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- PREPARATION OF ELEMENTS AND THEIR FRACTURE TESTS -

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For the High Temperature Engineering Test Reactor, HTTR, the control rod sleeve is made of Alloy 800H for which a particular process is imposed when the reactor needs to be scrammed. The process rises from the limited heat resistance of the metallic alloy. The less restricted operation of the reactor as well as the even more reliable integrity of the control rod would be attained if there would be the control rod more resitant to high temperature and neutron irradiation.

In this respect the use of C/C composite for the control rod for HTGR is believed to be aspired, though there still remain a few difficulties to be overcome towards the actrual use of the material in the reactor: evaluation of the effect of neutron irradiation on its properties, fabrication of the elements, structural integrity of the elements, etc. This report summarizes the results which have been obtained as of March 1996 in the course of the development of the C/C composite control rod.

Materials used were pitch- or PAN-based fiber-reinforced 2-dimensional carbon composites, from which preforms of the elements of a cotrol rod were fabricatd. The preforms were carbonized at 1000℃ after being impregnated with pitch. Then they were graphitized at 3000℃, followed by a purification treatment with halogen. The elements included the pellet holder, lace truck and pin. The pin was fabricated by the fiber laminating technique. A control rod is to consist of pellet holders which are connected by the lace trucks with pins.

Various strength tests were carried out on these elements. An irradiation of the elements made of PAN-based material was performed in JRR-3 at 900±50℃ to a neutron fluence of 1×10^{25} n/m^2 (E>29fJ). As for the strength tests on the elements, there were some differences between PAN- and pitch-based composites: In general, elements made of PAN-based composite showed the more plastic behavior before they fractured, whereas those of pitch-based material behaved in the more brittle manner.

Fracture tests of the irradiated elements showed that frature load and fracture displacement enough for assuring the integrity of the control rod structure were maintained even after the irradiaion. It was also found that if the applied load was parallel ot the fiber felt plane both fracture load and strain increased, whereas the load increase and strain decrease were observed for the applied load against the plane.

Keywords: C/C Composite, Control Rod, HTTR, Irradiation, Strength

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炭素複合材料を用いたＨＴＴＲ用制御棒の開発（Ｉ）

― 制御棒要素の試作と破壊強度試験 ―

日本原子力研究所東海研究所材料研究部
衛藤　基邦・石山新太郎・宇賀地弘和

（1996年７月８日受理）

ＨＴＴＲの制御棒管はアロイ800Hで作られており、スクラム時には、その耐熱性を考慮して、制御棒操作は低温部（反射体領域）及び高温部（炉心領域）への二段階挿入で行われる。このような制御棒挿入時の制限を軽減するため、より耐熱性に優れた炭素複合材料を用いた制御棒の開発を進めた。炭素複合材料を使って実際に設計製作するためには、材料特性に及ぼす中性子照射の影響の解明、制御棒要素の製作と健全性の評価等多くの項目について研究開発を進める必要がある。本報告は炭素複合材料を用いた制御棒の開発を目的として製作した制御棒要素の照射前後の強度試験の結果をまとめたものである。

ＰＡＮ系及びピッチ系繊維を原料とする２Ｄ炭素複合材料を用いた。予備成型材にピッチ含浸処理を施した後、1000℃で炭素化処理、さらに3000℃で黒鉛化処理した。高純度化はハロゲンガスを用いて行った。制御棒要素としてペレットホールダ、レーストラック及びピンを製作した。これらの要素について種々の強度試験を行った。照射試験はＰＡＮ系材料についてのみＪＲＲ－３を用いて行った。照射温度及び照射量は、各々900±50℃、$1\mathrm{x}10^{25}\mathrm{n/m^2}(E>29\mathrm{fJ})$である。

ＰＡＮ系及びピッチ系材料を用いた各要素非照射材の強度試験の結果、一般にＰＡＮ系材料で製作した要素の方が破壊までの変形量が大きいことが分かった。ＰＡＮ系材料製要素の照射後試験の結果、各要素の破壊強度と変形量は制御棒の健全性を保つのに十分であることが明らかになった。また、負荷荷重が要素の繊維フェルト面に平行である場合、破壊荷重、破壊までの変形量ともに増加すると、垂直の場合には、破壊荷重の増加と変形量の減少が観察されることが分かった。

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1. INTRODUCTION

High Temperature Engineering Test Reactor(HTTR) has been under construction at JAERI, Oarai Research Establishment since 1990 and is supposed to attain criticality by the end of 1997. As is shown in Fig. 1 the core of the reactor consists of arrays of stacked fuel or replaceable reflector blocks made of graphite. In the normal operation conditon the maximum temperature of the graphite blocks will be around 1300°C when the temperature of the helium coolant at the outlet is 950°C [1]. To control the reactivity control rods are to be inserted into appropriate holes in the core and reflector blocks. In the event of a scram the control rods are inserted into the core taking advantage of the gravity. Nine out of 16 pairs of control rods in the reflector region are inserted immediately at the time of scram, while the other seven pairs in the core region are to be inserted 40 minutes later when the tempeature of the outlet coolant becomes lower than 750°C. The two step inserting method is employed mainly for the purpose of preventing the control rod sleeves in the core region from overheating. Fig. 2 shows an overview of the present control rod system to be applied for the HTTR. In the system the control rod sleeves are made of Alloy 800H.

Since the carbon-carbon (C/C) composite material is believed to be the more heat resistant than Alloy 800H, it would be lessen the restriction imposed on the reactor control procedure, if the control rod sleeve which accommodates boron carbide/carbon pellets is to be made of the material. The prominence of the C/C composite has been widely recognized as a high-strength heat-resistant structural material. Effect of neutron irradiation on thermal, mechanical and other properties of the composite has been fairly extensively examined by a number of investigators[2-9] so that the data on the material have been accumulated fairly abundantly enough to commence to think about its application to high temperature components of nuclear facilities.

In consideration of this situation a concept of control rod to be made of C/C composites has been proposed by the present authors in quest of the even better performance of the control rod for the HTTR. On the basis of the concept several elements for the control rod have been prepared from C/C composites and their mechanical strengths have been tested both before and after neutron irradiation. This paper summarizes the results of fracture tests of the elements to evaluate the applicability of the material to the control rod, aiming at the further development of the more heat-resistant control rod.

2. EXPERIMENTAL

PAN(polyacrylonytrile)-based or pitch-based carbon fibers were used for the preparation of elements for the control rod. These elements were shown in Fig. 3. The

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PAN(polyacrylonytrile)-based or pitch-based carbon fibers were used for the preparation of elements for the control rod. These elements were shown in Fig. 3. The textile preforms were enforced two-dimensionally. After being impregnated with pitch, the preforms were carbonized at 1000°C and graphitized at 3000°C. Then they were purified with halogen gas. The pellet holder and the lace truck were preformed by the combination of the cross-knitting and filament winding techniques. The fiber lamination technique was employed for the fabrication of pins. Fig. 4 shows a schematic for the control rod when these elements are assembled.

The strength of these elements was measured at room temperature in air using a screw-driven tensile test machine. Compressive tests of the pellet holder were carried out in its radial direction or axial one, as shown in Fig. 5(a). The lace truck was tensile-tested in a manner shown in Fig. 5(b). Bending strength of the pins was measured by three point method in either the against or parallel direction, as is shown in Fig. 5(c). Cross-head speed for these tests was 0.5 mm/min. Some of these elements were irradiated at 900± 50°C in JRR-3 to a maximum fluence of 1 x 10^{25} n/m^2(E>29fJ).

3. RESULTS AND DISCUSSION

3.1 Comparison between the PAN- and pitch-based materials

Fig. 6 shows the results of the bending test of pins made of PAN- or pitch-based material. The fracture strain for each material in the present case is several times larger than that for most materials which have been examined before[8-10]. The strength is also larger or comparable to that of the materials previously investigated. The fracture strength and strain of the materials in the present study seem to be comparable to those of PAN-based materials examined by other researchers who prepared hybrid C/C composites with surface treated fibers[11]. Comparing the present materials with each other, the pitch-based material showed the less brittle behavior, though the fracture strain seemed to be large enough even for the PAN-based material. The parallel specimens, which imply that their felt plane was parallel to the applied load, endured fairly larger stress comparing with the against specimens, whereas the fracture strain seems to be larger for the latter. This suggests that the felt plane bends more readily when the load is applied against itself.

Fig. 7 shows the results of the fracture test of pellet holders. Here, the ordinate and abscissa represent the load and displacement of the cross-head, respectively. It is seen in Fig. 7(a) that the axial fracture load does not differ much between the two materials, whereas the amount of deformation seems to be a little larger for PAN-based material than for the pitch-based one. The radial fracture load is much larger for the PAN-based material than for the pitch-based, which is seen in Fig. 7(b). On the basis of its larger fracture load and deformability observed in the case of radial fracture test, only PAN-based material was chosen for the irradiation experiment, textile preforms were enforced two-dimensionally. After being impregnated with pitch, the preforms were carbonized at 1000°C and graphitized at 3000°C. Then they were purified with halogen gas. The pellet holder and the lace truck were preformed by the combination of the cross-knitting and filament winding techniques. The fiber lamination technique was employed for the fabrication of pins. Fig. 4 shows a schematic for the control rod when these elements are assembled.

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3.2 Results of the irradiation experiment on the PAN-based material

(1) Pins

Fig. 8 shows stress-strain curves obtained from the bending test of pins made of PAN-based material. Loads were applied to the specimens either (a)parallel to or (b)against the felt plane. It is to be noted that the strength increased very pronouncedly when the load was parallel to the plane. This is probably because the strength of the specimen parallel to the felt plane would be less influenced by the weakening of the bond between the felt planes which might be caused by the irradiation. Though there was no large difference in the bending strength and fracture strain between the parallel and the against specimens before irradiation, they behaved diferently after irradiation. As is seen in Fig. 8(b) for the against specimens, the bending strength increased after irradiation, whereas the fracture strain decreased, which seems to be characteristic of irradiated carbon materials. In fact, the most materials examined in the previous experimant showed this general trend[8,9].

Peak stress(bending strength) versus fracture strain is plotted in Fig. 9 for both irradiated and unirradiated pins of PAN-based material. It is to be noted in this figure that the strength of parallel specimens increases with increasing fracture strain, whereas that of against specimens decreases with increasing fracture starain. It is interesting to find that the solid lines drawn in the figure fit to the data points for both irradiated and unirradiated specimens.

(2) Lace trucks

The results of the fracture test of lace trucks are shown in Fig. 10, where one can see that both strength and displacement, which was estimated from the movement of the cross-head, are larger for the irradiated specimens than for the unirradiated. A tendency similar to that observed for the pin was also found for the lace truck, which is shown in Fig. 11, since the felt plane of the lace truck was parallel to the loading axis, i.e., the strength increases with increasing fracture starain, though in this case the strain is not the one derived from the strain gage but from the displacement of the cross-head of the tensile test machine.

(3) Pellet holder

Load versus displacement curves for pellet holders in the cases of axial and radial loadings are shown in Figs. 12 and 13, respectively. It is to be noted that the fracture load for the axial loading seems to increase after the irradiation, even though the curve is rather rugged, which is probably because of the inclination to the buckling during the test. In the case of radial loading, fairly large decrease in the fracture load as well as in the fracture displacement was observed after the irradiation, though the values of both load and displacement seem to be large enough to apply the material to the holder.

(4) Dimensional changes caused by the irradiation

Results on the dimensional changes caused by the irradiation are shown in Fig. 14 for three kinds of elements. The dimensional changes of the control rod elements here are rather large comparing with those of the C/C composite materials, which have been reported in the references[8,9]. In an irradiation condition similar to the present case, dimensional reduction larger than 1 % was caused only for a uniaxial material and most materials showed reduction smaller than 0.3 %[8,9]. However, the dimensional reduction of about 2 % caused in the present irradiation conditon is believed to be tolerable for the design of the control rod of C/C composites. Moreover, it is very probable that the future modification or improvement of the material, of the preparation technique of preforms and/or of the heat treatment procedure would increase the stability of the irradiated elements. Along this line, the elements the concept of which is different from that for the ones tested in the present study were also considered and prepared for the future evaluation. This is shown in Fig. 15.

(5) Young's modulus

Fig. 16 shows changes in the Young's modulus of irradiated elements. Here, the data on the pin are derived from stress-strain curves obtained using strain gages during the bending test. The rest of the data are relative values calculated from the load-displacement curves.

The results on pins which indicate that there is a rather large difference in the modulus change between the parallel and the against specimens are reasonable if the fact that the against specimen can bend more readily than the parallel specimen is taken into account, i.e., the lower modulus of the against specimens. The apparent decrease in the modulus of the lace truck may be fortuitious in consideration of the method of calculation described above.

4. CONCLUSIONS

Elements for the control rod to be used in the HTTR were prepared from two kinds of carbon-carbon composites, pitch-based and PAN-based, aiming at the development of the more heat-resistant control rod which is believed to lessen the restriction on the operation and shut-down processes of the HTTR. The elements included pellet holder, lace truck and pin.

Main conclusions are;

(1) For the unirradiated elements those of PAN-based material showed the more deformable behavior than those of the pitch-based.

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Main conclusions are;

(1) For the unirradiated elements those of PAN-based material showed the more deformable behavior than those of the pitch-based.

(2) Elements of PAN-based material irradiated at 900°C to a fluence of 1×10^{25} n/m^2 (E>29fJ) maintained the fracture load and fracture displacement which may well be believed to be enough to assure the integrity of the control rod made of the material.

(3) The dimensional changes of the elements were at most less than 2 % after the irradiation.

(4) The strength and fracture strain increased after the irradiation when the applied load was parallel to the felt plane, whereas the strength increase and strain decrease were observed for the load applied against the plane.

References

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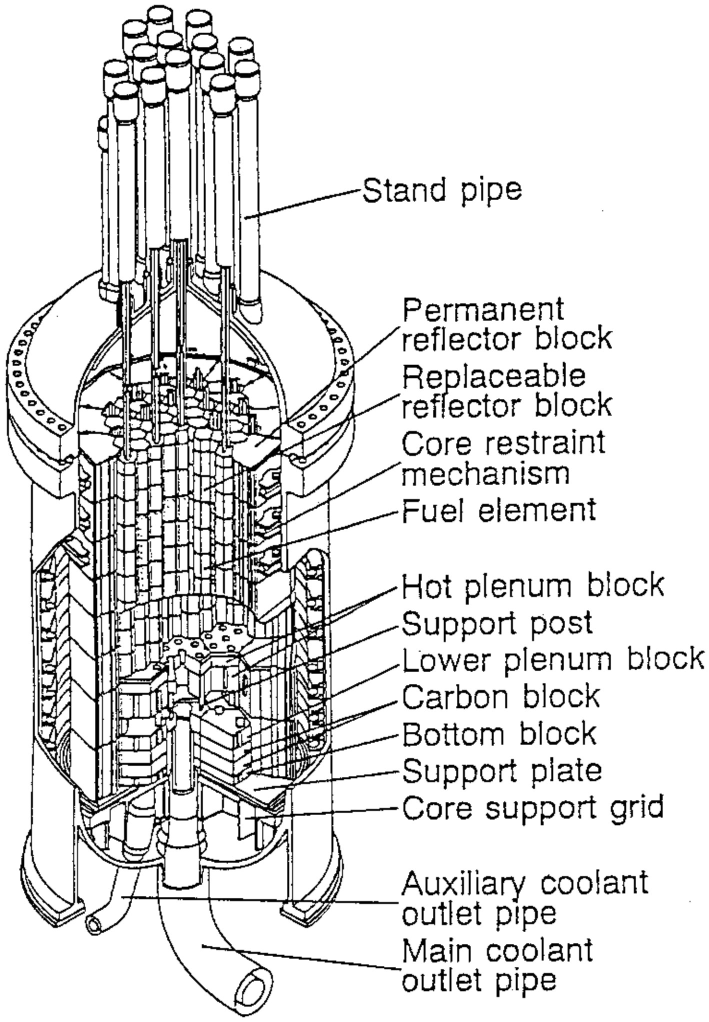


Fig. 1 A schematic for the HTTR pressure vessel and core.

Thermal power	30 MW
Outlet coolant temperature	850°C/950°C
Inlet coolant temperature	395°C
Fuel	Low enriched UO_2
Fuel element type	Prismatic block
Direction of coolant flow	Downward-flow
Pressure vessel	Steel
Number of main cooling loop	1
Heat removal	IHX and PWC (parallel loaded)
Primary coolant pressure	4 MPa
Containment type	Steel containment
Plant lifetime	20 years

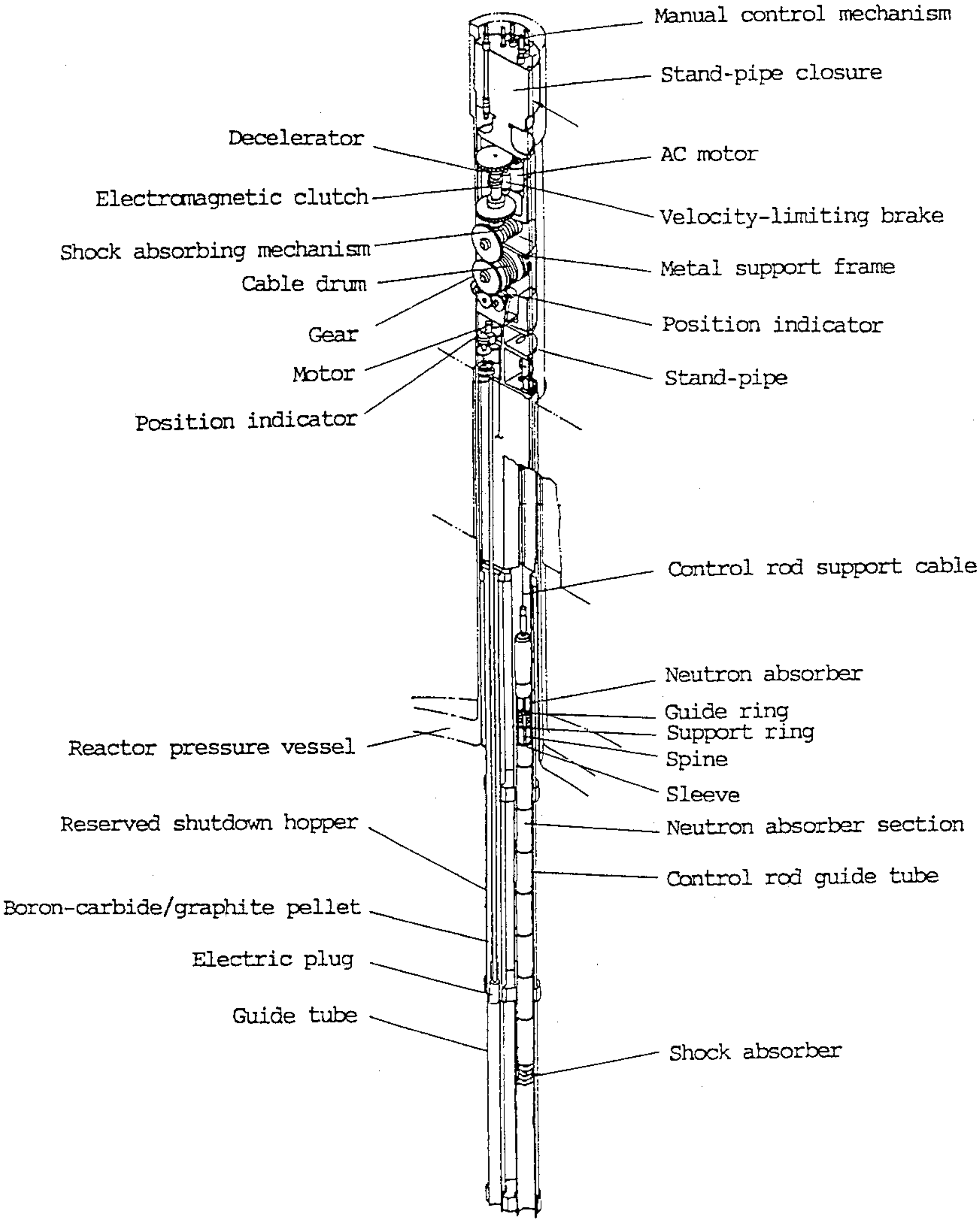


Fig. 2 Reactivity control system of the HTTR

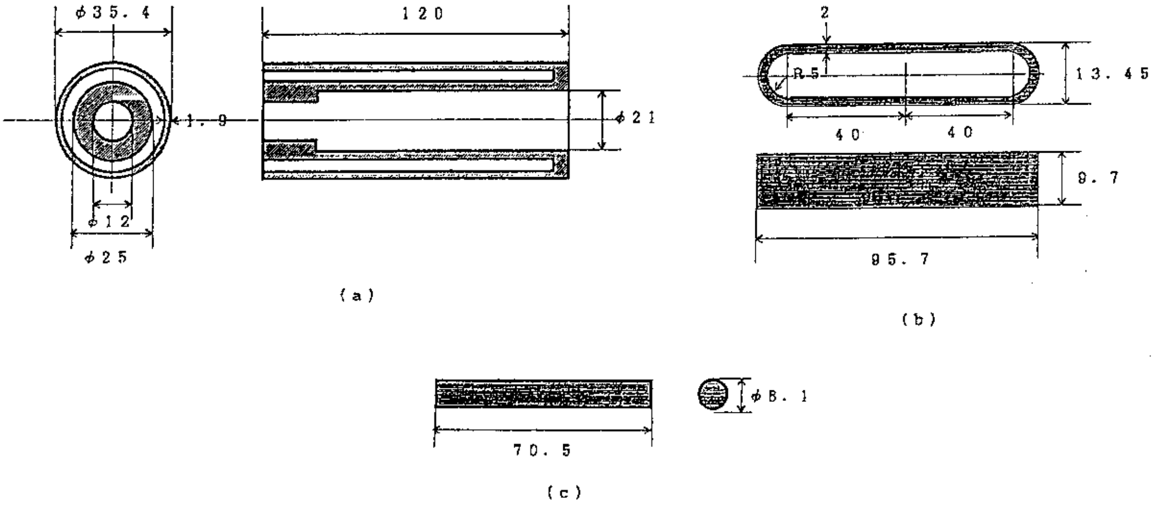


Fig. 3 Control rod elements prepared and tested in the present study.

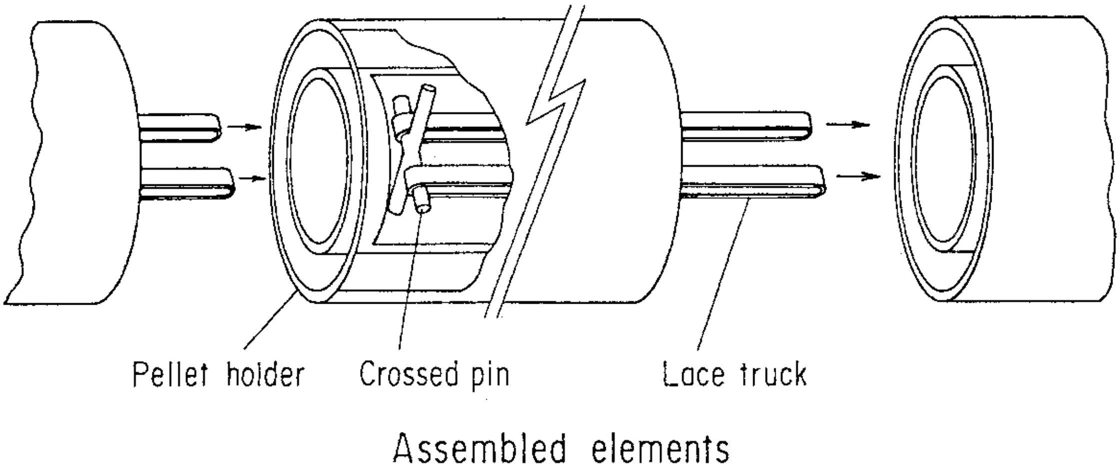
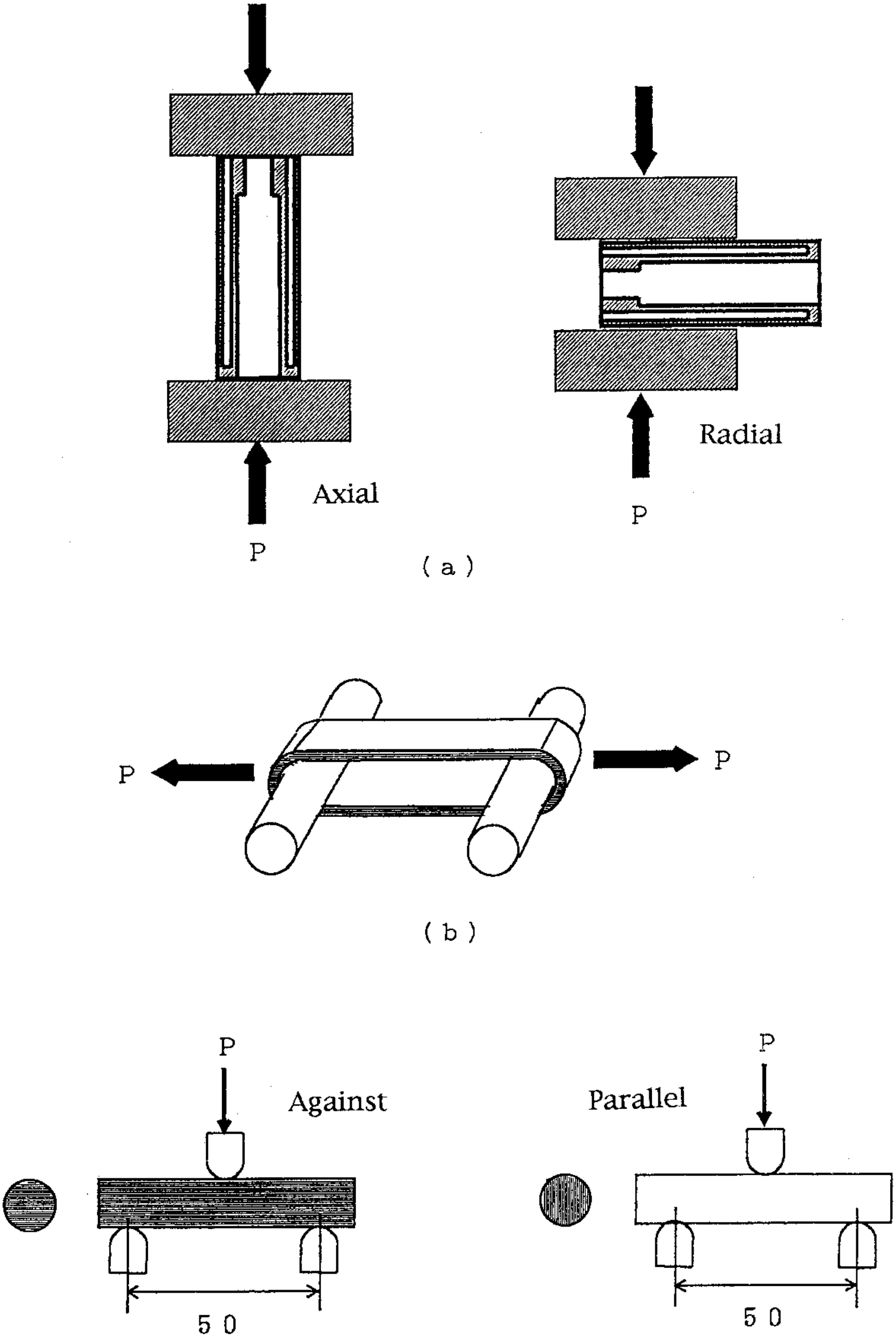


Fig. 4 A schematic for the control rod consisting of the elements.



(a)

(b)

(c)

Fig. 5 Method of mechanical strength tests.

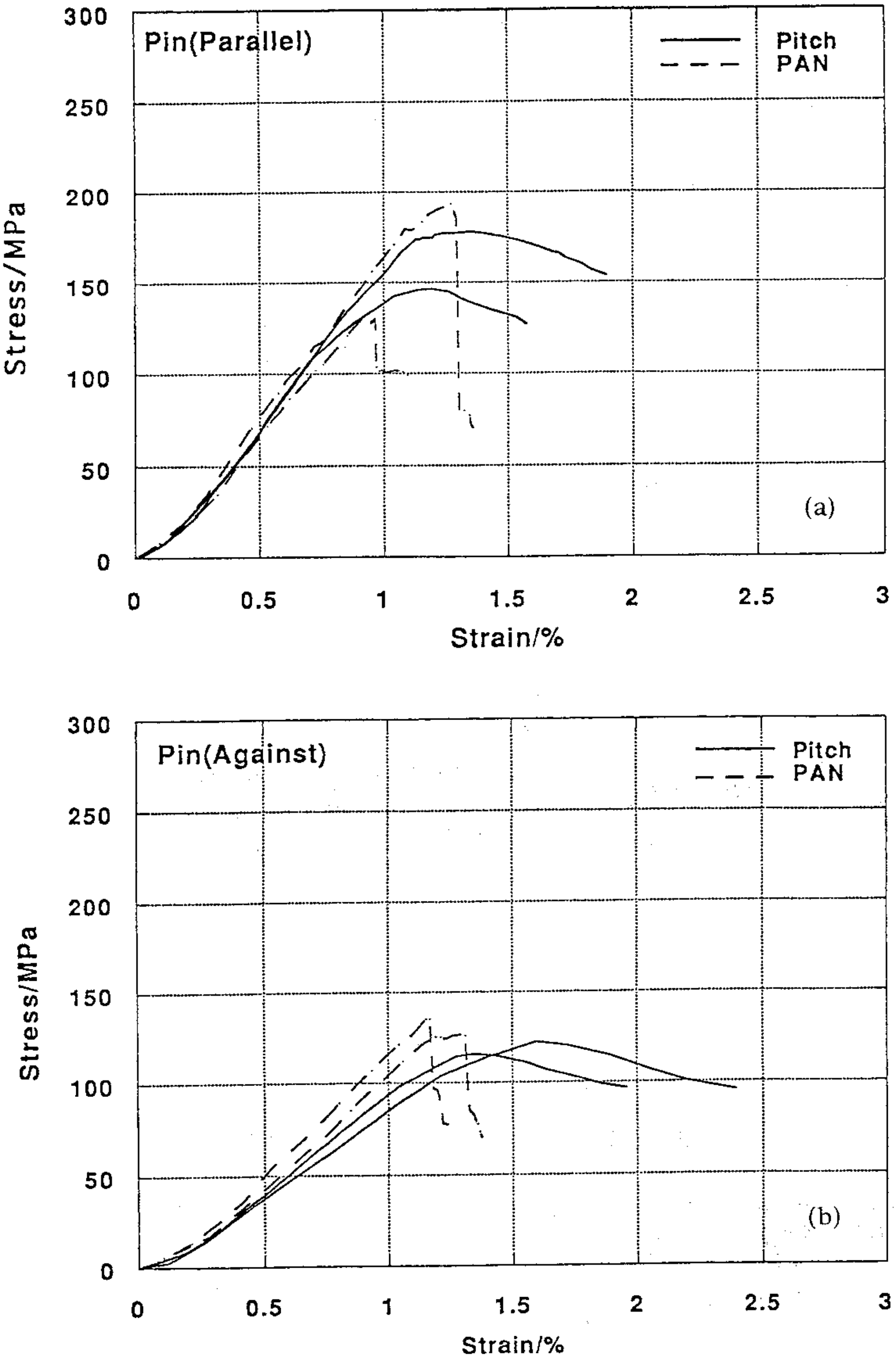
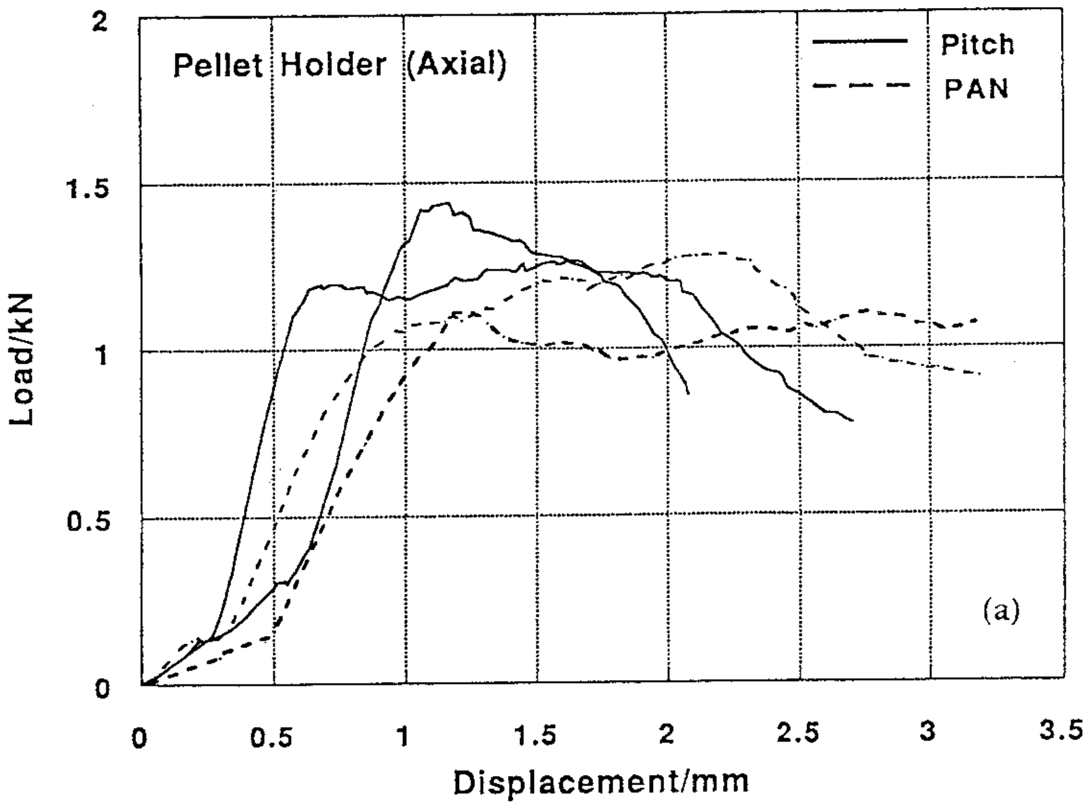


Fig. 6 Stress-strain curves obtained during the bending test of pins of pitch- or PAN-based material (a)parallel to or (b)against the felt plane.



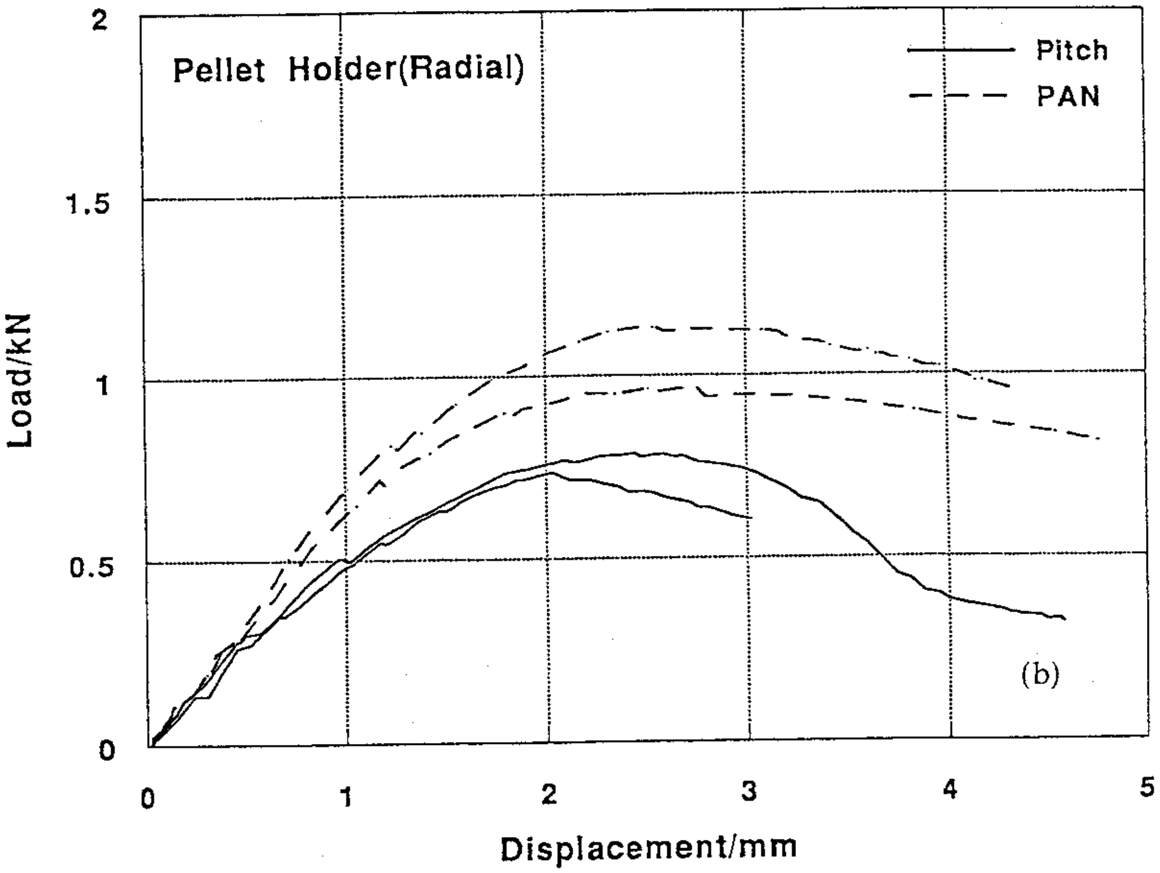


Fig. 7 Load-displacement curves obtained during the strength tests of pellet hoders of pitch- or PAN-based material with the load applied (a)axial or (b)radial.

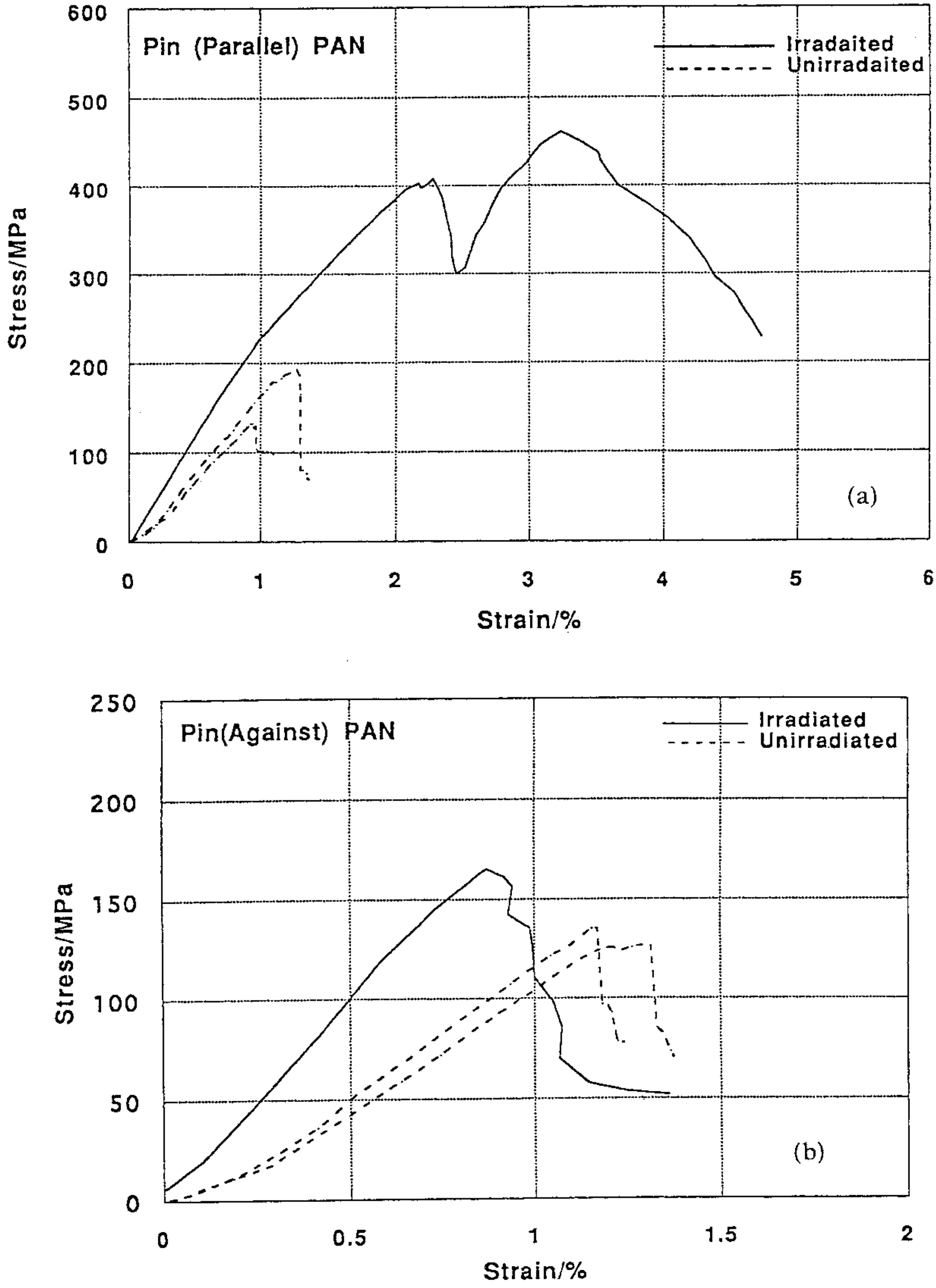


Fig. 8 Stress-strain curves for irradiated or unirradiated pins of PAN-based material with the applied load (a)parallel to or (b)against the felt plane.

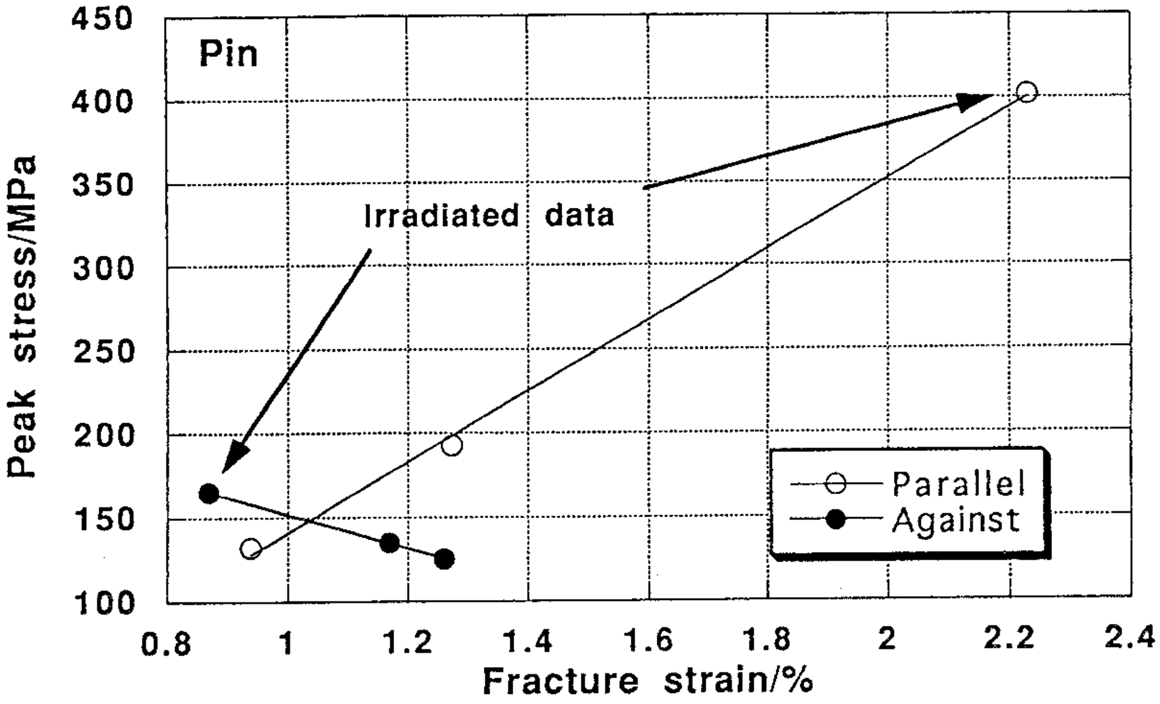


Fig. 9 Peak stress(bending strength) versus fracture strain curves for irradiated or unirradiated pins of PAN-based material.

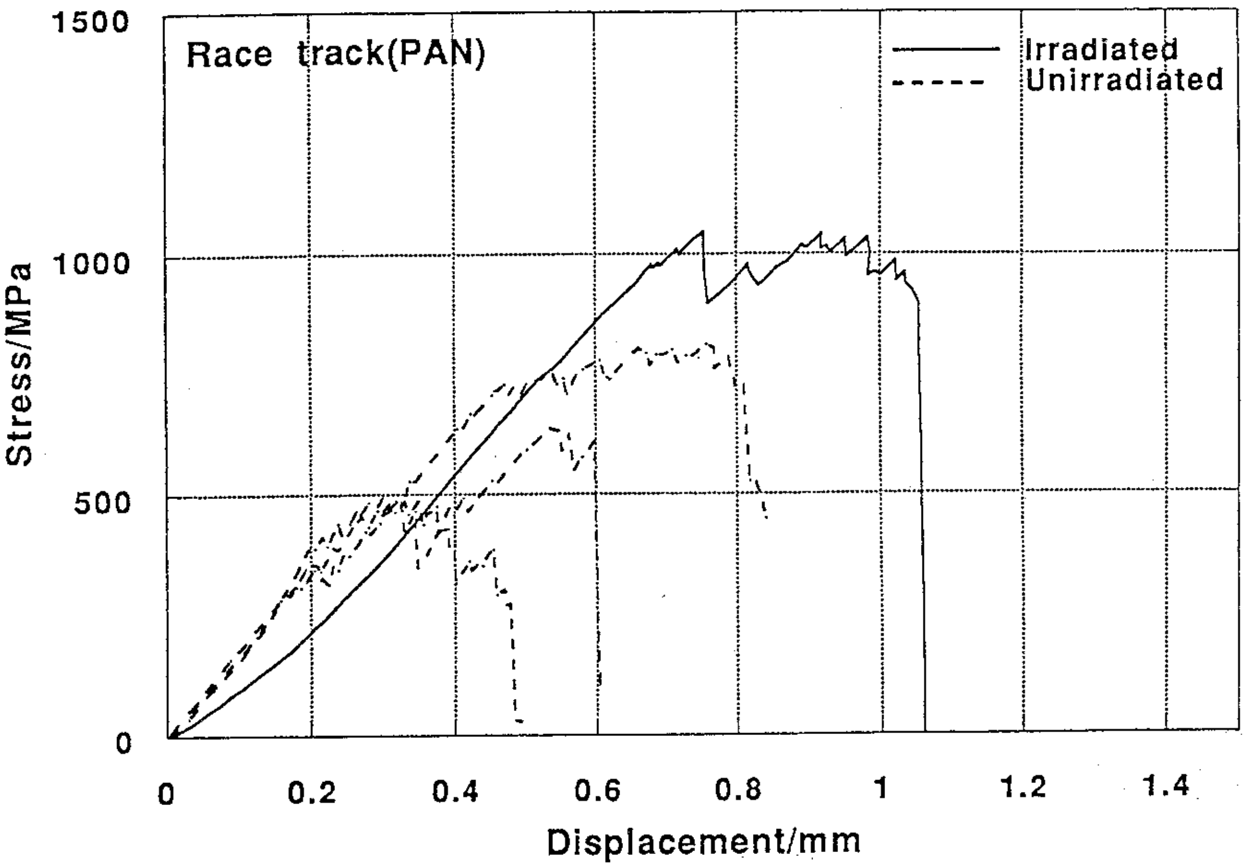


Fig.10 Stress-displacement curves for lace trucks of the PAN-based material irradiated or unirradiated.

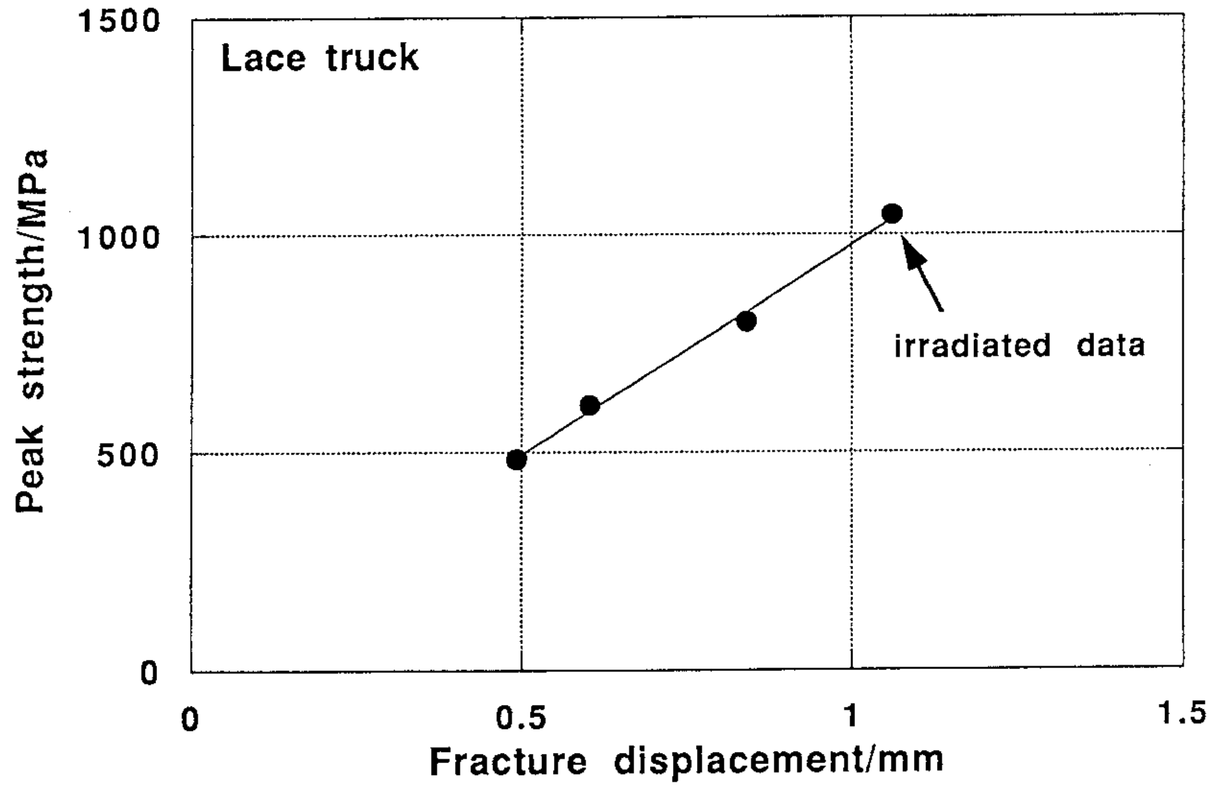


Fig.11 Peak stress(strength) versus fracture displacement plots for lace trucks.

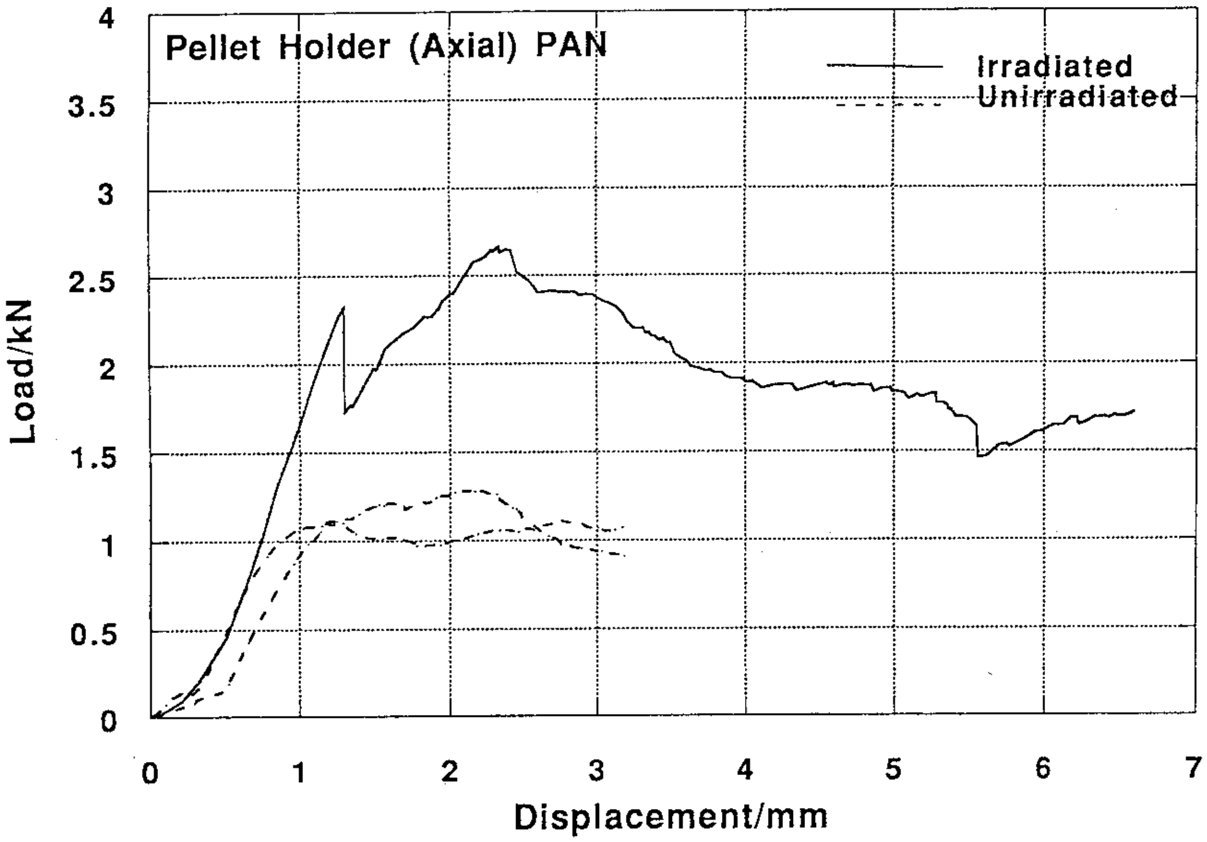


Fig.12 Load-displacement curves for pellet holders of PAN-based material loaded axially.

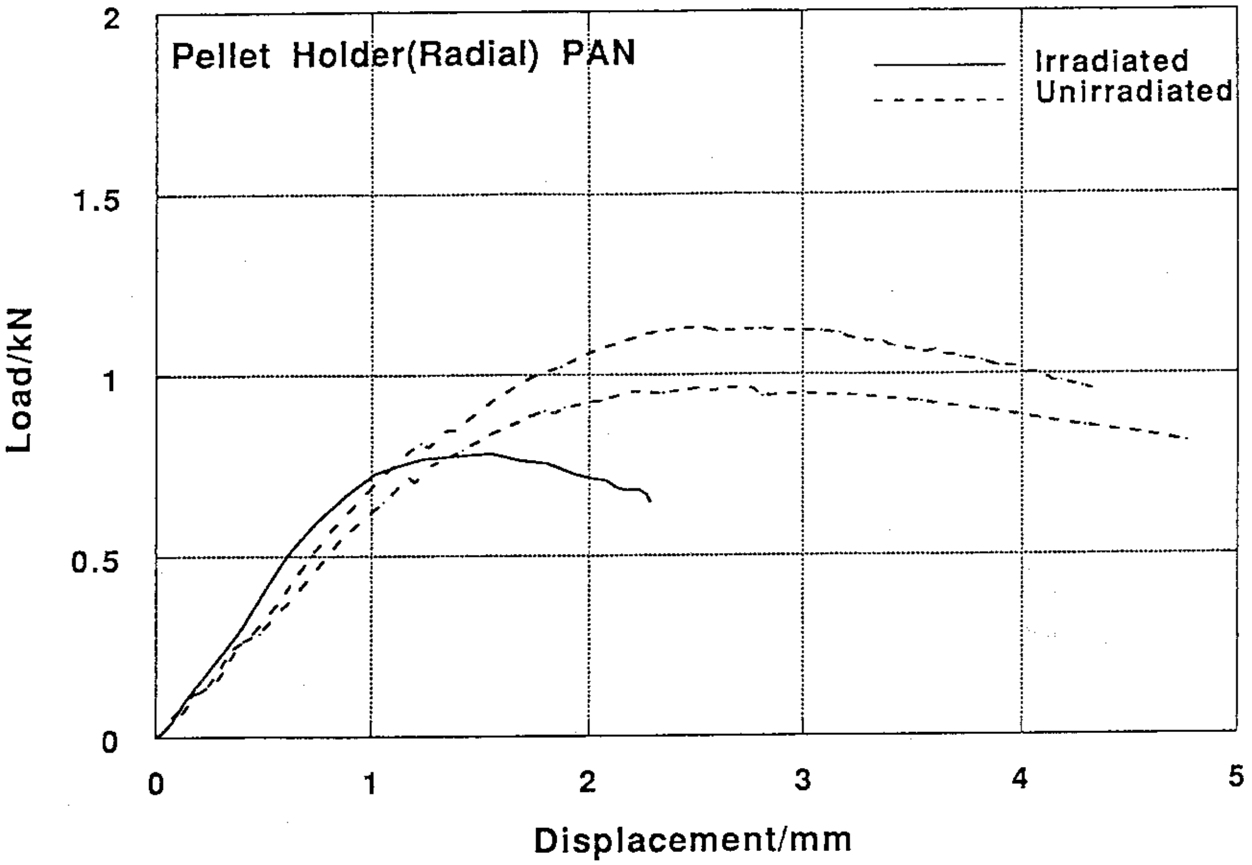


Fig.13 Load displacement curves for pellet holders of PAN-based material loaded radiallv.

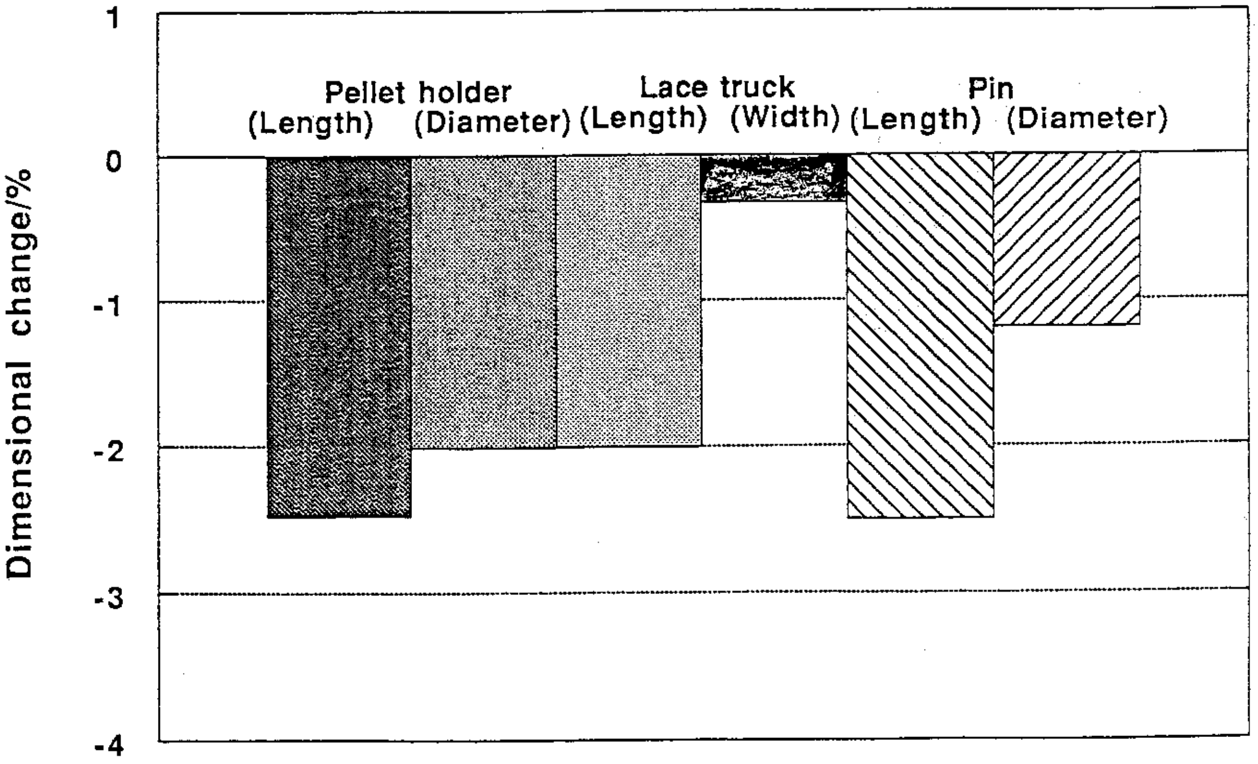
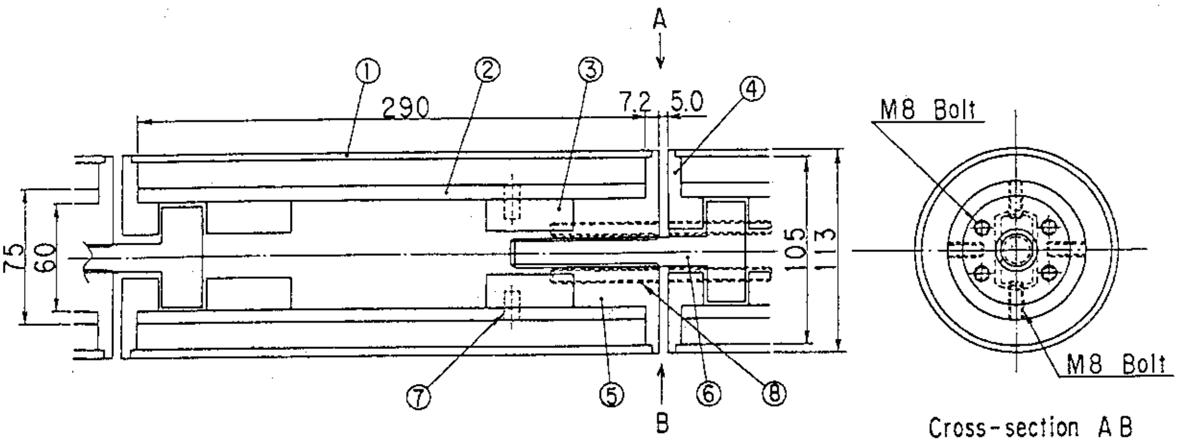


Fig.14 Dimensional changes of the elements after the neutron irradiation.



① Holder outer tube
② Holder inner tube
③ Lid support plate
④ Upper lid
⑤ Lower lid
⑥ M16 Bolt
⑦ M8 Bolt
⑧ M8 Bolt

Fig.15 A new concept of the control rod.

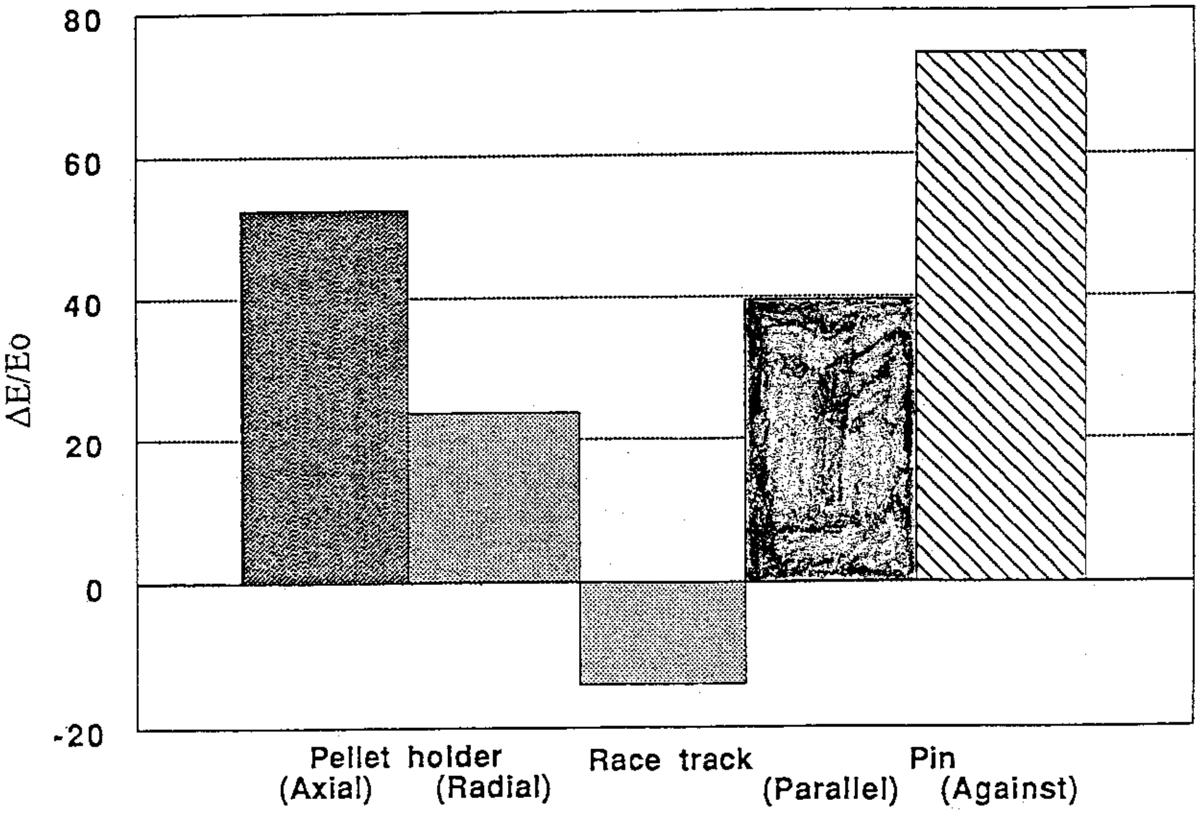


Fig.16 The modulus changes of the elements after the neutron irradiation.