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An Evaluation of Three Partially Volatile Neutron Shields for High-Performance Shipping Casks

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AN EVALUATION OF THREE PARTIALLY VOLATILE NEUTRON
SHIELDS FOR HIGH-PERFORMANCE SHIPPING CASKS*

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ABSTRACT

The thermal stability and mechanical behavior of three partially volatile candidate neutron shield materials have been evaluated. The results indicate that silicone based rubbers, impregnated with elemental boron or boron carbide, Boro-silicone 236 and Bisco NS-I respectively are more thermally stable than are borated beechwoods, e.g., Permalin JN. Mechanical property measurements indicated however that the compressive strength of the borated beechwood is 10 to 48 times higher than that of the silicone-based rubbers. The compressive strengths of the borated beechwood and boron carbide impregnated silicone rubber were substantially more sensitive to test temperature than was the compressive strength of the boron impregnated silicone rubber. Finally the compressive strengths and energy absorbing capability of the boron impregnated silicone rubber is not affected by prior thermal exposure at 425K for 1000h.

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CONTENTS

	<u>Page</u>
INTRODUCTION	7
EXPERIMENTAL PROCEDURES	9
Materials	9
Thermal Stability	9
Mechanical Properties	9
EXPERIMENTAL RESULTS AND DISCUSSION	15
Thermal Stability	15
Mechanical Behavior	23
SUMMARY AND CONCLUSIONS	37

ILLUSTRATIONS

<u>Figure</u>		<u>Page</u>
1	Tensile Configuration for Permali JN.	11
2	Sample Orientation for Permali JN.	13
3	Thermo-gravometric analysis of candidate neutron shield materials. Heating rate: 10K/min.	16
4	Isothermal weight loss for (a) Boro-silicone 236, (b) NS-I and (c) Permali JN. Temperatures as indicated.	20
5	Comparison of isothermal weight loss measurements for Boro-silicone 236, NS-I and Permali JN at (a) 375K, (b) 425K and (c) 475K.	24
6	Tensile Failure in Permali JN.	30
7	Compressive Failure in Permali JN., LT Orientation.	31
8	Influence of Test Temperature on the Compressive Properties of Permali JN. LT Orientation.	33
9	Influence of Test Temperature on the Compressive Strength of Boro-Silicone 236 and Bisco NS-I.	34
10	Influence of Test Temperature on Absorbed Energy of Boro-Silicone 236 and Bisco NS-I.	35

TABLES

<u>Table</u>		<u>Page</u>
1	Chemical Analysis of Partially Volatile Candidate Neutron Shield Materials Prior to Thermal Exposure	10
2	Temperature for Achieving 5 Percent Weight Loss in Constant Heating Rate (10K/min) Evaluation of Candidate Neutron Shield Materials	19
3	Temperature for Achieving 5 Percent Weight Loss From TGA and Isothermal Measurements	27
4	Chemical Analysis of Partially Volatile Candidate Neutron Shield Materials Following Thermal Exposure	28
5	Room Temperature Mechanical Properties of Permali JN	29

INTRODUCTION

Many spent-fuel shipping casks in use throughout the world utilize water or ethylene glycol/water mixtures as the neutron shielding material. In either case the shield material is usually contained in a cavity near the outer surface of the cask. As such, the probability of retaining the liquid neutron shield in a severe accident is low. For such casks, the designer usually is required by his certificating authority to assume in his safety analysis that the shield material is lost during an accident. The radiation level external to the package following an accident must not exceed that which is allowed by regulation. In the IAEA Safety Series No. 6, paragraphs 229 and 532, an increase in external dose from 10 mrem/h, 2 m from the external surface of the package to 1000 mrem/h, 1 m from the surface of the package is allowed following the "tests for demonstrating ability to withstand accident conditions in transport."

At least three problems associated with the use of volatile liquids as neutron shielding materials in shipping casks have been identified. First, for casks such as might be needed for the shipment of high-burnup, short-cooled spent fuel from liquid metal fast breeder reactors (LMFBR), the external dose rate following exposure to the accident-related tests may exceed that allowed by the regulations if all of the neutron shielding material is lost. Specifically, this occurs if the design of the cask has been optimized about a balanced shielding of both gamma rays and neutrons. Complete loss of the neutron shield following an accident requires that the designer add extra gamma shielding. This results in significant increases in cask weight and overall dimensions. Second, the use of volatile neutron shields may often complicate the packaging design and operation, since

expansion volumes and pressure relief systems must be provided to prevent over-pressurization during normal use and exposure to fire environments. Third, irrespective of the material being carried, the loss of all the neutron shielding material in an accident results in a more difficult and potentially more hazardous accident recovery procedure.

Recently a study was initiated to evaluate the feasibility of fabricating neutron shields for spent fuel shipping casks using commercially available materials which would retain at least part of their neutron shielding capability following exposure to a 1075K, 30-minute thermal test. This concept utilizes interstitially located layers of a neutron shielding material and metallic heat transfer fins. Various materials which could either be preformed to specific shapes and inserted into the neutron shield cavities or could be "injected" into the cavities and solidified in-situ were examined. These preliminary evaluations resulted in the selection of three candidates: (a) a boron loaded silicone rubber, Boro-silicone rubber 236, a product of Reactor Experiments, Inc.; (b) a boron carbide loaded silicone rubber Bisco NS-I, a product of Brand Industrial Services Inc., and (c) a boron oxide impregnated cross-laminated beechwood, Permali JN, a product of the Permali Industrial Group. This report describes studies performed to assess the thermal stability and mechanical behavior of these three candidate materials.

EXPERIMENTAL PROCEDURES

Materials

The three candidate neutron shield materials examined were received in the form of 5.1 cm thick x 51 cm square blocks. Table 1 lists the H, C and B contents of the as-received material. The Boro-silicone 236 contained 1.6 wt. pct. B in the form of elemental boron, the Bisco NS-I contained 16.1 wt. pct. B in the form of B_4C , and the Permali JN contained 2.1 wt. pct. in the form of B_2O_3 . In addition the Boro-silicone 236 contained approximately 16 wt. pct. Al apparently as an aluminum oxide filler.

Thermal Stability

The thermal stability of the Boro-silicone 236, Bisco NS-I and Permali JN was initially evaluated using thermo-gravometric analysis (TGA), with more detailed long time stability being established using standard isothermal weight change (loss) procedures. The former (TGA) procedure involved a measurement of the percent weight loss which occurred during isochronel (constant rate) heating of the test sample. Atmosphere control during heating also made it possible to consider the possible influence of an environment, e.g., air versus argon, on the material's thermal stability.

Results of the TGA measurements were used to establish the temperatures for the isothermal weight loss measurements. Duplicate weight change samples 1.27 cm sq. x 5.1 cm high, having a surface to volume ration of 0.177, were then exposed at selected temperatures in an air environment for times up to 2024 h.

Mechanical Properties

Triplicate tensile samples, as shown in Fig. 1, were used to evaluate the tensile properties of Permali JN. These samples contained 10-11 plies

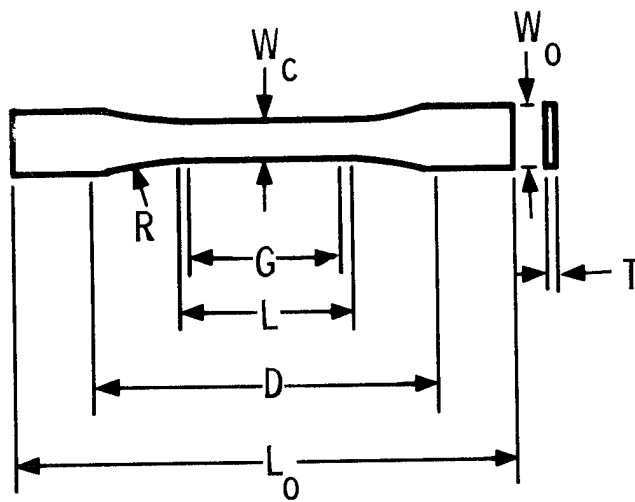
Table 1

Chemical Analysis of Partially Volatile Candidate
Neutron Shield Materials Prior to Thermal Exposure

<u>Candidate Material</u>	<u>Weight Percent</u>		
	<u>H</u>	<u>C</u>	<u>B^(a)</u>
Boro-silicone 236 ^(b)	4.5	11.4	1.6
Bisco NS-I	3.3	18.1	16.1
Permali JN	5.6	43.0	2.1

(a) Boron present as elemental boron, B_4C and B_2O_3 for Boro-silicone 236, Bisco NS-I and Permali JN, respectively.

(b) Also contained approximately 16 wt. pct. Al as aluminum oxide.



DIMENSIONS (mm):

W_c	19	D	114.3
W_0	28.7	L_0	246.4
G	50	R	76.2
L	57.2	T	12.7

Fig. 1. Tensile Configuration for Permalloy JN

and were removed from both the LT and TL ply orientations, the latter as defined in Fig. 2. Measurements of the compressive properties of Permali JN, as well as Boro-silicone 236 and Bisco NS-I utilized samples having a length to diameter/side ratio of 2 with the length being 2.54 cm. The compressive properties of the three candidate neutron shield materials were determined at a strain rate of 10^{-4} sec^{-1} over the temperature range 233 to 473K, with all specimens having been soaked at temperature for at least 0.5h prior to testing.

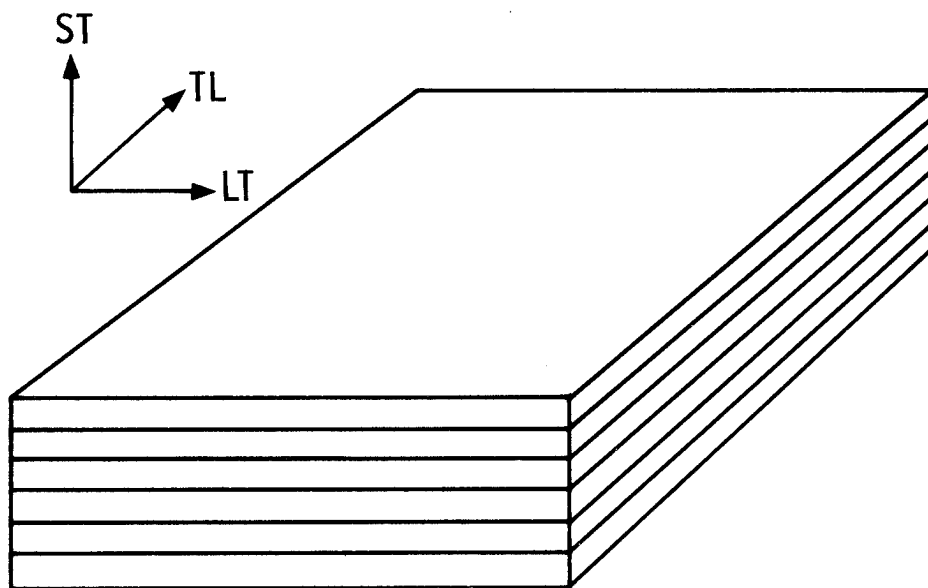


Fig. 2. Sample Orientation for Permali JN

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EXPERIMENTAL RESULTS AND DISCUSSION

Thermal Stability

Fig. 3 presents the results of the thermo-gravometric analysis of the three candidate materials. These results indicate that the silicone based materials, Boro-silicone 236 and Bisco NS-I, are stable to $\sim 475\text{K}$, with a gradual weight loss occurring above this temperature. In contrast Permali JN is unstable at all temperatures above ambient, with significant instability beginning at approximately 475K . This latter degradation is so severe that at temperatures exceeding 975K essentially all of the material has been volatilized.

Another measure of the relative thermal stability of the candidate materials can be obtained using the thermo-gravometric analysis results by establishing the temperature to which the materials must be heated at a constant heating rate to achieve a pre-selected weight loss. Table 2 presents such a comparison for a 5 percent weight loss. This table shows that Bisco NS-I is the most stable material under both inert (argon) and oxidizing (air) environments. It should be noted, however, that the thermal stability of Bisco NS-I appears to be more sensitive to cover-gas environment than is Boro-silicone 236. The temperature for attaining a 5 percent weight loss in Boro-silicone 236 is essentially independent of the cover-gas, while that for Bisco NS-I decreases by $\sim 50\text{K}$ by changing from an inert (argon) atmosphere to an oxidizing (air) environment.

The final method used to assess the long term thermal stability of the candidate neutron shield materials involved isothermal weight-loss measurements in an oxidizing (air) environment. These results, Fig. 4, show that, contrary to the thermo-gravometric analysis observations, long term exposure of Boro-silicone

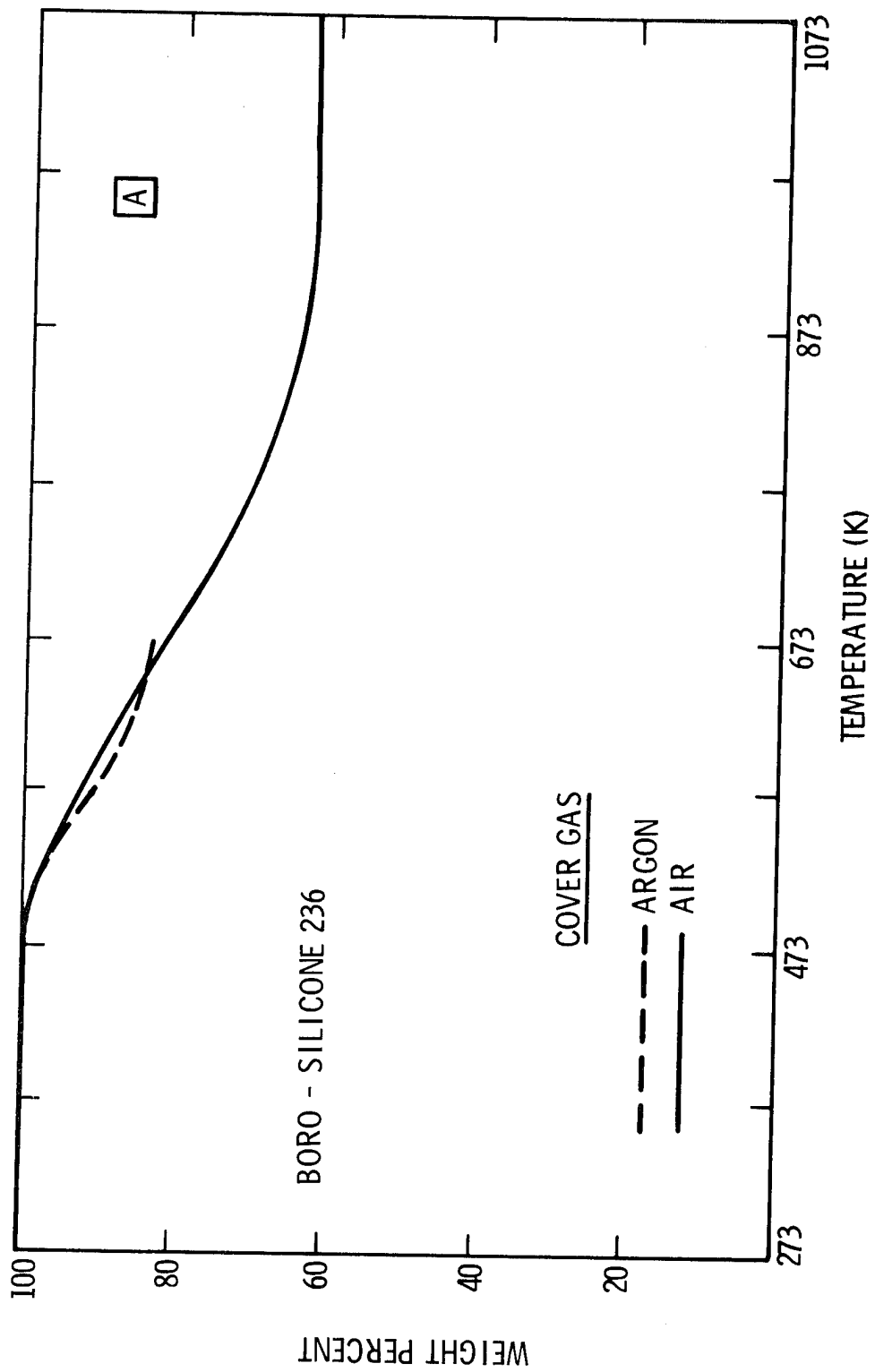
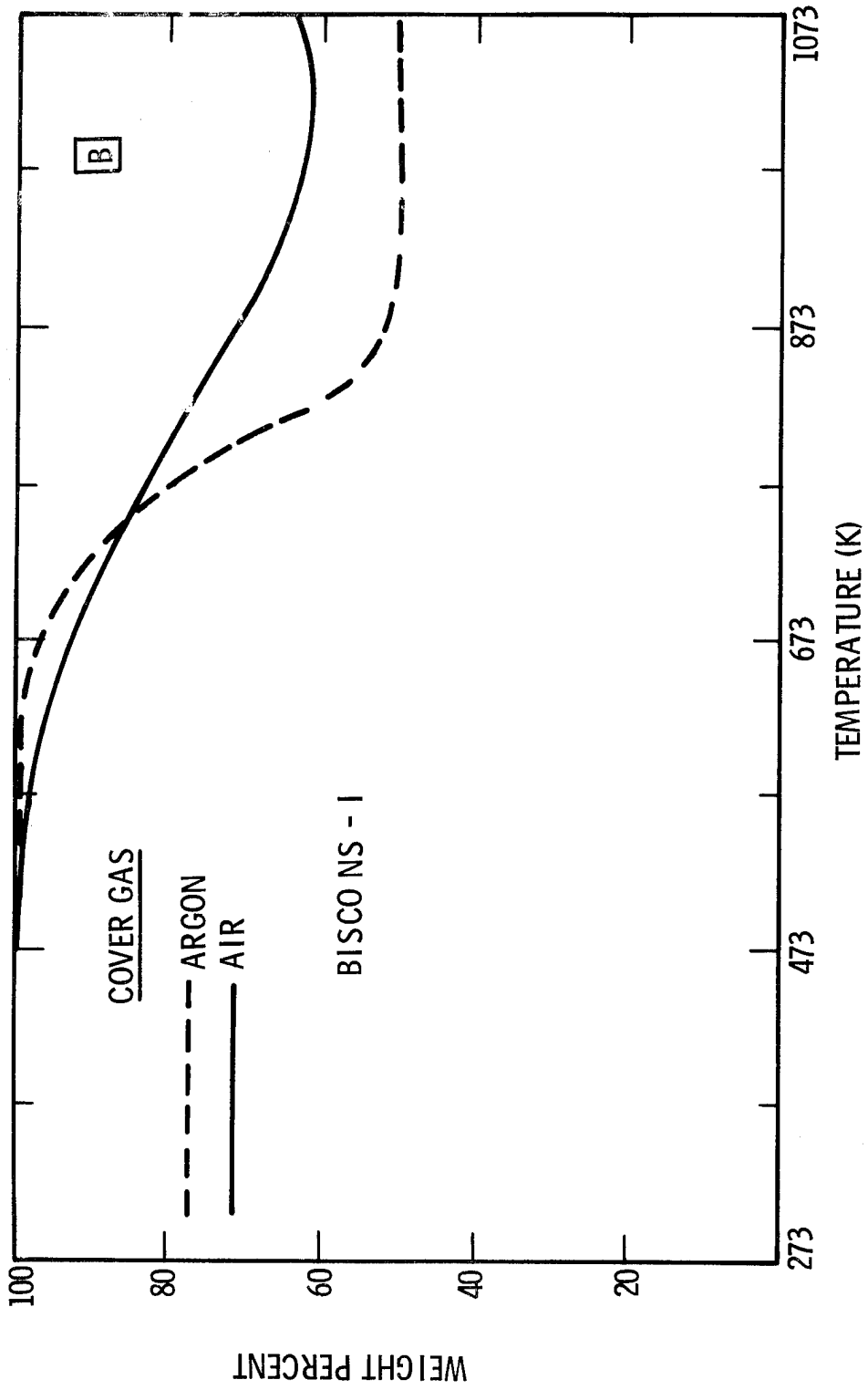


Fig. 3. Thermo-gravometric analysis of candidate neutron shield materials.
Heating rate: 10K/min.



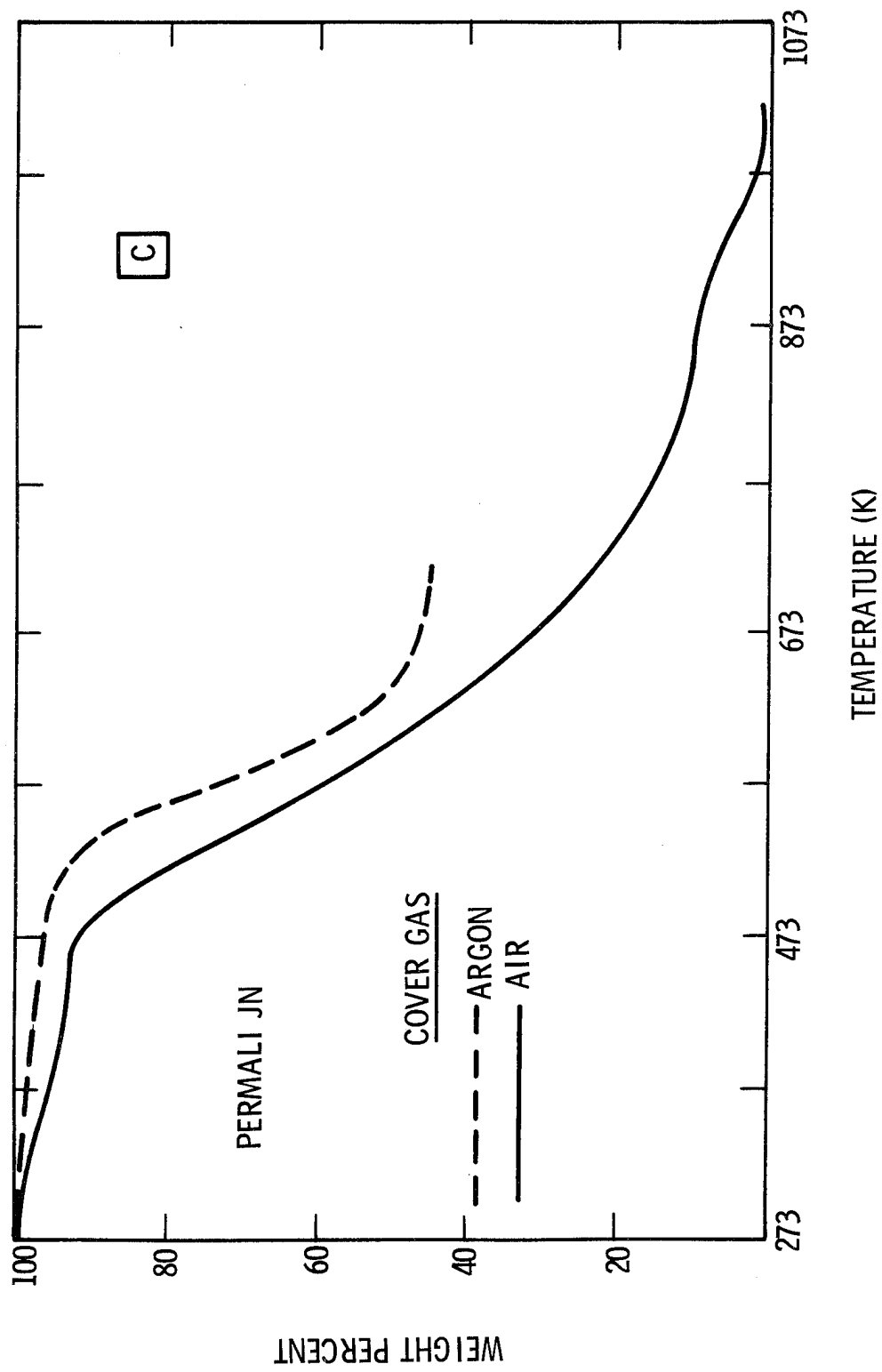


Table 2

Temperature for Achieving 5 Percent Weight Loss in Constant Heating Rate
(10K/min) Evaluation of Candidate Neutron Shield Materials

<u>Candidate Material</u>	<u>Air Atmosphere Temperature (K)</u>	<u>Argon Atmosphere Temperature (K)</u>
Boro-silicone 236	552	547
Bisco NS-I	641	694
Permali JN	388	503

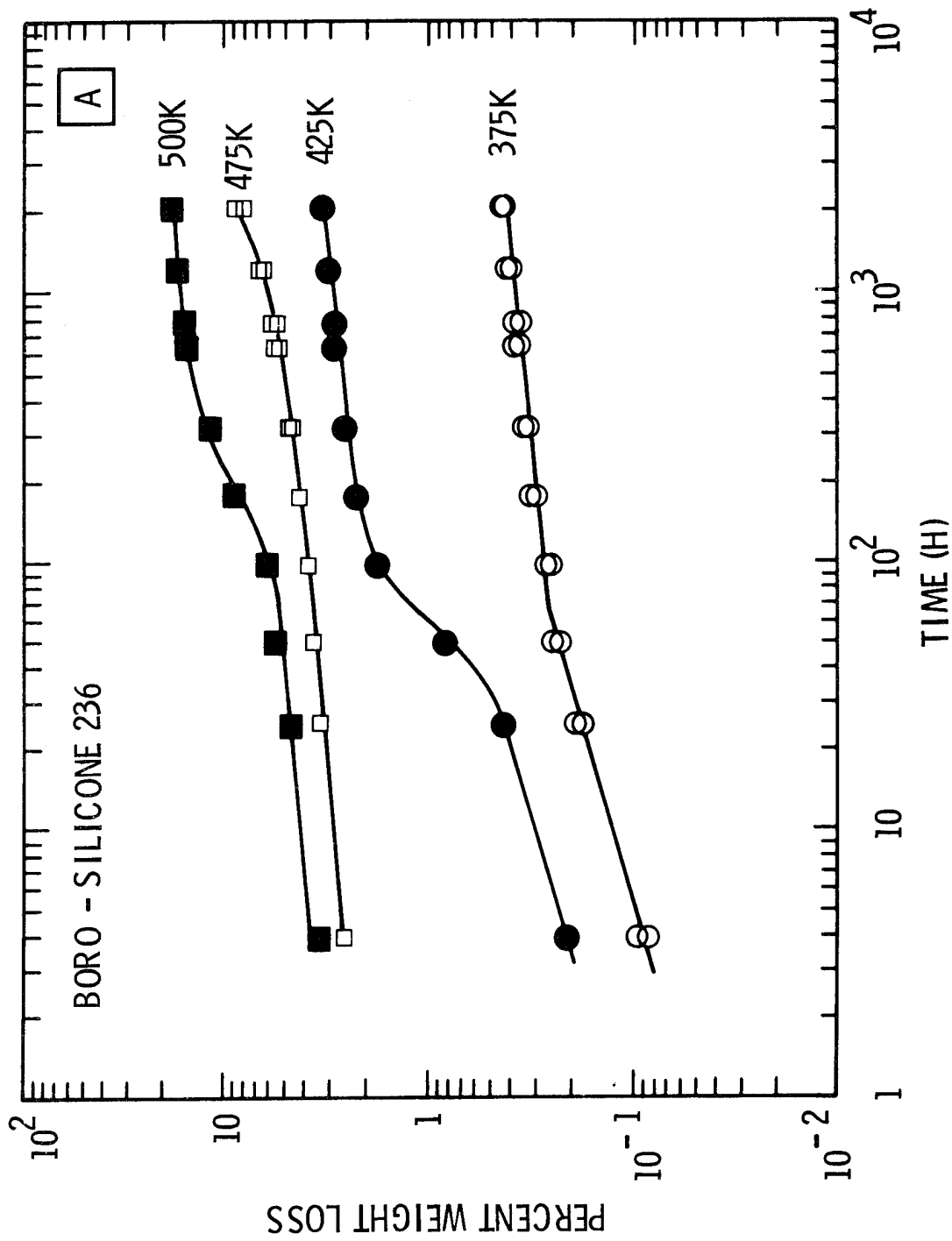
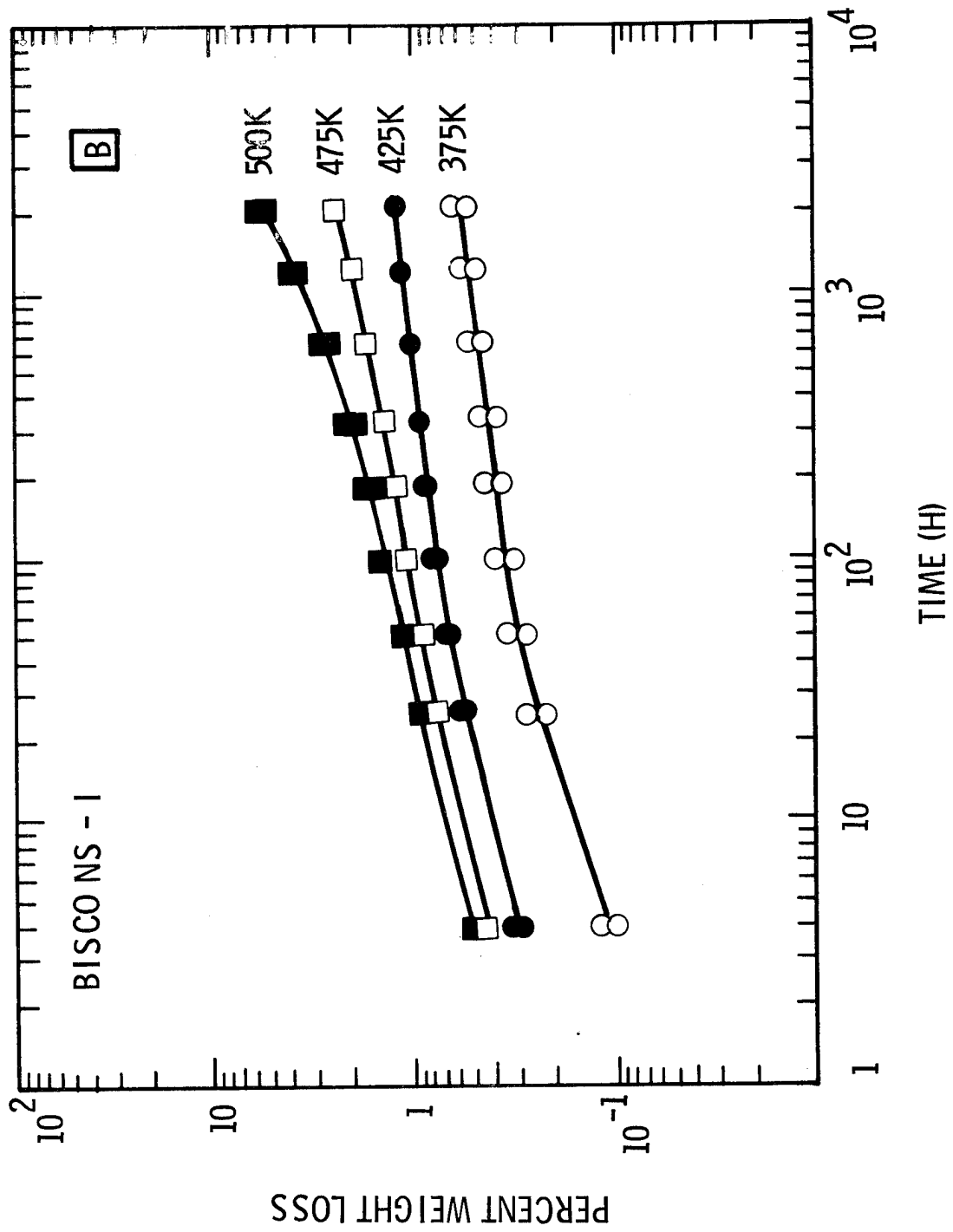
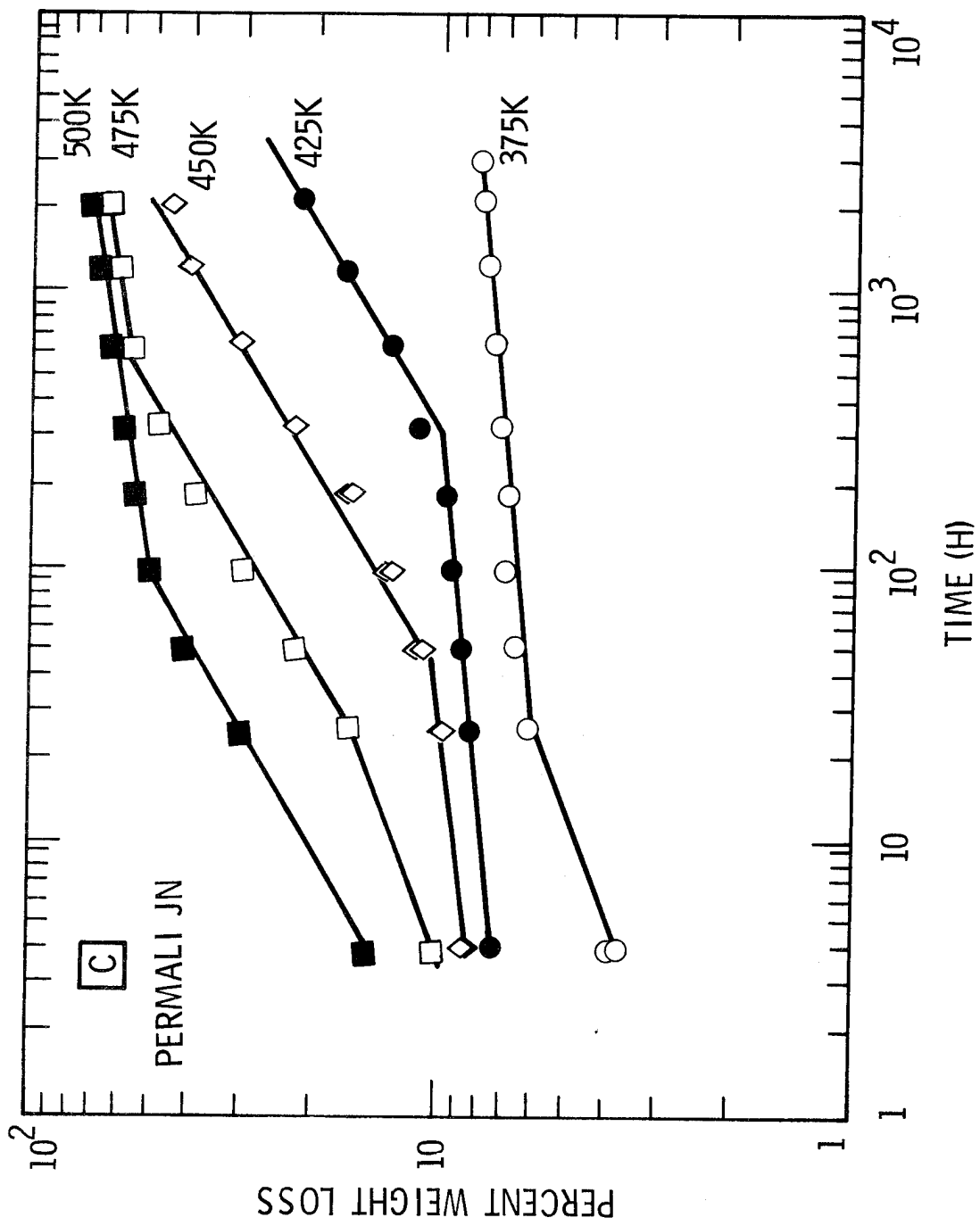


Fig. 4. Isothermal weight loss for (a) Boro-silicone 236, (b) NS-I and (c) Permalin JN. Temperatures as indicated.





236 and Bisco NS-I below 475K can lead to degradation, although the percentage weight losses observed were quite low, typically less than 2 percent after 2024 h exposures. Fig. 5 presents a direct comparison, at constant temperature, of the three candidate neutron shield materials. In each case the silicone based materials were again more stable than the Permali JN. These results also indicate that inclusion of time dependent aging effects leads to a decrease in the maximum continuous operating temperature for the three candidate materials relative to that which would have been obtained by only considering the thermal-gravometric analysis results. Table 3 presents such a comparison for the three materials at the 5 percent weight loss level. The isothermal observations as shown in Table 3 also tend to reduce the differences between the two silicone based materials. Finally, chemical analysis, Table 4, indicated that the weight losses observed after isothermal exposure in an air environment appeared to be related to a reduction in the hydrogen content of the three candidate neutron shield materials.

Mechanical Behavior

The results of the room temperature tensile and compressive measurements of Permali JN are summarized in Table 5. Tensile loading of this material results in an elastic failure, the final failure mode involving tearing of the beechwood cross-ply as shown in Fig. 6. This failure mode is in contrast with the shear type failures observed when Permali JN was compressively loaded in the LT direction, Fig. 7. Table 5 shows that the tensile and compressive properties of the Permali JN examined in this study were, contrary to manufacturing specification for a cross-ply laminate, functions of orientation. Indeed the observed orientations effects were quite large and would have to be considered in any neutron shield design.

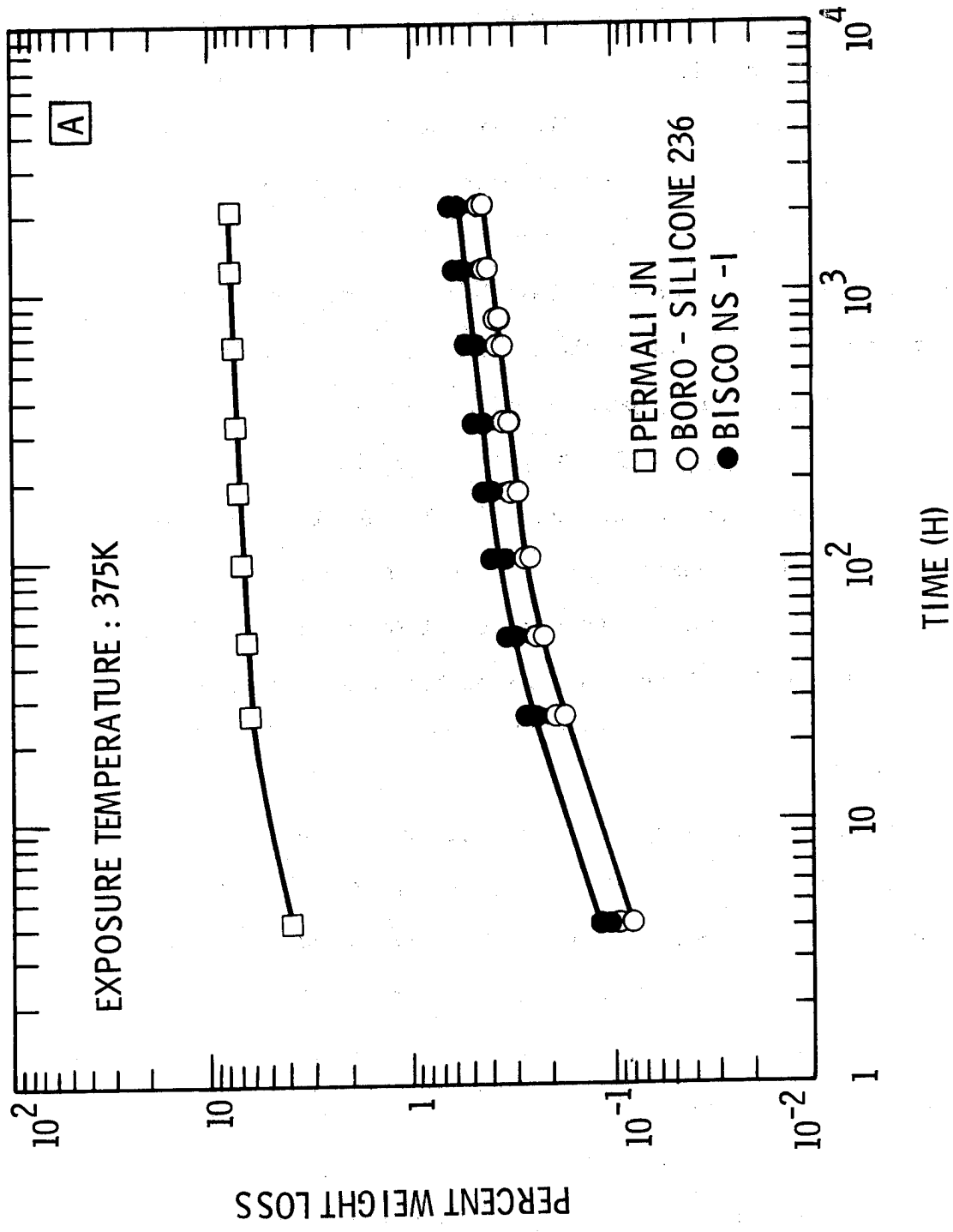
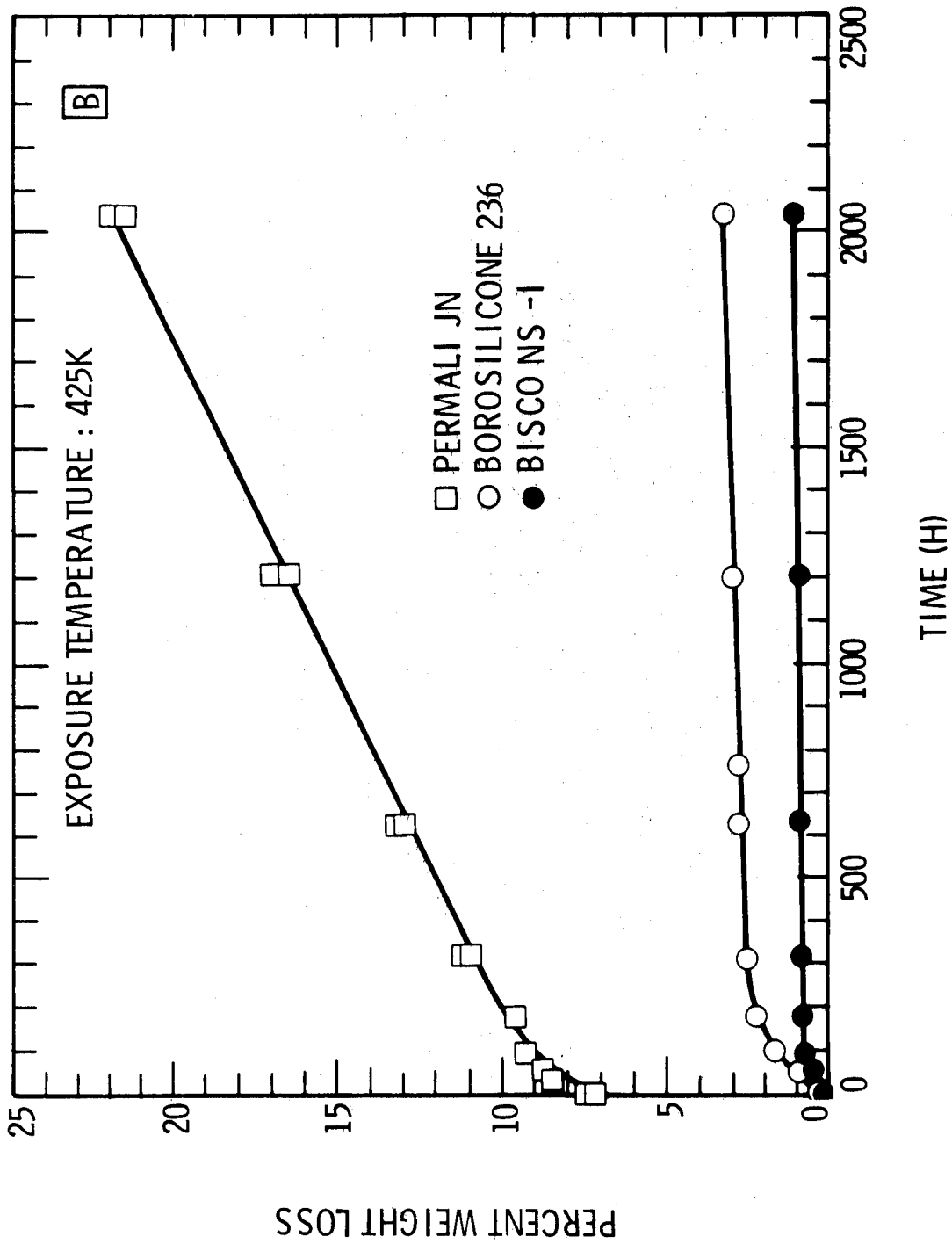
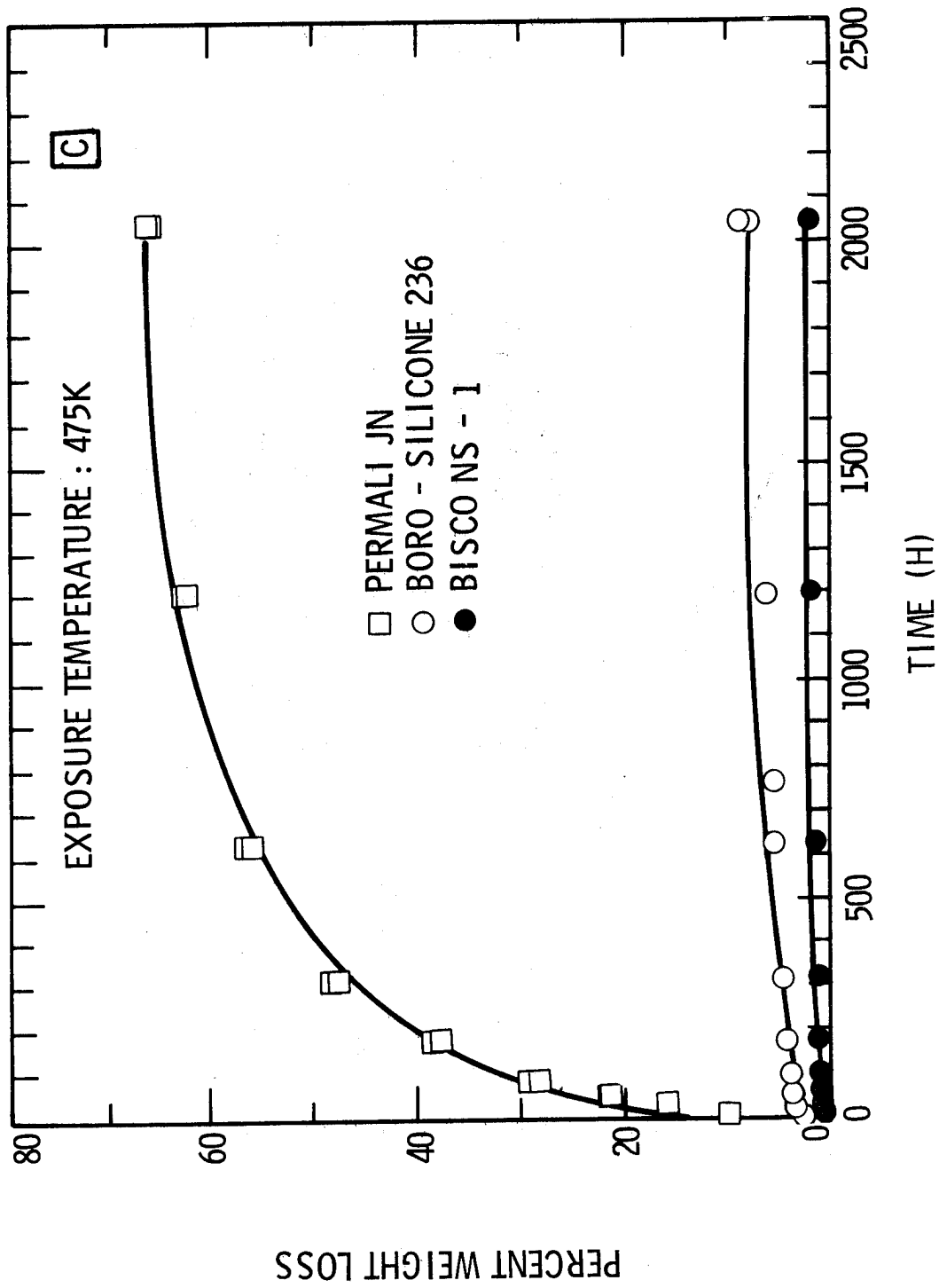


Fig. 5. Comparison of isothermal weight loss measurements for Boro-silicone 236, NS-I and Permal JN at (a) 375K, (b) 425K and (c) 475K.





C

Table 3

Temperature for Achieving 5 Percent Weight Loss
From TGA and Isothermal Measurements⁺

<u>Candidate Material</u>	<u>TGA Temperature (K)</u>	<u>Isothermal Temperature (K)</u>
Boro-silicone 236	552	~ 450
Bisco NS-I	641	~ 475
Permali JN	388	< 375

⁺ Air atmosphere

Table 4

Chemical Analysis of Partially Volatile Candidate Neutron Shield
Materials Following Thermal Exposure[†]

<u>Candidate Material</u>	<u>Exposure Temperature (K)</u>	<u>Weight Percent</u>		
		<u>H</u>	<u>C</u>	<u>B</u>
Boro-silicone 236	298	4.5	11.4	1.6
	375	4.5	11.2	1.9
	425	4.1	10.9	1.6
	475	4.1	11.5	1.3
	500	3.2	10.6	1.7
Bisco NS-I	298	3.3	18.1	16.1
	375	3.2	17.8	17.2
	425	3.1	18.2	16.8
	475	3.0	17.6	17.3
	500	3.0	17.7	17.7
Permalin JN	298	5.6	43.0	2.1
	375	5.5	44.2	2.1
	425	4.7	45.0	2.5
	450	3.6	44.6	3.5
	475	2.3	43.8	5.3
	500	1.8	42.4	6.2

[†]Exposed 2024 h at indicated temperatures

Table 5

Room Temperature Mechanical Properties of Permalloy JN^(a)

	<u>Orientation</u>	<u>$\sigma_{0.5}^{(b)}$ (MPa)</u>	<u>$\sigma_u^{(c)}$ (MPa)</u>	<u>Y^(d) (GPa)</u>
Tension	LT	--	86	16.1
	TL	--	49	12.1
Compression	LT	91	124	11.0
	ST	112	190	2.7

(a) Strain rate: 10^{-4} sec⁻¹, as-received condition.

(b) $\sigma_{0.5}$: flow stress at 0.005 strain

(c) σ_u : ultimate strength

(d) Y: Young's modulus



Tensile Axis

Fig. 6. Tensile Failure in Permalloy JN

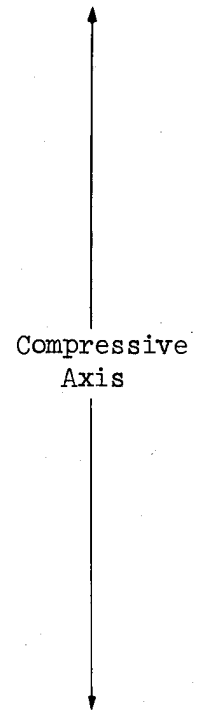


Fig. 7. Compressive Failure in Permalloy JN.
LT Orientation.

Further examination showed that the compressive properties of Permali JN were also quite sensitive to test temperature, Fig. 8. For example, decreasing the test temperature from 400K to 233K tripled the compressive strength of Permali JN. Finally, there appeared to be some increase in the compressive strength of Permali JN when the test temperature was increased from 400 to 450K.

Comparison of the results shown in Fig. 9 for Boro-silicone 236 and Bisco NS-I with those for Permali JN, Fig. 8, indicated that the compressive strengths of the silicone based rubbers were substantially below those of Permali JN. For example the room temperature compressive strength of Bisco NS-I was approximately 12 MPa and that of Boro-silicon 236, 2.5 MPa, factors of 10 and 48 below that of Permali JN. The compressive strength of Boro-silicone 236, as compared to both Permali JN and Bisco NS-I, was relatively insensitive to test temperature. Finally, Fig. 9 shows that prior thermal aging, in this case for 1000h at 425K, had little effect on the compressive behavior of Boro-silicone 236 and only effected that of Bisco NS-I at 233K.

One final comparison between the two silicone-based candidate neutron shield materials is presented in Fig. 10. Here the energy absorbed during compressive straining to a total strain of 15 percent has been plotted against test temperature. These results, which are typical of all strain offsets examined, indicate that (a) the energy absorption capability of these silicone rubbers was essentially independent of test temperature, (b) Boro-silicone 236 will absorb approximately twice the energy to a fixed strain than will Bisco NS-I, and (c) that aging, again for 1000h at 425K, does not effect the energy absorption capability of either Boro-silicone 236 or Bisco NS-I.

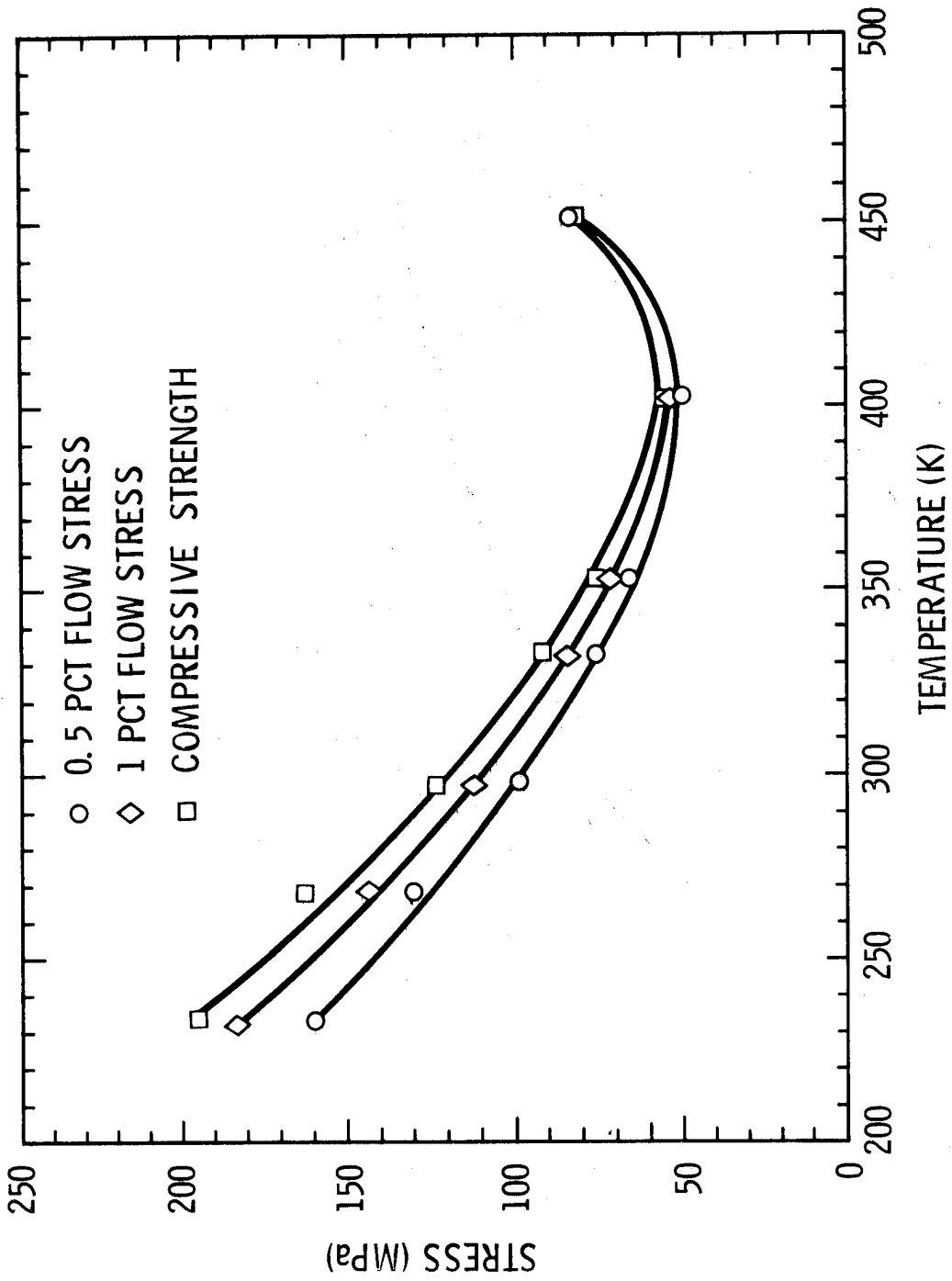


Fig. 8. Influence of Test Temperature on the Compressive Properties of Permalloy JN - LT Orientation

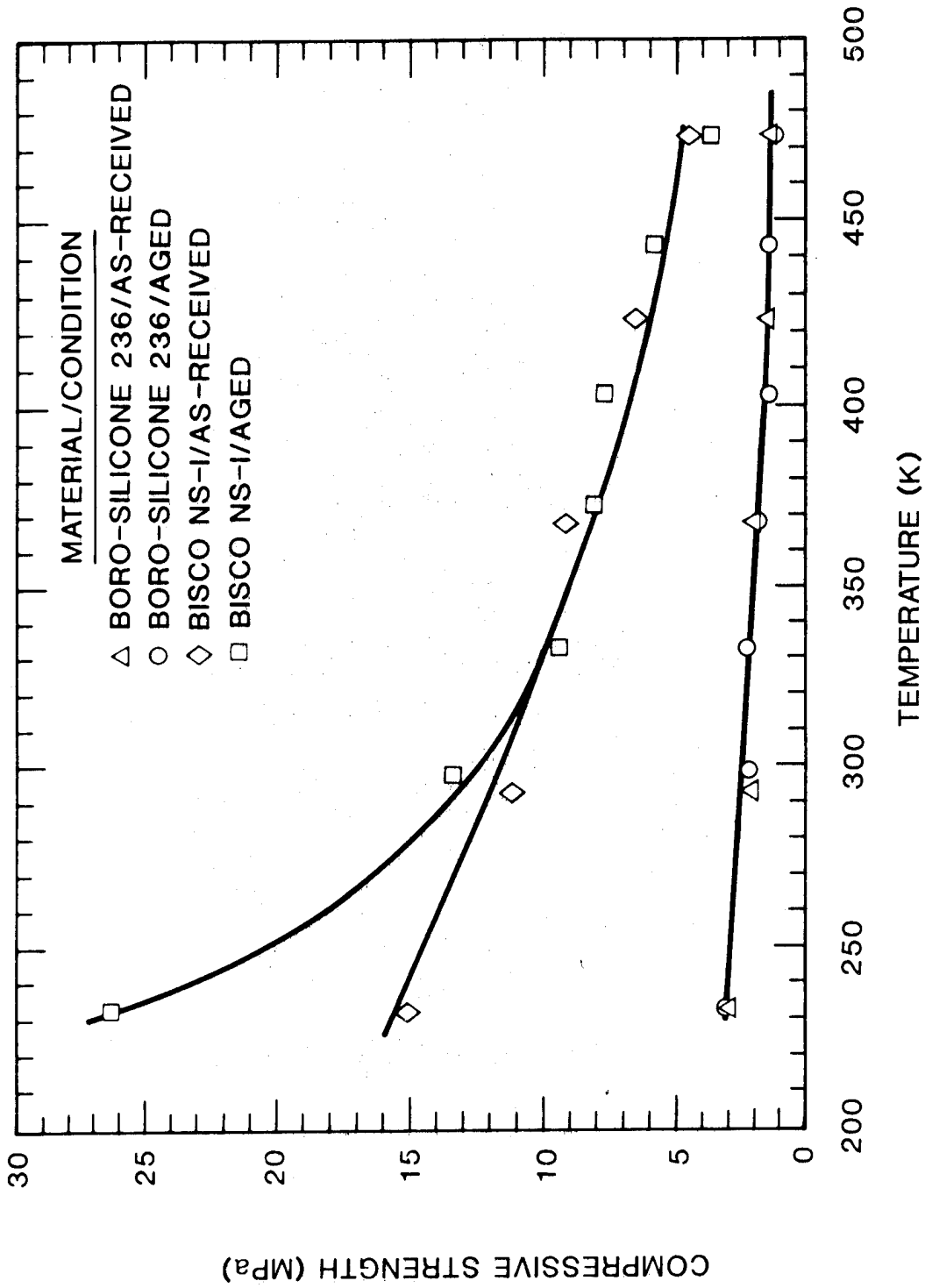


Fig. 9. Influence of Test Temperature on the Compressive Strength of Boro-Silicone 236 and Bisco NS-I.

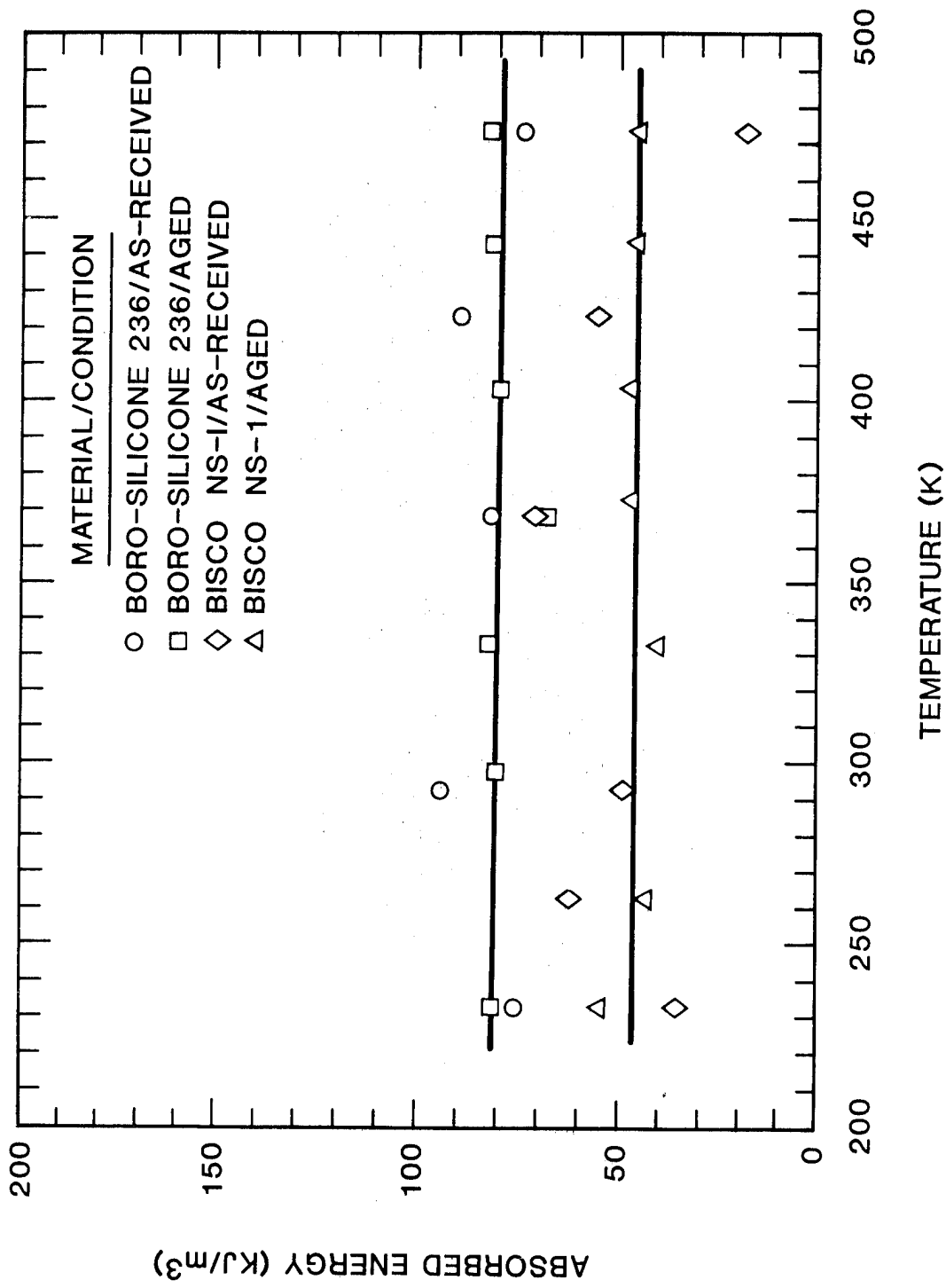


Fig. 10. Influence of Test Temperature on Absorbed Energy of Boro-Silicone 236 and Bisco NS-1.

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SUMMARY AND CONCLUSIONS

The present investigation of three partially volatile candidate neutron shield materials has shown that silicone-based rubbers, e.g., Boro-silicone 236 and Bisco NS-I, are more thermally stable than borated beechwood, Permali JN. Under oxidizing (air) conditions these materials have upper continuous operating temperatures of 450K (Boro-silicone 236), 475K (Bisco NS-I) and < 375K (Permali JN).

Mechanical property measurements have shown that the compressive strength of Permali JN is typically 10-48 times higher than either Boro-silicone 236 or Bisco NS-I. In addition, the compressive strengths of Permali JN and Bisco NS-I increase markedly with decreasing temperature, while that of Boro-silicone 236 is relatively insensitive to temperature. Finally it has been shown that Boro-silicone 236 will absorb approximately twice as much energy during compressive straining than will Bisco NS-I and that prior thermal exposure (aging) and test temperature has little effect on this energy absorbing capability.

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