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# Statistical Modeling of soft magnetic composite's permeability

Frédéric Mazaleyrat

*SATIE CNRS UMR 8029, Ecole normale supérieure Paris-Saclay,*

*Université Paris-Saclay, Gif-sur-Yvette, France.*

## Abstract

Derivation of the demagnetizing coefficients of non ellipsoidal solids or composite media is one of the most difficult problems in electromagnetism. For a single solid object, exact solutions may be derived for various simple shapes involving quite complicated mathematics but never without ambiguity since the coefficient depends on the susceptibility of the material. For composite medium, usually called soft magnetic composites (SMC) or powder cores, essentially two approaches are used both based on spatial periodicity hypothesis: the non magnetic grain boundary model and the effective medium theory. Actually, the first one works only for dense materials and the second works at low concentration. Both need fitting of two parameters, the inner demagnetizing factor and the particle's susceptibility, just because the periodicity hypothesis is never verified for magnetic filling factors larger than 20%. A breakaway model is proposed based on the computation of mathematical esperance of the statistical distribution of magnetic chains and the subsequent determination of demagnetizing coefficient of an ellipsoid of equivalent aspect ratio. The model is collated with permeability data from the literature for spherical or non-spherical particles based SMCs and shows a excellent agreement with only one or even without fitting parameter in the whole concentration range.

## I. INTRODUCTION

From the beginning of electromagnetism science the concept of demagnetizing field remained one of the most difficult. Already touched experimentally by Michael Faraday about 1845, Maxwell get deeper inside by deriving the expression of the demagnetizing factor of revolution ellipsoid. However, they where effectively computed in a large range of aspect ratio only in 1945 by Stoner [1] and the demagnetizing factors of general ellipsoids were calculated by Osborn [2] the same year. For other shapes (cylinders, prisms) some approximated expressions were known or deduced from experiment by various authors (Du Bois, Ewing, Tanakadaté, in the 1885-1895 decade, see *e.g.* [3],) but because the demagnetizing field is not homogeneous in a non ellipsoidal body, the demagnetizing factor depends on the susceptibility of the material. Demagnetizing coefficients of cylinders where exactly calculated by D.X. Chen only in 1991 and improved in 2006 including negative susceptibilities [4, 5], that of rectangular prisms by Aharoni for  $\chi = 0$  in 1998 [6], by D.X. Chen for  $\chi \neq 0$  [7] in 2006. Non exhaustively, different recent contribution may be quoted to show how the problem is current: tensorial approach, for a collection of ellipsoids with a distribution of aspect ratio [8]; computation by finite element method and subsequent mathematical

fitting [9]; microscopic approach, by Monte Carlo simulations of model spin Hamiltonians [10]; derivation from Newtonian potential [11]; 2D reluctance modeling [12]; superposition principle applied to magnetic/non-magnetic disks stacks [13]; mixing of two powders with different shapes [14]; discussion on the demagnetization in hard magnetic solid bodies is still controversial [15, 16].

Concerning the focus of this paper, *i.e.* the problem of modeling the magnetic permeability of heterogeneous media, despite considerable work done since the XIX<sup>th</sup> century, no model has been established addressing composites with arbitrary particle shapes, and with concentrations from 0 to 1 with good accuracy compared to experiment. The models used can be classified in two radically different approaches: the reluctance model and the effective medium theory (EMT).

The reluctance model is often called non magnetic grain boundary model (NMGB) [17] though the result is similar to that of a magnetic ring with an air gap. Indeed, applying Hopkinson's law, a similar result was already obtained by J.A. Ewing to determine the gap of cut cores [18].

The EMT model is based on the classical concept originally developed by Rudolf Clausius [19] which consist in considering a dielectric media as an electric dipole located in a spherical cavity inside an homogenized medium. The concept was developed by Garnett [20] to model the optical properties of glasses with metal inclusions by in 1904 [21] and Bruggeman for the effective permittivity or conductivity of mixed media in 1935 [22]. Despite the age of the theory, there is still very active researches in this fields particularly in various fields such as homogenization of layered media with periodicity smaller than wave length [23], permittivity of non linear media [24], quantum dot arrays in graphene [25], electro-magnetic behavior of cement paste [26] or magneto-electrics [27].

EMT has been applied to SMCs by Mattei et al. [28] introducing the effect of particle shape [29]. They have shown the existence of a magnetic percolation threshold at 15% [30] and proposed different validations and improvements [31–34]. More recently a tensorial approach was proposed to take cubic magnetocrystalline anisotropy of magnetic particles into account [35] and a solution was given by Skomski for 2 types of particles [14]. However, all these studies were validated with relatively low particles concentrations ( $< 50\%$ ). Good agreement with the EMT approach has been shown studying up to 80% [29] keeping the concentration dependent demagnetizing coefficient as a fitting parameter [36–38].

The main problem of these models is that they consider an isolated particle in a medium so they are applicable only to diluted media (filling factor is below the percolation threshold, typically 20% for spheres). For interacting particles media the effect of the demagnetizing factor has to be

fitted.

In this paper the two existing models will be first reviewed. A revision of the reluctance models will be proposed and it will be compared to the homogeneization model and experimental data from low to high concentration, showing the limits of these models. In the next section, a new approach is proposed in which the SMC is not not considering periodic but as a statistical distribution of spherical particles forming magnetic percolation paths which mean length depends on the filling factor. Then the demagnetizing factor is computed from which the effective permeability is deduced and the results are compared with experimental data in the whole range of concentration. Finally, the model is extended to non spherical particles.

## II. CURRENT MODELS

### II.1. Reluctancy based models

The reluctance model, is from far the simplest. The principle is to consider the powder as a magnetic circuit with a gap. One should consider a simple cubic pack of cubes having a edge  $D$  and separated by a non magnetic layer of thickness  $\delta$ . The total reluctance of the circuit is the sum of the particle and the gap reluctances:

$$\mathcal{R} = \frac{D}{\mu_0\mu_r D^2} + \frac{\delta}{\mu_0 D^2} \quad (1)$$

Writing the magnetomotive force according to Hopkinson's law  $nI = \mathcal{R}\Phi$  with  $\Phi = BD^2$  and  $nI = H(D + \delta)$ , one easily gets the effective permeability :

$$\mu_{eff} = \frac{D}{D + \delta} \cdot \frac{\mu_r}{1 + \frac{\delta}{D}\mu_r} \quad (2)$$

which is equivalent to the result given in [17] apart form the unimportant geometrical factor under the condition  $D \gg \delta$ . The ratio  $\delta/D$  is actually a demagnetizing coefficient associated to this geometry, it is related to the filling fraction  $x$  by:

$$N = \frac{\delta}{D} = (x^{-1/3} - 1) \quad (3)$$

Experience shows that NMGB model agrees well with MnZn ferrites which are composed of MnZn grains with CaO ultra-thin insulating layer though, usually, the magnetic barrier is estimated from the model. Typically this barrier is about few nm as shown by TEM in high permeability

Mn-Zn ferrite [40]. However, in Mn-Zn ferrites, the magnetic fraction is over 95% and each grain is separated by the paramagnetic layer. In the usual form, the model has major drawbacks:

- it systematically underestimate the effective permeability if the fraction is lower than 90%
- it has no physical meaning for small fractions because when  $\delta > D$ ,  $N > 1$ , *i.e.*  $x < \frac{1}{8}$ .
- $\mu_{eff} \rightarrow 0$  when  $x \rightarrow 0$
- $\mu_{eff} < 0$  if  $\frac{\delta}{D} > 1 - \frac{1}{\mu_r}$
- SMCs have imperfect insulation, which implies the presence of magnetic contact between particles.

A more general approach would deduce the applied field from Ampere's theorem  $H(D + \delta) = H_i D + H_g \delta$  where the subscript  $i$  is for internal and  $g$  for gap. By continuity of the normal component of  $B$ , one can write that  $\mu_0 H_g = B_i = \mu_0 H_i + M$ . Combining these two equations leads to:

$$H = H_i + \frac{\delta}{D + \delta} M \quad (4)$$

Substituting  $M = \chi H_i$  and putting  $B = \mu_0 \mu_{eff} H = \mu_0 (1 + \chi) H_i$  yields a slightly different expression of the effective permeability

$$\mu_{eff} = \frac{1 + \chi}{1 + \frac{\delta}{D + \delta} \chi} = \frac{1 + \chi}{1 + (1 - x^{1/3}) \chi} \quad (5)$$

Within the limit of small gap and large susceptibility this expression is equivalent to Eq. (2), but now it can be seen that the demagnetising coefficient has a different expression and is never greater than 1. Also, the effective permeability  $\mu_{eff} \rightarrow 1$  if  $\delta \gg D$ . This correction of the NMGB model improves the physical meaning and the range of applicability of the model. It is well known that the problem with NMGB model is due to the fact that the main assumptions (*i*) the uniformity of the field in the gap is clearly not justified if the filling fraction is not very close to one, (*ii*) the regular cubic packing, *i.e.* 1D approximation, is unrealistic.

A 2D extension of the model has already been proposed by the author to apply to bcc packing of square flakes [12]. Despite good agreement with nanocrystalline flakes composites, it lacks by generality since it works only for oriented square flakes with in-plane field.

## II.2. Homogeneization models

The concept of homogeneisation was first applied to dielectrics. In the original model, each individual electric dipole is supposed to be located in a spherical cavity in an uniformly polarized homogeneous medium corresponding to the mean field produced by the contributions of all dipoles. The effective permittivity deduced from this concept is given by Clausius-Mossotti relation (see e.g. [41].) There are different improvements of the model such as Garnett's, to take into account the permittivity of the host medium, or Bruggeman's, which considers two (or more) particles in an effective medium. Among the different variants of the model [42], Bruggeman's is mostly used for magnetic composites [29] and usually referred as effective medium theory (EMT). In its scalar form, the effective permeability  $\mu_{eff}$  is defined from the relative permeability of each media  $\mu_i$ , their concentration  $c_i$  and their demagnetizing coefficients  $N_i$  by the expression:

$$\sum_i \frac{c_i N_i (\mu_i - \mu_{eff})}{\mu_{eff} + N_i (\mu_i - \mu_{eff})} = 0 \quad (6)$$

In the present case, the material has 2 components only one of which is ferromagnetic,  $\mu_1 = 1 + \chi$ ,  $\mu_2 = 1$ ,  $N_1 = N_2 = N$ ,  $c_1 = x$  and  $c_2 = 1 - x$ . Written in terms of susceptibility, the above equation is simplified as a second order equation:

$$(1 - N)\chi_{eff}^2 + [1 + (N - x)\chi] \chi_{eff} - x\chi = 0 \quad (7)$$

which has only one physical solution

$$\chi_{eff} = \frac{-[1+(N-x)\chi] + \sqrt{[1+(N-x)\chi]^2 + 4(1-N)x\chi}}{2(1-N)} \quad (8)$$

For larger concentrations,  $\chi_{eff}$  has the same order of magnitude as  $\chi_i$  so the last term of Eq. (7) is negligible and a trivial solution is

$$\lim_{x \rightarrow 1} \mu_{eff} = 1 + \frac{(x - N)\chi - 1}{1 - N} \quad (9)$$

On the over edge, for low concentrations, first order expansion of Eq. (8) yields

$$\lim_{x \rightarrow 0} \mu_{eff} = 1 + \frac{x\chi}{1 + (N - x)\chi} \quad (10)$$

furthermore if  $N \gg x$  - *i.e.* any particle shape except needles - and  $\chi_i \gg 1$ :

$$\mu_{eff} \approx 1 + \frac{x}{N} \quad (11)$$

As mentioned in the introduction, the problem is that  $N_{eff}$  has to be fitted from experiment. Mattei has proposed an expression of  $N_{eff}$  but it depends on the magnetic contrast  $\alpha = (\mu_i - \mu_{eff})/\mu_{eff}$  leading to non explicit solution of the problem [31].

Furthermore, the EMT can't be applied for large particle fractions because the volume fraction itself is limited by the shape of the particles. In the case of spheres, the highest density is 74% for hcp or fcc packing, 63.6% for random close packing. and even 53.6% for random loose packing with a connectivity of 4 [43]. Indeed, it is impossible to keep the particles spherical above the limit of 60% so for any larger  $x$ ,  $N$  is changed.

### II.3. Comparison of the models with experiment

Concerning high permeability SMCs, it is observed that none of these models can fit the data without a coefficient adjustment corresponding to unreasonable particle geometries. In Fig. 1 (top box) reasonable agreement is found in a very limited range by fitting the demagnetizing factor and the susceptibility. For NMGB model, it has to be reduced — compared to a sphere — by a factor 8 which may be justified qualitatively by the contact between particles. However the experimental agreement remains poor. For EMT, a good fitting is found for  $N = 0.9$  which seems quite unreasonable since  $N$  is expected to be close to  $1/3$  and this value would corresponds to a prolate ellipsoid of  $r = 5$  with field parallel to short axis. Furthermore, the permeability is below 10 for  $x < 0.8$ . Besides the models disagree on the value of the susceptibility up to a factor of 5 or more. For low concentrations (Fig. 1 bottom box), EMT is in very good agreement with experience but needs two fitting parameters, here  $N = 0.27$  and  $\chi = 15$ . In contrast, NMGB needs no fitting parameter, here  $\chi = 1000$ , but 10 times less or more wouldn't make appreciable difference. Again the experimental agreement is poor for NMGB.

This clearly shows that the two existing models hardly agree on the value of  $N$  and  $\chi$ . Furthermore, there is a gap for fractions between 60 and 90% where none of these models apply whereas powder cores have mostly a magnetic particle concentration in this gap.



### III. MODEL BASED ON THE STATISTICAL MEAN CHAIN APPROXIMATION (SMCA)

#### III.1. Equivalent chain length

In order to take into account the random nature of real SMCs, we first consider an infinite chain of spheres composed of a fraction  $x$  of magnetic particles,  $1 - x$  of non magnetic particles or voids, both of diameter  $D$ . If the voids are randomly distributed, the chain is actually composed of series of magnetic and non-magnetic monomers, dimers, trimers, etc, separated by voids.

For example, if  $x = 0.5$ , the random chain can be decomposed in ordered chains alternating single particles/voids units, particle/voids dimers, particle/voids trimers, etc. with associated probability as illustrated in Fig. 2. In contrast an ordered chain would be composed of alternating single particles and voids.

In order to obtain the average chain length as a function  $x$ , the number of chains composed of  $n$  particles has to be calculated as a function of  $x$ .

The event  $A_n$  corresponding to a chain of  $n$  particles surrounded by 2 voids has the following probability to be realized:

$$P_n(A_n) = x^n(1 - x)^2 \quad (12)$$

where  $x^n$  corresponds with the particles and  $(1 - x)^2$  with the 2 voids.

The sum over all chain lengths is calculated from Eq. (12)

$$\begin{aligned} (1 - x)^2 \sum_{n=1}^{+\infty} x^n &= (1 - x)^2 \left( \sum_{n=0}^{+\infty} x^n - 1 \right) \\ &= (1 - x)^2 \left( \frac{1}{1 - x} - 1 \right) \\ &= (1 - x)x \end{aligned} \quad (13)$$

so Eq. (12) can be normalized with respect to Eq. (13)

$$P_n(A_n) = (1 - x)x^{n-1} \quad (14)$$

The average chain length is the expected value of Eq. (14)

$$\begin{aligned}
\mathbb{E}[x] &= \sum_{n=1}^{+\infty} nP_n(A_n) \\
&= (1-x) \sum_{n=1}^{+\infty} nx^{n-1} \\
&= (1-x) \frac{1}{(1-x)^2} \\
&= \frac{1}{1-x}
\end{aligned} \tag{15}$$

So finally, for any value of the filling fraction  $x$ , the aspect ratio of the mean chain takes a remarkably simple form:

$$r = \frac{\langle \ell \rangle}{D} = \frac{1}{1-x} \tag{16}$$

were  $D$  is the diameter of the spheres. Going back to the example when  $x = 0$ ,  $r = 2r$  is found instead of 1 for an ordered chain.

If  $x \rightarrow 0$ , the chains are single particles, and if  $x \rightarrow 1$ , the chain is infinite.

In 3 dimensions, voids are no more distributed along the chain but in the volume. In a simple cubic packing, voids around a magnetic particle can be localized in the 3 directions. However, if the magnetic field is homogeneous and directed parallel to one of the cube edges, we may consider only the voids along a 1D chain, this is to say that percolation path perpendicular to the field is not taken into account. This situation is totally different from that of electrical percolation. Indeed, in the scheme Fig. 3 (a) shows the electrical percolation path and (b) the magnetic path for the same geometry providing the susceptibility is relatively small which is often the case for small particles. So path (b) would be equivalent to (c). If the voids are distributed with the same probability in the 3 directions, the equivalent chain will have an aspect ratio:

$$r = \frac{\langle \ell \rangle}{D} = \frac{1}{1-x^{1/3}} \tag{17}$$

### III.2. Effective demagnetizing factor

Now the problem is to determine the demagnetizing factor associated with the equivalent chain. Here a classical assumption is made saying that a chain composed of  $r$  particles (see Fig. 4a) exhibits a demagnetizing factor equivalent to a prolate ellipsoid of aspect ratio  $r$  (see Fig. 4c)

which is known from Eq. (17). Eventually this model avoids the problem of compaction limits since particles may be distorted inside a chain with limited effect on  $N_{eff}$  (see Fig. 4b.)

Using one of the well known expression of the demagnetising factor of a prolate ellipsoid (here as formulated by Stoner [1]),

$$N(r) = \frac{1}{r^2 - 1} \left[ \frac{r}{\sqrt{r^2 - 1}} \ln(r + \sqrt{r^2 - 1}) - 1 \right] \quad (18)$$

one can easily compute the equivalent demagnetizing factor as a function of the filling fraction thru Eq. (17). This computation is compared with effective demagnetizing coefficients determined experimentally by Forrer with iron balls [44] in Fig. 5. In this experiment Forrer considered that the demagnetizing factor can be determined as the inverse of the initial slope of the anhysteretic curve,  $\chi_{an}$  which is equivalent to the slope of the major hysteresis loop at  $H = H_c$ . For low concentrations as  $N$  is low, the loop may be not saturated so the  $\chi_{an}$  may be underestimated and by consequence  $N$  may be overestimated, which may explain the discrepancy between the present model and the measurements. On the other side, the demagnetizing coefficient of a  $n$ -spheres chain is obviously larger than the ellipsoid having an aspect ratio of  $n$  and the difference is more visible for small  $n$ . This may explain why the gap between the model and the experiment is larger for small  $x$ , but evidently there is a good agreement between the two without any fitting parameter.

### III.3. Permeability of magnetic spheres composites

Let us consider a SMC composed of spherical magnetic particles dispersed in a zero susceptibility matrix such as a para or diamagnetic binder. The field  $H_a$  required to apply to the material to achieve the internal field  $H_i$  is increased by the demagnetizing field according to:

$$H_a = H_i + N_{eff}M = (1 + N_{eff}\chi)H_i \quad (19)$$

where  $N_{eff}$  is the effective demagnetizing factor of the equivalent average chain determined statistically. The mean magnetization of the SMC can be written

$$M = \chi_{eff}H_a = x\chi H_i \quad (20)$$

Substitution of Eq. (19) in Eq. (20) leads to

$$\mu_{eff} = 1 + \frac{x\chi}{1 + N_{eff}\chi} \quad (21)$$

This expression makes physical sense since for  $x \rightarrow 0$  and  $\chi \rightarrow \infty$  it leads exactly the same as EMT Eq. (11) – where EMT works well –, and since for  $x \rightarrow 1$  it leads exactly the same expression as modified NMBG Eq. (5) – where NMBG works well – except for the new definition of the demagnetizing factor.

A computation of the effective permeability as a function of the filling factor is shown for different values of the susceptibility in Fig. 6. It is remarkable that for  $x = 0.5$  and susceptibilities above 99, the present model give an effective permeability about 10 which is significantly larger than that given by NMBG (only 2) and contrarily to EMT the linear behavior expected for  $x > 1/3$  is not observed but an exponential one up to  $x \approx 0.5$ . A useful empirical approximation may be written:

$$\mu_{eff} = e^{4.4x} \quad \forall x \leq 0.5 \quad \& \quad \chi \geq 299 \quad (22)$$

Compared with experiment as shown in Fig. 7, a very good fit of Weidenfeller’s [38] and Minot’s [39] low permeability composites is obtained by only adjusting the susceptibility, contrarily to EMT that needs also  $N$  to be fitted. Note that with the present model  $\chi = 21$  and 150 whereas  $\chi = 16$  and 52 with EMT.

For large concentrations, comparing with commercial data (iron and permalloy based SMCs) shown in Fig. 8 highlights the efficiency of the model. Though significant spread is observed – partly due to the uncertainty on  $x$  values – data are best fitted with  $\chi = 999$  for low concentrations and  $\chi \rightarrow \infty$  for high concentrations. This may be partly justified by the fact high density SMCs are usually based on larger particles, thus having higher susceptibility [45].

For dense media, because particle chains are very long, a 1D model should be sufficient. If  $x \rightarrow 1$  the SMC should be equivalent to a cylinder of aspect ratio  $r = 1/(1-x)$ . Its demagnetising coefficient can be approximated using the fluxmetric demagnising coefficient of a cylinder with  $r > 20$ ,  $N_{eff} \approx \frac{1}{2r^2}$ , so Eq. (21) becomes

$$\mu_{eff} = 1 + \frac{x\chi}{1 + \frac{1}{2}(1-x)^2\chi} \quad \forall x > 0.95. \quad (23)$$

Surprisingly, the approximated model fits better the data, but one has to be cautious because of the relatively large scattering of data and also because particles were assumed to be spherical and mono-dispersed in the model based on Statistical Mean Chain Approximation (SMCA) which is clearly not the case in dense SMCs because the particles are not spherical and hot pressed (see *e.g.*[46].)

### III.4. Permeability of non-spherical magnetic particles composites

If the particles are not spherical Eq. (18) should be replaced by that of the corresponding shape (*e.g.* that of a cylinder as computed by Chen [4].) However, expressions using complete elliptic integrals are rather difficult to implement. A simple approximation would be to consider that the aspect ratio dependence for any shape is the same as that of a prolate ellipsoid. In other words, if  $N_p$  is the demagnetizing coefficient of the particle, that of the chain of aspect ratio  $r$  would be  $3N_p N_{eff}(r)$  verifying that in diluted media  $3N_p N_{eff}(1) = N_p$ . This leads to the general expression:

$$\mu_{eff} = 1 + \frac{x\chi}{1 + 3N_p N_{eff}\chi} \quad (24)$$

A parametric study of this equation is given in Fig. 9. Qualitatively these curves look like in [29] except the curvature which is more pronounced at same  $N_p$ .

To collate with experimental data, a fitting of  $N_p$  with  $\chi \rightarrow \infty$  is displayed Fig. 10. The best fit of experimental curves is obtained for  $N_p = 0.5$  corresponding to  $r \approx 1.8$ , a realistic result considering the micrographs of SMCs [46] showing usually non equiaxis particles. In order to demonstrate the validity of Eq. (24) in a more straightforward way, comparison with SMC constituted of Finemet flake has been displayed in Fig. 11. The selected data correspond to particles sieved with a  $80 \mu\text{m}$  mesh, pressed in a resin and measured with a field perpendicular to pressure. Considering each particle as a  $20 \times 80 \times 80 \mu\text{m}^3$  square prism,  $N_p$  was calculated according to [6] (field applied in plane) and Eq. (24) was applied directly considering  $\chi \rightarrow \infty$  without any kind of fitting. Compared with EMT, one would find an effective demagnetizing coefficient slightly higher ( $\approx 0.22$ ) which is self-contradictory since in EMT  $N_{eff} \leq N_p$  and  $\chi \approx 200$  which would be almost one order of magnitude too small for Finemet, one of the softest materials ever made ( $\chi > 15000$ ). The only limitation of the model is that non-spherical particles must be oriented. This condition is however easily satisfied with flakes based composites since compaction naturally orients flakes perpendicular to pressing direction.

## IV. CONCLUSION

A new model for the permeability of SMCs has been successfully developed based on simple statistical consideration and classical concepts in electromagnetism. The main strength of the model is the predetermination of the effective demagnetizing coefficient directly from the filling fraction and the approximate shape of the particles. For SMCs with a filling fraction less than

60% even the exact knowledge of the intrinsic susceptibility is not necessary providing it's value is greater than 100. Experimental collation is very good with composites made from spherical particles, but also for that based on flake-shaped particles like milled nanocrystalline ribbons. Because the main parameter of the model  $N_p$  is determined independently and the second one  $\chi$  doesn't require accurate determination – only the order of magnitude is needed – the Statistical Mean Chain Approximation model is able to predict the permeability of SMCs made from any particle shape and any magnetic material. A small adjustment of  $N_p$  may be necessary only in the case of powders having indefinite particle shape.

To compare the models, the parameters of the models are indicated in Table I. In some cases, parameters are obtained directly from the model (noted C in the table) and in some cases they must be fitted. An assessment of the quality of the fit is given in column  $Q$  together with an assessment of the physical meaning ( $\Phi$ ) of the value of the parameters. The NMGB model makes good physical sense but needs adjustment of  $N_{eff}$  for dense SMCs and the fitting is poor. On the over side, EMT usually yields good fitting but the two parameters must but adjusted, especially  $N_{eff}$  that may be lower than calculated for low concentrations and much higher for high concentrations. In contrast, the present model gives very good fit whatever is the concentration or the shape of the particles and requires at maximum one parameter to be fitted,  $\chi$  for low SMC made from micrometric spherical particles or  $N_p$  when the particle shape is undefined, or even no fitting parameter in the case of composite made from nanocrystalline flakes for which  $\chi$  may be measured on a ribbon or wound toroid before crushing or even set to infinity. It is thought that the present model based on a statistical approach is more versatile, robust and physically coherent, the reason is clearly because usual models are based on a spacial periodicity hypothesis which is never verified in practice.

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| material                  | model | parameters |          |          | scores |        |     |
|---------------------------|-------|------------|----------|----------|--------|--------|-----|
|                           |       | $N_{eff}$  | $\chi$   | $N_p$    | $Q$    | $\Phi$ | $F$ |
| 3 $\mu$ m spheres         | NMGB  | C          | $\infty$ | $\times$ | P      | P      | 0   |
|                           | EMT   | 0.27       | 15       | $\times$ | VG     | VG     | 2   |
|                           | SMCA  | C          | 20       | $\times$ | VG     | VG     | 1   |
| High density SMCs         | NMGB  | C/8        | 8000     | $\times$ | P      | P      | 2   |
|                           | EMT   | 0.9        | 1000     | $\times$ | VG     | P      | 2   |
|                           | SMCA  | C          | $\infty$ | 0.5      | VG     | VG     | 1   |
| 80 $\mu$ m                | NMGB  | C          | $10^4$   | $\times$ | G      | VG     | 0   |
| Nanocrystalline composite | EMT   | 0.22       | 100      | C        | P      | P      | 2   |
|                           | SMCA  | C          | $\infty$ | C        | VG     | VG     | 0   |

TABLE I. Comparison of the models.  $Q$  is quality of the fit;  $\Phi$  the physical coherence,  $F$  is the number of fitting parameters. C means predetermined by calculation, numbers are fitted values, except the susceptibility of nanocrystalline flakes which was measured before crushing of the ribbon.

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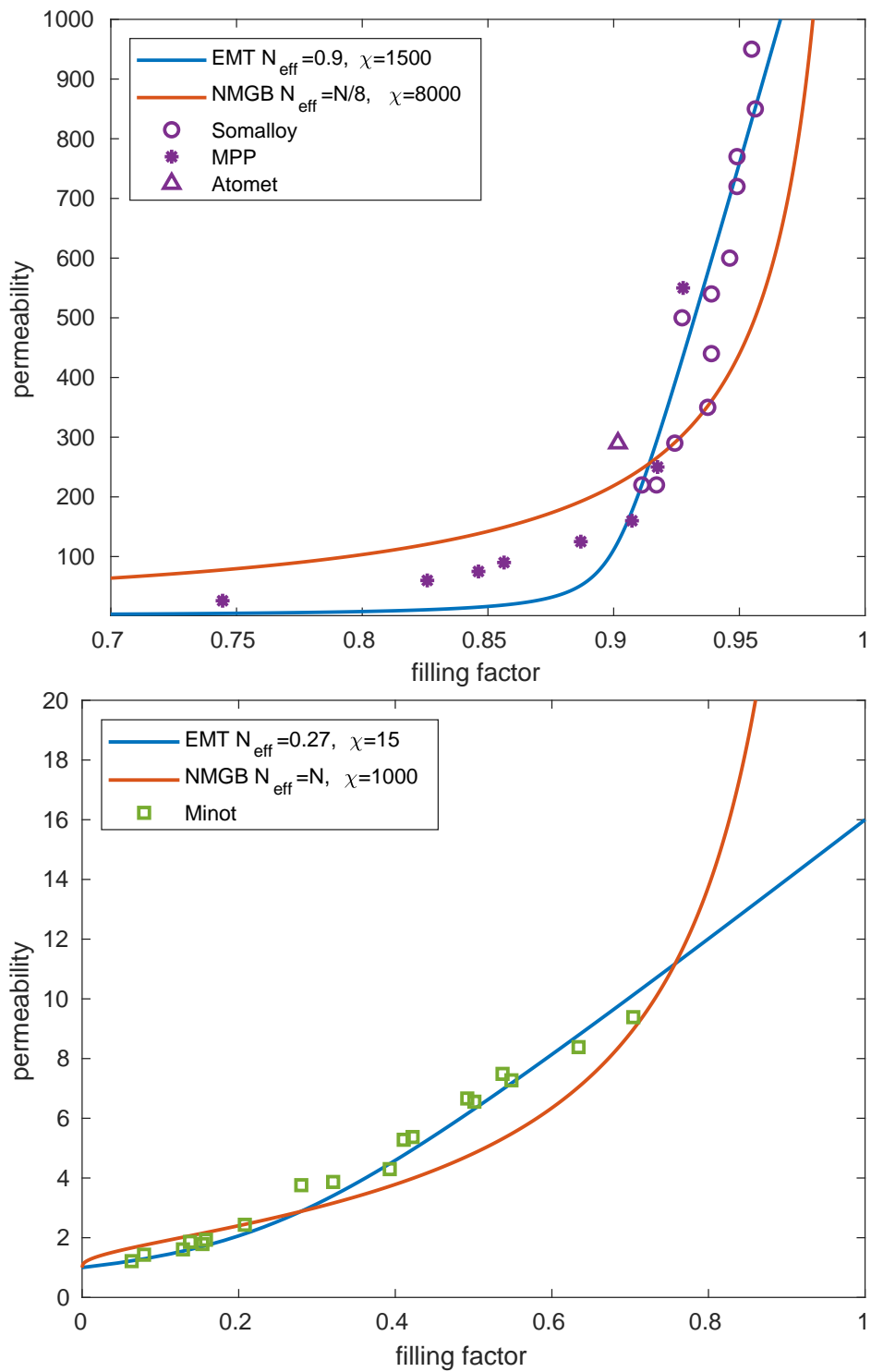


FIG. 1. NMGB and EMT models compared to values of some high permeability commercial SMCs (top box) and to an experimental composite composed of  $3\mu\text{m}$  iron spheres [39]. Somalloy and Atomet are trade mark of Höganäs and Rio Tinto respectively.

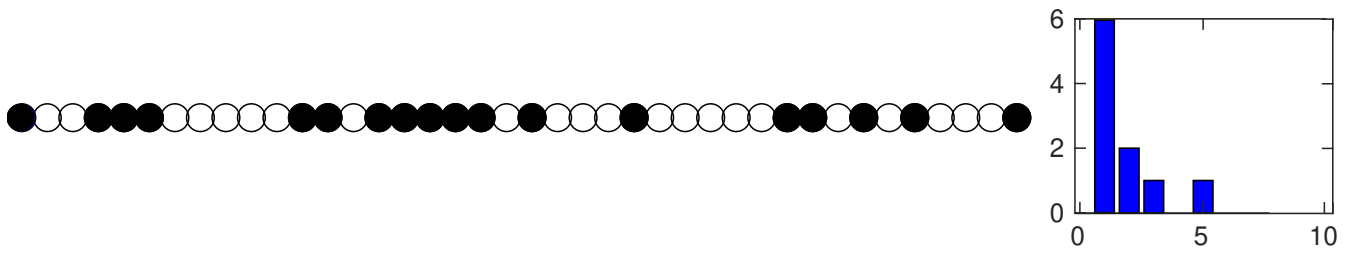


FIG. 2. Example of a random chain generated by a uniform probability with a threshold of 0.5 – black and white balls are equiprobable – and the corresponding histogram.

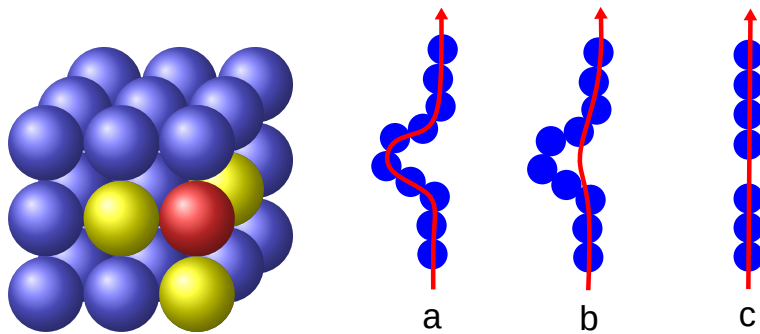


FIG. 3. 3 possible position of a magnetic particle (red ball) nearest void (yellow balls). Scheme of a percolation path (a), the mean magnetic path for same geometry (b), the 1D equivalent magnetic path (c).

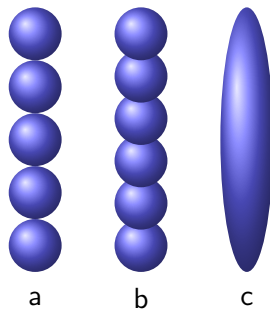


FIG. 4. Schemes of (a) a hard sphere chain, (b) a soft sphere compressed chain, (c) the equivalent ellipsoid.

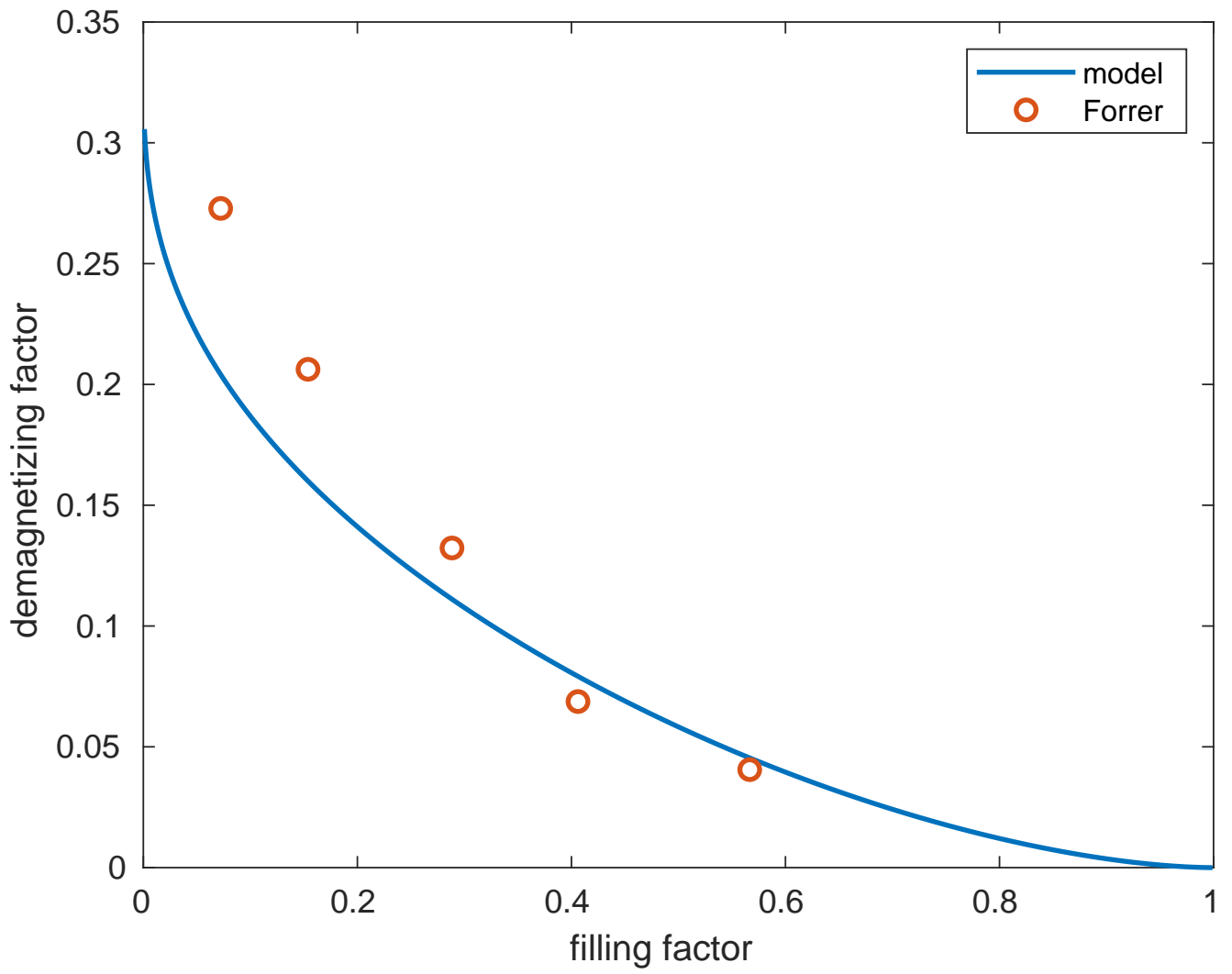


FIG. 5. Average demagnetizing factor as a function of filling fraction (data are from Ferrer [44])

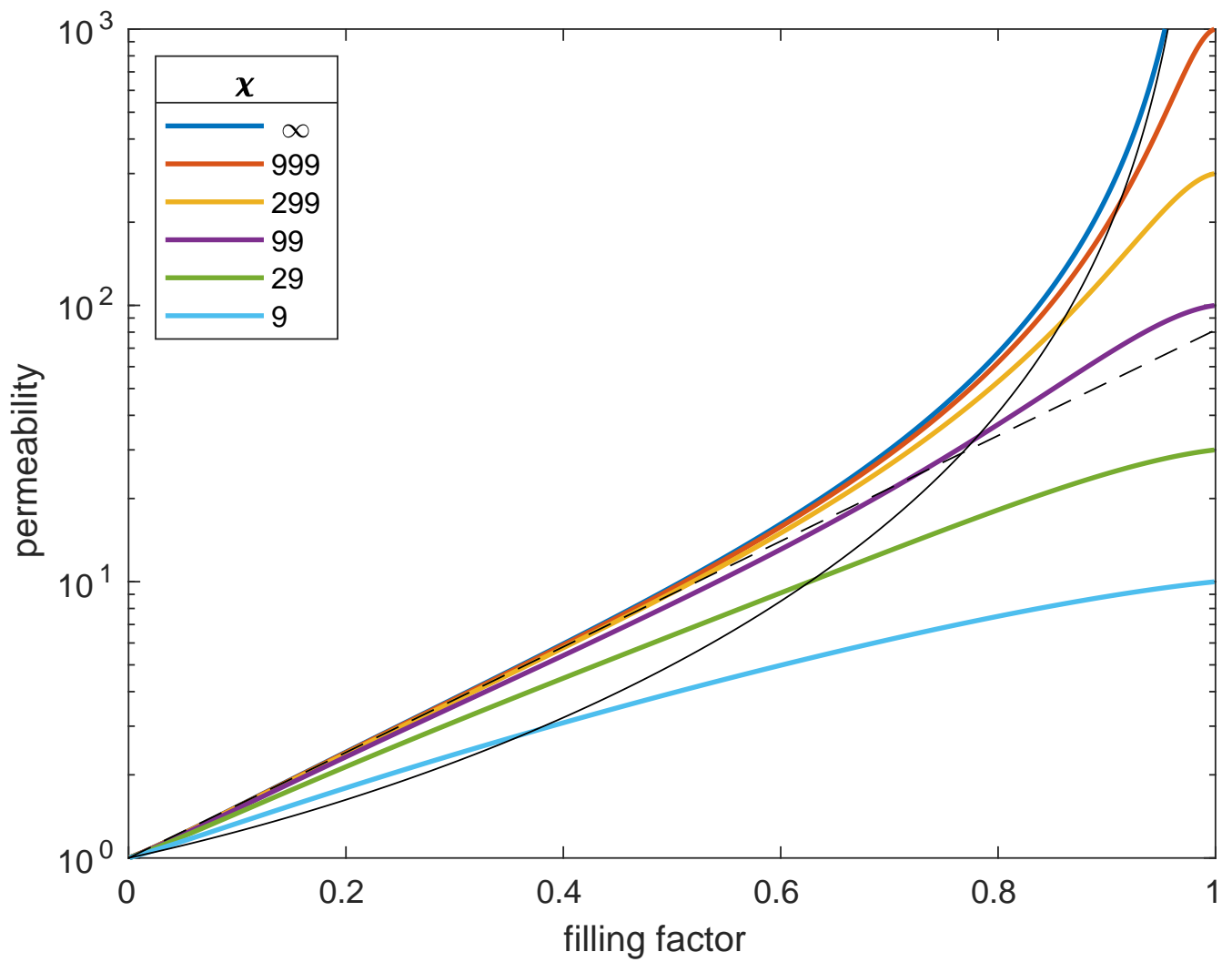


FIG. 6. Effective permeability dependence on the filling factor parametrised by the susceptibility according to the present model. (--) exponential approximation Eq. (22), (—) long cylinder approximation Eq. (23).

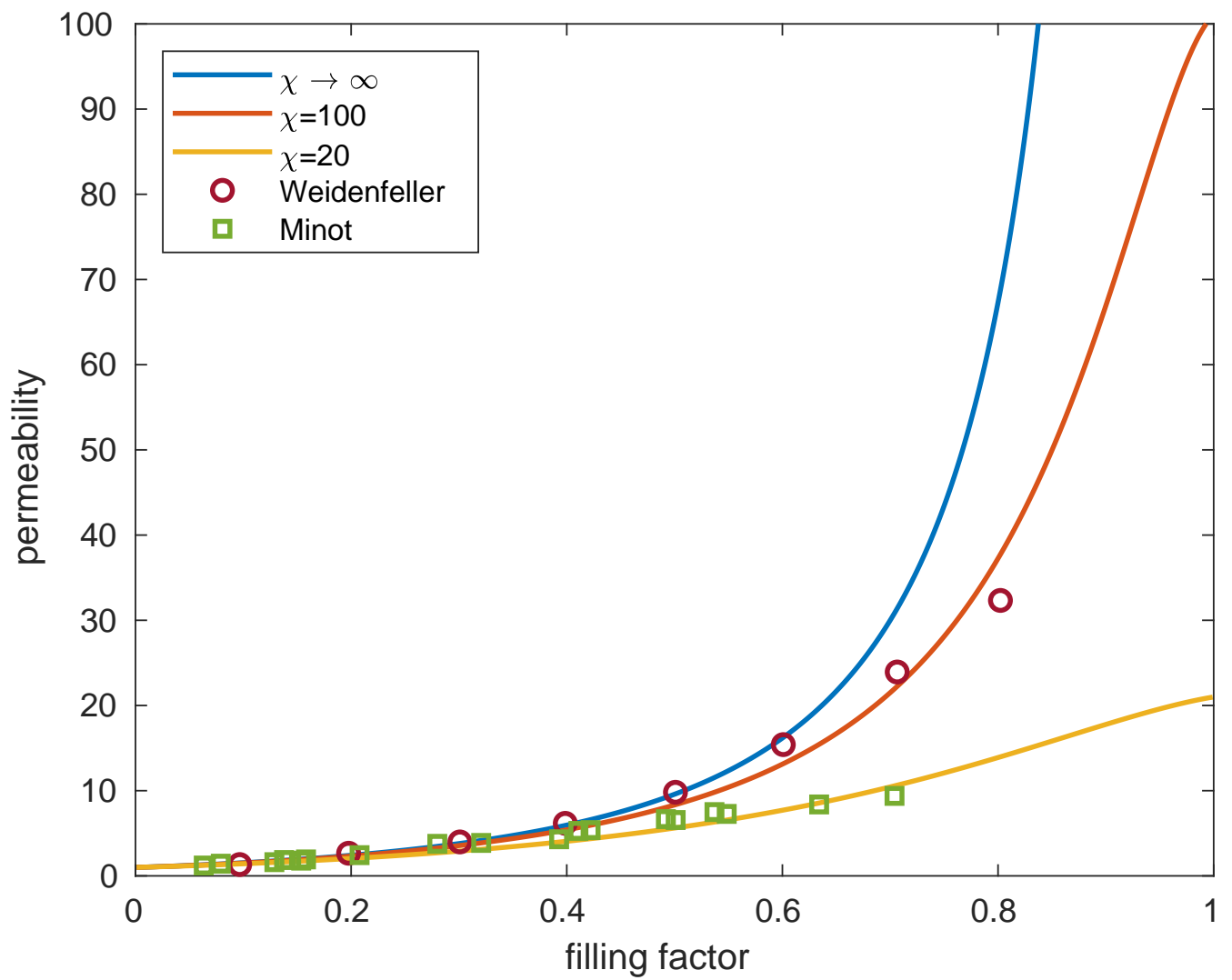


FIG. 7. Comparison of the model with experience for low concentration SMCs based on spherical particles (top box). Data are from Weidenfeller [38], Minot [39] .

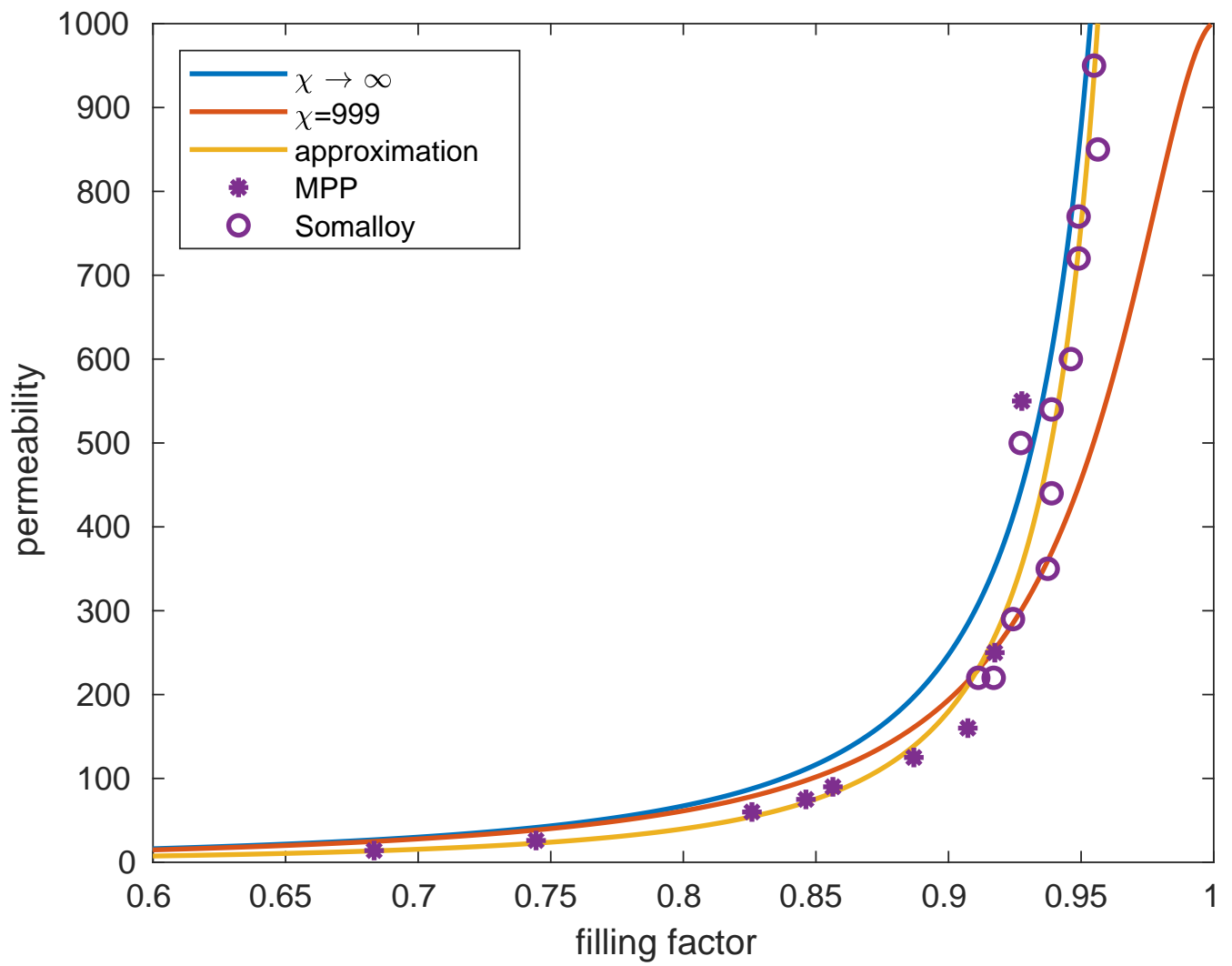


FIG. 8. Comparison of the model with experience for high concentration SMCs based on nearly random shaped particles (bottom box). Data are from technical documentations from Höganäs and Magnetics

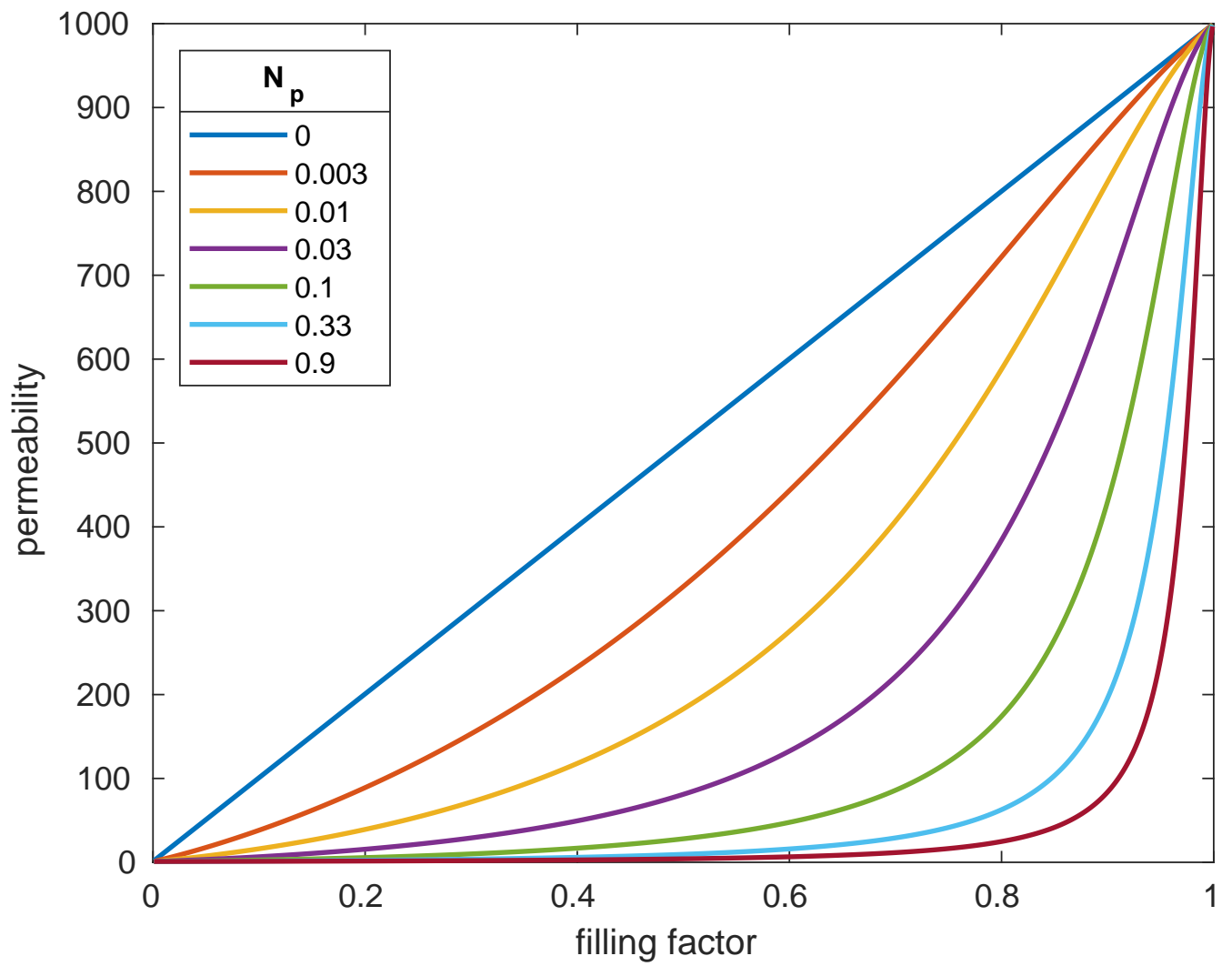


FIG. 9. Effective permeability dependence on the demagnetising coefficient of the particles according to the present model computed for  $\chi = 999$ . Demagnetising coefficients correspond to aspect ratio of  $\infty$ , 32, 15, 7.5, 3, 1, 1/30, respectively



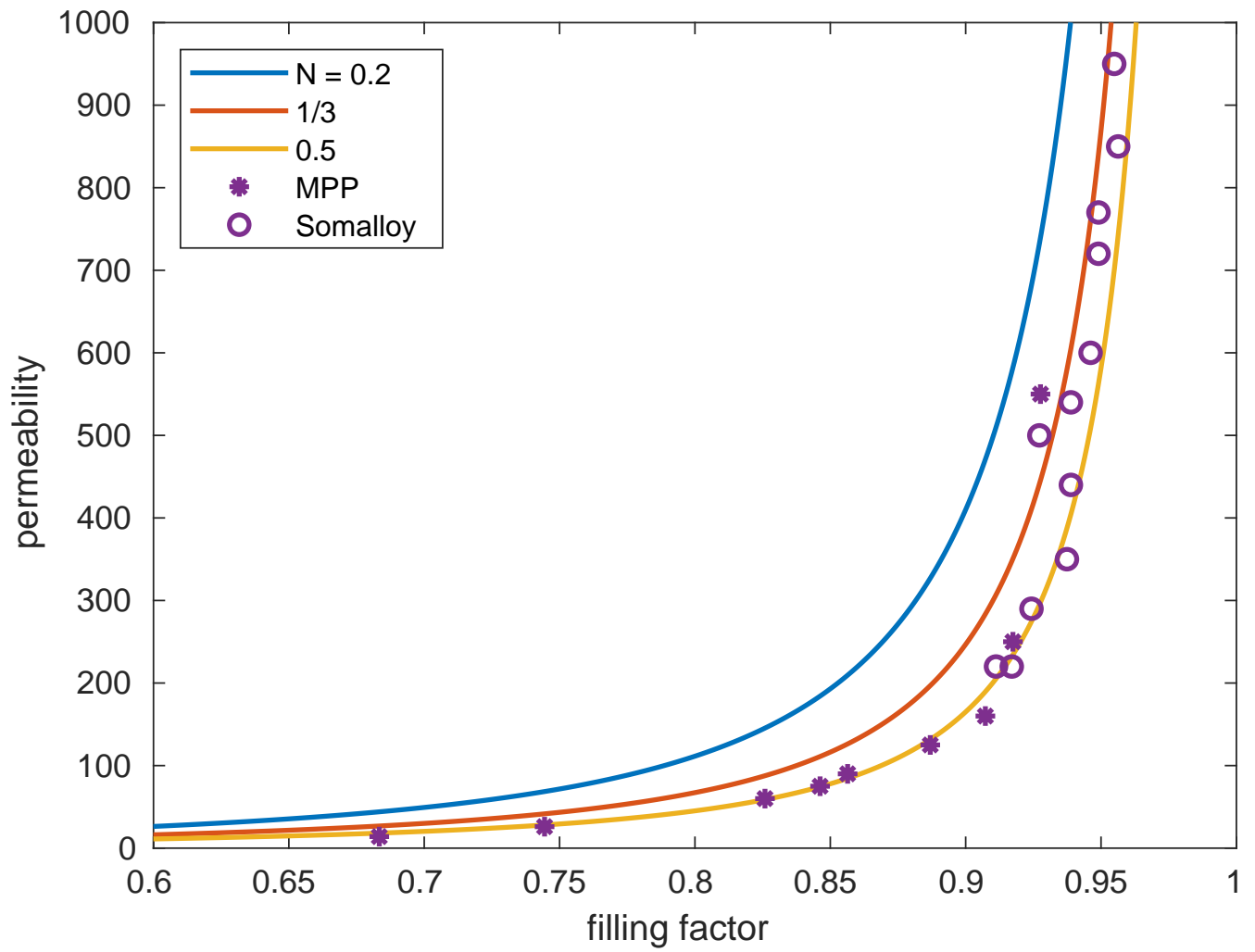


FIG. 10. Effect of the shape of the particles on the model for  $\chi \rightarrow \infty$  compared to high density SMC.

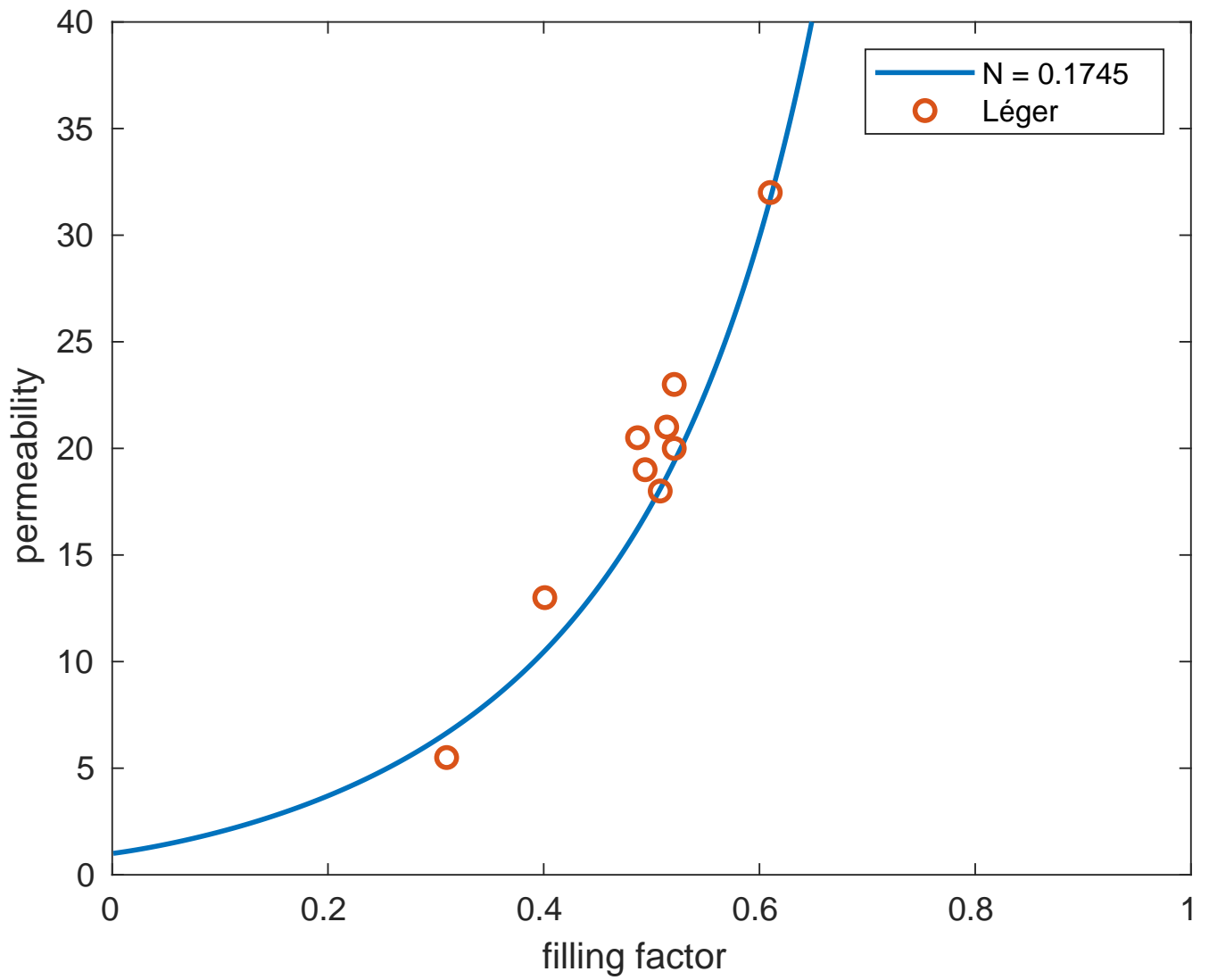


FIG. 11. Permeability of Finemet SMCs as a function of filling fraction. Particles are  $20 \times 80 \times 80 \mu\text{m}^3$  Finemet flakes. Data from V. Léger [12], demagnetizing coefficient of rectangular prism is from [6].