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New Materials for Loudspeaker Diaphragms and Cones — An Overview —

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Desirable properties of materials for loudspeaker diaphragms are high specific Young's modulus E/ϱ (E: Young's modulus, ϱ: densintry), large flexural rigidity EI (I: moment of inertia of section) and large internal loss $\tan\delta$. In order to obtain large flexural rigidity, honeycomb structured diaphragms or porous metal diaphragms have been used. For the purpose of realizing ideal loudspeaker diaphragms which can be easily produced, however, the material itself should have high values of E/ϱ and $\tan\delta$.

This paper is a review on recent developments of materials for loudspeaker diaphragms in Japan. There are beryllium and boronized titanium diaphragms which are suitable for the high frequency loudspeakers because of their high specific Young's modulus, and carbon fiber reinforced olefin and polymer-graphite composite diaphragms which are suitable for the low frequency loudspeakers because of their high specific Young's modulus and internal loss and also production easiness of large diaphragms.

1. INTRODUCTION

Acoustical performances of loudspeakers are mainly dependent upon the physical properties of the vibrating systems. The diaphragm is the principal factor for the performance of the loudspeaker within the vibrating system. There are three desirable properties for materials of loudspeaker diaphragms, as follows,

1. large specific Young's modulus E/ϱ to get wider frequency range,
2. large flexural rigidity EI to reduce harmonic distortions,
3. large internal loss $\tan\delta$ to suppress breakups of the diaphragms.

From the above points of view, paper cones produced from natural pulps have been used as loudspeaker diaphragms for 50 years. This is because that paper cones satisfy the above three requirements somehow. However, there are large deviations of properties among mass-produced paper cones because they are produced one by one through many processes such as beating, felting, drying and pressing. Moreover, it is not so easy to obtain stable paper cones because they are sensitive against humidity. In this situation, new diaphragms for loudspeakers, which have superior and stable properties over paper cones and can be mass-produced, have been desired for many years.

In order to improve the properties of paper cones, newly developed high Young's modulus fibers; inorganic fibers such as glass fiber, carbon fiber and almina fiber, or organic fibers such as polyamid fiber, have been mixed into natural pulps. Because of the weakness of cross linking forces between reinforcing fibers and natural pulps, however, much progress has not been achieved yet.

Recently, some inorganic materials such as beryllium and boron with high specific Young's modulus have been developed and used as diaphragms for mid-range and high frequency loudspeakers. Composite diaphragms such as carbon fiber reinforced olefin diaphragm and polymer-graphite composite diaphragm with high specific Young's modulus and proper internal loss have also been developed and used for low frequency loudspeakers.

There are developments concerning honeycomb structured diaphragms and porous metal diaphragms which provide low frequency loudspeaker diaphragms with large flexural rigidity. However, there seems some production difficulties in these diaphragms.

In this paper, recent developments in materials for loudspeaker diaphragms in Japan are presented. These are beryllium and boronized titanium diaphragms for mid-range and high frequency loudspeakers, and carbon fiber reinforced olefin and polymer-graphite composite diaphragms for low frequency loudspeakers. This paper describes also their production processes, physical properties and performances of the loudspeakers using the new diaphragms.

2. NEW DIAPHRAGMS WITH HIGH SPECIFIC YOUNG'S MODULUS

Materials with high specific Young's modulus are desirable for diaphragms of mid-range and high frequency loudspeakers because wider frequency range is the most important factor for these loudspeakers. Beryllium and boron are suitable materials for this purpose. And these physical properties are shown in Table 1. Unfortunatelly, it is nearly impossible to make diaphragms of pure beryllium or pure boron by mechanical processes because of their hardness and low ductility and their activity in higher temperature state. Relatively inexpensive mass production processes of beryllium and boron diaphragms have been developed. These diaphragms have several superior properties comparing with ordinary diaphragms.

2.1 Beryllium diaphragm

2.1.1 Production process

There are several methods such as chemical vapor deposition, frame jet spraying and electro-plating to pro- produce beryllium diaphragms. However, these methods are not practical because of their low productivity. Taking account of low melting temperature and high vapor pressure of beryllium, Yuasa et al. have adopted vacuum deposition method[1].

Fig. 1 shows the vacuum deposition apparatus for

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massproduction of beryllium diaphragms. The substrate should satisfy the following requirements, such as easiness in forming, stability in high temperature atmosphere, and easiness in removing from the deposited beryllium. After several tests, Yuasa et al. found that a chemical dissolution of the substrate is the most practical method to remove the substrate considering these requirements, they adopted copper as the substrate material. The deposition process is as follows,

1. put beryllium ingots produced in a vacuum induction furnace into a hearth,
2. put copper substrates mounted to substrate holders into the vacuum deposition chamber, apply negative electrical potential of 1000 ~ 2000Volts, and heat the substrates up to 300°C ~ 500°C,
3. evacuate the vacuum chamber up to $3 \times 10^{-3} \sim 8 \times 10^{-4}$Pa,
4. focus the electron beam onto the beryllium ingot to evaporate it in a deposition rate of 1 ~ 2 μm/min,
5. after deposition, heat the substrates up to 600°C to anneal and sinter the diaphragms,
6. chemically dissolve the copper substrates away from the beryllium diaphragms by an aqueous solution of nitric acid,
7. deposit SiO_2 to protect the beryllium surfaces.

Fig. 2 is the experimental results of relations between substrate temperature and Young's modulus of the beryllium diaphragm. Fig. 3 are scanning electron micrographs of deposited beryllium surfaces at various substrate temperatures. This figure shows that the crystalline graine size grows up as the temperature increases. The substrate temperature during the deposition process has been decided to 300 °C ~ 500°C because the beryllium diaphragm becomes brittle when the substrate temperature exceeds 600°C.

Fig. 4 shows a thickness variation over the beryllium diaphragm. A substantially uniform thickness can be obtained by adjusting direction of the substrate holder, mechanisms of rotation and revolution of the substrate holder and the electrical potential applied to the substrate.

2.1.2 Characteristics of loudspeakers with beryllium diaphragms

Fig. 5 shows a comparison of sound pressure level frequency responses of 2.5cm dome type high frequency loudspeakers with the same shape beryllium diaphragm and titanium diaphragm.

By the use of beryllium diaphragms, mid-range and high frequency loudspeakers with wide and flat frequency responses and excellent transient responses can be obtained.

2.2 Boronized titanium diaphragm

A development of boron deposited titanium diaphragm by a vacuum deposition method has been reported[2]. However, the method doesn't seem suitable to be used as a mass production process of boron deposited diaphragms, because boron has exceedingly high melting temperature and its vapor pressure is very low.

Utilizing the activity of boron at high temperatures, a diffusion process of boron into steel has been recently established. In the case of boronizing of titanium, however, titanium is apt to become brittle because titanium reacts easily with oxigen and nitrogen at temperatures over 800°C. Tsukagoshi et al. have developed a boronizing process adaptable for titanium and succeeded in mass-producing the high specific Young's modulus diaphragms at a reasonable cost[3].

2.2.1 Boronizing process

Substrates for this boronizing process should have low density, easy formability and small deformation at high temperatures. Accordingly titanium is the most suitable material for the substrate.

Fig. 6 shows a new apparatus to produce boronized titanium diaphragms. Titanium diaphragms are buried into a mixed powder of boron (70wt%), carbon (20wt%) and sodium carbonate (10wt%) within the graphite case. The chamber is evacuated up to 1 Pa to prevent titanium from oxidation, and then the graphite case is heated by a high frequency induction furnace. Carbon and sodium carbonate functions as an activator and oxidation suppressor for boron and titanium. Activated boron diffuses into the titanium and boronized layers are produced at the titanium surfaces.

Fig. 7 shows the Young's modulus of a 25μm boronized titanium diaphragm as a function of treating temperatures and treating times. Proper treating temperature is determined from 1000°C to 1200°C because diffusing rate is too slow under 1000°C and substrates deform over 1200°C. Fig. 8 is an X-ray diffraction pattern of the boronized titanium diaphragm at 1200°C for 10 minutes. Only titanium borides in the forms of TiB and TiB_2 are recognized. Fig. 9 is a scanning electron micrograph of the same specimen. A result of atomic analysis of boron and titanium by an electron probe microanalizer along the marked line is shown in Fig. 10. As a result of these analyses, it is recognized that boronized layers of TiB_2 are produced on both surfaces of the substrate, and spikes of TiB are developed towards the center of the substrate. These spikes prevent the layer from exfoliation. Several physical properties of typical boronbized titanium and pure titanium are listed in Table 1. The boronized titanium has 2.6 times higher specific Young's modulus than that of pure titanium.

2.2.2 Characteristics of loudspeakers with boronized titanium diaphragm

Fig. 11 shows a comparison of sound pressure level frequency responses of 2.5cm dome type high frequency loudspeakers with a same shape boronized diaphragm and titanium diaphragm. The high frequency resonance of the boronized diaphragm reaches 37kHz which is 1.6 times higher than that of titanium one. The ratio of the high frequency resonances almost coincides with the ratio of speeds of sound in both materials. Fig. 12 shows a sound pressure level frequency response of 12cm cone type mid-range loudspeaker using the boronized titanium diaphragm. The frequency response is fairly flat and smooth in the frequency range from 300Hz to 12kHz and the harmonic distortion is very low in the frequency range from 500Hz to 7 kHz.

As the results, using the newly developed boronizing process, we can achive,

1. diaphragm with 2.6 times higher specific Young's modulus than that of titanium,
2. diaphragm with a reasonable cost because an inexpensive amorphous boron and relatively simple apparatus can be used, and large number of diaphragms can be treated at a time.

Boronized titanium diaphragm can provide mid-range and high frequency loudspeakers with wide and smooth frequency response and low harmonic distortion.

3. NEW DIAPHRAGMS WITH HIGH SPECIFIC YOUNG'S MODULUS AND INTERNAL LOSS

For the applications to low frequency loudspeaker diaphragms, some new materials are desirable, which can be mass-produced into large diaphragms and provide a wide frequency range of pistonic motion and a smooth roll off frequency response above the cut off frequency. For this purpose, materials with high specific Young's modulus and large internal loss are required. Moreover, to get diaphragms with small variations and stability against various weather conditions, synthesized materials should be used.

Recently, there have been many researches and de velopments on the composite materials which consist of plastic matrices and inorganic fillers. And these composite materials have excellent properties and can be mass-produced in a reasonable cost. Examples of the developments are on the carbon fiber reinforced olefin diaphragm by Niiguchi et al. which consists of synthesized pulp and carbon fiber, and polymer-graphite composite diaphragm by Tsukagoshi et al. which consits of polyvinylchloride resin and graphite flakes.

3.1 Carbon fiber reinforced olefin diaphragm

Niiguchi et al. have developed carbon fiber reinforced (CFR-) olefin diaphragm using high density polyethylene with lowest density within polymers as a matrix and high modulus carbon fiber as a filler[4].

3.1.1 Production process

Synthesized pulp of the high density polyethylene have been used as a matrix. In order to adapt to a wet process, the pulp is treated to be given a hydrophilicity. High modulus carbon fiber with small radius has been selected as a filler so as to keep good formability. Physical properties of the high modulus carbon fiber are shown in Table 1.

Fig. 13 is the mass production process of CFR-olefin diaphragms. Beaten and fibrilized pulp and chopped carbon fiber are mixed. From this mixture, continuous composite sheets are produced by felting process same as in the ordinary paper. Various kinds of loudspeaker diaphragms can be produced continuously from the sheet by the apparatus shown in Fig. 14. The composite sheet as felted has low Young's modulus because of weak cross linking forces between fibers. Through the hot forming process, however, a composite diaphragm with strong structure and light weight is produced because the synthesized pulp is melted and tightly links to the carbon fiber. Therefore, CFR-olefin diaphragm has a higher Young's modulus and stability against heat and humidity comparing to the paper cone. Fig. 15 is the scanning electron micrographs of the composite sheet before and after the heat treatment. After the heat treatment, carbon fibers are buried completely into the matrix.

Fig. 16 shows the relation between Young's modulus and density of the CFR-olefin diaphragm and the weight content of carbon fiber. Fig. 17 shows the variation of internal loss by the weight content. Taking acount of the above mentioned results and the formability, 20wt% of carbon fiber content seems optimum. Table 1 indicates the physical properties of CFR-olefin diaphragm at 20 wt% of carbon fiber. CFR-olefin diaphragm has three times larger specific Young's modulus comparing to that of paper cone.

3.1.2 Characteristics of loudspeakers with CFR-olefin diaphragms

Fig. 18 shows a comparison of sound pressure level frequency responses of 10cm cone type loudspeakers with the same shape CFR-olefin diaphragm and paper cone.

By the use of CFR-olefin diaphragm, a flat frequency response and a low harmonic distortion characteristic have been realized.

3.2 Polymer-graphite composite diaphragm

Tsukagoshi et al. have developed polymer-graphite (PG) composite diaphragm having a graphite-like structure and a high Young's modulus[5]. The structure is based on completely different idea from that of other conventional fiber reinforced plastics.

3.2.1 Production process

Graphite is one form of the carbon crystal and has a laminar structure. The layers of the laminar structure consist of strongly linked many hexagonal rings of carbon atoms as shown in Fig. 19. And this structure gives an extremely high Young's modulus. Because graphite is easily exfoliated between the layers by applying shearing forces, thin graphite flakes can be easily produced. The graphite-like structure has been realized in PG composite by orienting the graphite flakes parallel to the surfaces of the diaphragm. Polyvinylchloride was selected as a matrix, because it can strongly adhere graphite flakes by a strong polarity, inspite of poor adhesiveness of graphite.

Fig. 20 shows the flow chart of the production process of PG composite diaphragms. The most important processes are mixing and orientation. In the mixing process, delamination of the graphite flakes is caused by strong sheering force applied to PG composite material, consequently fresh and active surfaces of the graphite flakes can be rigidly bound with polyvinylchloride. In the orientation process, through repeated rollings, the graphite flakes are oriented parallel to the surfaces of a PG composite sheet, and results in a graphite-like structure and a high specific Young's modulus. Various shapes of loudspeaker diaphragms can be produced from PG composite sheets by vacuum forming.

Fig 21 shows the relation between Young's modulus of a PG composite sheet and mixing ratio of graphite to polyvinylchloride matrix by weight. Considering the above results and the formability, optimum mixing ratio is about two. A PG composite sheet has a higher sound velocity than that of titanium or aluminum and has a comparable internal loss to that of a cone paper as shown in Table 1.

Fig. 22 shows the relation between Young's modulus and the orientation of graphite flakes. Reed specimens are prepared by slicing at various inclined angles θ from the surface of a block produced by lamination of PG composite sheets. This figure shows that a specimen parallel to the surface has the maximum Young's modulus. The scanning electron micrograph of the fracture cross section of a PG composite sheet is shown in Fig. 23. A laminar structure parallel to the surface is observed. According to the above mentioned results, it was confirmed that the high Young's modulus of the PG composite sheet is originated from the extremely high orientation of graphite flakes. Fig. 24 shows the decay patterns of free vibration of PG composite, aluminum and cone paper reed. The decay of vibration in PG composite is faster than that in aluminum and is comparable to that in cone paper. Therefore, it is apparent that PG composite has a relatively large internal loss.

3.2.2 Characteristics of loudspeakers with Polymer graphite composite diaphragm

Because PG composite has a good formability, it can be formed into various shapes and sizes of diaphragms for either low frequency or high frequency loudspeakers.

Fig. 25 shows a 40cm low frequency loudspeaker using a PG cone with corrugations and a PG dust cap. Fig. 26 shows a comparison of sound pressure level frequency responses of 40cm low frequency loudspeakers with the same shape PG composite diaphragm and paper cone. The low frequency loudspeaker with PG composite diaphragm realizes flat frequency response and lower harmonic distortion.

Fig. 27 shows a comparison of sound pressure level frequency responses of 2.5cm dome type high frequency loudspeakers with the same shape PG composite diaphragm and titanium one. High frequency resonance of the loudspeaker with PG composite diaphragm is about 20% higher than that with titanium diaphragm. This ratio is proportional to the ratio of sound velocities of both materials. The peak in the frequency response of the loudspeaker with PG composite diaphragm at the high frequency resonance almost disappears.

As discussed above, by the use of PG composite, superior diaphragms with high specific Young's modulus, large internal loss, and stability against humidity can be obtained in various shapes and sizes at a lower cost comparable to paper cone.

4. CONCLUSION

This paper presents the production processes, physical properties and performances of the newly developed diaphragms in Japan, such as beryllium diaphragm, boronized titanium diaphragm, and corbon fiber reinforced olefin diaphragm and polymer-graphite composite diaphragm.

The former two diaphragms are suitable for mid-range and high frequency loudspeaker because of their high specific Young's modulus. The latter two are applicable to various loudspeakers because of their high specific Young's modulus and relatively large internal loss. In the latter two materials, specific sheets such as carbon fiber reinforced olefin sheets and polymer-graphite composite sheets are produced at first, and then the sheets are formed into diaphragms with various shapes and sizes.

Polymer-graphite diaphragms furnish loudspeakers which can respond faithfully to pulsive signals included in digital audio signals without a delay or a distortion. This means the loudspeaker can produce high fidelity sounds. Moreover, the polymer-graphite diaphragms can offer loudspeakers which would be necessary in the future digital audio era.

However, newer materials and structures for loudspeaker diaphragms which have lighter weight and larger flexural rigidity should be developed continuously.

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(4) H. Niiguchi and M. Ieki, "Reinforced Olefin Diaphragms for Loudspeakers", National Technical Report, vol. 7, pp.970—977, (1979), in Japanese.

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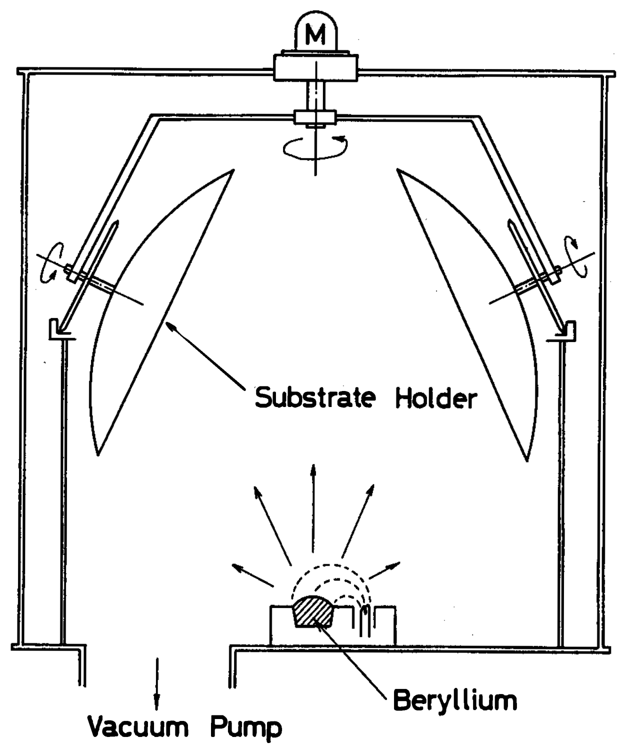

Fig. 1 Vacuum deposition apparatus for mass production of beryllium diaphragms.

Table 1 Physical properties of materials for diaphragms.

Material	Young's modulus E, x10^{10}N/m^2	Density ϱ, x10^3kg/m^3	Specific modulus E/ϱ, x10^7(m/s)^2	Sound velocity $\sqrt{E/\varrho}$, x10^3m/s	Internal loss tanδ, –	Melting point °C
Beryllium	28	1.85	15	12	0.002	1284
Boron	40	2.34	17	13	0.002	2225
Aluminum	7.0	2.7	2.6	5.1	0.002	660
Titanium	10	4.5	2.2	4.7	0.002	1668
Boronized titanium	25	4.5	5.6	7.5	0.002	
Carbon fiber	23	1.74	13	11.5		
CFR-olefin	0.37	0.45	0.82	2.9	0.025	
Polymer-graphite	7.0	1.8	3.9	6.2	0.05	
Cone paper	0.1~0.2	0.5	0.2~0.4	1.4~2.0	0.02~0.05	
Graphite Glass	35	1.4	25	5.0	.005	

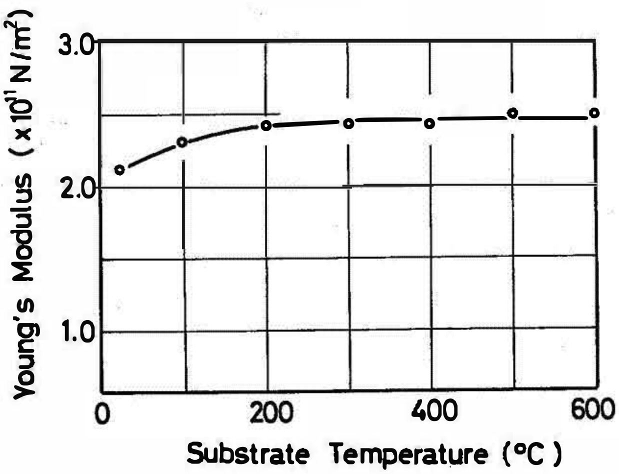

Fig. 2 Relation between substrate temperature and Young's modulus of beryllium diaphragm.

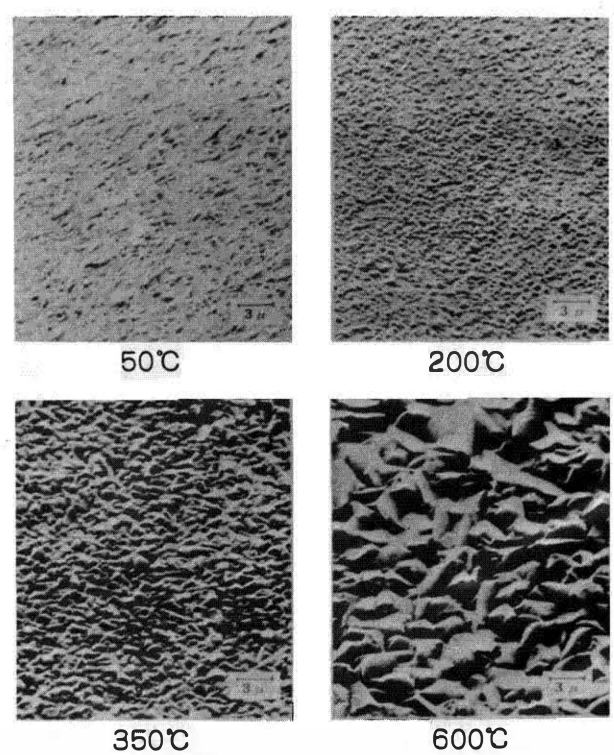

Fig. 3 Scanning electron micrographs of deposited beryllium surfaces at various substrate temperatures.

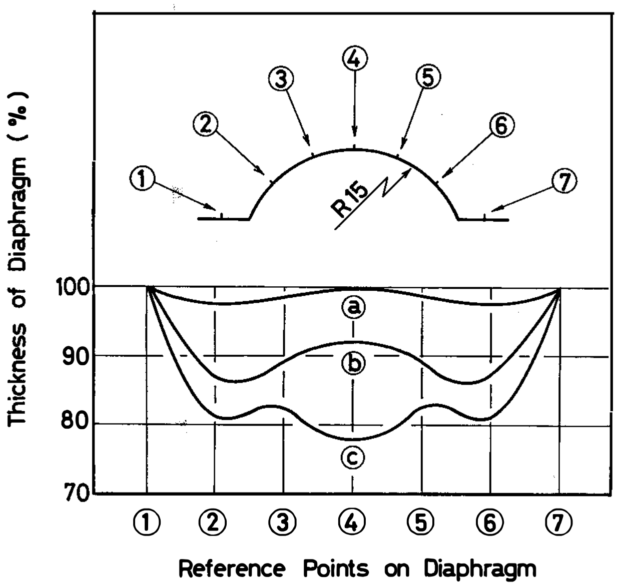

Fig. 4 Thickness variation over diaphragms.
a) Deposited beryllium diaphragm.
b) Pressed aluminum diaphragm.
c) Pressed beryllium diaphragm.

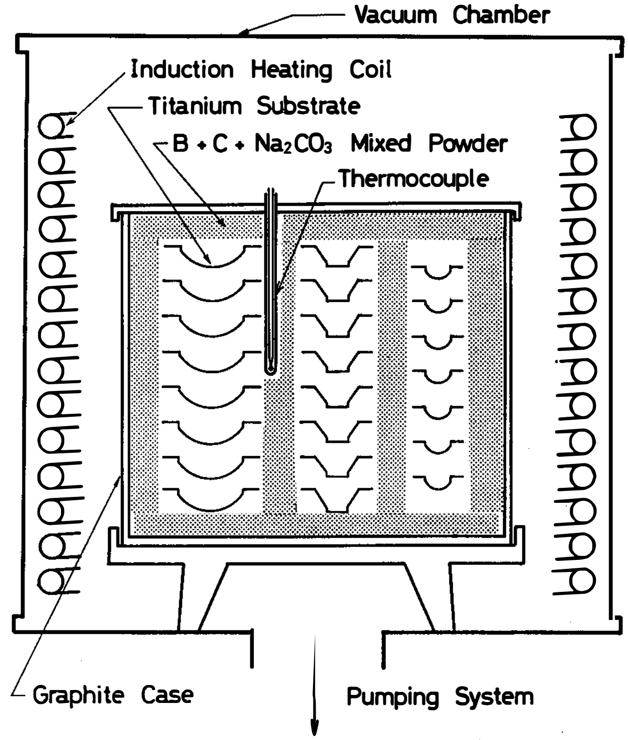

Fig. 6 Apparatus for production of boronized titanium diaphragms.

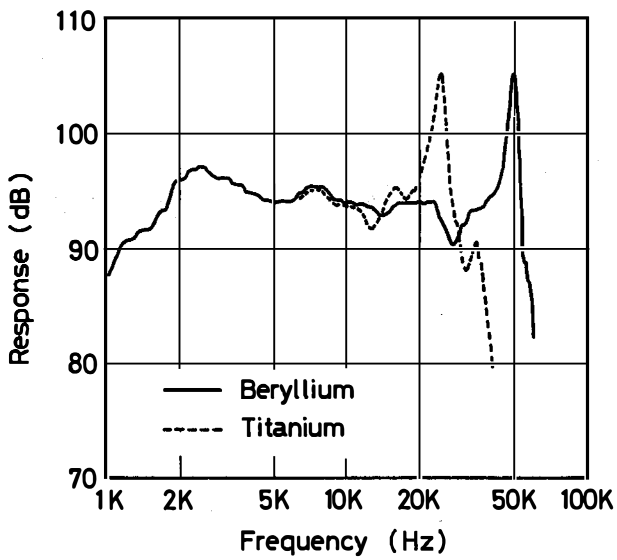

Fig. 5 Comparison of sound pressure level frequency responses of 2.5cm dome type high frequency loudspeakers with beryllium diaphragm (a), and titanium diaphragm (b).

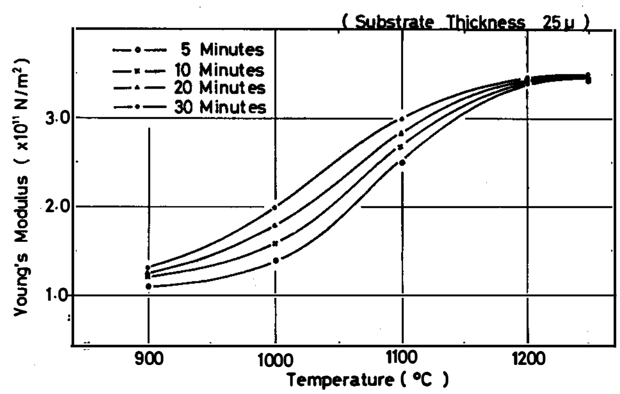

Fig. 7 Young's modulus of 25μm-thick boronized titanium diaphragm as function of treating temperatures and treating times.

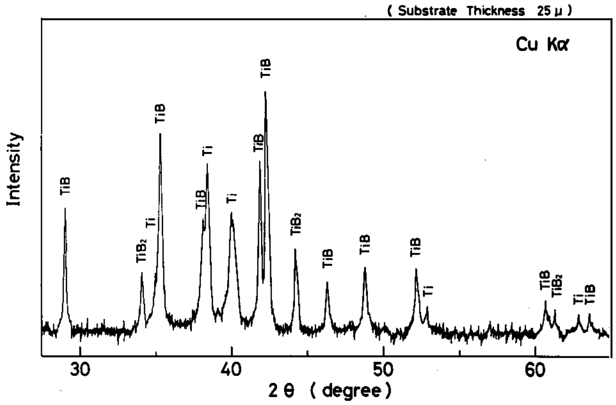

Fig. 8 X-ray diffraction pattern of boronized titanium diaphragm at 1200°C for 10 minutes.

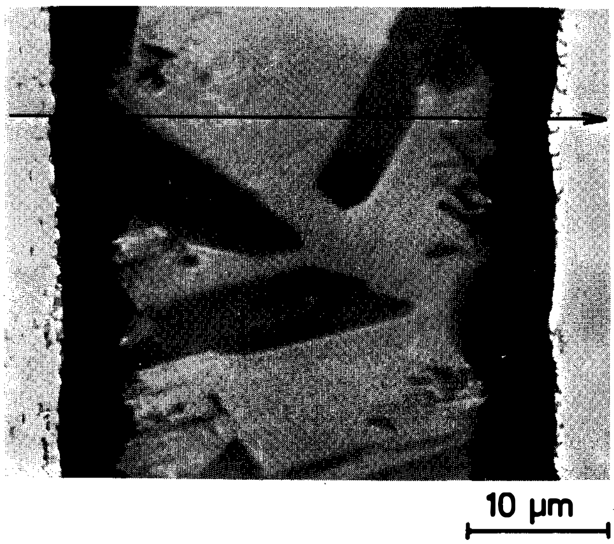

Fig. 9 Scanning electron micrograph of cross section of boronized titanium (1200°C, 10 min.).

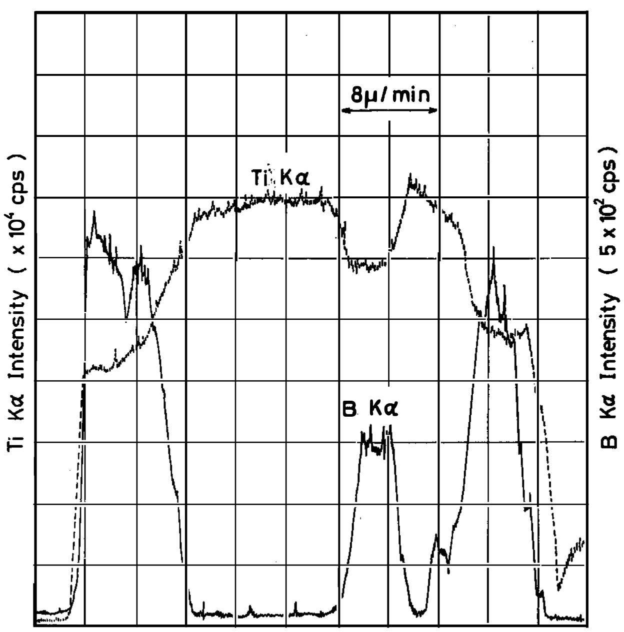

Fig. 10 Atomic analysis of boron and titanium by electron probe microanalyzer along the line marked on Fig. 9.

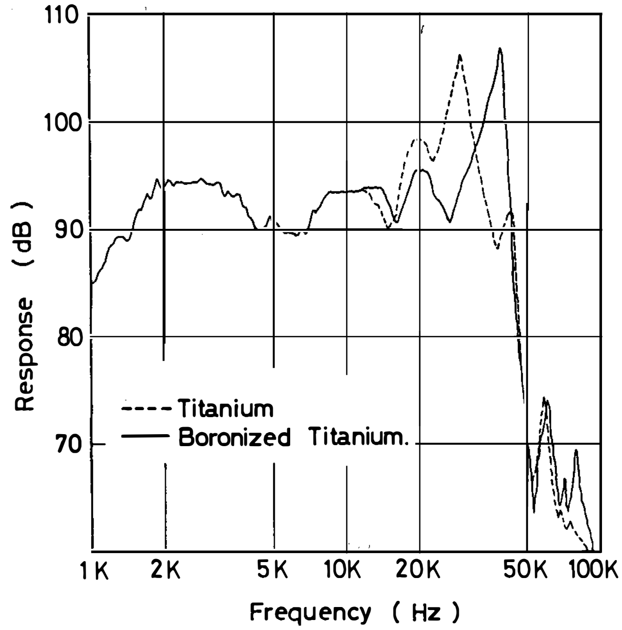

Fig. 11 Comparison of sound pressure level frequency responses of 2.5cm dome type high frequency loudspeakers with boronized titanium diaphragm (a), and titanium diaphragm (b).

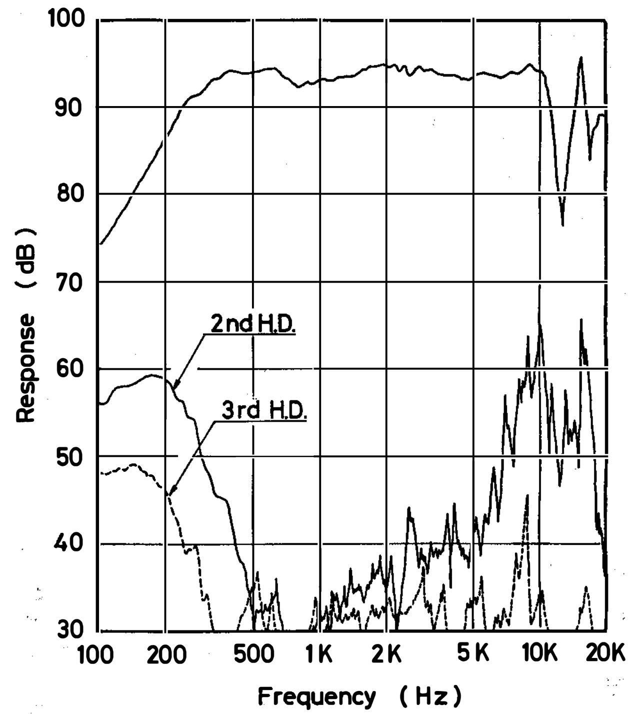

Fig. 12 Sound pressure level frequency response of 12cm cone type mid-range loudspeaker with boronized titanium diaphragm.

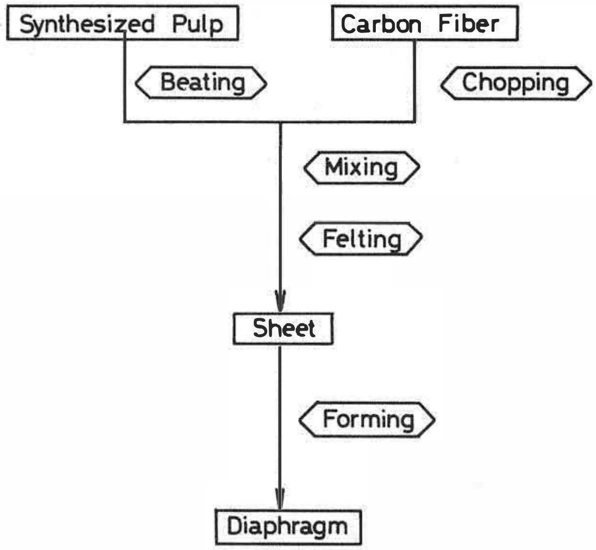

Fig. 13 Flow chart of mass production process of CFR-olefin diaphragms.

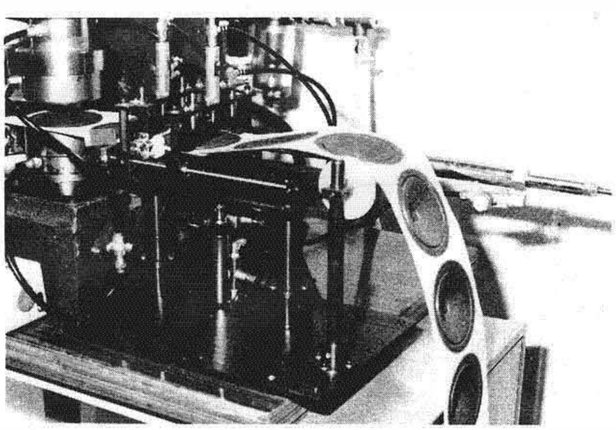

Fig. 14 Continuous mass production apparatus of CFR-olefin diaphragms.

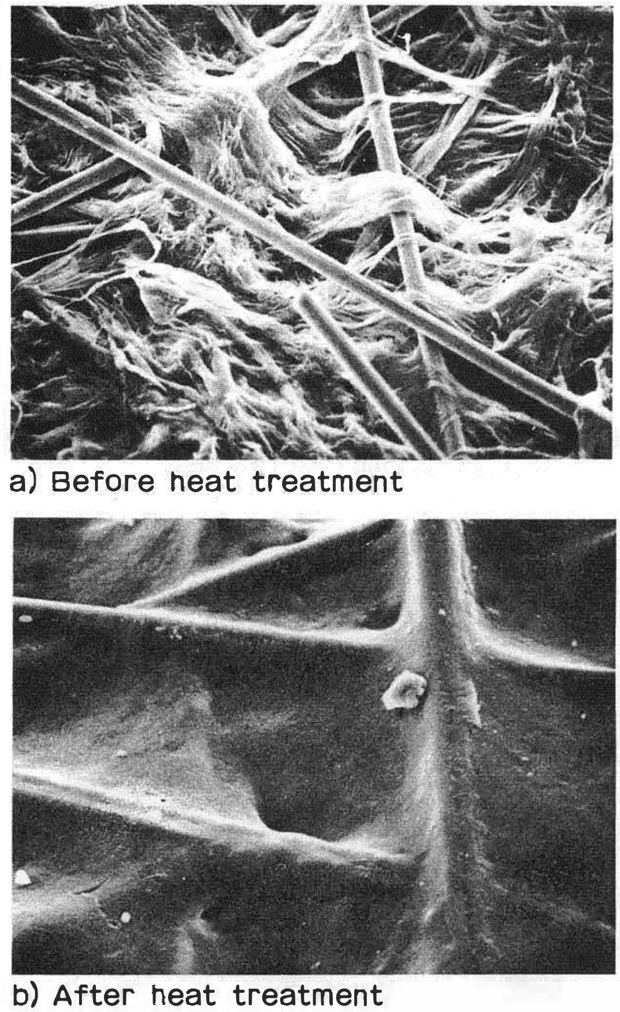

Fig. 15 Scanning electron micrographs of CFR-olefin sheets before (a), and after heat treatment (b).

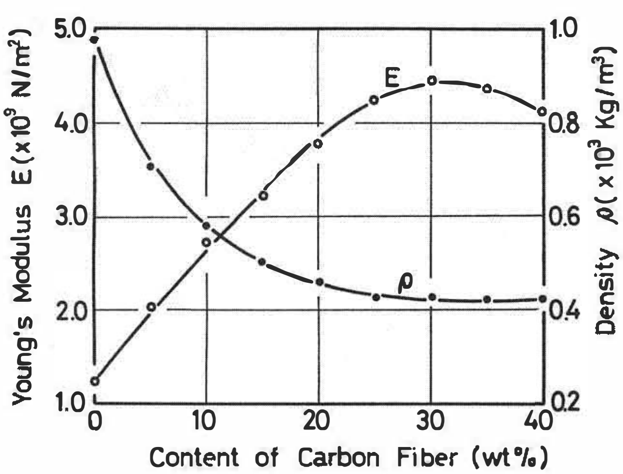

Fig. 16 Relation between Young's modulus and density of CFR-olefin diaphragm, and weight content of carbon fiber

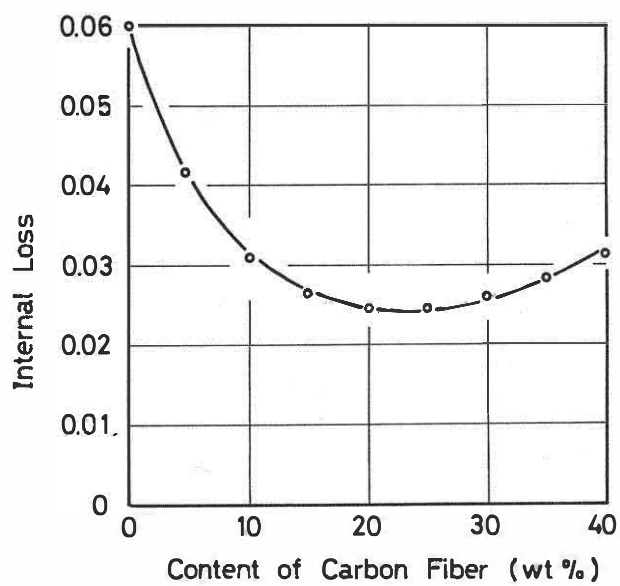

Fig. 17 Relation between internal loss of CFR-olefin diaphragm and weight content of carbon fiber.

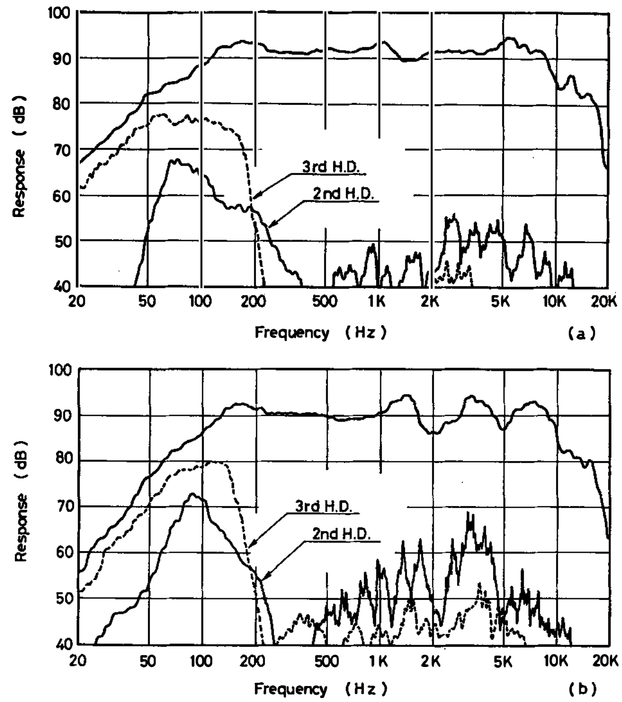

Fig. 18 Comparison of sound pressure level frequency responses of 10cm cone type loudspeakers with CFR-olefin diaphragm and paper cone.

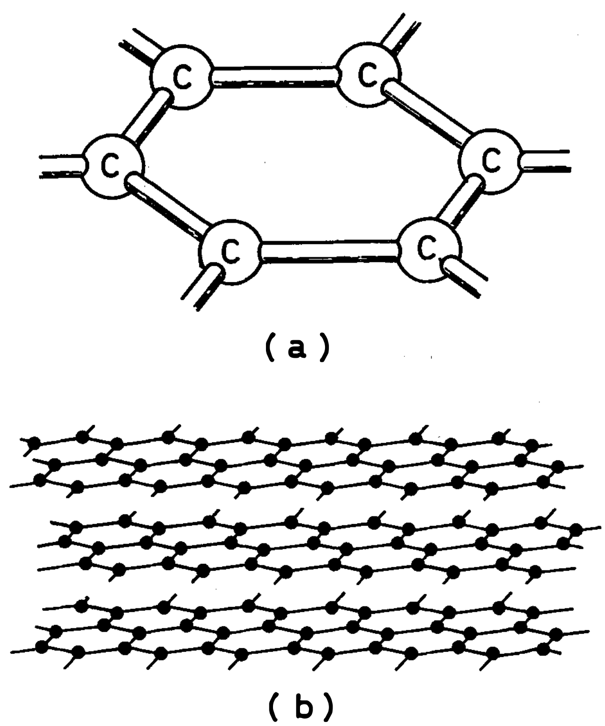

Fig. 19 Schematic models of bonding of carbon atoms (a), and laminar structure of graphite crystal (b).

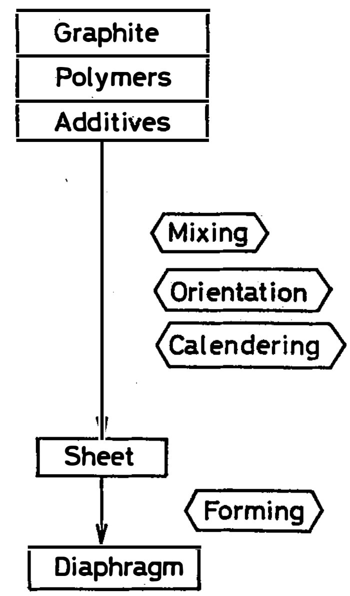

Fig. 20 Flow chart of PG composite diaphragm production.

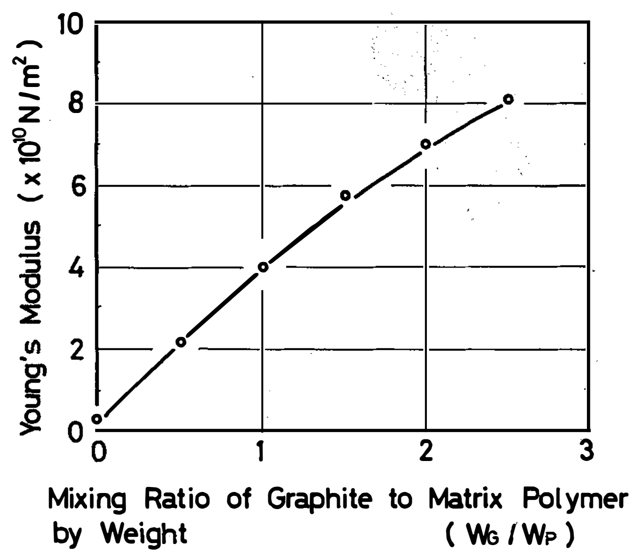

Fig. 21 Relation between Young's modulus of PG composite sheet and mixing ratio of graphite to PVC matrix by weight.

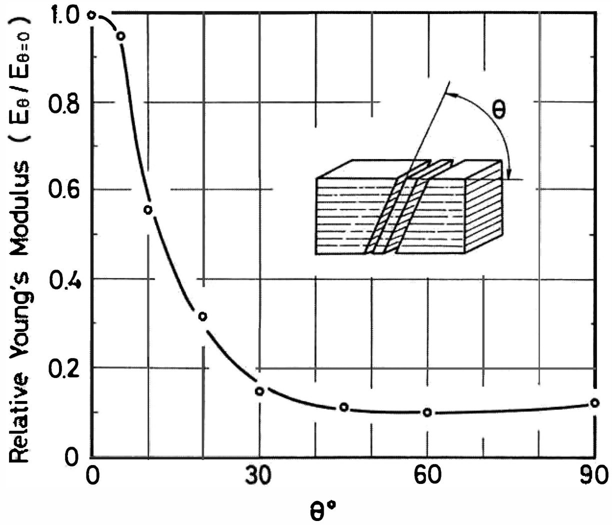

Fig. 22 Variation of Young's modulus by orientation of graphite flakes.

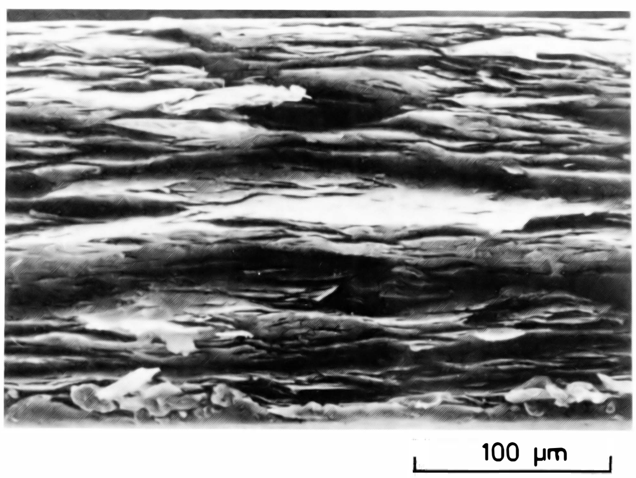

Fig. 23 Scanning electron micrograph of fracture cross section of PG composite sheet.

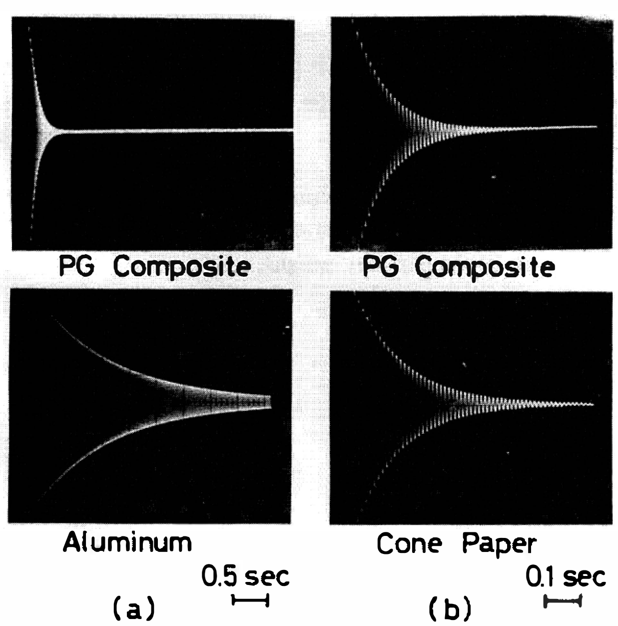

Fig. 24 Comparison of decay patterns of free vibration.
(a) PG composite and aluminum
(b) PG composite and cone paper

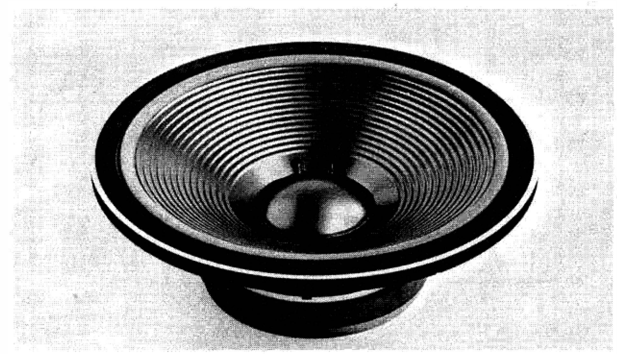

Fig. 25 40cm low frequency loudspeaker with PG composite diaphragm.

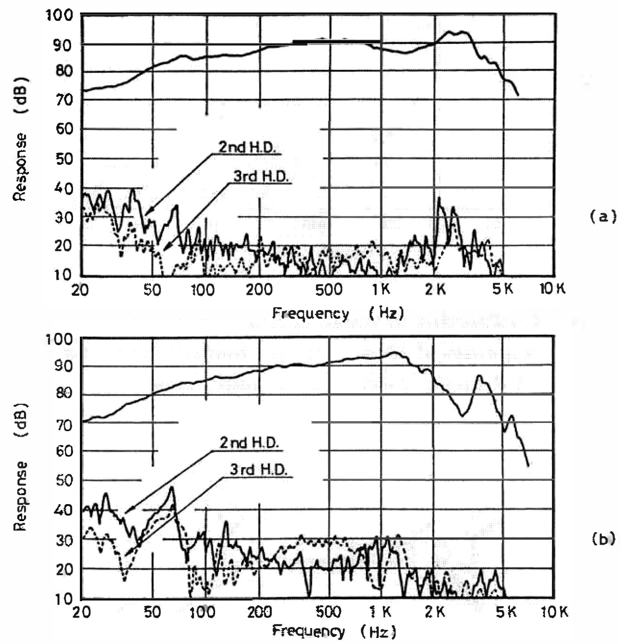

Fig. 26 Comparison of sound pressure level frequency responses of 40cm low frequency loudspeakers with PG composite diaphragm (a), and paper cone (b).

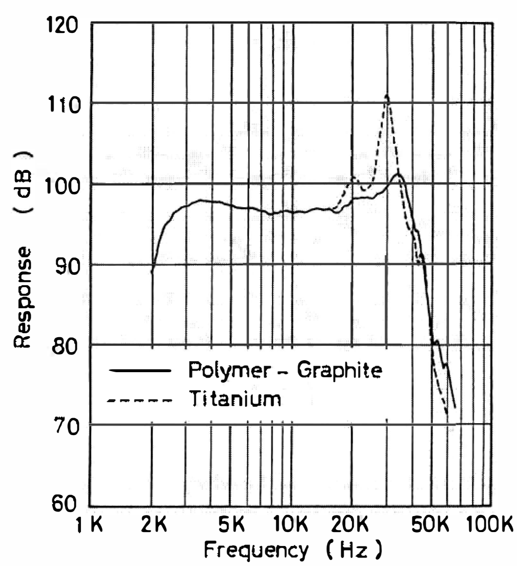

Fig. 27 Comparison of sound pressure level frequency responses of 2.5cm high frequency loudspeakers with PG composite diaphragm (a), and titanium diaphragm (b).