THE PROCESSING AND APPLICATIONS
OF
SINTERED NdFeB-BASED
PERMANENT MAGNETS

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ABSTRACT

Rare earth permanent magnets account for more than 25% of the total world permanent magnet market, valued at over £1000 millions in 1988 and projected to increase at 10% per annum over the coming years.

Since their introduction in 1983, NdFeB - based permanent magnets have rapidly become established as a preferred option in a wide range of permanent magnet devices and applications. This paper outlines the powder processing of these materials and compares and contrasts alternative processing routes. Typical application areas are described and the improvement and future developments of NdFeB - based magnets are considered.

1. INTRODUCTION

The discovery in 1983 of a high performance iron-rich rare earth permanent magnet (REPM) is probably the most significant advance in permanent magnetism since the development of ferrites in the 1950's.

At present, all high performance iron - based REPM's are composed largely of the tetragonal $Nd_2Fe_{14}B$ phase. Since the announcement of permanent magnets based on $Nd_2Fe_{14}B$ by workers of Sumitomo Special Metals [1] and Delco Remy (GM Corporation), [2] the search for alternative phases has been extremely active. Whilst many man-years have been spent on this search, as yet no substitute novel compound has been identified.

Several alternative processing methods have been developed to produce useful permanent magnets from NdFeB - based compositions. These are:

i)   Conventional REPM powder metallurgical processing,

ii) Melt spinning followed by bonding, cold pressing and / or hot pressing,

iii) Hot working of cast alloy.

Subsequent commercialisation around the world has concentrated heavily on the powder metallurgical route, or a variant, and several magnet producers are manufacturing NdFeB magnets, some under licence from Sumitomo Special Metals. Out of the 15 major sintered REPM producers in the western world over 50% have now obtained licences from Sumitomo Special Metals to manufacture and market sintered NdFeB - based permanent magnets. Philips are one of the licenced manufactures.

Permanent magnets today are used in a wide range of industrial, domestic, automotive and aerospace applications. Their special technological importance derives from the ability to act contactlessly on ferromagnetic material, either by attraction or repulsion, and to provide a permanent magnetic flux with no energy input and hence at no operating cost.

Figure 1 shows the total permanent magnet market split by material type. This total market is valued at £1050 millions in 1988. The market, in terms of volume, is totally dominated by the ferrite class of material (>97%). However, in terms of value the rare earth permanent magnet share is significant (>25%).

Two significant areas of market growth have occured. Firstly, the introduction of ferrites in the early 1950's has led to market domination today. Secondly, market penetration by rare earth permanent magnets has been dramatic since their introduction in the early 1970's. The growth rate of the total market is around 7% per annum with the rare earth magnet sector growing in excess of 10% per annum. It is predicted that future usage of permanent magnets will expand faster than the historical rate, with ferrite and rare earth permanent magnets being the dominant materials.

The improvement in primary magnetic properties of the NdFe - based magnets over the existing high energy SmCo - based magnets is illustrated in Table 1.
As a comparison, the properties of a typical ferrite permanent magnet, the type of permanent magnet material produced in the largest tonnage, are included in the table. Of course, not only performance of the permanent magnet material but also the costs of production determine their relative market shares. Figure 2 shows the relative production costs of typical magnet geometries for four rare earth materials and a ferrite magnet. The comparison is made on the basis of unit mass. As can be seen the ferrite material is an order of magnitude cheaper than the rare earth materials.

A similar cost comparison is given in Figure 3 this time on the basis of unit maximum energy product. In this case sintered NdFeB is much closer to ferrite. Interestingly the cost / performance ratio for bonded NdFeB is rather poor in this comparison. This suggests that applications for this type of material will be only were benefit can be gained from the special forming possibilities of injection moulding or where thin magnet sections are required.

Philips Components Limited manufacture a wide range of REPM grades based on either the $Nd_2Fe_{14}B$ system or $(NyDy)_2Fe_{14}BNb$ system and marketed under the trade name Neodure. Table 2 illustrates, the typical magnetic characteristics of the Neodure grades currently available.

2. MICROSTRUCTURE

The important properties characterising a permanent magnet component are:

Remanence, Br,
Polarization coercivity, HcJ,
Maximum energy product, (BH) max,
and,
Temperature dependence of the Br and HcJ, Tk

All these parameters are largely influenced by the microstructure and phase chemistry. Hence the processing route used to fabricate the permanent magnet is critical in determining the final magnetic performance.

The requirements of a rare earth transition metal compound to be a potential permanent magnet material are:

i)   The compound must be ferro-or ferri-magnetic with a high Curie temperature, Tc,

ii) room temperature magnetisation, Ms, must be high

and

iii) the compound must have a large magnetocrystalline anisotropy, HA, with a magnetically unique crystallographic axis.

The relationship between these so called primary
magnetic properties and the characteristic parameters
of a permanent magnet, the secondary properties, is
shown diagramatically in Figure 4. To date powder
metallurgical processing has yielded the optimum
microstructure resulting in the best translation from
primary to secondary magnetic properties.

3. THE POWDER METALLURGICAL PROCESSING OF RARE EARTH PERMANENT MAGNETS.

The processing of the various types of rare earth
permanent magnet materials can be carried out by a
broadly similar powder metallurgical process. In this
section the process steps are outlined.

The high reactivity of the rare earths and their alloys
and the critical dependence of the magnetic properties
on the chemical composition requires the effective
suppression of contamination during the alloy
preparation and subsequent powder metallurgical
processing.

In particular oxidation of the rare earth components by
O_2 / H_2O must be kept to a minimum through all fine
powder handling and heat treatment stages.

The main process steps taken during the production of
REPM's are shown in Figure 5. The general process
consists of alloy preparation, pre-milling, milling,
composition control and adjustment, particle alignment
and pressing, sintering and heat treatment, machining,
coating and finally magnetising.

3.1 ALLOY PREPARATION

Rare earth - 3d transition metal alloys can be produced
by two methods on an industrial scale. These are
vacuum induction melting and calciothermic reduction.

The vacuum induction method has the advantage that it
can produce a wide range of rare earth alloy
compositions with very low oxygen contents (<200 ppm).

The calciothermic production of rare earth alloys was developed independently by two groups working at General Electric, U.S. (3) and Th. Goldschmidt, W. Germany (4). In the General Electric process cobalt powder, calcium granules and rare earth oxide powder are blended together and then reacted under hydrogen at 1423K. The reaction can be represented by the equation:

$$R_2O_3 + 10Co + 3Ca \xrightarrow[\text{3 hours}]{1423K, H_2} 2RCo_5 + 3CaO$$
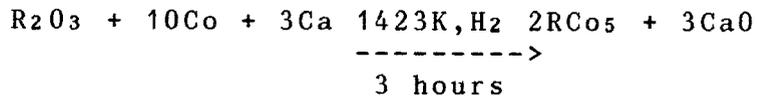
After cooling, the excess calcium and calcium oxide are removed from the reacted product by reacting with moist nitrogen, then washing with water and dilute acid. This process is known as the reduction - diffusion (R - D) process.

The Goldschmidt process, known as co-reduction, is a variation of the R - D process in that the reaction is carried out under vacuum at 1273K and both cobalt powder and cobalt oxide powder are used as raw materials.

The main advantages of the calciothermic reduction process are the use of rare earth oxide as raw material and the direct production of alloy powder suitable for milling. However, both the oxygen content and calcium content are higher than alloys prepared by melting.

3.2 PRE-MILLING

Depending on the method used to prepare the alloy, the material may require a size reduction stage prior to final milling. For example, after vacuum melting and casting, the NdFeB alloy is in the form of chill cast lumps. These are crushed, under a nitrogen atmosphere in a high energy hammer mill, to a particle size range <500 micron.

An alternative chemical method of pre-milling the alloy has been developed in co-operation with the group of Prof. Rex Harris at the University of Birmingham. This process is known as hydrogen decrepitation.

It has been shown that NdFeB alloys react readily with hydrogen at moderate pressures exhibiting a strongly exothermic reaction (5). Measurements of the desorption behaviour of hydrogen from a $Nd_{16}Fe_{76}B_8$ alloy indicated the vacuum degassing consisted of two stages whereby hydrogen was first desorbed from the $Nd_2Fe_{14}B$ matrix phase below 300°C with the remainder being evolved from the Nd rich phase at higher temperatures (6).

The formation of two hydrides is consistent with the observed decrepitation behaviour where the initial activation process corresponds with the hydriding of the intergranular Neodymium - rich material resulting in transgranular fracture. This is followed by the hydriding of the matrix phase with the attendant transcrystalline cracking of the individual crystallites.

This technique of hydrogen decrepitation is exploited to produce a friable pre-milled material with a particle size less than 500 microns directly suitable as input material for the fine milling stage.

3.3 MILLING

The object of milling the rare earth permanent magnet alloys is to produce a narrow size distribution of single crystal particles, i.e. individual particles containing no grain boundaries and therefore only one preferred axis of magnetisation. In addition, sufficient particle surface area must be present for high sinter reactivity.

For single phase magnets, where the coercivity is controlled by domain nucleation and wall pinning at grain boundaries, the particle size and particle surface condition play a critical role in determining the coercivity of the sintered magnet. The critical parameters to be controlled during milling are particle size, particle size distribution, damage to crystal structure and oxidation.

The milling process can be carried out by either ball milling in an organic liquid under an inert gas e.g. attritor milling in cyclohexane, or by jet milling. The jet mill is a dry milling process in which the attrition action is mainly due to high velocity particle - particle collision.

3.4 COMPOSITION CONTROL AND ADJUSTMENT

As previously mentioned the magnetic properties of rare
earth permanent magnets are critically dependent on the
chemical composition. Any contamination, particularly
oxidation, of the alloy occurring during processing
depletes the alloy of the rare earth components. This
results in a shift in composition to the T - rich side
of the phase diagram, which may cause a magnetically
unfavourable phase distribution.

For NdFeB the composition range for optimum magnetic
properties is less critical than that for $SmCo_5$. The
critical R content for NdFeB magnets is about a factor
of 10 greater than the critical range for $SmCo_5$
magnets. This enables NdFeB magnets, with careful
processing control, to be prepared by the single powder
method.

3.5 PARTICLE ALIGNMENT AND PRESSING

To obtain a powder compact with maximum magnetisation
the powder particles are magnetically aligned and
pressed such that the easy axes of magnetisation of the
particles are parallel.

The powder compaction is performed by die pressing or
by isostatic pressing. In the first method, the
aligning magnetic field is set up in the cavity of a
non magnetic die with its axis lying either in the
direction of pressing or at right angles.

In large scale production multi-impression tooling is
generally used. High homogeneous field levels are
required to produce a high level of uniformly aligned
particles.

The applied field can be D.C. or some combination of
D.C. and pulse. The degree of alignment is influenced
by particle shape and the particle size distribution,
magnitude of aligning field and pressing pressure. The
pressing pressure should be sufficient to give the
powder compact enough mechanical strength to withstand
handling but not high enough to cause particle
misorientation.

Isostatic pressing is normally carried out on powders
pre aligned in a pulsed magnetic field of a level 3 or
4 times that used in uniaxial die pressing.

This improves the degree of particle alignment, which
is then maintained during isostatic pressing and
results in higher Br and (BH) max values than for die
pressed pieces.

3.6 SINTERING AND HEAT TREATMENT

The sintering of rare earth permanent magnets is
carried out in inert gas atmospheres, reducing
atmospheres or under vacuum.
The sintering treatment should result in a magnet with
a high density, and for magnets with a nucleation and
grain boundary pinning coercivity mechanism, no
appreciable grain growth.

A constant and well defined sintering temperature is
necessary in order to ensure the magnet has no open
porosity which could lead to oxidation and ageing
during use. This requires the density to be greater
than 95% of theoretical density.

3.7 MACHINING

During the sintering operation the pressed product
volume reduces to the final magnet body. This
shrinkage depends upon production factors and the
final magnet shape and size. This results in some
variation in magnet size and therefore a machining
operation is necessary.

Rare earth permanent magnets are in general hard and
brittle, although NdFeB magnets are rather tougher and
less susceptible to breakage and chipping.

Magnetic chucks are not generally used to hold pieces
down directly. Small series items are fastened by
special adhesives to steel backing plates, then ground
on conventional grinding machines fitted with either
silicon carbide or diamond grinding wheels.

Large series production is ground on double disc
machines where the pieces are moved between two
grinding wheels set the required distance apart. Small
blocks tend to be slit using diamond impregnated
wheels.

Machined surfaces are required either to give the
required magnetic contact with the associated
components in the final assembly or for dimensional
reasons.

3.8 COATING

Magnets based on the $Nd_2Fe_{14}B$ intermetallic are
susceptible to corrosion and require a corrosion
resistant coating for normal applications. The optimum
coating and coating technology has to fulfil several
criteria:

i)   The coating must be thin to minimise air gaps in
     the magnetic circuit.

ii) The coating must be uniform and provide complete
     coverage of all magnet surfaces.

iii) The coating must be glueable to other parts of the
     magnetic circuit.

iv) The coating technology must evolve little or no
     hydrogen.

v)   The coating technology has to be applicable to a
     wide range of magnet shapes and sizes.

Philips have invested several man-years of research and
development effort focussed on identifying an optimum
coating and applying the technology to Neodure magnets.

The conventional techniques of electro-, electroless
and immersion plating of metallic layers e.g. Sn, Ni,
Cu, duplex systems, on Neodure were all found to be not
applicable or shown not to meet our corrosion
reliability requirements. In addition a range of
physical vapour deposition e.g. sputtering, thermal
evaporation, and chemical vapour deposition processes
were studied.

The outcome of these extensive investigations has led
to the application of two coating processes to
Neodure. These are:-

i)   Electropaint (epoxy) deposited by a cathodic
        process. This is a multi-stage process consisting
        of surface cleaning and pretreatment, cathodic
        electropainting, washing and stoving steps. A
        coating thickness of 23 ± 5 microns is obtained
        from this process, which can only be applied to
        jigged components. This process is more suited to
        large products were the cost of individually
        jigging components. is not prohibitive.

ii)   Aluminium deposited by an ion vapour deposited
        (IVD) process. This multi-stage process consists
        of surface cleaning and pre-treatment, plasma
        cleaning and ion deposition under vacuum, post
        coating treatment and chemical conversion steps.
        Products can either be barrel or jig coated and a
        coating thickness of 10 ± 3 microns results. This
        process is applicable to all component sizes.

Philips Components are the only European manufacture to
currently provide large production series Neodure
magnets coated with IVD aluminium.

The IVD aluminium process was originally developed by
the aerospace industry to provide a solution to the
challenge of protecting their products and components
from corrosion and its deleterious effects. The
application of IVD aluminium to Neodure has been
optimized by engineers at Philips Components. In
particular product specific surface pre-treatment, post
treatment and chemical conversion processes have all
been developed to provide an environmentally clean and
high performance corrosion protection finish.

Aluminium is particularly suited as a protective layer
for Neodure since they both exhibit similar
electropotentials.

Hence, small surface defects do not result in
accelerated galvanic corrosion of the substrate as is
the case with other dissimilar metallic layers e.g. Sn,
Ni, Cu. In fact the extremely rapid and stable oxide
formation typical of aluminium, promotes the sealing of
any small surface scratches in the coating.

3.9 MAGNETISATION

Since most rare earth permanent magnets combine relative recoil permeabilities close to unity and high coercivity, then, they may be magnetised prior to assembly without flux loss.

However, because of the difficulty of handling magnetised and brittle material it is usual for the user to carry our magnetisation during the system assembly.

4. APPLICATIONS

The earliest commercial applications of rare earth magnets were in stepper motors for electronic watches, in the replacement of AlNiCo magnets in electron beam focussing systems such as travelling wave tubes and in some medical applications. Subsequent applications made greater use of the potential in weight and volume reduction made possible by the high magnetic energy available.

Typical new applications were in magnetic bearings, servomotors, switches and actuators. The real breakthrough happened after 1975 when many new types of motor, generator, couplings, etc, were designed to use rare earth permanent magnets. Over the past few years, the application of rare earth permanent magnets, particularly in the Far east, in audio-visual and other consumer products has started to open new market opportunities.

Recently, a new market opportunity has developed due to the growth in Information Technology. Disk file systems have been developed which utilise a voice coil motor actuator to position the read /write head over the hard disk. The use of rare earth permanent magnets in these devices has enabled access times to be dramatically reduced.

It is impossible here to describe in detail all the many applications in which rare earth permanent magnets are expected to provide an innovative edge. The major applications are listed in Table 3 and characterized by their dependence on flux density B.

5. CONCLUSION

The powder metallurgical processing of REPM's produces materials with the highest known magnetic properties. At their present stage of development NdFeB magnets are distinctly superior to SmCo materials in terms of their performance at room temperature. However, at temperatures in excess of 150°C they are inferior.

In order to ensure NdFeB magnets find applications in as many fields of modern technology as possible their operating temperature must be extended to 200°C without risk of demagnetisation.

Improvements in the temperature dependence of magnetic properties requires both an increase in the Curie temperature and improved thermal stability of the coercivity.

The improvement in temperature stability of Neodure magnets is illustrated in Figures 6,7 & 8. These show the thermally reversible demagnetisation characteristics for the first generation ternary RES 270 grade, the multicomponent RES255 grade and the latest high temperature RES257 grade. With these developments maximum continuous operating temperatures have been increased from around 100°C upto 180°C.

Corrosion and oxidation problems are mainly associated with the free Nd present in the grain boundary eutectic phase. It may be possible to reduce the amount of free Nd by the addition of Co or a heavy rare earth which forms a rare earth compound in favour of free Nd in the eutectic phase. However, currently available coatings e.g. Ion vapour deposited aluminium, have guaranteed performance in excess of 200°C.

Finally, increase usage of all REPM's will be achieved by the economics of reducing manufacturing costs during powder metallurgical processing. Since all rare earth permanent magnets exhibit high values of HcJ this results in magnetic circuit design requiring components with short magnetic axes. Such components present particular problems in minimising grinding allowances. This requires the careful control of pressing conditions and powder dosing in the die cavities, the minimisation of distortion during sintering and heat treatment and the choice of suitable finishing methods. Similarly, because of the high raw material costs, the process control at all process steps must be such that a zero defect failure rate is guaranteed.

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WORLD PERMANENT MAGNET MARKET
1988
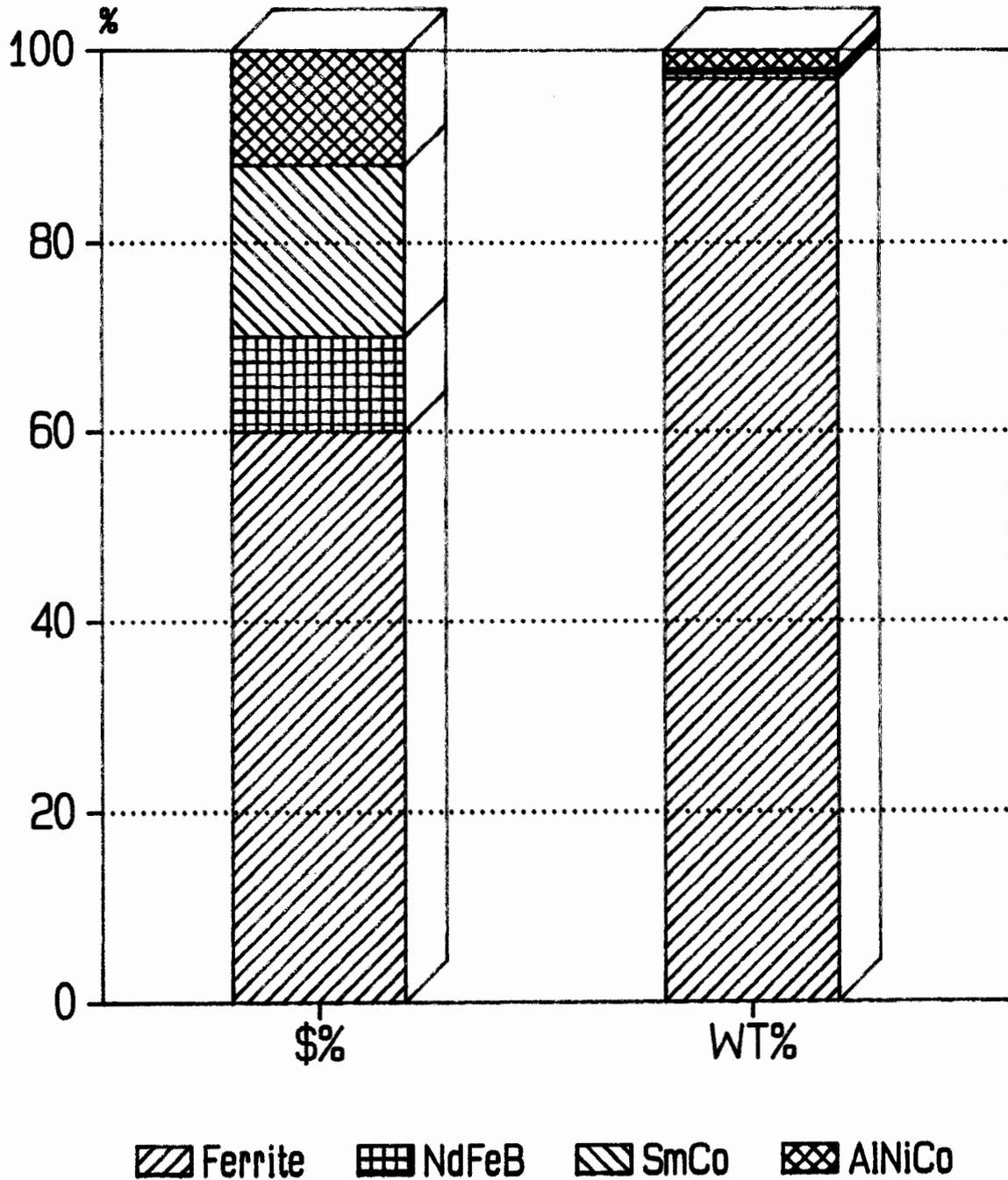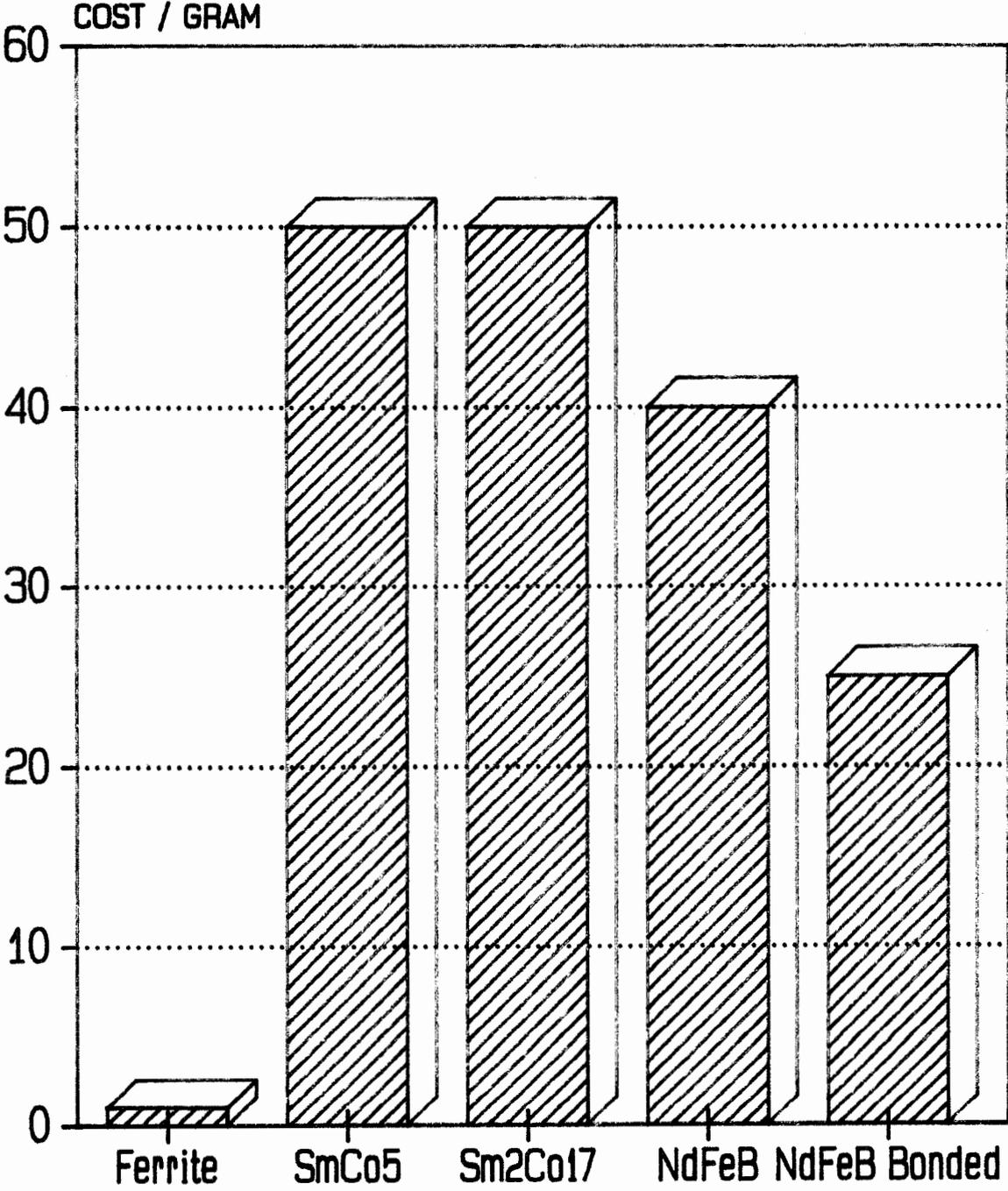

FIGURE 1

COST COMPARISON OF VARIOUS PERMANENT MAGNETS
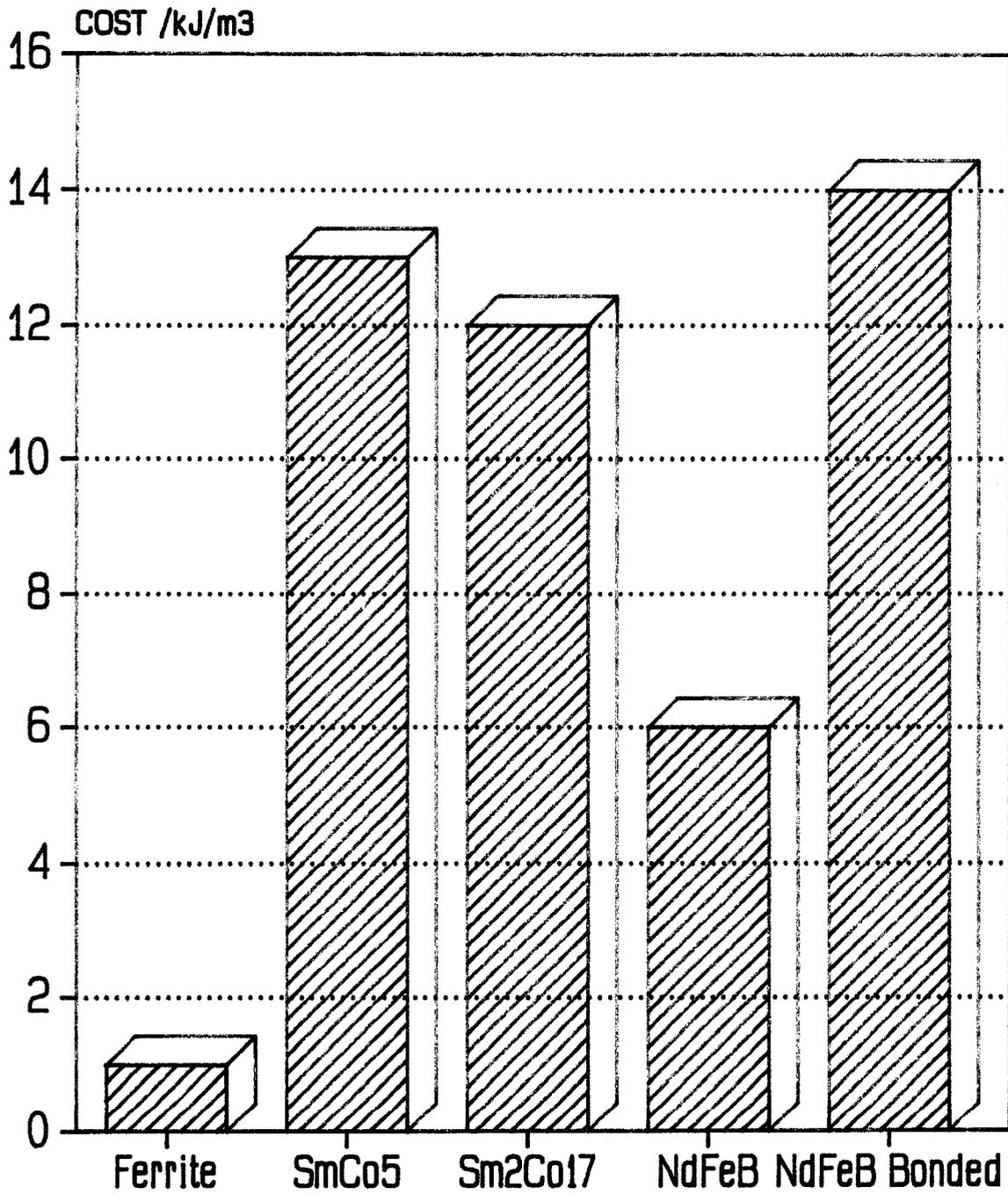

FIGURE 2

COST COMPARISON OF VARIOUS
PERMANENT MAGNETS

FIGURE 3
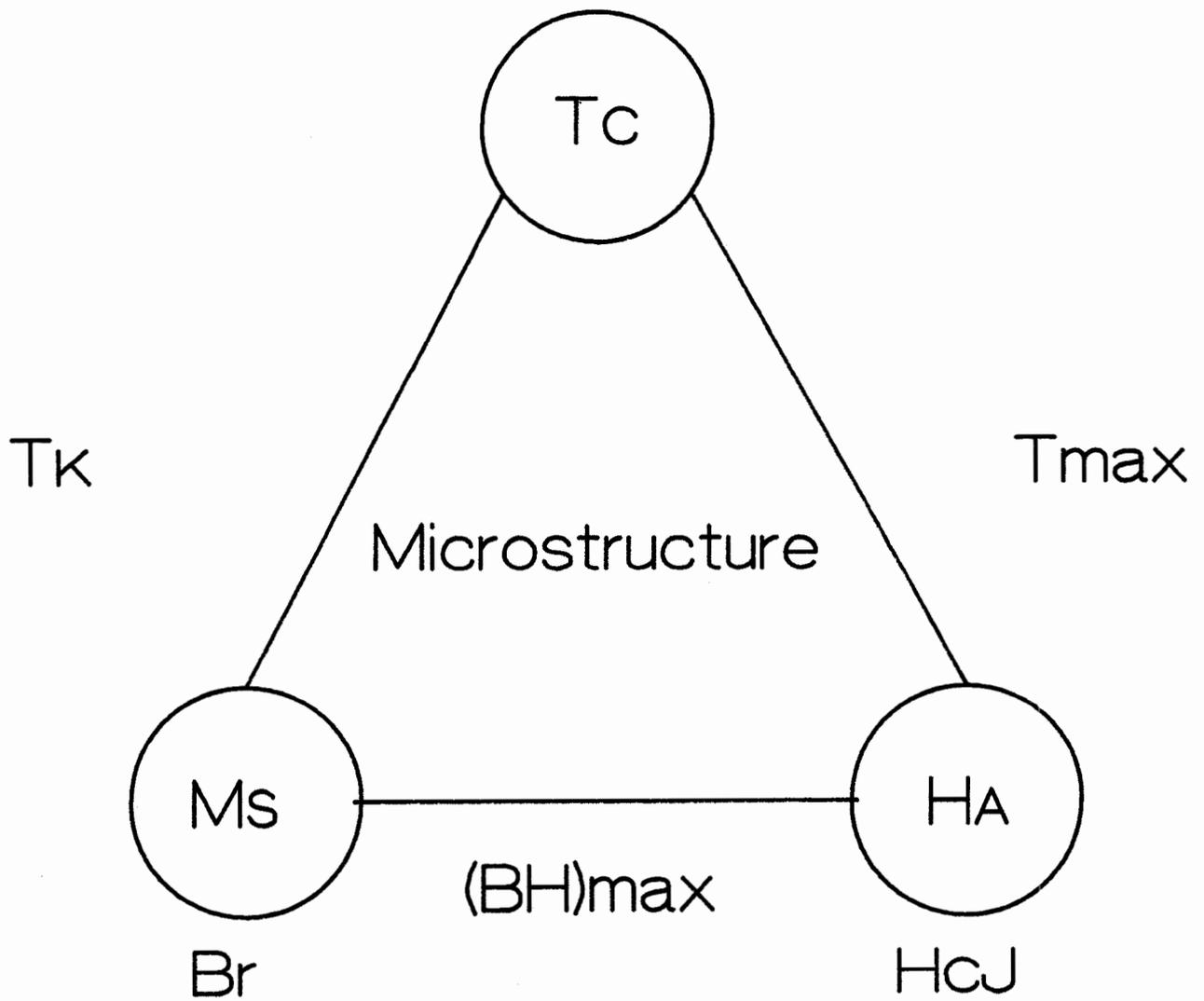
FIGURE 4

THE REPM MANUFACTURING PROCESS

Nd-Fe-B

Nd-Fe-B cast alloy

Decrepitation

Jet milling

SmCo

Co-reduced powder

Milling

Blending

Pressing and particle alignment

Vacuum sintering and heat treatment

Machining

Coating/Assembly

Inspection, magnetising and packing

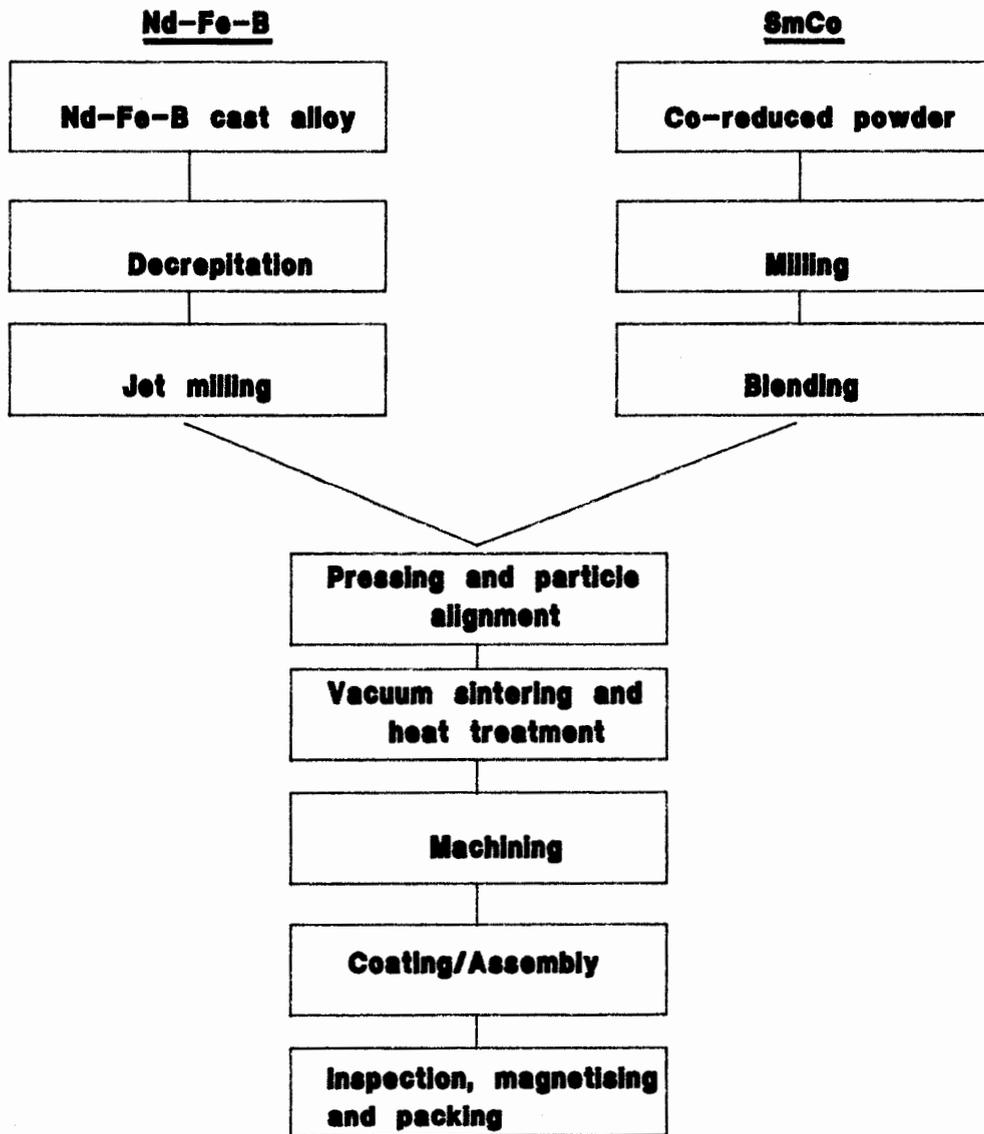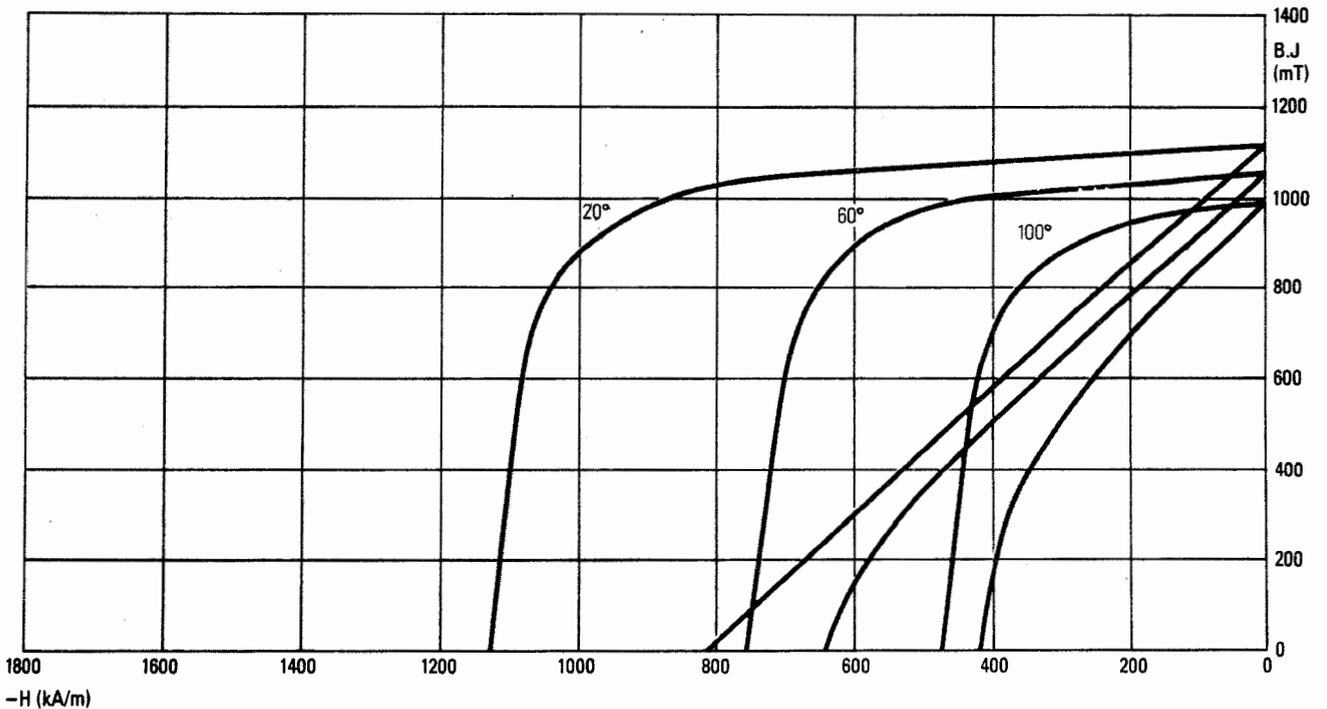**FIGURE 5**

Philips Components

RES 270 NEODURE
RARE EARTH
MAGNETS

NEODURE* sintered magnets are based on a compound of neodymium, iron and boron (Nd-Fe-B). RES270 is a specific grade within a range of NEODURE materials.

DEMAGNETISATION CURVES

Typical characteristics at 25°C

		Typ.	Min
REMANENCE	Br	1100mT	1050mT
COERCIVITY	HcB	700 kA/m	650 kA/m
INTRINSIC COERCIVITY	HcJ	1000 kA/m	750 kA/m
MAXIMUM ENERGY PRODUCT	(BH) Max	215 kJ/m^3	
		(27 MGOe)	

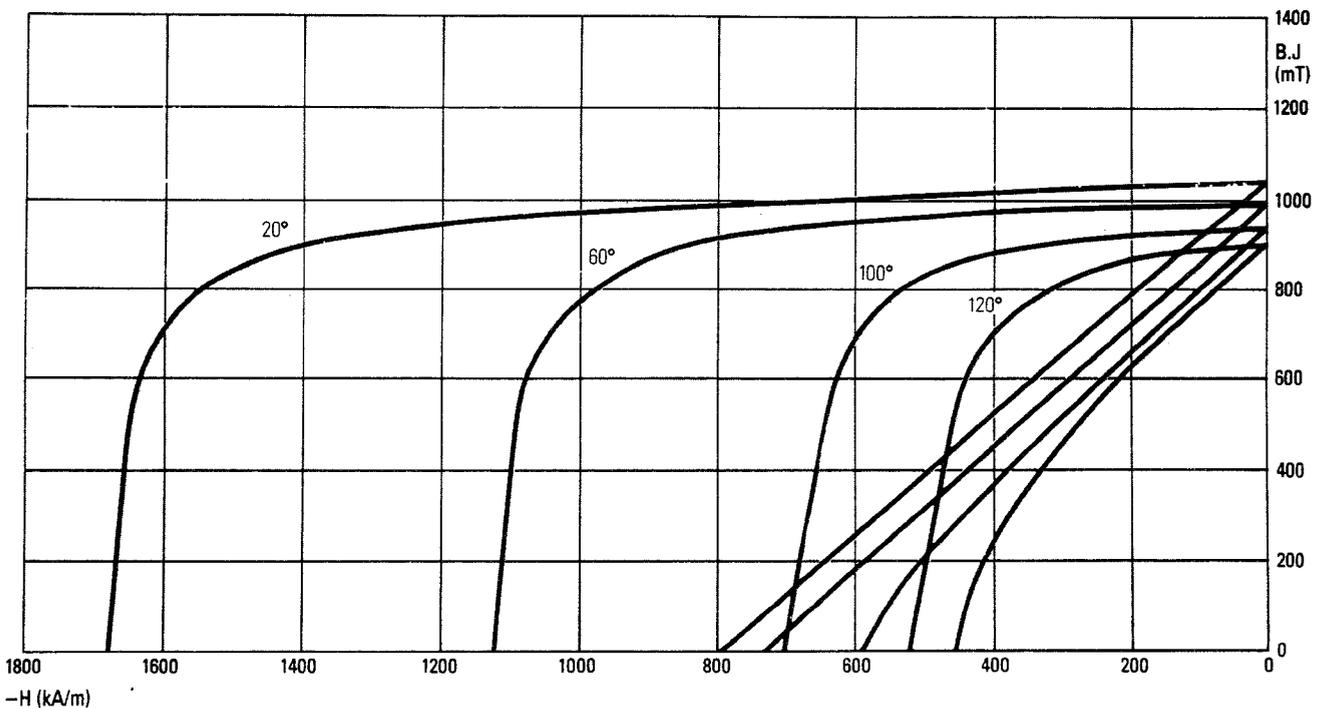*Manufactured under license to SUMITOMO SPECIAL METALS.

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FIGURE 6

Philips Components

RES 255 NEODURE
RARE EARTH
MAGNETS

NEODURE* sintered magnets are based on a compound of neodymium, iron and boron (Nd-Fe-B). RES255 is a specific grade within a range of NEODURE materials.

DEMAGNETISATION CURVES

Typical characteristics at 25°C

		Typ.	Min
REMANENCE	Br	1050mT	1000mT
COERCIVITY	HcB	750 kA/m	700 kA/m
INTRINSIC COERCIVITY	HcJ	1500 kA/m	1200 kA/m
MAXIMUM ENERGY PRODUCT	(BH) Max	200 kJ/m³	
		(25 MGOe)	

*Manufactured under license to SUMITOMO SPECIAL METALS.

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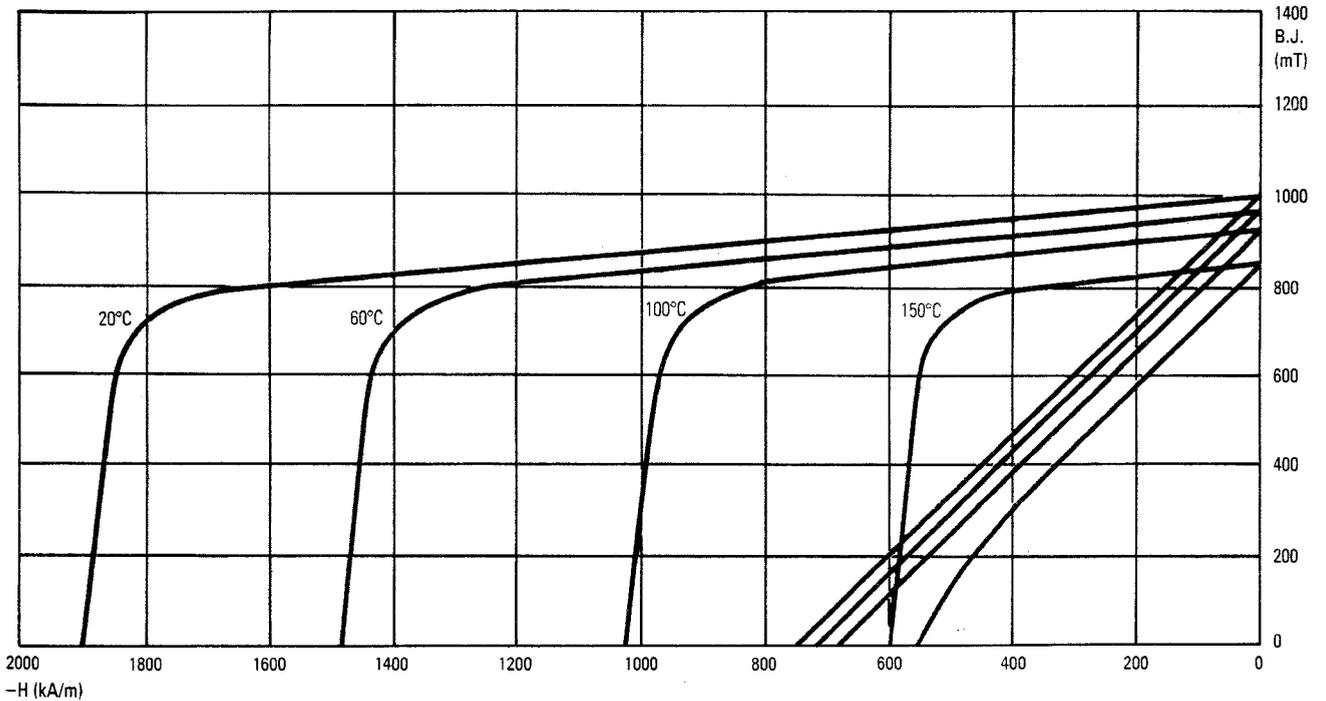FIGURE 7

Philips Components

RES 257 NEODURE
RARE EARTH
MAGNETS

NEODURE* sintered magnets are based on a compound of neodymium, iron and boron (Nd-Fe-B) RES257 is a specific grade within a range of NEODURE materials.

DEMAGNETISATION CURVES

Typical characteristics at 20°C

		Typ.	Min
REMANENCE	Br	1000mT	950mT
COERCIVITY	HcB	750 kA/m	700 kA/m
INTRINSIC COERCIVITY	HcJ	1900 kA/m	1800 kA/m
MAXIMUM ENERGY PRODUCT	(BH) Max	188 kJ/m³	
		(23.5 MG Oe)	

* Manufactured under license to SUMITOMO SPECIAL METALS.

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FIGURE 8

EXAMPLES OF APPLICATIONS

APPLICATION GROUP	EXAMPLES	SYSTEM FUNCTION PROPORTIONAL TO
ACOUSTIC TRANSDUCERS	LOUDSPEAKERS HEADPHONES TELEPHONE RECEIVERS MICROPHONES PICKUPS	B
PERMANENT MAGNET MOTORS & GENERATORS, ELECTRO-MECHANICAL TRANSDUCERS	D.C. MOTORS E.C. MOTORS SYNCHRONOUS MOTORS STEPPING MOTORS GENERATORS LINEAR MOTORS MOVING-COIL MOTORS TORQUE TRANSMITTERS	B
MAGNETO-MECHANICS	COUPLINGS SEPARATORS ATTACHMENT SYSTEMS MAGNETIC CONVEYORS BEARINGS TRANSPORT SYSTEMS	B^2
MAGNETIC FIELD AND FOCUSSING SYSTEMS	DIPOLES QUADRUPOLES HEXAPOLES UNDULATORS WIGGLERS CIRCULATORS MASS SPECTROMETERS NMR SYSTEM	B

TABLE 3

STANDARD NEODURE GRADES

GRADE	REMANENCE Br	COERCIVITY		MAXIMUM BH PRODUCT (BH) MAX (kJ/m3)
		HcB(kA/m)	HcJ(kA/m)	
RES 270	1.10	700	1000	215
RES 300	1.15	750	1000	240
RES 350	1.20	800	1000	280
RES 255	1.05	750	1500	280
RES 275	1.10	800	1500	215
RES 305	1.15	850	1500	240
RES 257	1.00	750	1900	190

TABLE 2

COMPARISON OF VARIOUS PERMANENT MAGNET MATERIALS

COMPOSITION	Nd2Fe14B	SmCo5	SrFe12O19	
MATERIAL GRADE	RES270	RES190	FXD380	
Br typical	1.1	0.89	0.39	T
BH max typical	215	154	28.2	kJ/m3
HcJ typical	1000	1100	275	kA/m
Temperature coefficient of Br (20 to + 150°C)	-0.13	-0.04	-0.2	%/K
Temperature coefficient of HcJ (20 to 150°C)	-0.6	-0.05	0.34	%/K
Recoil permeability	1.05	1.05	1.1	
Curie point	310	720	450	°C
Max continuous operating temperature.	120	250	350	°C
Density	7.4	8.3	4.75	x10 3 kg/m3

TABLE 1