291-8

LOW-LEVEL WASTE PACKAGE AND ENGINEERED-BARRIER STUDY

QUARTERLY PROGRESS REPORT

APRIL - JUNE 1988

P. Soo
J. H. Clinton
L. Milian

AUGUST 1988

NUCLEAR WASTE AND MATERIALS TECHNOLOGY DIVISION

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ABSTRACT

The sulfate-attack tests for various Portland cement based mortars were completed this reporting period. It is believed that the poor performance of a mortar containing silica fume cannot be attributed to the larger number of big pores in this material. Some other unidentified mechanism appears to be responsible.

Creep tests are continuing on Marlex CL-100 high density polyethylene which is being used as a high-integrity container material. In-test gamma irradiation in air at 5×10^3 rad/h is beneficial at higher stress levels since it leads to a slower creep rate and a higher ductility compared to non-irradiated material. At lower stresses (≤10.34 MPa, 1500 psi) irradiation appears to be detrimental.

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ACKNOWLEDGMENTS

The authors gratefully acknowledge Ms. A. Lopez for the preparation of this report.

1. INTRODUCTION

Since the publication of NRC Rule 10 CFR 61, "Licensing Requirements for Land Disposal of Radioactivity Wastes," and the NRC Technical Position on Waste Form, there has been action by industry to develop improved low-level waste forms, containers, and engineered barriers. Over the last several years the NRC received a large number of Topical Reports for review as a part of license applications for waste forms, containers, and engineered barriers. During review of the reports, it was recognized that the data provided by the vendors are usually insufficient or questionable. It was also recognized that conventional test methods, such as ASTM test procedures, may not be applicable to certain waste package materials and that analytical procedures have not been established to interpret the test data with respect to the performance objectives in the regulation.

The objective of this research project is to develop an adequate data base for performance review of low-level waste package materials identified in vendors' topical reports and to provide a basis for technical guidance to States and applicants. This project will also review and improve, if needed, the existing tests methods for application to materials and to the design of waste packages and engineered barrier concepts. Methods will be developed to extrapolate short-term test data to long-term performance of waste packages as required in the regulation.

To date, five research tasks have been specified by NRC and BNL. They include:

<u>Task 1</u>: Development of Work Plan.

<u>Task 2</u>: Mechanical and Chemical Stability of Concrete-Based Structural Materials.

<u>Task 3</u>: Degradation Mechanisms in High-Density Polyethylene (HDPE).

<u>Task 4</u>: Biodegradation of Ion-Exchange Media.

<u>Task 5</u>: Development of HDPE Testing Protocol.

Task 1 has been completed. Work in Task 2 is at an advanced stage, but will be significantly curtailed after this reporting period because of reduced funding. Task 3 is continuing on the long-term creep behavior of HDPE. Data from this study will be used in Task 5 which began this quarter. Experimental work in Task 4 has been completed and a Topical Report is being printed.

2. MECHANICAL AND CHEMICAL STABILITY OF CEMENT-BASED STRUCTURAL MATERIALS

Three types of cementitious material were prepared for this study, including Portland I and V, and a formulation prepared from information received from Ontario Hydro. This is designated Ontario Hydro-type cement mortar (OHCM), although it should be stated that such laboratory-sized specimens may not accurately simulate the actual material. These materials are used for both sulfate-attack and gamma-irradiation tests. Details of specimen preparation and testing were given in a previous quarterly report (WM-3291-5, 1987).

2.1 Sulfate - Attack Tests

These are accelerated tests to determine the susceptibility of cementitious barrier materials to deterioration from sulfates which are present in soils in contact with the cement or from sulfate-containing waste. The BNL procedure is based on that developed by Kalousek (1976). He showed that sulfate attack effects could be accelerated by a factor of eight if alternate wet-dry cycling of samples was adopted in place of continuous immersion in sulfate solution. The drying cycle evaporates water in the cement matrix and allows fresh sulfate solution to enter during reimmersion. Without the drying period, sulfate would penetrate more slowly into the cement pores by a diffusional process. Four replicate mortar bars were used for both the sulfate-attack tests and their corresponding controls.

The BNL immersion/drying cycle is:

Step 1: Immersion of specimens in 2.1% Na_2SO_4 solution (or deionized water for the control tests) at room temperature for 16 h.

Step 2: Forced-air drying of the specimens for 7 h 40 min at 54 ± 1°C.

Step 3: 20 min cooling of the specimens in still air.

Step 4: Repeat Step 1 through 4.

All testing begins with an immersion cycle with specimens (measuring 25.4 x 2.54 x 2.54 cm) placed in plastic containers of Na_2SO_4 solution (or deionized water). Glass rods are placed on the bottoms of the containers to assure solution contact on all sides of the test bars. During weekends the samples are left in the immersion cycle and they accumulate 64 h of soaking during this period. Sulfate-induced deterioration of concrete is caused by sulfate interacting with tricalcium aluminate in the cement paste to form a constituent with a larger volume. This causes volume increases in the cement and leads to cracking and failure of the concrete. Length-change measurements are typically used to estimate the degree of sulfate attack. Table 2.1, taken from the last quarterly report, shows that sulfate is most deleterious to the OHCM type formulation which is surprising since it is specially designed to provide resistance to this form of degradation. It was speculated that increased porosity in the cement matrix could be on explanation based on easier access of sulfate to the interiors of the test bars (WM-3291-7, 1988). Photographs of cross-sections of the concrete test bars were taken after the cyclic immersion tests had been completed to check whether there were major

Table 2.1 Length increase measurements on cement mortar bars exposed to alternate wet-dry cycling in 2.1% Na_2SO_4 solution or deionized water.

Specimen	Test Solution	Test Time(d)	Number of Cycles	Initial Dial Gage Reading (in)	Final Dial Gage Reading (in)	Length Change(in)	% Change(1)
Portland I							
1	DIW	71	48	0.1810	0.1820	0.0010	0.01
2	DIW	71	48	0.1785	0.1790	0.0005	0.01
4	DIW	71	48	0.1770	0.1775	0.0005	0.01
5	DIW	71	48	0.1740	0.1745	0.0005	0.01
1	DIW	180	115	0.1810	0.1820	0.0010	0.01
2	DIW	180	115	0.1785	0.1795	0.0010	0.01
4	DIW	180	115	0.1770	0.1780	0.0010	0.01
5	DIW	180	115	0.1740	0.1745	0.0005	0.01
8	Na_2SO_4	71	48	0.0470	0.0715	0.0245	0.25
9	Na_2SO_4	71	48	0.0335	0.0580	0.0245	0.25
10	Na_2SO_4	71	48	0.1780	0.2000	0.0220	0.22
11	Na_2SO_4	71	48	0.1800	0.2010	0.0210	0.21
8	Na_2SO_4	180	115	0.0470	0.2480	0.2010	2.01
9	Na_2SO_4	180	115	0.0335	0.2320	0.1985	1.99
10	Na_2SO_4	180	115	0.1780	0.3730	0.1950	1.95
11	Na_2SO_4	180	115	0.1800	0.3710	0.1910	1.91
Portland V							
1	DIW	69	45	0.1605	0.1595	-0.0010	-0.01
2	DIW	69	45	0.1625	0.1615	-0.0010	-0.01
3	DIW	69	45	0.0470	0.0460	-0.0010	-0.01
4	DIW	69	45	0.0340	0.0330	-0.0010	0.01
1	DIW	166	105	0.1605	0.1610	0.0005	0.01
2	DIW	166	105	0.1625	0.1630	0.0005	0.01
3	DIW	166	105	0.0470	0.0475	0.0005	0.01
4	DIW	166	105	0.0340	0.0340	0	0
5	Na_2SO_4	69	45	0.1845	0.1895	0.0050	0.05
6	Na_2SO_4	69	45	0.1790	0.1835	0.0045	0.05
7	Na_2SO_4	69	45	0.0270	0.0335	0.0065	0.07
8	Na_2SO_4	69	45	0.0550	0.0605	0.0055	0.06
5	Na_2SO_4	166	105	0.1845	0.2180	0.0335	0.34
6	Na_2SO_4	166	105	0.1790	0.2125	0.0335	0.34
7	Na_2SO_4	166	105	0.0270	0.0745	0.0475	0.48
8	Na_2SO_4	166	105	0.0550	0.0990	0.0440	0.44
Ontario Hydro Type							
2	DIW	74	44	0.1710	0.1770	0.0060	0.06
4	DIW	74	44	0.0445	0.0515	0.0070	0.07
5	DIW	74	44	0.1600	0.1665	0.0065	0.07
6	DIW	74	44	0.1525	0.1580	0.0055	0.06
2	DIW	159	100	0.1710	0.1815	0.0105	0.11
4	DIW	159	100	0.0445	0.0565	0.0120	0.12
5	DIW	159	100	0.1600	0.1720	0.0120	0.12
6	DIW	159	100	0.1525	0.1630	0.0105	0.11
7	Na_2SO_4	74	44	0.0250	0.0690	0.0430	0.43
8	Na_2SO_4	74	44	0.0410	0.0875	0.0465	0.47
9	Na_2SO_4	74	44	0.1565	0.1900	0.0335	0.34
11	Na_2SO_4	74	44	0.0300	0.0800	0.0500	0.50
7	Na_2SO_4	159	100	0.0250	0.3030	0.0278	2.78
8	Na_2SO_4	-	-	0.0410	-(2)	-	-
9	Na_2SO_4	159	100	0.1565	0.3915	0.235	2.35
11	Na_2SO_4	-	-	0.0300	-(2)	-	-

Note:

(1) Based on an effective gagelength of 10.00 in.

(2) No measurement performed because mortar bar fractured during immersion/oven transfer.

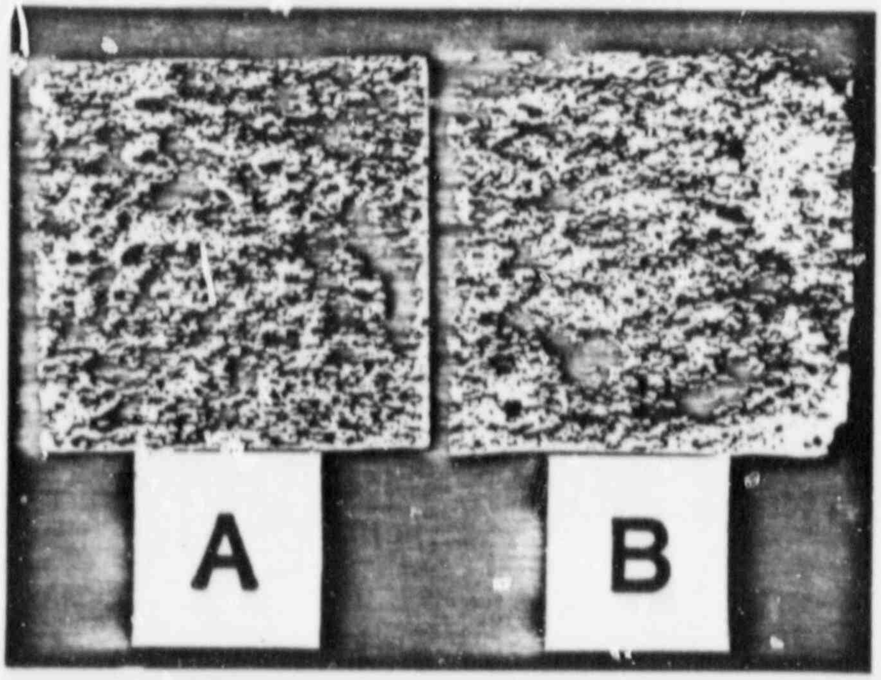

Figure 2.1 Pore structure in Portland I cement mortar bars after 115 wet-dry cycles in deionized water (A) and sulfate solution (B). Mag. 2X.

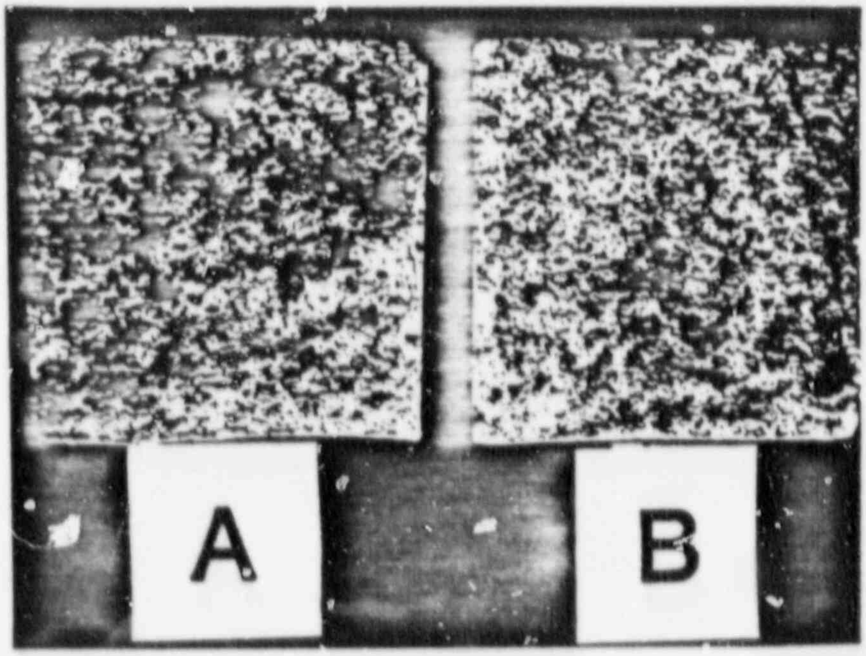

Figure 2.2 Pore structure in Portland V cement mortar bars after 105 wet-dry cycles in deionized water (A) and sulfate solution (B). Mag. 2X.

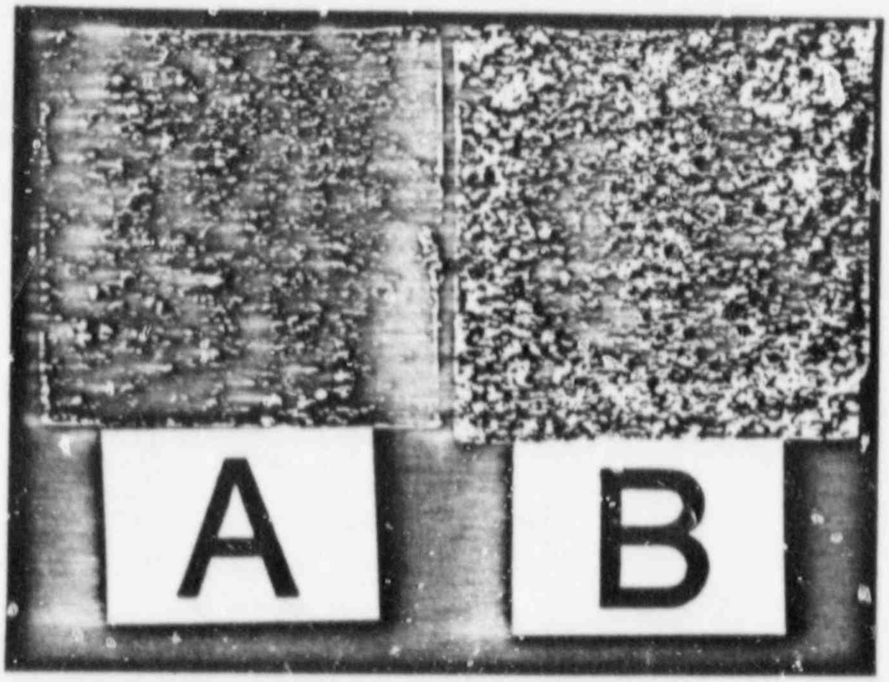

Figure 2.3 Pore structure in OHCM bars after 100 wet-dry cycles in deionized water (A) and sulfate solution (B). Mag. 2X.

variations in porosity among the three materials. Figures 2.1 through 2.3 show these results for test bars that were immersion cycled in deionized water and bars cycled in the sulfate test solutions. Portland I and V cement mortars show very similar types of porosity, viz., very small pores with a number of much larger pores distributed throughout the section. For the OHCM the pore structure is similar but the larger pores are slightly more numerous and larger than those for Portlands I and V cement mortars. However, they are not interconnected and do not appear to provide rapid flow pathways for sulfate solution. Thus, excessive porosity in the OHCM samples does not offer a satisfactory explanation for the poor resistance to sulfate attack. In fact, there is evidence that air entrainment in cement could be beneficial with respect to sulfate attack (Lea, 1971). At this time there is no satisfactory explanation for the poor performance of the OHCM. Additional work will not be expended in this effort because of lack of funding.

3. DEGRADATION MECHANISMS IN HIGH-DENSITY POLYETHYLENE (HDPE)

3.1 Overview of Research Activities

High-density polyethylene is currently being used as a high-integrity container material for low-level wastes. Because of the need for such containers to maintain their structural integrity for at least 300 years (NRC Technical Position on Waste Form) potential failure/degradation modes must be determined for a range of environmental conditions. These include consideration of mechanical stress, gaseous/liquid environments within and external to the container, and the gamma radiation field. In some instances it is necessary to test under conditions more aggressive than those anticipated under shallow-land burial conditions so that failure or degradation modes can be more quickly identified and their relative importance assessed.

A combination of simple inexpensive tests (stressed U-bend samples) and more sophisticated uniaxial creep tests are being used to define the ranges of conditions for which mechanical failure/degradation is important. The creep test environments include Igepal CO-630, turbine oil and liquid scintillation fluid as well as air and deionized water (DIW), the control environments. Igepal CO-630 is a surfactant specified in standard ASTM tests for environmental stress cracking. Turbine oil is a possible constituent of low-level waste generated at reactor power plants, and is used in the current tests because of its known detrimental behavior to many types of plastic. Liquid scintillation fluids are not likely to be disposed of in burial sites at this time because of more stringent controls on their disposal. However, they are being evaluated here because they are representative of the class of organic solvents containing toluene and xylene. As such they will give valuable insights regarding a type of potential failure or degradation mode of HDPE.

In addition to the above-mentioned creep tests, the effect of gamma irradiation on mechanical properties is being studied. U-bend samples of HDPE are being irradiated in the BNL gamma irradiation test facility to check for crack initiation and propagation, and the creep of in-test-irradiated HDPE was recently initiated to quantify creep behavior.

A description of the various subtasks is given below.

3.2 Crack Initiation and Propagation in a Gamma-Radiation Environment

Crack initiation and propagation is important in stressed HDPE containers because of the anticipated embrittlement by gamma irradiation. A simple inexpensive test was developed at BNL involving the use of static "U-bend" samples exposed to air and gamma radiation. It involved the evaluation of miniature U-bends manufactured from HDPE strips measuring 10.2 x 1.27 x 0.32 cm (4" x 0.5" x 0.125"). Holes were drilled at distances of 1.27 cm (0.5") from each end of the strips so that nuts and bolts could be used to hold the ends of the strip together when the U-bends were made. The specimens were prepared with the outer surfaces of the U-bends in three different conditions:

Type I - the as-received oxidized condition, which will have "natural" cracks present, as a result of bending, Type II - as above, but with 10 mils of the oxidized surface removed with emery paper prior to bending. No cracks were formed during bending,

Type III - the as-received "non-oxidized" surface which also does not crack during bending.

Table 3.1 shows the test matrix for the U-bend irradiation tests.

Table 3.1 Test matrix for crack-propagation studies on irradiated Marlex CL-100 miniature U-bend specimens.

Gamma Dose Rate (rad/h)	Outer Surface Condition of U-Bend		
	Oxidized Surf. Present (Type I)	Oxidized Surf. Removed (Type II)	Non-Oxidized Surf. Present (Type III)
0	8[(1)]	8	3
1.4×10^3	8	8	8
8.4×10^3	8	8	8
4.4×10^5	8	8	8

(1) Number of replicate specimens.

Figure 3.1 shows the U-bend specimens mounted on aluminum frames to facilitate irradiation. At the time the photograph was taken, the batches of specimens (A, B, and C) had accumulated 2.1×10^6 rad, 1.3×10^7 rad, and 6.7×10^8 rad, respectively. At the time of examination this reporting period, the doses have reached 1.3×10^7 rad, 9.5×10^7 rad, and 3.1×10^9 rad, respectively. Figures 3.2 through 3.9 show sketches of cracks in the apex regions of Type I specimens for the various irradiation conditions. Crack patterns immediately after specimen bending are shown together with the patterns after the given irradiation doses. To obtain statistical data on the cracking behavior, the number of cracks in Type I specimens were counted. The results are given in Table 3.2. Large cracks are defined as those with a length greater than one-half of the specimen width (i.e., >0.64 cm). A small crack is one with a length less or equal to one-half of the specimen width. The number of cracks given in Table 3.2 are the totals for each batch of 8 replicate specimens.

The numbers of small starting cracks in the unirradiated control batch were higher than for the other three batches. The majority of these cracks were present in Specimens 46 and 48 and, probably, were caused by small differences in the bending technique for these two specimens during fabrication into U-bend configurations. Nevertheless, the data given in Table

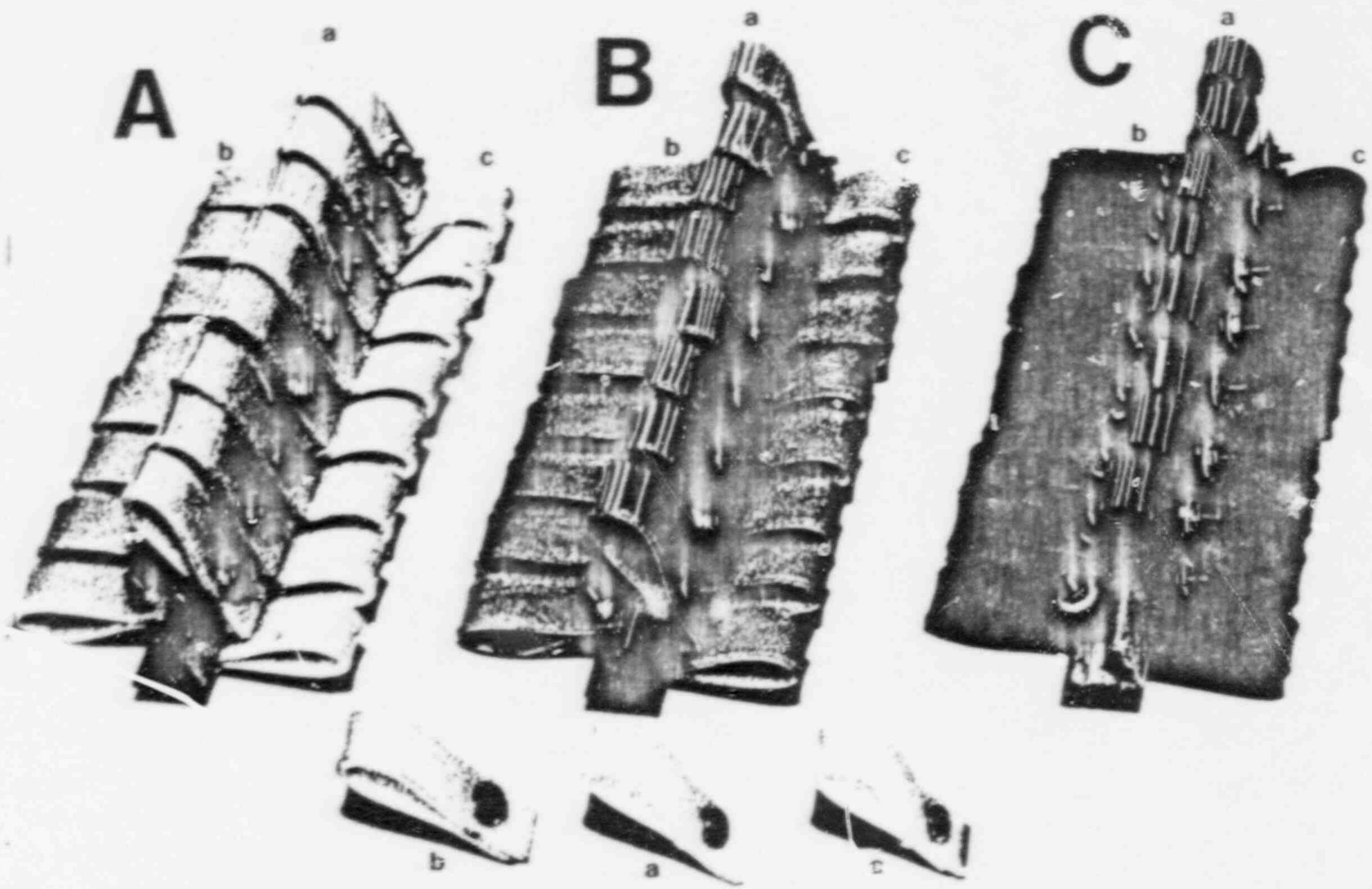

Figure 3.1 Appearance of Type I (a), Type II (b), and Type III (c) Marlex CL-100 HDPE U-bend specimens gamma irradiated to 2.1×10^6 rad (A), 1.3×10^7 rad (B), and 6.7×10^8 rad (C). Individual unirradiated Type I, Type II, and Type III control specimens are shown at the bottom of the figure. Magnification 0.8 X.

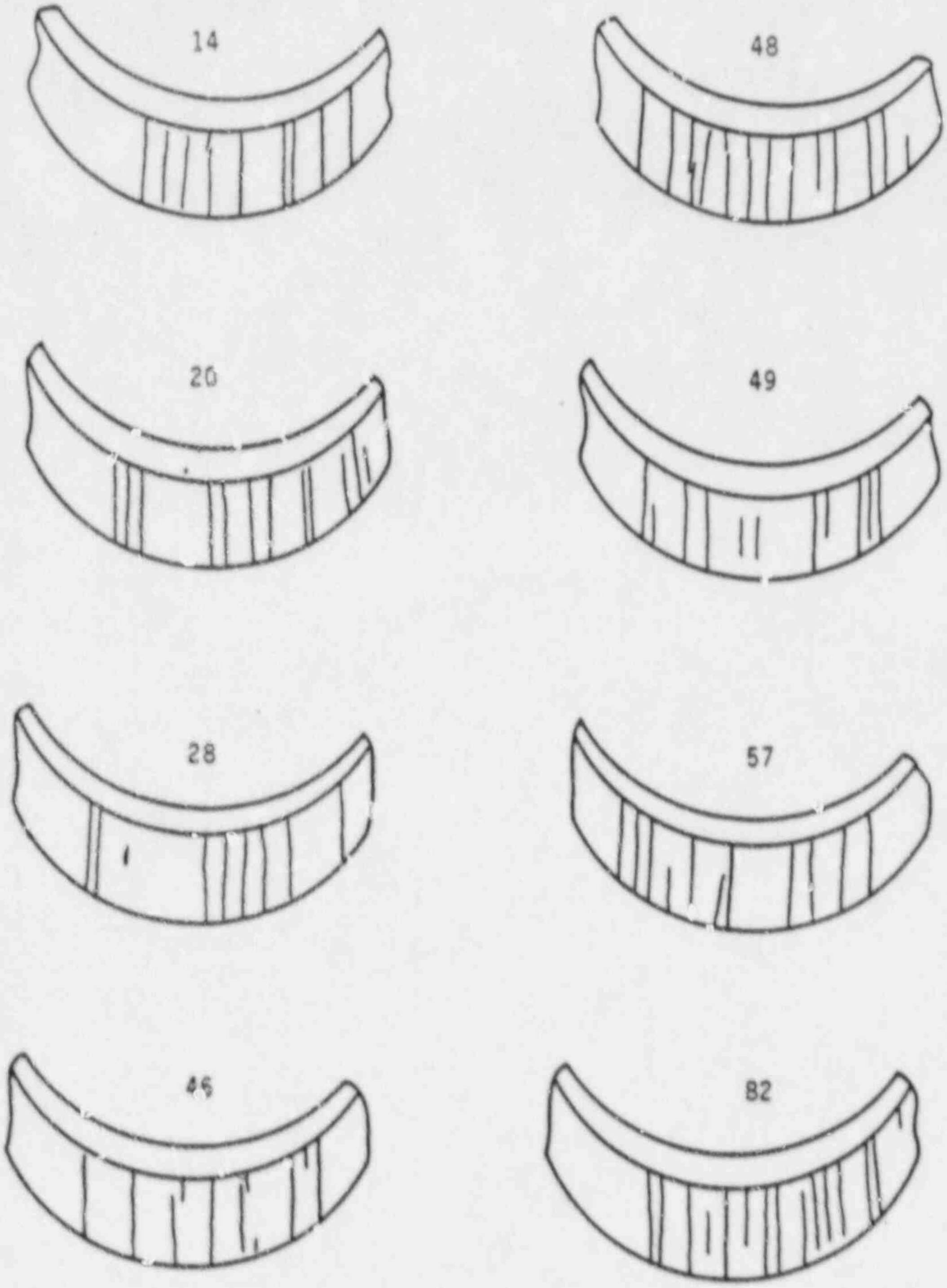

Figure 3.2 Crack patterns in as-prepared Type I Marlex CL-100 HDPE U-bend samples. Specimens are unirradiated controls. Specimen numbers given above each sketch.

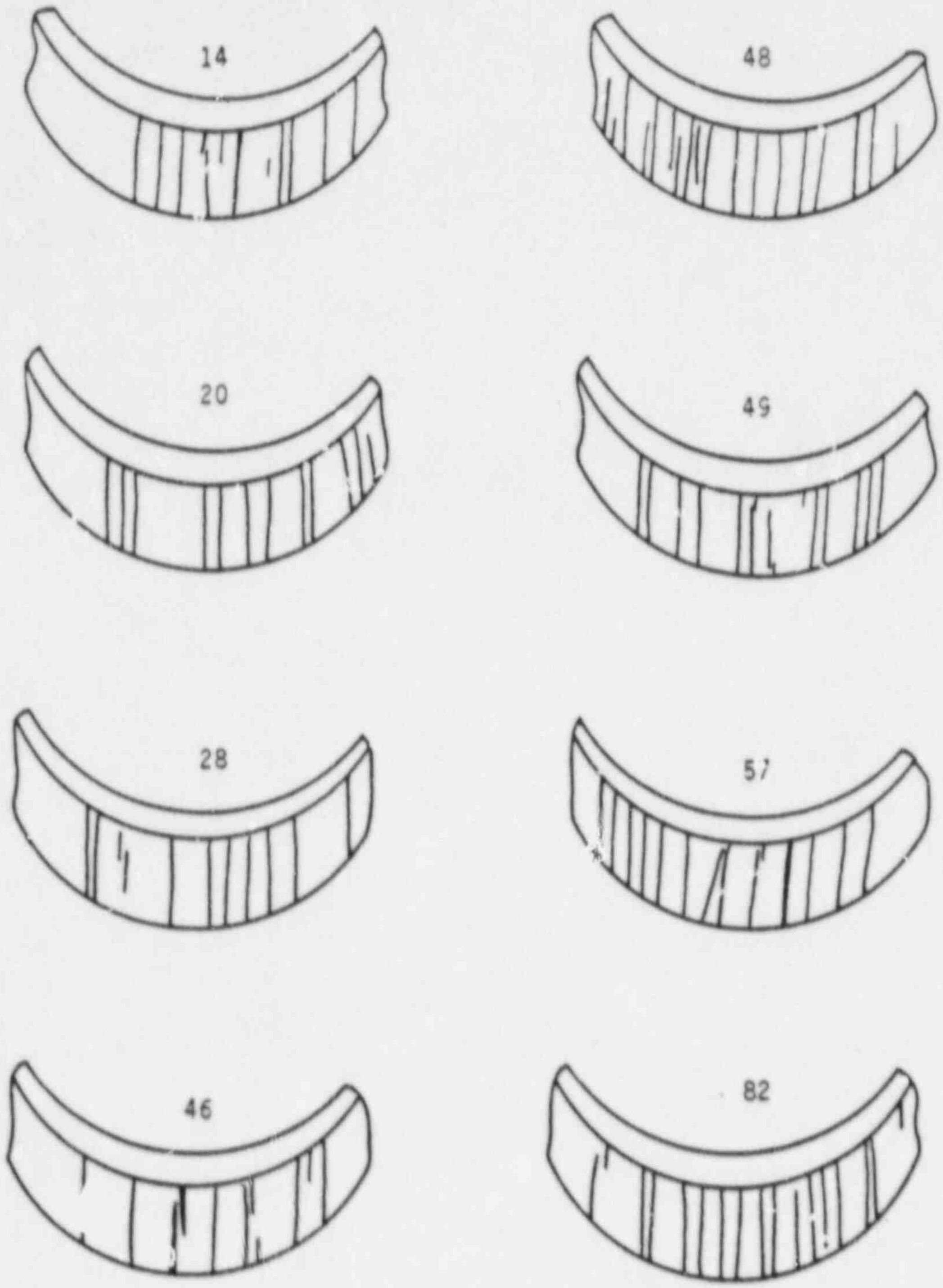

Figure 3.3 Crack patterns in unirradiated Type I Marlex CL-100 HDPE U-bend samples held at 10 C for 530 d in air. Specimen numbers given above each sketch.

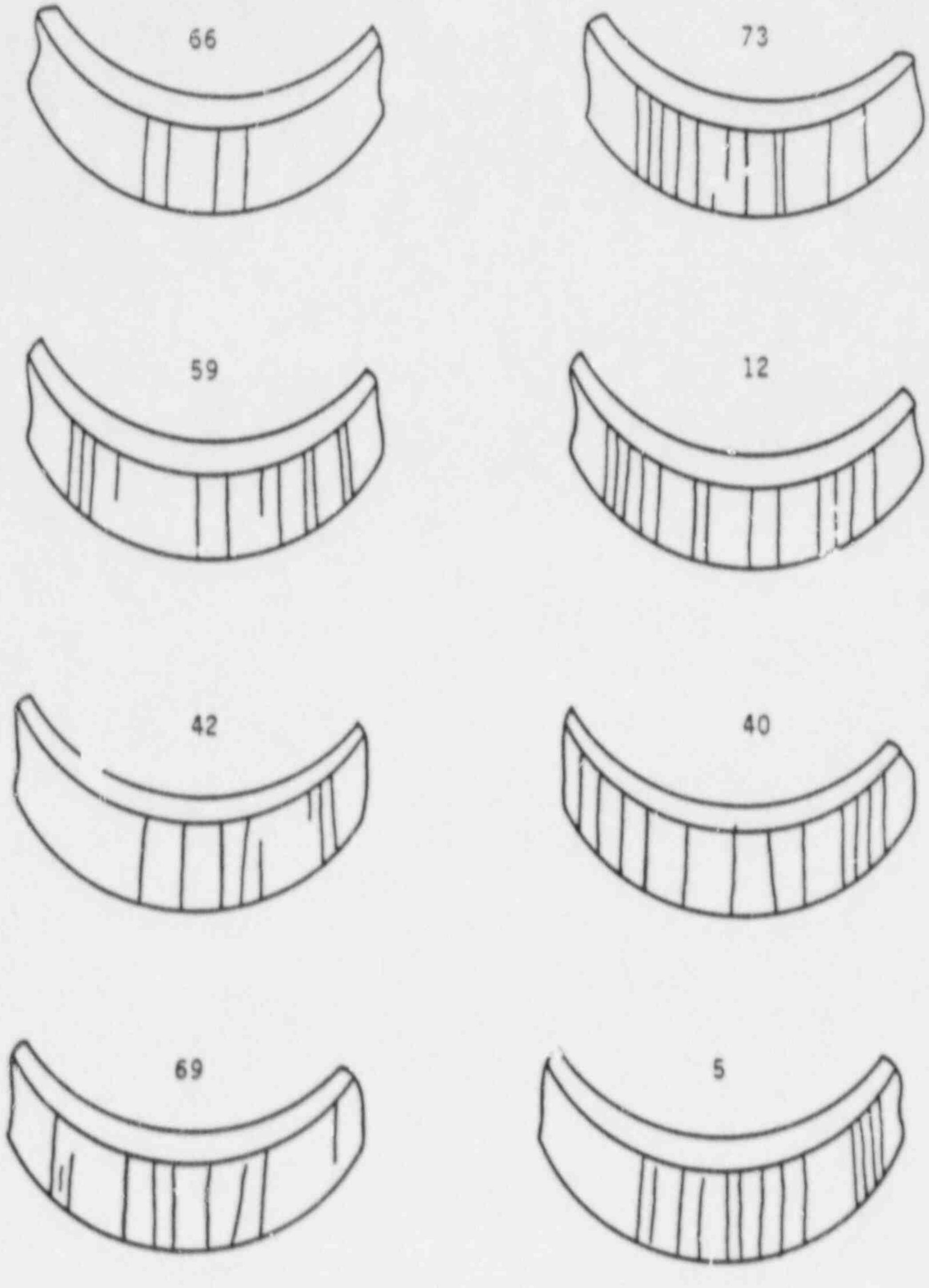

Figure 3.4 Crack patterns in as-prepared Type I Marlex CL-100 HDPE U-bend samples prior to gamma irradiation at 1.4×10^3 rad/h. Specimen numbers given above each sketch.

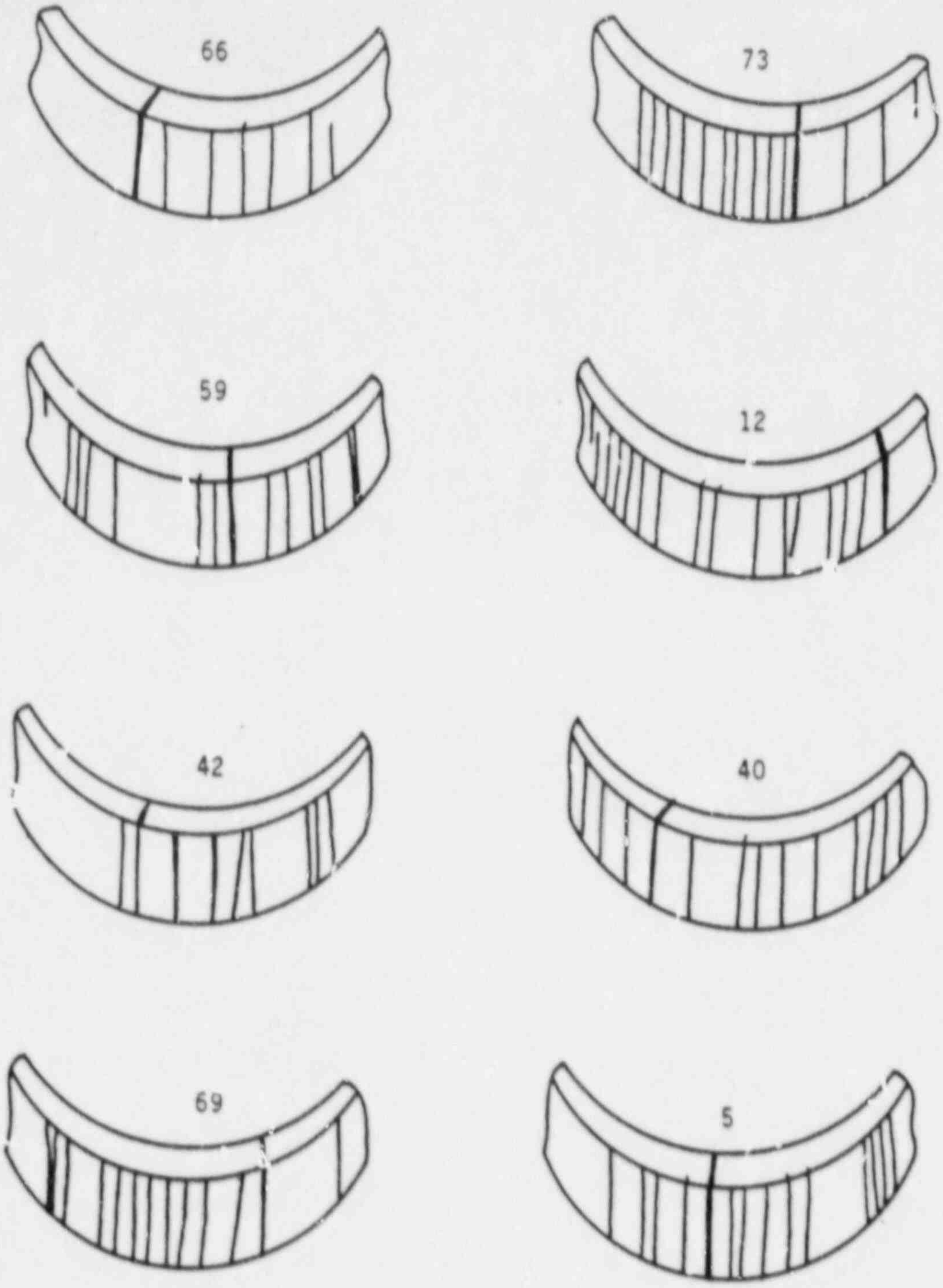

Figure 3.5 Crack patterns in Type I Marlex CL-100 HDPE U-bend samples after gamma irradiation to 1.3×10^7 rad at a dose rate of 1.4×10^3 rad/h. Specimen numbers given above each sketch.

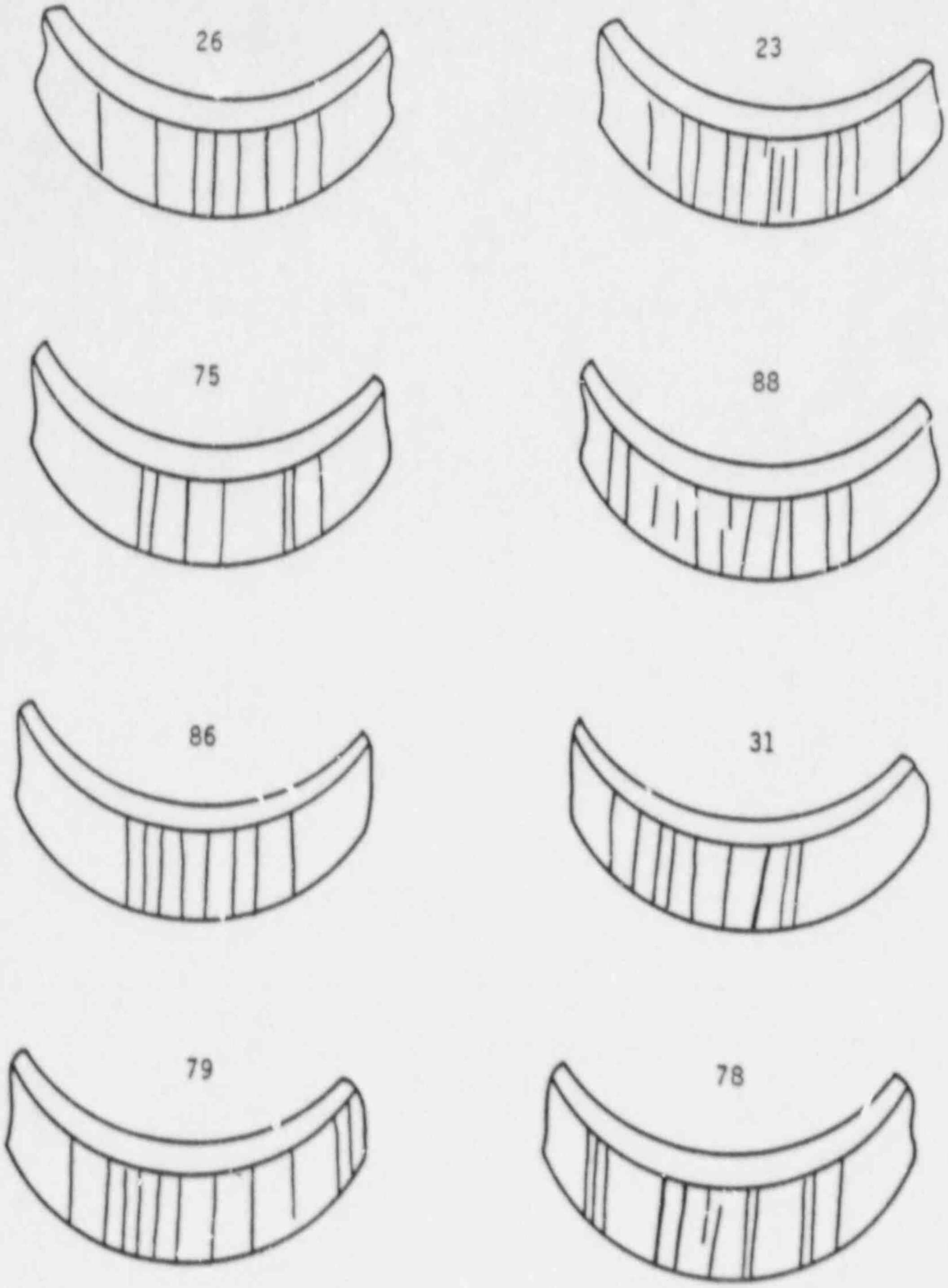

Figure 3.6 Crack patterns in as-prepared Type I Marlex CL-100 HDPE U-bend samples prior to gamma irradiation at 8.4×10^3 rad/h. Specimen numbers given above each sketch.

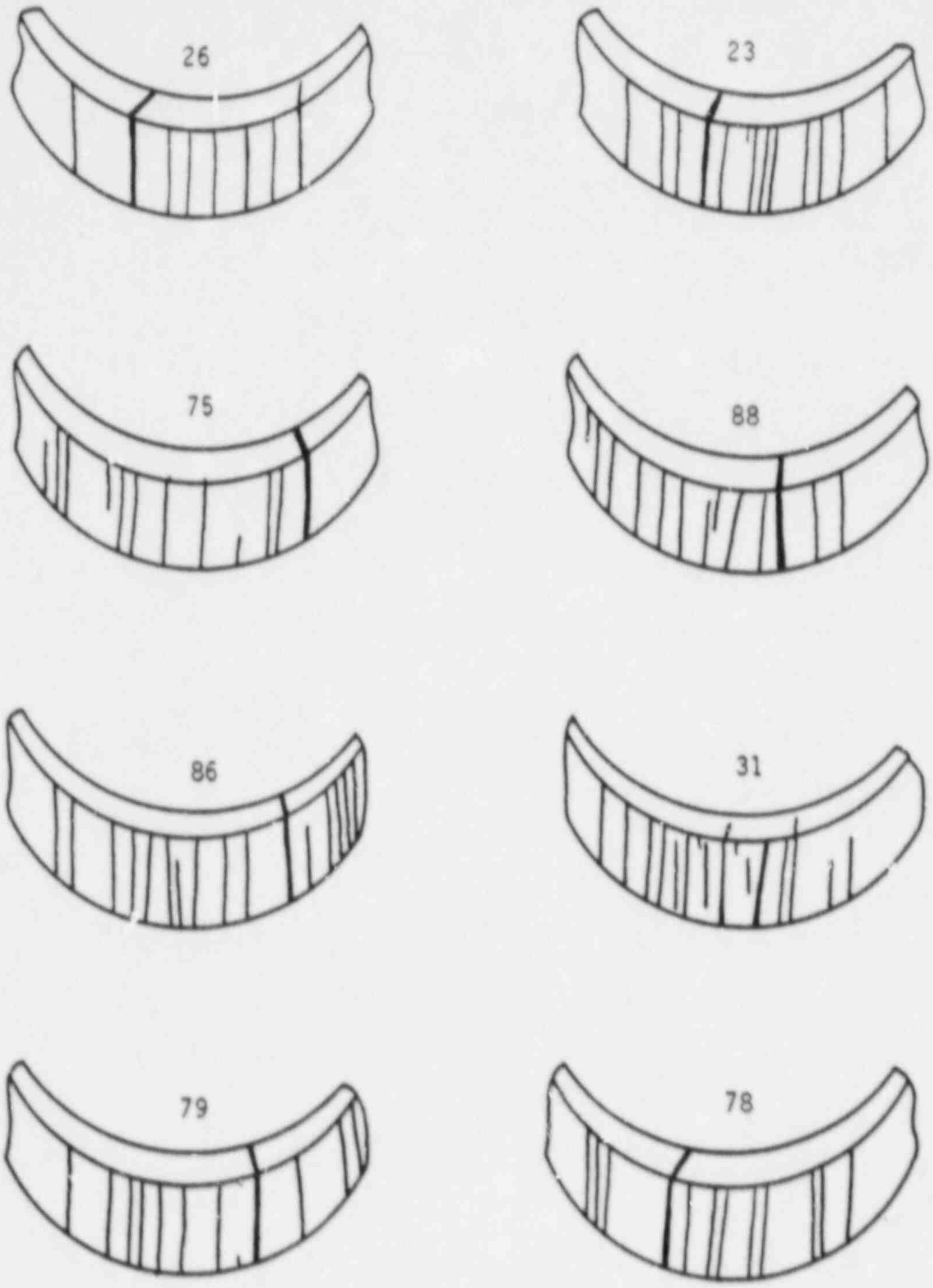

Figure 3.7 Crack patterns in Type I Marlex CL-100 HDPE U-bend samples after gamma irradiation to 9.5×10^7 rad at a dose rate of 8.4×10^7 rad/h. Specimen numbers given above each sketch.

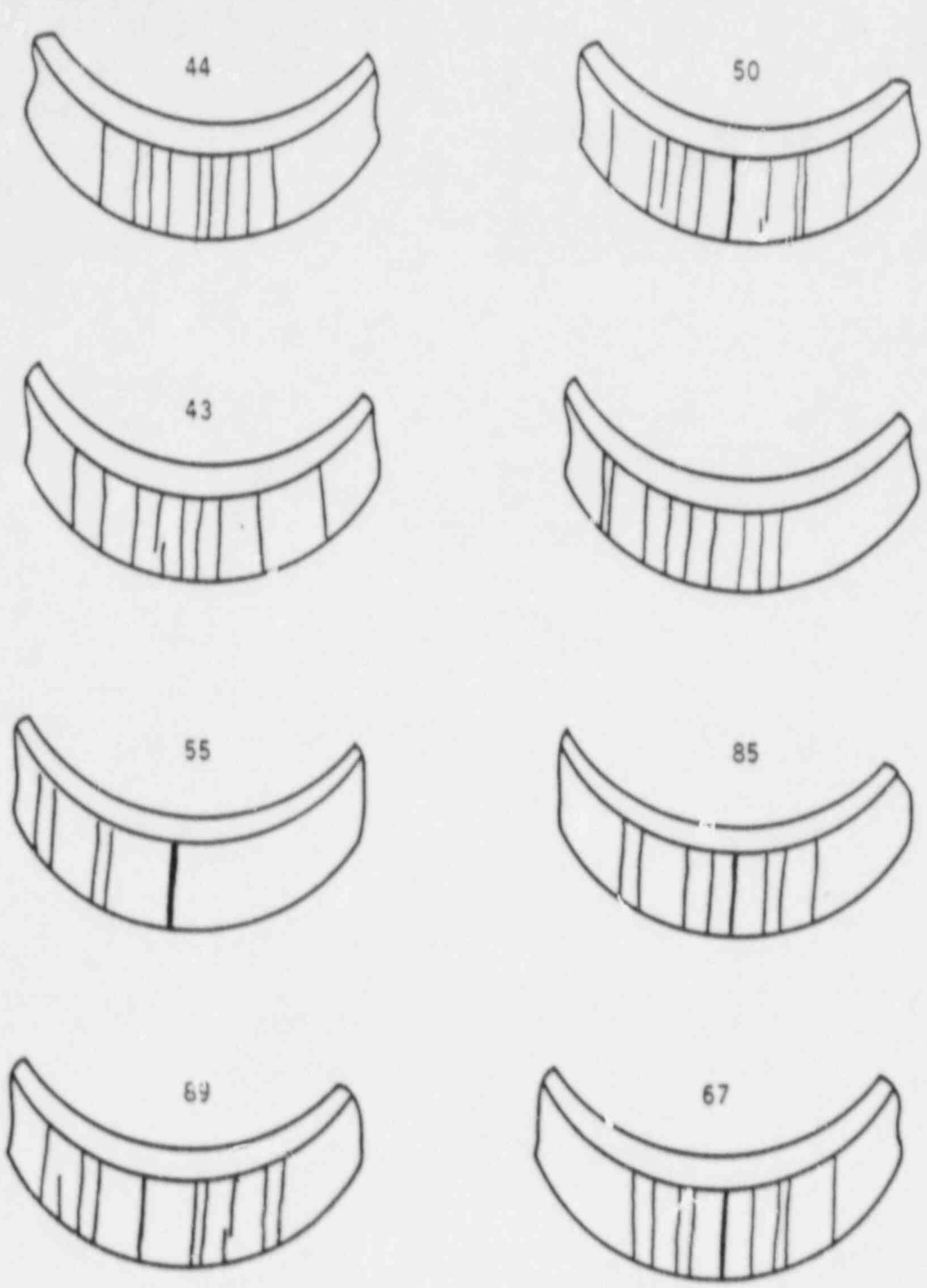

Figure 3.8 Crack patterns in as-prepared Type I Marlex CL-100 HDPE U-bend samples prior to gamma irradiation at 4.4×10^5 rad/h. Specimen numbers given above each sketch.

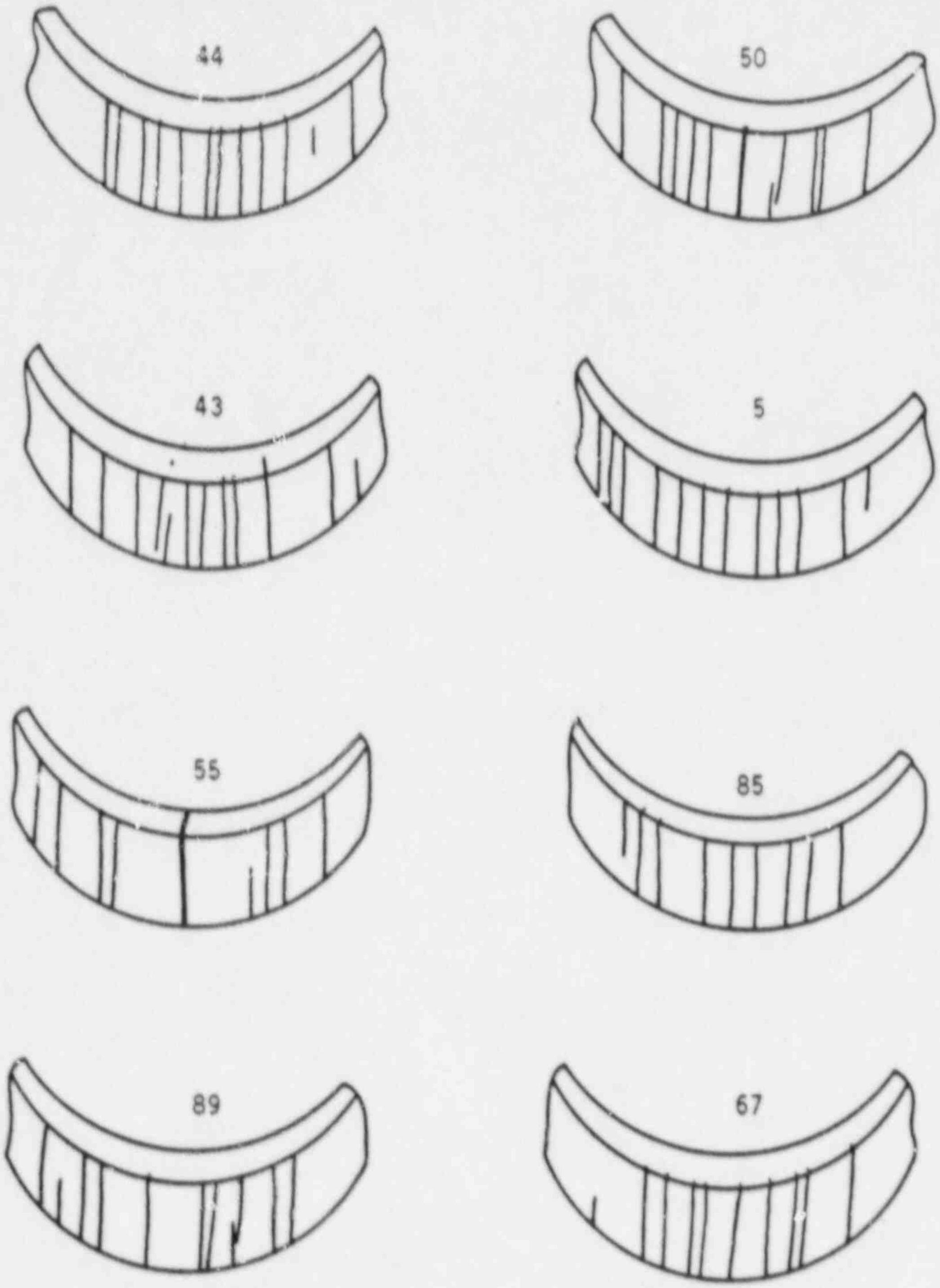

Figure 3.9 Crack patterns in Type I Marlex CL-100 HDPE U-bend samples after irradiation to 3.1×10^9 rad at a dose rate of 4.4×10^5 rad/h. Specimen numbers given above each sketch.

Table 3.2 Crack initiation and propagation in Type I HDPE U-bend specimens exposed to gamma irradiation.

Irradiation Dose	Cracks Before Irradiation			Cracks After Irradiation			Percent Change in Numbers of Cracks			Full Penet. Cracks	Near Full Penet. Cracks
	Large	Small	Total	Large	Small	Total	Large	Small	Total		
Unirradiated Controls	84	11	95	95	15	110	13	36	16	0	0
1.3×10^7 rad (at 1.4×10^3 rad/h)	81	3	84	97	3	100	20	0	15	7	1
9.5×10^7 rad (at 8.4×10^3 rad/h)	78	2	30	94	8	102	21	300	28	7	1
3.1×10^9 rad (at 4.4×10^5 rad/h)	69	3	72	83	3	86	21	0	19	1	0

3.2 for the numbers of large and small cracks in Type I specimens, irradiated at different dose rates, show well-defined trends. For example:

- The full-penetration cracks which completely fracture the U-bend specimens into two pieces occur almost exclusively in the low and intermediate gamma dose rate environments.

- Unirradiated specimens show the smallest increase in the total numbers of cracks (16 percent) and no deep cracks were observed.

- The intermediate dose rate (8.4×10^3 rad/h) gives the large percent increase in the total number of cracks.

These irradiation tests are continuing. However, in the future, there may be slower changes in crack densities in the Type I specimens since a large number of specimens have completely fractured, leading to losses in the tensile stresses which are responsible for crack initiation and propagation. Future studies will mainly be focused on Type II and Type III specimens which, to date, only show fine cracking (WM-3291-6, 1988).

3.3 Uniaxial Creep Behavior in Selected Environments

Creep tests are continuing at a test temperature of 20°C (68°F) using a simple dead-load system. Strains are measured using LVDTS (linearly variable differential transducers). Rates of creep, ductility-at-failure, and weight increase in the specimens caused by the absorption of the test liquids during creep are all measured. Tables 3.3 and 3.4 show the results accumulated to date. The data for the "old HDPE" were obtained from an earlier batch of material, and the remainder are for a newer supply purchased about 2 years ago. Current work is on the newer material, except for the irradiation-creep study which is described in Section 3.4.

Figures 3.10 and 3.11 show the latest stress-rupture and creep-ductility plots for tests in air, deionized water, scintillation fluid, turbine oil and Igepal. There appears to be reasonably well-defined threshold stresses below which failure should not occur in the latter three environments, but for air and water longer-term testing will be needed to define this threshold. Note in Figure 3.11 that at the lowest stress levels (less than 8 MPa) the ductility of the HDPE is low and apparently approaching an embrittlement regime. This is not the case, however, for scintillation fluid for which ductilities in the range of 90-110 percent are observed.

Figures 3.12 and 3.13 show the beneficial effects of removing the oxidized surface layer from HDPE. In Igepal the rupture times are increased at all stress levels compared to as-received material. The threshold stress for creep is also increased by about 1·2 MPa (175 psi). In the case of scintillation fluid, removal of the oxidized layer mainly increases the threshold stress. The failure times at the higher stress levels are quite similar for non-oxidized and as-received HDPE.

Table 3.3 Creep test data for Marlex CL-100 HDPE tested in air and deionized water at room temperature.

Test Number	Specimen Condition	Test Environment	Stress (MPa)	Stress (psi)	Failure Time (h)	Elong. at Break (%)
381	As rec.	Air	13.79	2000	4.0	56.0
382	As rec.	Air	13.79	2000	0.98	56.6
367	As rec.	Air	13.10	1900	6.8	56.8
369	As rec.	Air	13.10	1900	[illegible].8	46.6
374	As rec.	Air	12.76	1850	5.4	46.2
365	As rec.	Air	12.41	1800	41.0	74.6
359	As rec.	Air	12.41	1800	11.3	50.0
366	As rec.	Air	12.10	1750	52.5	76.0
358	As rec.	Air	11.72	1700	80.3	79.6
362	As rec.	Air	11.65	1690	28.5	5[illegible].0
361	As rec.	Air	11.03	1600	457	79.7
360	As rec.	Air	10.86	1575	212	85.4
350(a)	As rec.	Air	10.62	1540	166	72.9
350(b)	As rec.	Air	10.62	1540	502	55.0
300	As rec.	Air	10.34	1500	662	61.6
315	As rec.	Air	10.34	1500	761	58.2
363	As rec.	Air	10.1[illegible]	1475	4023	60.4
357	As rec.	Air	10.00	1450	3821	55.1
380	As rec.	Air	10.00	1450	2455	66.8
377	As rec.	Air	9.83	1425	5173	53.5
316	As rec.	Air	9.65	1400	5378	36.6
364	As rec.	Air	9.31	1350	1819	71.9
391	As rec.	Air	9.13	1325	[illegible]610	38.9
355	As rec.	Air	8.96	1300	3100	37.0
390	As rec.	Air	8.96	1300	2808	33.7
321	As rec.	Air	8.27	1200	7740	16.2
388	As rec.	Air	7.93	1150	>5400	-
322	As rec.	Air	7.24	1050	>14400	-
387	(1)	Air	13.79	2000	>5092	>673
385	(1)	Air	12.41	1800	>5257	>598
338	(1)	Air	11.03	1600	2319	585
323	(1)	Air	10.34	1500	7704	248.9
320	(1)	Air	8.27	1200	Ongoing	-
337	As rec.	DIW	11.03	1600	112	58.6
347	As rec.	DIW	10.69	1550	57	54.5
301	As rec.	DIW	10.34	1500	2027	95.5
302	As rec.	DIW	9.65	1400	5854	54.5
334	As rec.	DIW	8.27	1200	Ongoing	-
339	(1)	DIW	11.03	1600	206	221.6
327	(1)	DIW	10.34	1500	452	85.2
326	(1)	DIW	8.27	1200	Ongoing	-
306	(2)	DIW	10.34	1500	1154	57.5
310	(2)	DIW	9.65	1400	[illegible]264	53.5

NOTES:

1. 10 mils removed from oxidized surface of specimen.
2. 10 mils removed from non-oxidized surface of specimen.

Table 3.4 Creep test data for Marlex CL-100 HDPE tested in various environments at room temperature.

Test Number	Specimen Condition	Test Environment	Stress (MPa)	Stress (psi)	Failure Time (h)	Elong. at Break (%)	Weight Change (% per test day)
370	As rec.	Oil	11.03	1600	45.9	92.1	0.19
305	As rec.	Oil	10.34	1500	128	60.3	0.06
345	As rec.	Oil	8.96	1300	102	43.4	0.02
348	As rec.	Oil	8.96	1300	168	51.1	0.04
352	As rec.	Oil	8.62	1250	198	54.5	0.01
325	As rec.	Oil	8.27	1200	1502	36.3	-
396	As rec.	LSF	12.41	1800	7.9	35.0	0.72
379	As rec.	LSF	11.72	1700	9.6	94.8	-
378	As rec.	LSF	11.38	1650	10.5	49.3	1.47
[illegible]	As rec.	LSF	11.03	1600	33.1	89.9	0.34
[illegible]	As rec.	LSF	10.34	1500	14	83.5	0.25
309	As rec.	LSF	9.65	1400	35	98.0	0.15
341	As rec.	LSF	8.27	1200	50	98.5	1.40
333	As rec.	LSF	7.24	1050	340	95.8	0.02
383	As rec.	LSF	6.89	1000	1602	111.4	-
394	As rec.	LSF	6.72	975	Ongoing	-	-
330	(1)	LSF	9.65	1400	29	76.0	-
332	(1)	LSF	8.27	1200	85	216.0	-
331	(1)	LSF	7.24	1050	Ongoing	-	-
313	(2)	LSF	10.34	1500	12	55.0	0.28
311	(2)	LSF	9.65	1400	31	77.0	0.21
42	Old HDPE	LSF	10.34	1500	12	86.1	-
55	Old HDPE	LSF	9.65	1400	35	37.8	-
62	Old HDPE	LSF	8.27	1200	280	100.7	-
392	As rec.	Igepal	12.41	1800	3.1	52.3	0.27
393	As rec.	Igepal	11.72	1700	17.7	51.8	0.23
401	As rec.	Igepal	11.72	1700	20.7	49.1	-
371	As rec.	Igepal	11.03	1600	45.6	54.7	0.13
303	As rec.	Igepal	10.34	1500	65	49.1	0.06
304	As rec.	Igepal	9.65	1400	106	54.8	0.03
346	As rec.	Igepal	8.96	1300	128	47.3	0.02
324	As rec.	Igepal	8.27	1200	1194	22.0	-
389	As rec.	Igepal	8.10	1175	Ongoing	-	-
106	Old HDPE	Igepal	12.41	1800	8	74.8	-
105	Old HDPE	Igepal	12.41	1800	9	69.2	-
107	Old HDPE	Igepal	11.72	1700	50	87.4	-
108	Old HDPE	Igepal	11.72	1700	47	54.6	-
72	Old HDPE	Igepal	10.34	1500	216	68.7	-
113	Old HDPE	Igepal	10.34	1500	366	62.2	-
114	Old HDPE	Igepal	10.34	1500	372	79.2	-
84	Old HDPE	Igepal	10.34	1500	95	115.3	-
340	(1)	Igepal	10.34	1500	312	97.3	0.04
329	(1)	Igepal	9.65	1400	476	59.4	-
328	(1)	Igepal	9.29	1350	12778	31.2	-
314	(2)	Igepal	10.34	1500	56	71.4	0.09
312	(2)	Igepal	9.65	1400	130	50.4	0.0[illegible]

NOTES:
1. 10 mils removed from oxidized surface of specimen.
2. 10 mils removed from non-oxidized surface of specimen.

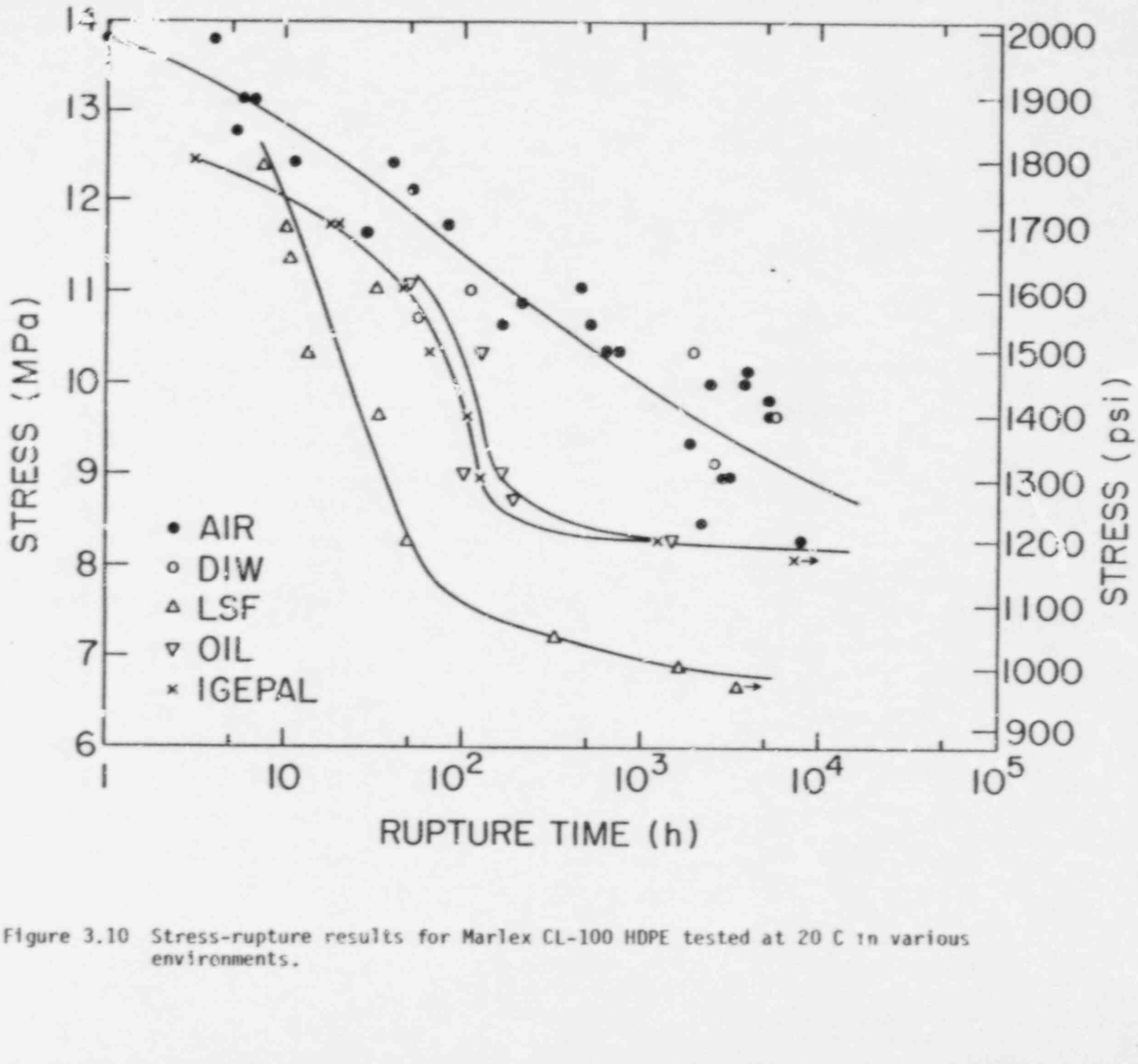

Figure 3.10 Stress-rupture results for Marlex CL-100 HDPE tested at 20 C in various environments.

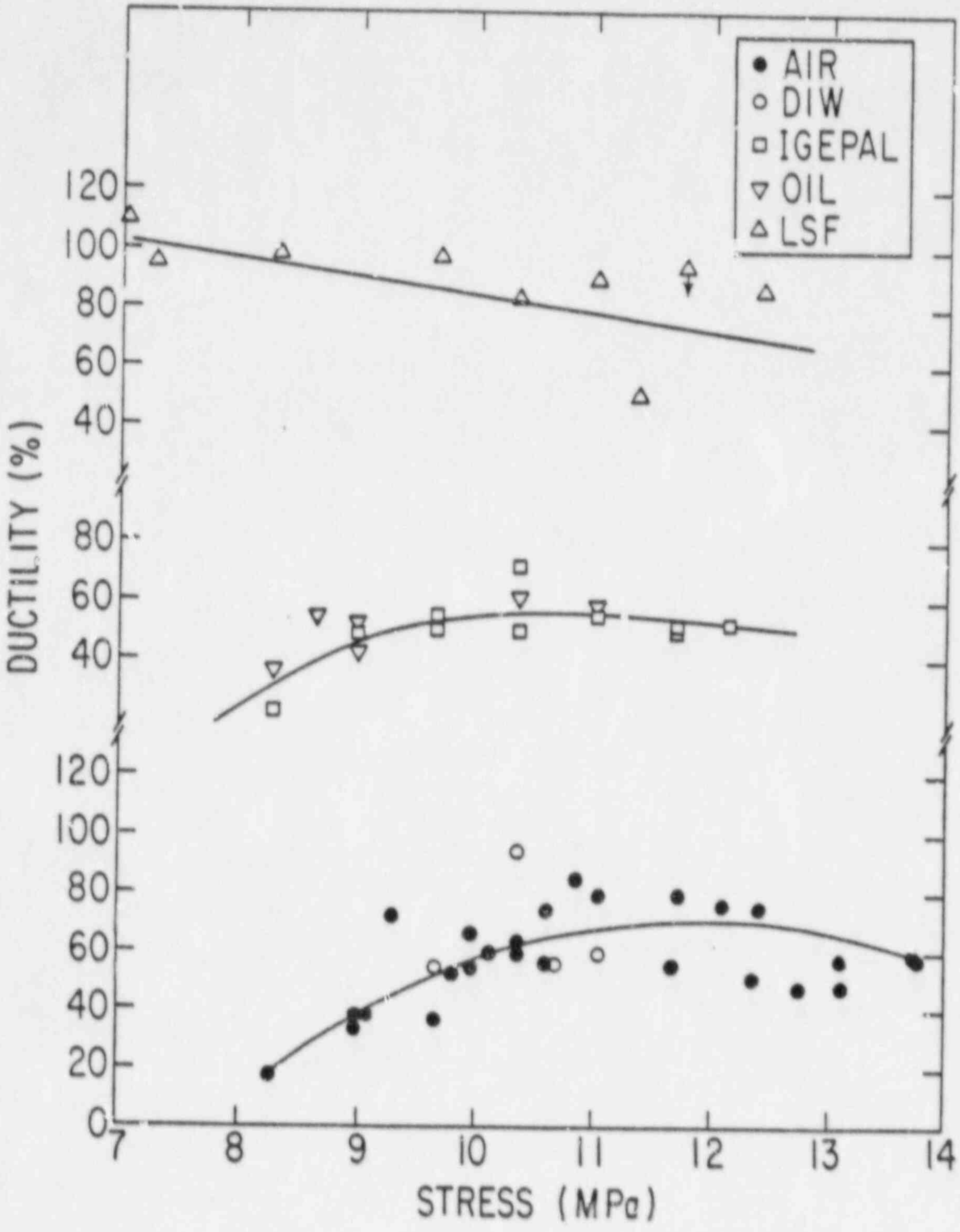

Figure 3.11 Elongations at failure for Marlex CL-100 HDPE tested at 20 C in various environments.

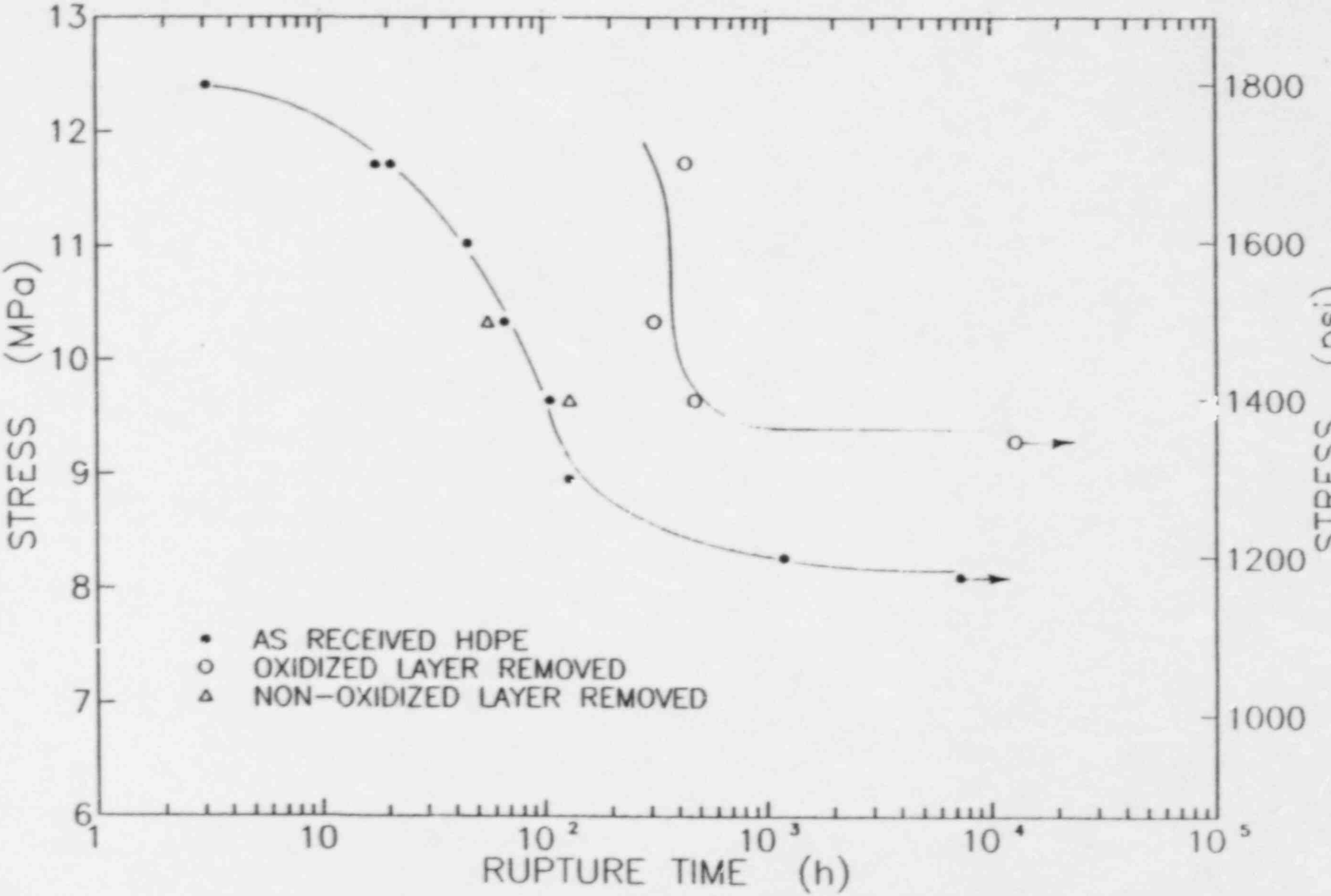

Figure 3.12 Effect of removing the oxidized and the non-oxidized surface layers on the stress rupture of Marlex CL-100 HDPE in Igepal CO-630.

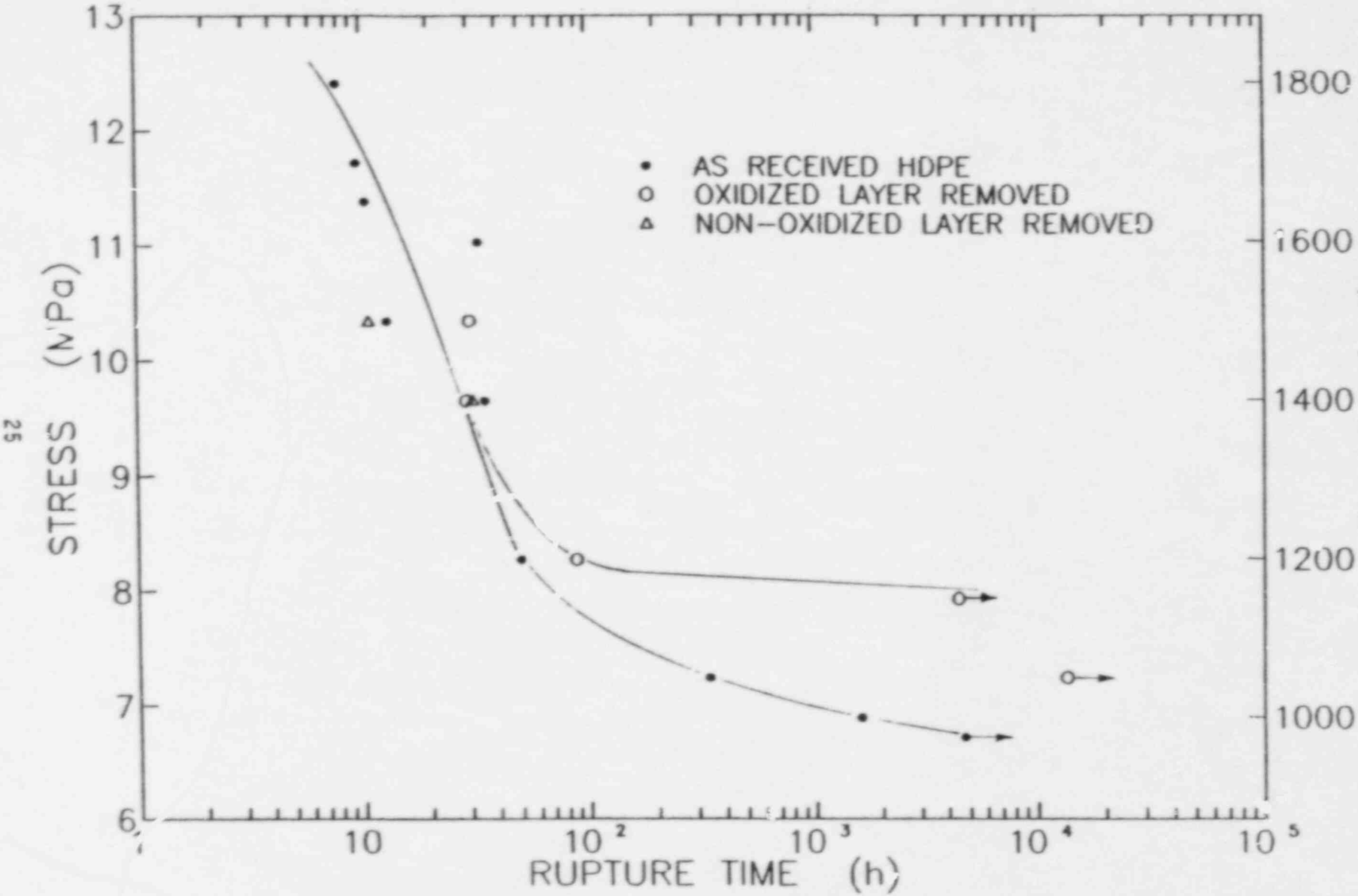

Figure 3.13 Effect of removing the oxidized and the non-oxidized surface layers on the stress rupture of Marlex CL-100 HDPE in liquid scintillation fluid.

3.4 Irradiation-Creep Behavior

A new series of uniaxial creep tests was started this quarter to determine the effects of in-test gamma irradiation on the creep rate and ductility of HDPE. Some earlier preliminary studies carried out at BNL were inconclusive since one indicated that the creep rate was increased by irradiation, whereas the other seemed to show the opposite (NUREG/CR-3898, 1984; NUREG/CR-4607, 1986). Very few tests were carried out and no definite conclusions could be drawn. Table 3.5 shows the basic test matrix for the new series of tests which cover the ranges for the earlier programs. The material used in this effort was from the "old batch" of HDPE used in the earlier work. Table 3.6 shows results to data from the irradiation-creep work. Some of the non-irradiated tests were conducted much earlier in this program and, therefore, the data cover a broader range of stress conditions than the four stresses listed in Table 3.5.

Figure 3.14 shows stress-rupture data for the in-test irradiated HDPE together with esults from unirradiated controls. At the higher stress levels the failure times are significantly larger for irradiated samples. At lower stress levels, gamma irradiation appears to become detrimental, and the failure time begins to decrease below that for unirradiated controls.

Table 3.5 Irradiation-creep test matrix for HDPE

Test Medium	Stress		γ Flux (rad/h)
	(MPa)	(psi)	
Air	12.58	1825	0
Air	12.58	1825	5×10^3
Air	12.58	1825	3×10^4
Air	11.72	1700	0
Air	11.72	1700	5×10^3
Air	11.72	1700	3×10^4
Air	11.07	1600	0
Air	11.07	1600	5×10^3
Air	11.07	1600	3×10^4
Air	10.34	1500	0
Air	10.34	1500	5×10^3
Air	10.34	1500	3×10^4

Table 3.6 Irradiation-creep data for Marlex CL-100 HDPE

Test	Material Cond.	Dose Rate(rad/h)	Stress (psi)	Stress (MPa)	Failure time(h)	Elong. (%)
405	As fab.	5×10^3	1825	12.58	27.5	206.1
403	As fab.	5×10^3	1700	11.72	209.7	326.2
406	As fab.	5×10^3	1600	11.03	119.8	8[illegible]7
404	As fab.	5×10^3	1500	10.34	Ongoing	-
384	As fab.	0	2000	13.79	1.8	67.6
368	As fab.	0	1900	13.10	5.3	46.6
375	As fab.	0	1850	12.76	18.6	(3)
351	As fab.	0	1825	12.58	5.8	82
344	As fab.	0	1700	11.72	47	127.6
343	As fab.	0	1600	11.03	127	92.0
342	As fab.	0	1500	10.34	2544	96.3
317	As fab.	0	1500	10.34	7514	93.4
31[illegible]	As fab.	0	1400	9.65	>15300	-
386	As fab.	0	1200	8.27	>6300	-

Examination of the creep curves measured for various stress levels show in detail the effects of irradiation (Figures 3.15 through 3.18). At lower stress levels 10.34 and 11.03 MPa (1500 and 1600 psi) the irradiated specimens show a far lower creep rate (Figures 3.15 and 3.16). For the three failed specimens, the ductility values fell between about 80-95 percent. On the other hand, at stresses of 11.72 and 12.58 MPa (1700 and 1825 psi) the irradiation-creep curves show a final very fast rate of creep, associated with high creep strain, before failure occurs. Examination of the specimens while they are creeping shows that this stage of creep is converted with "necking" of the HDPE (local thinning of material in the gagelength). Usually, necking causes intense local plastic deformation leading t failure. In the case of these irradiation-creep specimens, material in the necked regions becomes

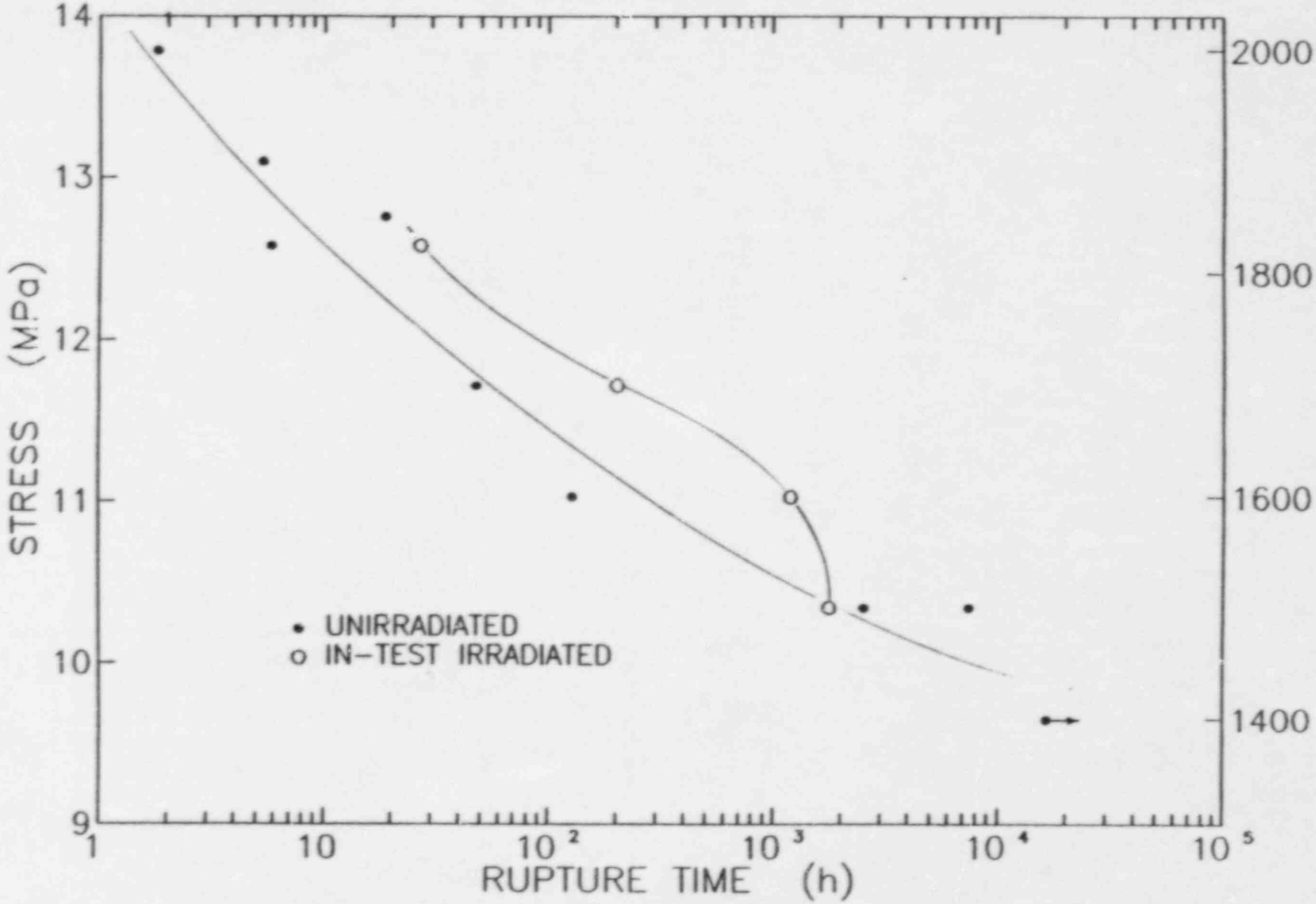

Figure 3.14 Effect of in-test gamma irradiation at 5×10^3 rad/h on the stress-rupture behavior of Marlex CL-100 HDPE.

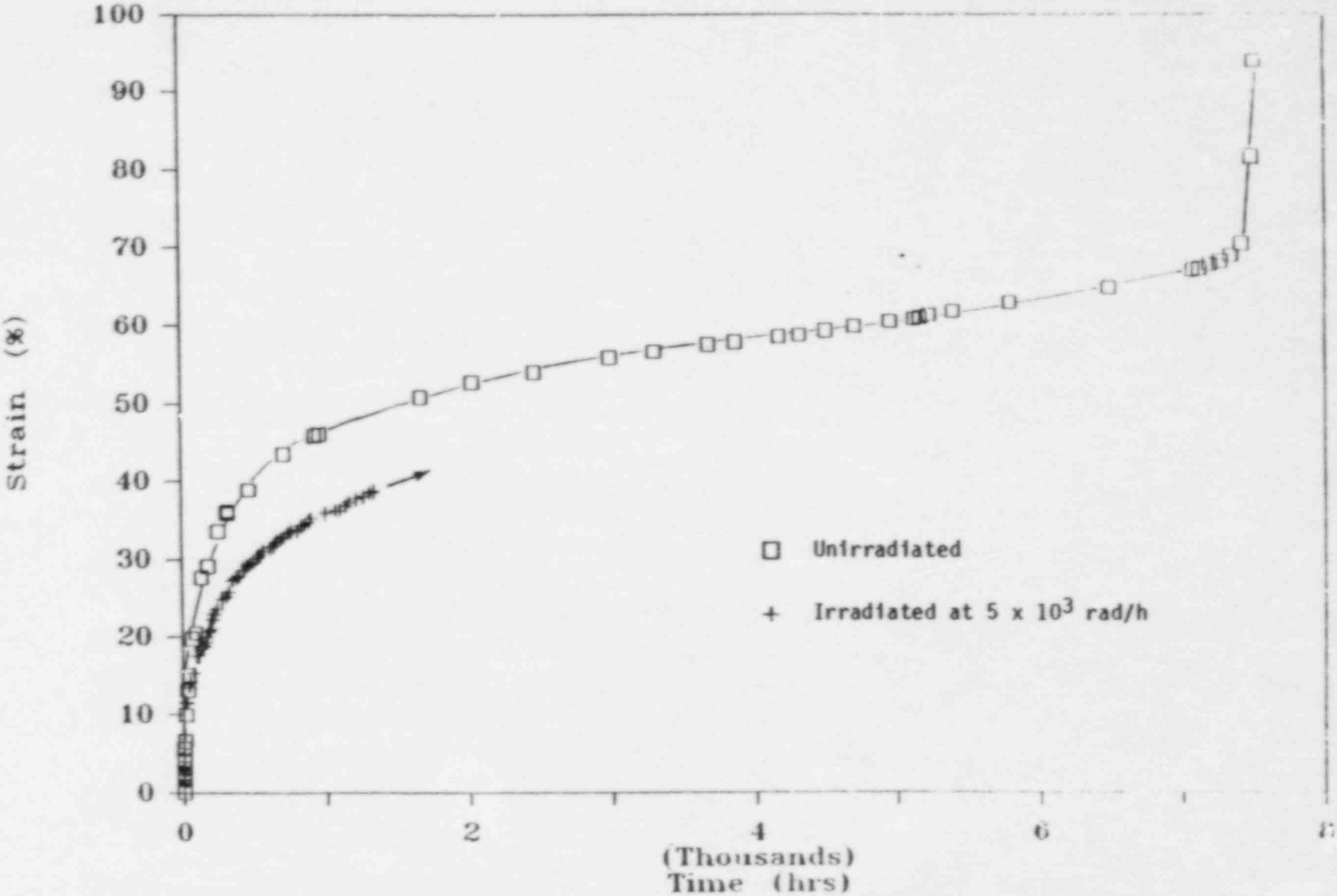

Figure 3.15 Effect of in-test gamma irradiation on the creep of Marlex CL-100 HDPE at a stress of 10.34 MPa (1500 psi).

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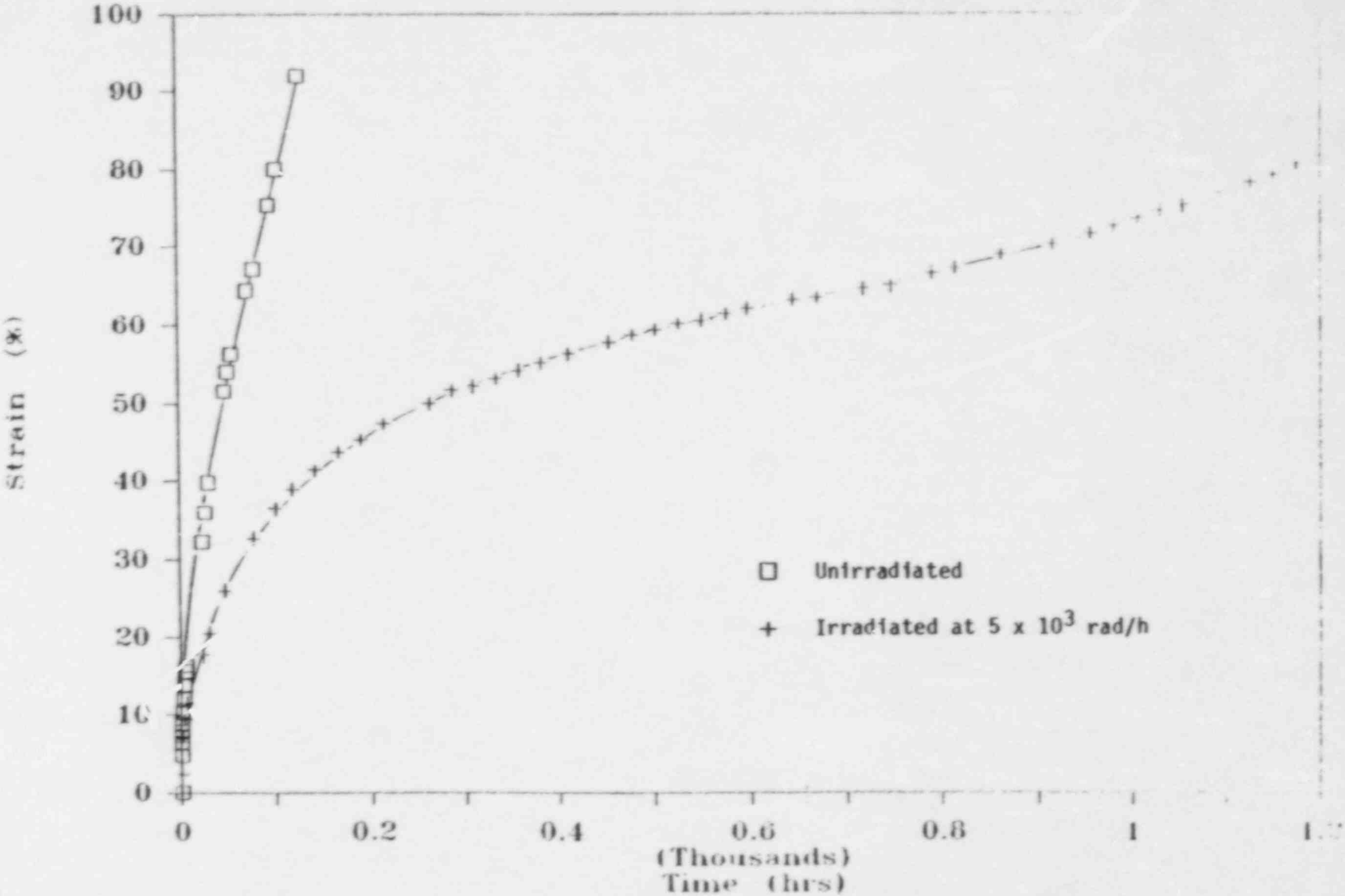

Figure 3.16 Effect of in-test gamma 'rradiation on the creep of Marlex CL-100 HDPE at a stress of 11.03 MPa (1600 psi).

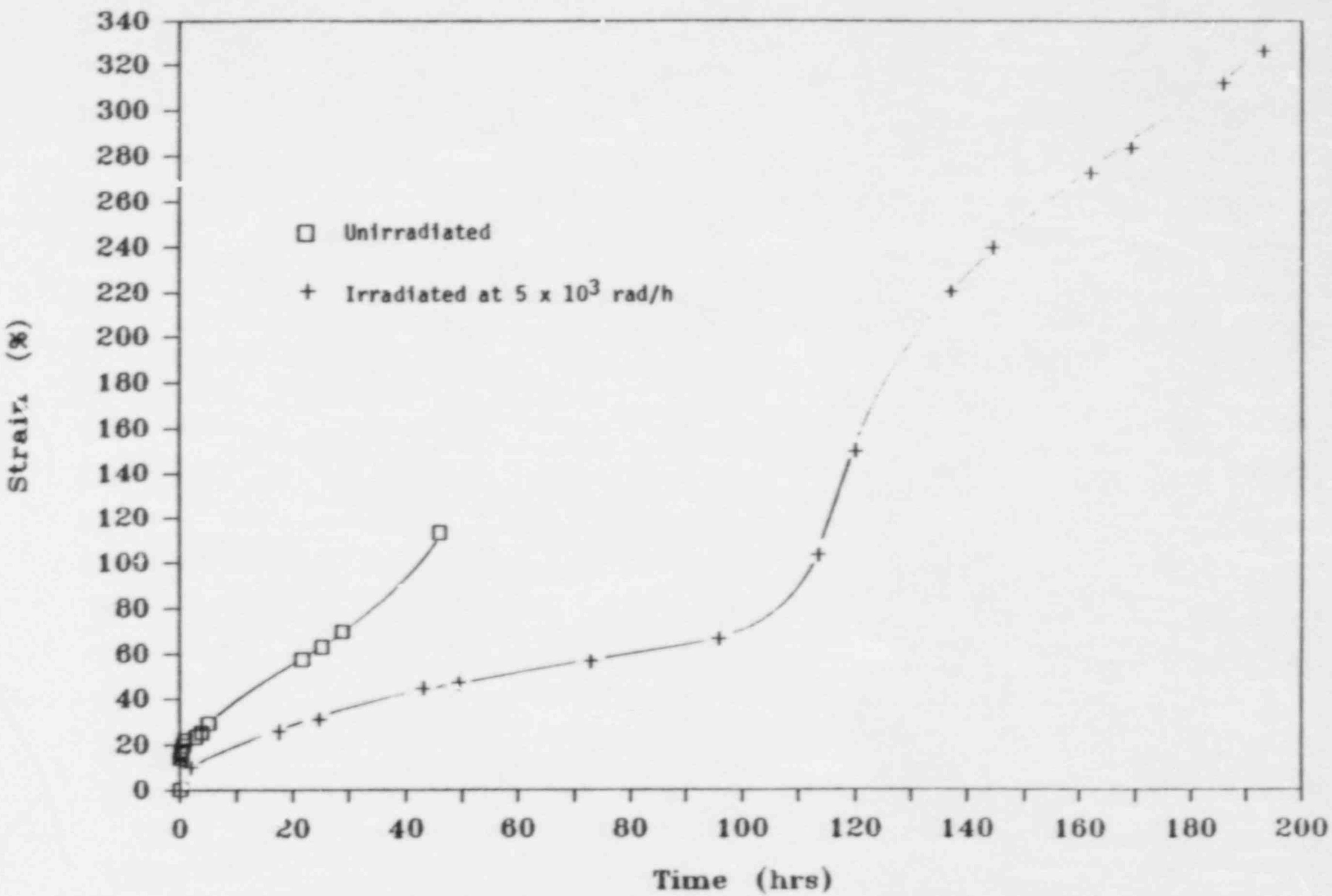

Figure 3.17 Effect of in-test gamma irradiation on the creep of Marlex CL-100 HDPE at a stress of 11.72 MPa (1700 psi).

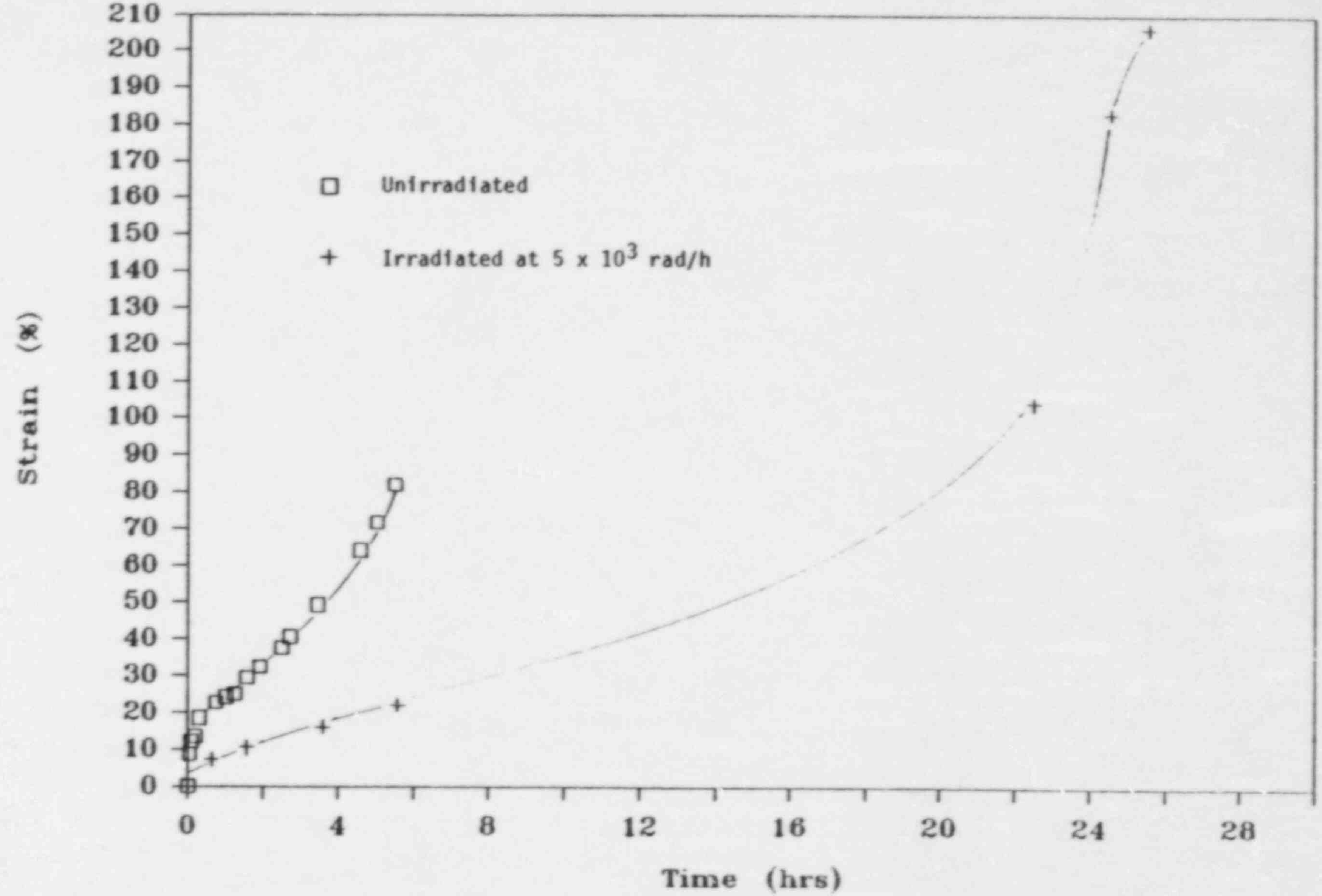

Figure 3.18 Effect of in-test gamma irradiation on the creep of Marlex CL-100 HDPE at a stress of 12.58 MPa (1825 psi).

hardened and the rate of additional plastic deformation decreases. New deformation is forced to spread to areas adjacent to the neck, giving an extended necked region. Over a period of time elongations of several hundreds of percent may be achieved.

Clearly, in-test gamma irradiation at a level of 5 x 10^3 rad/h is beneficial to the creep of HDPE. It decreases the creep rate and, at the higher test stresses used in this program, essentially triples the rupture ductility compared to unirradiated controls.

In the next reporting period, new irradiation-creep tests will be initiated for the higher dose rate of about 3 x 10^4 rad/h specified in Table 3.5.

3.5 Testing Protocol for HDPE

To determine failure modes and failure behavior of HDPE, a series of tests will be required. Several applicable tests have been formalized by ASTM and are in general use. New or modified procedures (such as the BNL U-bend test) may be used to study special aspects of plastics behavior.

Table 3.7 is a list of testing protocols for identifying and quantifying mechanical failure/degradation modes in HDPE. Some of them have been used in the current program and others are of obvious benefit in characterizing the mechanical behavior of HDPE. A more detailed description of their usage will be given in the next report for this effort.

Table 3.7 Available tests for evaluating failure/degradation modes in HDPE

Failure/Degradation Mode	Test Methods	Scope of Test Method
Environmental Stress-Cracking	ASTM D 1693	General scoping test to determine susceptibility of material to cracking under the action of a local multiaxial stress and a surface-active liquid (Igepal CO-630).
	ASTM D 2552	More quantitative test than D 1693 to determine failure time of material under a given stress and a surface-active agent (Igepal CO-630)
	BNL U-bend test	General scoping test, similar to ASTM D 1693, but designed to quantify crack initiation and propagation in surface oxidized material under a static tensile stress. Various test environments may be used.
Irradiation Embrittlement	BNL U-bend test	General scoping test to determine crack initiation and propagation in material under a static tensile stress.
	ASTM D 638	Standard tensile test to measure tensile strength (at yield or break), elongation (at yield or break), and the modulus of elasticity. Comparison of properties for non-irradiated and pre-irradiated material will quantify degree of embrittlement
	ASTM D 2990	Standard tensile, compressive, and flexural creep test to quantify creep rates and ductilities. Effects of irradiation may be quantified by comparing properties of non-irradiated and pre-irradiated material or, more preferably, comparing properties of in-test irradiated material and corresponding non-irradiated methods.

Table 3.7 Available tests for evaluating failure/degradation modes in HDPE (Continued)

Failure/Degradation Mode	Test Methods	Scope of Test Method
Ductile Failure	ASTM D 638	Standard tensile test to measure strength and ductility. May be used to test in various environments.
	ASTM D 790	Standard test to measure flexural (bending) properties of bar specimens. Test continued until fracture or until 5% [illegible]ximum fiber strain is reached.
	ASTM D 2990	Standard test for creep under tensile, compressive, or flexural conditions. May be used to test in various environments.
Low-Stress Creep Embrittlement	ASTM D 2990	Standard test that can be used to evaluate low-ductility creep failure under tensile and flexural conditions for various environments. Depending on test conditions, very long-term testing may be required. Higher temperatures may possibly be used to accelerate time for failure, so that failure times and ductilities may be extrapolated to service conditions.
	ASTM D 2991	Standard test for stress relaxation at constant strain level. This test is useful to evaluate crack initiation/propagation under low stress conditions. In particular, it will show how temperature, environment, and irradiation influence the rate of change in residual stresses in a plastic.
Impact Embrittlement	ASTM D 3029	Standard test to determine energy to fracture plastic by high speed falling weight. It is a valuable procedure when

Table 3.7 Available tests for evaluating failure/degradation modes in HDPE (Continued)

Failure/Degradation Mode	Test Methods	Scope of Test Method
		used to measure low-temperature impact energy since it is a good indication of the degree of crosslinking.
Liquid Absorpsion	ASTM D 570	Test specifically developed to measure amount of water absorbed by plastic in a given time at a given temperature. It should be very useful for a range of liquids pertinent to HDPE usage.

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