structural factors controlling crack formation. For example, the use of single phase bi- and tri-crystals having well-defined geometries is recommended for studies of intergranular crack nucleation, growth and interconnection when the constituent crystals are behaving in a nonlinear viscous manner. Similarily, for studies of crack formation at second phase particles such as inclusions, specimens containing a single particle in a mono- or bi-crystal matrix should be useful.

Finally, the representation of the failure behavior in $\dot{\epsilon}-T-\epsilon$ space may be a useful adjunct to the framework within which a general theory of failure at elevated temperatures can be developed.

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Preparation of $SmCo_5$ Permanent Magnets

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The technology of making $SmCo_5$ magnets with energy products of 20×10^6 GOe is described. This technology consists of the preparation of a powder by grinding, aligning the powder particles in a magnetic field, and cold pressing by applying a uniaxial deformation at 20 kbar. The permanent magnetic properties of the $SmCo_5$ powders are shown to vary appreciably with various applied chemical treatments and with the composition of the alloy. The intrinsic coercive force appears to decrease with time but remains so large that the demagnetization of the induction is hardly affected. Some properties of the magnets are discussed.

INTRODUCTION

Recently the possibility of making permanent magnets from RCo_5 compounds (R = Rare earth) has been recognized.[1,2] These compounds crystallize in the hexagonal $CaZn_5$ structure. After grinding and etching some of them possess a very high coercive force, combined with a high saturation magnetization. To get a high remanence and BH_{max}, it is necessary to compact the obtained powder; the permanent magnet properties are also strongly dependent on the alignment of the powder particles. Hence one needs a process that gives the material a high density without affecting the orientation of the particles. With conventional pressing techniques relative densities of approximately 85% can be achieved; by applying uniaxial deformation at a pressure of 20 kbar, however, we were able to obtain a relative density of 95%, without destroying the orientation; energy products of 20×10^6 GOe were attained.[3,4] The present paper gives some details of the preparation of the powders and of the compacting process.

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[1] K. Strnat, G. Hoffer, J. Olson, W. Ostertag, and J. J. Becker, J. Appl. Phys. **38**, 1001 (1967).

[2] W. A. J. J. Velge and K. H. J. Buschow, J. Appl. Phys. **39**, 1717 (1968); see also Z. Angew. Phys. **26**, 157 (1969).

[3] K. H. J. Buschow, W. Luiten, P. A. Naastepad, and F. F. Westendorp, Philips Tech. Rev. **29**, No. 11, 336 (1968).

[4] F. F. Westendorp and K. H. J. Buschow, Solid State Commun. **7**, 639 (1969).

PREPARATION OF $SmCo_5$ POWDERS

The $SmCo_5$ intermetallic compound was melted in an arc furnace or a ceramic crucible by means of inductive heating. Some difficulties arise due to samarium evaporation during melting so that, if the true stoichiometric compound $SmCo_5$ is required, it is necessary to use a slight excess of Sm. For each sample we determined the lattice constants and estimated the composition by interpolation between the lattice constants of $SmCo_5$ and $SmCo_6$.[5]

For the introduction of high coercive forces the material can be ground in a mortar, in a ball mill, or by vibration milling using little glass balls, etc. Some results are shown in Fig. 1. For all powders we observed a pronounced decrease of the $_IH_c$ with time after these treatments. In our experiments to impede this "aging" of the $_IH_c$ we tried to stabilize the material by electroless nickel plating. Surprisingly, we observed a pronounced increase of the $_IH_c$ by this treatment, e.g., from 11 000 to 22 000 Oe. It was found that various etching treatments in acids had a similar effect. In Fig. 1 the increase by etching in a saturated solution of citric acid in water at 80°C is indicated. It can be seen that the increase of the $_IH_c$ is of the same order-of-magnitude as observed for

[5] K. H. J. Buschow and A. S. van der Goot, J. Less-Common Metals **14**, 323 (1968).

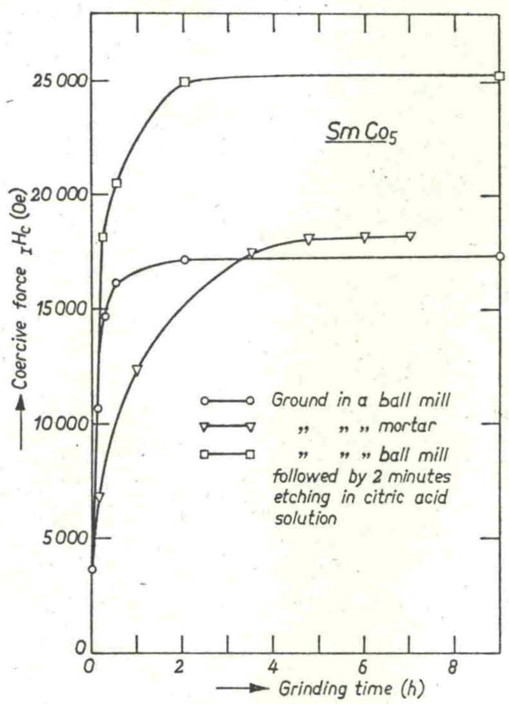

FIG. 1. Coercive force after various grinding and etching treatments.

electroless nickel plating. Actually, the plating is carried out in a weakly acid solution ($p\text{H}\approx 4.5$), so that etching may not be excluded. These observations strongly suggest that the coercivity is determined by surface nucleation of Bloch walls. Other experiments, however, indicate that the demagnetization curve is rather determined by wall pinning.[6] The aging phenomenon reported above is shown in Fig. 2 for 200C°. An important fact is that the $_IH_c$ remains nearly constant after a steep initial decrease. At room temperature aging proceeds much slower. It is possible to stabilize a magnet by heating it above the temperature at which it is used.

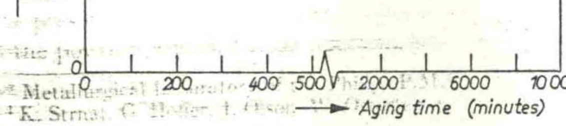

FIG. 2. "Aging" of $SmCo_5$.

[6] F. F. Westendrp and H. Zijlstra Solid state Commun. **7**, 857 (1969).

TABLE I. Remanence σ_R and saturation magnetization σ_s ($\text{emu}\cdot\text{g}^{-1}$), at various stages of the pressing process.

	σ_R	σ_s(16 000 Oe)
Powder	93	93
Green pill	85	87
Pressed 10 kbar	85	87
20 kbar	83	85.5
To 95% density	83	85.5

The saturation magnetization of the powder, and hence the remanence of the magnets, can be enhanced by increasing the Co content of the material.[4] The $SmCo_5$ compound has a homogeneity region which extends primarily to the higher Co concentration.[5] For a compound $SmCo_{5+x}$ $(0\leq x<1)$ the saturation magnetization will increase when x becomes larger because the phase becomes richer in Co and the moment of each Co atom increases. This latter fact is due to a decrease in electron concentration. The $_IH_c$ decreases somewhat at higher Co contents: For $x=1$ the $_IH_c$ is about 40% lower than for $x=0$.

ORIENTING AND HYDROSTATIC PRESSING

Generally the $SmCo_5$ powder can be oriented almost ideally using rather low fields when immersed in a liquid (e.g., paraffin). Actually this is done for the measurements of the properties of the powder. The orientation of the powder particles in a magnet is less perfect due to the friction between the particles during the pressing process. The first step in this process consists of making a green pill in a magnetic field. The powder is contained in a small die, which is placed between the poles of an electromagnet, giving a field of 30 kOe. The powder is moderately compressed subsequently by a piston. After this the pill is removed from the die, packed in a rubber container, and subjected to a hydrostatic pressure up to 20 kbar; thus a compact body is obtained ready for further processing. The density obtained by this hydrostatic pressing as a

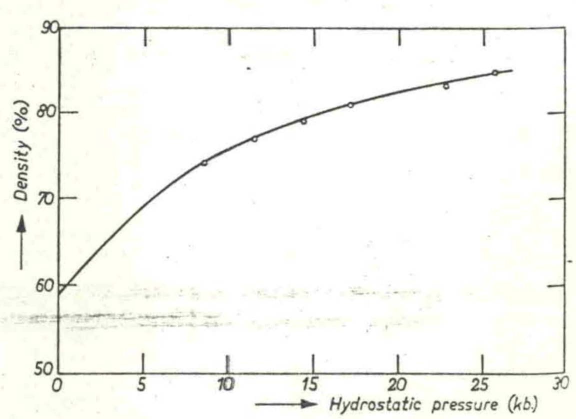

FIG. 3. Density of hydrostatic pressed $SmCo_5$.

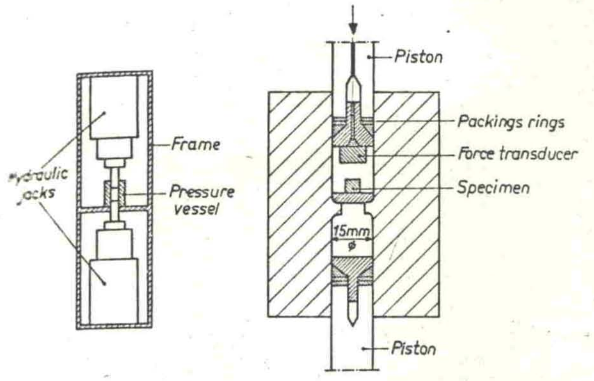

FIG. 4. (a) General view of the pressing equipment. (b) Dore up of the pressure vessel.

function of the pressure is shown in Fig. 3. The high pressure equipment is described below. The deviation from perfect orientation induced by the pressing process decreases the remanence (σ_R). This quantity is of primary importance for the permanent magnet properties. Its values throughout the pressing process are listed in Table I. It is found that the σ_R is hardly affected after the first step of the pressing process.

It can be concluded, therefore, that the final orientation of a powder particle is determined in the very first compacting treatment. We tried in several ways to enhance the orientation of the green pills. Vibrating the die in the orienting field proved to be rather successful: The remanence increases 8% by this treatment.

HYDROSTATIC PRESSING WITH UNIAXIAL DEFORMATION

The experiments have shown that the density of the magnets can be enhanced considerably by uniaxial deformation at high-fluid pressures.[3] The apparatus for this treatment is shown in Fig. 4. By means of two independently working hydraulic systems it is possible to move the pistons through the pressure vessel, in which process the upper piston deforms the specimen. The force exerted by the upper piston on the specimen

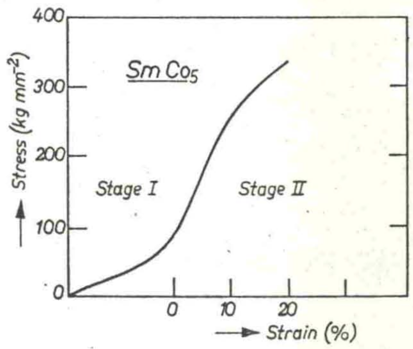

FIG. 5. Typical stress–strain curve recorded during a densification experiment for a $SmCo_5$ sample contained in a Pb–Hg container.

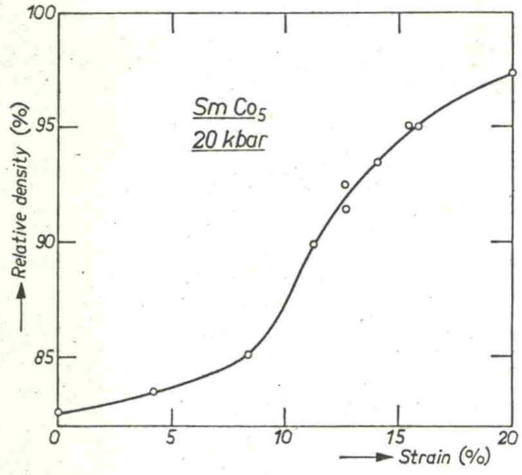

FIG. 6. Relative density versus strain of the pure material.

is measured with a special force transducer[7]: The deformation is measured outside the pressure vessel, thus making it possible to measure the stress–strain diagram of the specimen. The pistons are made of high-speed steel, hardened to R_c65. The pressure vessel is of the monoblock type and made of maraging steel with tensile strength of 180 kg/mm²; it is autofrettaged at 25 kbar. The pressure vessels never ruptured due to fatigue, one of the vessels being used a few hundred times. The pressure transmitting fluid is low-boiling petrol, or pentane. The seals are of the unsupported area type of Bridgman.[8,9]

In our first experiments the powder was packed in a thin nickel tube which was soldered tight. This method is not very suitable due to the rapid decrease of the $_IH_c$ at 300°C. In several ways we tried to make a tight container at room temperature. All plastic or rubber containers failed, probably because these materials become brittle at high pressures, and tear open during

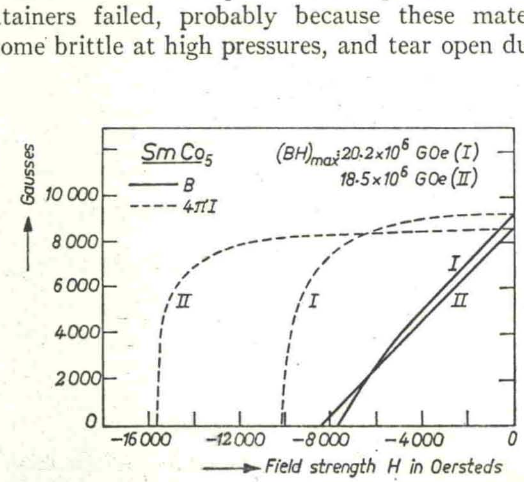

FIG. 7. Demagnetization curves of two types of $SmCo_5$ magnets.

[7] F. F. Westendorp and A. G. Rijnbeek (unpublished).
[8] P. W. Bridgman, Proc. Amer. Acad. Arts Sci. 49, 627 (1914).
[9] S. E. Babb, *Techniques of High-Pressure Experimentation* (John Wiley and Sons, Inc., 1966) chap. VI.

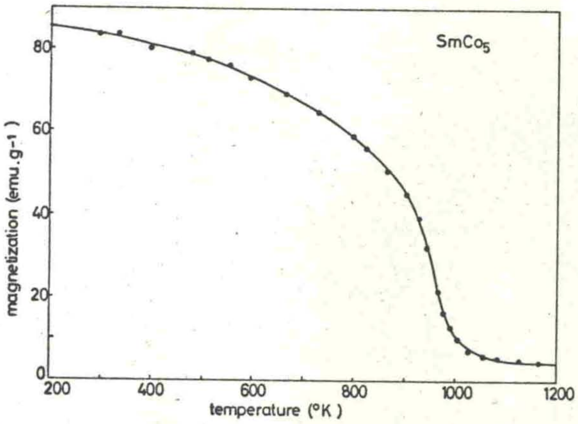

FIG. 8. Magnetization of $SmCo_5$ powder versus temperature obtained in a field of 5450 Oe.

the deformation. Finally, we found a successful way which is a variation of the Bridgman encapsulating method.[10]

The prepressed magnet is packed in lead foil which is wetted with mercury and kept at a moderate oil pressure for about 10 h in a rubber bag. During this time the lead and mercury form an alloy and the specimen is completely sealed in a ductile material.

In Fig. 5 a stress–strain curve is shown recorded during a densification experiment. The first slow rise (stage I) is due to the deformation of the lead. In stage II the strain is at least partially to be attributed to the densification. At the end of stage II the curve bends down somewhat, which is to be attributed to plastic deformation; this latter stage is far more pronounced with more ductile materials such as molybdenum.

The obtained relative density as a function of the total strain of the prepared specimen is shown in Fig. 6. At 20 kbar the specimen fractures after a strain of 20%. The maximum density we obtained was 97%. Although it is difficult to give a quantitative explanation of the described densification, the following facts are probably important. First, the mean compressive stress is enhanced by the deformation from 20 to about 30 kbar. Experiments with solid-pressure transmitting media, however, showed that after pressing at 40 kbar the relative density was only 86%. A possible explanation is that at a hydrostatic pressure of 20 kbar the pores can survive in spite of the very high surrounding stresses thanks to certain favorable stress distributions; they can collapse, however, when these distributions are disturbed by dimensional changes caused by the uniaxial plastic deformation.

[10] P. W. Bridgman, Proc. Amer. Acad. Arts Sci. **77**, 129 (1949).

SOME PROPERTIES OF THE $SmCo_5$ MAGNETS

In Fig. 7 typical demagnetization curves of types of $SmCo_5$ magnets are compared; one with a somewhat lower BH_{max} but with a very high ${}_IH_c$, and the other with high B_r and BH_{max} and lower coercive force. The first is made by using a low Co content and prolonged milling, followed by etching; the second is pressed from powder with a high Co content and which has been ground a relatively short time.

The magnet bodies resemble a solid metal; they can easily be machined by grinding and they do not corrode in air at room temperature. The magnetization versus temperature of $SmCo_5$ is shown in Fig. 8. The Curie point is 980°K. At room temperature the dependence of magnetization on temperature amounts to 5×10^{-5}/°C. As stated before, the material appears to be subjected to a slow decrease of coercivity. As to the possibility of practical application of the magnets, it is very important that the rate of this aging gradually decreases (Fig. 2) and remains so high that the $B=f(H)$ demagnetization curve is unaffected. Some physical aspects of the aging process will be reported elsewhere.[5]

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