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ISBN 1 899072 14 4

Euro PM2000
Soft Magnetic Materials Workshop

Modelling of the Infiltration Kinetics of Liquid Resin during Production of Soft Magnetic Composites (SMC)

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Abstract: Soft magnetic composites (SMC) based on metallic powders are material group with a wide range of application frequencies. The basic concept at the production of SMC is to add organic resin to the powder, after which pressing and curing serves as a binder and a surface insulation between the powder particles. This approach allows to reduce the eddy current loss and to obtain a strong connection between the powder particles.
One of the techniques used for production of powder-resin compacts is a layer by layer deposition of liquid resin and powder.
An equation for the modelling of the kinetics of liquid resin-metal infiltration into a porous compact is proposed. A solution describing the kinetics of infiltration is obtained. Different forms of the deposition of the liquid resin have been discussed. The model shows that the shortest time of infiltration is obtained when the liquid resin is deposited as droplets embedded into the powder deposit.

INTRODUCTION

During the last decade powder metallurgical processes have become increasingly important for production of magnetic materials, especially for the soft magnetic composites (SMC) based on metallic powders [1-5]. The latter is a material group with a wide range of application frequencies, which explains the constantly growing interest to these materials.
Unfortunately, classical methods for compaction of the powders are not applicable in this case. This is due to the fact that the excessive heating of the compact may lead to the deterioration or even to the disappearance of the valuable magnetic properties. Another problem is that, since the classical methods use metals or alloys as a binder, the powder particles are not insulated from each other. This imposes on additional problem, namely the increase of the eddy currents, which at high frequencies may lead to considerable power losses. Note that the dissipated power due to eddy currents P_e is proportional to the square of the applied frequency ϕ and inversely proportional to the specific resistivity ρ, $P_e \approx \phi^2 / \rho$. Thus the increase of the specific resistivity ρ leads to lower eddy currents

Therefore in order to avoid the problems mentioned above organic resins are used as a binder instead metallic powders. The resin after pressing and curing serves as a binder and a surface insulation between the powder particles [4,5]. This approach allows to reduce the eddy current loss and to obtain a strong connection between the powder particles [2-5].

One of the techniques used for production of powder-resin compacts is layer by layer deposition of liquid resin and powder. This approach allows the avoid the most drawbacks of the methods used until now since it is applied at low temperatures and is simple to use. This technique however arises several practical questions, which are directly connected to the quality of the final product. A first question is what pressure should be applied in order to drive the resin through the powder. In case of a too low pressure the density of the compact will be too low, which will deteriorate the magnetic properties of the final product. An excessive high pressure may lead to the closing of the capillaries between the powder particles and thus to deterioration of the infiltration kinetics e.g. rise of the porosity. The second question is the influence of the viscosity and the surface tension of the used resin on the infiltration kinetics [6]. In the present research we will introduce a criterion which will allow us to determine the influence of these two physical parameters. The third question is the way of the deposition of the resin: as will be shown below different ways of deposition lead to dramatic change of the time required for resin infiltration inside the powder compact. The last question concerns the flow of the liquid resin through the capillaries formed by the powder particles [7-10]. More specific the dependence of flow resistance on the fractional density *f*. Several models have been developed to describe this important characteristic of the powder. Most of these are obtained empirically and describe the flow of specific liquid through specific powder media [10].

The aims of the present research are:

-the development of a model of the infiltration kinetics,

-a selection of an appropriate permeability function that combines particle packing characteristics such as fractional density with the applied pressure,

On the basis of the model that will be developed, it will be possible to determine the most optimal way of the resin deposition. In the present investigation, three different forms of deposition will be considered: a plane, a cylinder and a spherical droplet. Finally the influence of the different physical parameters, e.g. surface tension, viscosity, fractional density etc. on the infiltration kinetics will be checked.

THE MODEL

In order to obtain the equations governing the infiltration kinetics and a criterion, which determines the importance of the different physical parameters on the infiltration kinetics a dimensional analysis is used [11,12]. The first step, according to this theory is the determination of the main parameters involved in the infiltration process and their dimensions. These are the dynamic viscosity η=[Nsm^{-2}], the surface tension σ [Nm^{-1}], the infiltration velocity v [ms^{-1}], the applied pressure gradient $\frac{dP}{dx}$ [Nm^{-3}] and some characteristic parameter of the powder compact such as the capillary length l [m] or the particle diameter D [m]. In the present analysis we choose the pressure gradient instead of the pressure because actually the pressure gradient is the driving force of the infiltration process. All five parameters of the infiltration process are expressed through three basic dimensions, namely, [m], [kg] and [s]. According to the π-theorem of the dimensional analysis, it thus can be stated that the number of dimensionless complexes, which can be constructed from the above parameters is two. Here, we skip the detailed description of the procedure and write directly the final result: $\frac{\eta v}{\sigma}$ and $\frac{l^2}{\sigma}\frac{dP}{dx}$. The combination of these complexes lead to the general dependence that describes the infiltration kinetics:

$$\frac{\eta v}{\sigma} = f\left(\frac{l^2}{\sigma}\frac{dP}{dx}\right) \qquad (1)$$

Thus we have two criterions which characterise the infiltration kinetics. The first, $\frac{\eta v}{\sigma}$, gives the relation between the friction and capillary forces and is not of special interest to our analysis. The second complex is the criterion for which we are looking for: it relates both driving forces of the infiltration process, namely the pressure gradient and the surface tension. In case that $\frac{l^2}{\sigma}\frac{dP}{dx} >> 1$ the infiltration process is driven by the pressure gradient; if $\frac{l^2}{\sigma}\frac{dP}{dx} << 1$, the surface tension force is dominant. In the latter case the capillary forces drive the infiltration process. It can be easily shown that the role of the surface tension for the infiltration during pressing of powders is negligible.

The equation describing the infiltration kinetics can be deduced from Eq.(1). Expand Eq.(1) into Taylor series and keep only the linear terms:

$$\frac{\eta v}{\sigma} = f(0) - f'(0)\frac{l^2}{\sigma}\frac{dP}{dx} + O^2 \qquad (1a)$$

At $\frac{dP}{dx} = 0$ the infiltration velocity is equal to zero (v=0), thus it follows that $f(0) = 0$ and Eq.(1a) can be rewritten as:

$$\frac{\eta v}{\sigma} = -f'(0)\frac{l^2}{\sigma}\frac{dP}{dx} \qquad (2)$$

Rearranging the members one obtains:

$$v = -f'(0)\frac{l^2}{\eta}\frac{dP}{dx} = -\frac{\alpha}{\eta}\frac{dP}{dx} \qquad (3)$$

The negative sign in the above equations indicates that the vectors of the pressure gradient and the infiltration velocity have opposite directions. Eq.(3) is the model equation for which we are looking for: it is known as Darcy's law (equation) and describes the infiltration kinetics into porous media [10]. The coefficient α is called permeability coefficient. The latter is a function depending on the packing characteristics of the porous media e.g. from the fractional density *f* and the dimensions of the powder particles *D* [10]. It can be noted that no general dependence of the permeability coefficient as a function of *f* and *D* is established until now. However it is well known that permeability is very sensitive to the porosity and the pore size. Small changes in either characteristic will induce major changes in the permeability coefficient.

In the present paper the Darcy's equation is applied to three different kinds of deposition of the resin: deposits with a planar, a cylindrical and spherical (droplets) geometry. For the sake of simplicity in the carried below analysis (for the all three cases studied) the following simplifying assumptions are made:

-pressure gradient is assumed to be a constant;

-fractional density *f* of the powder compact is a constant during the infiltration.

a. Planar geometry of the resin deposit

In this case it is assumed that the resin is deposited in the form of a layer with thickness

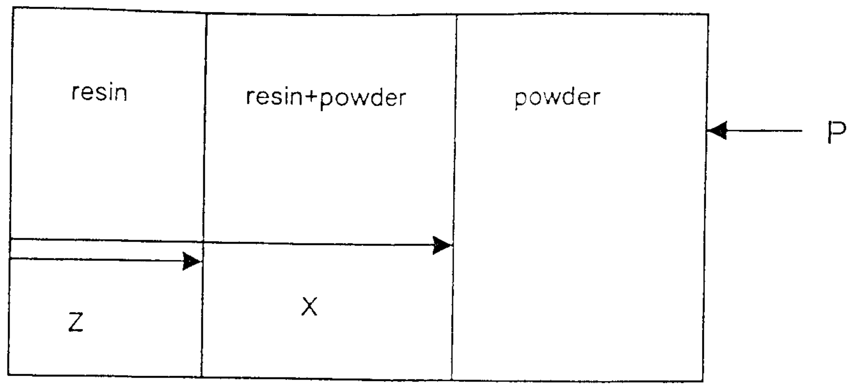

Fig. 1 Scheme of the geometry used in the model. The co-ordinate *x* shows the farthest distance reached by the resin at the time moment *t*. Co-ordinate z respectively, shows the resin-powder boundary at this time. At the right side is shown the direction of the applied pressure.

two equations: the kinetic (Darcy's) equation and the mass balance equation for the resin. The used below symbols are shown in Fig. 1:

$$\frac{dx}{dt} = \frac{\alpha}{\eta} \frac{P}{x - z} \tag{4a}$$

$$(1 - f)(x - z) + z = z_0 \tag{4b}$$

Here *x* is the co-ordinate of the farthest distance reached by the resin at time *t*, *z* is the co-ordinate of the resin-powder boundary at the same time and z_0 is the resin-powder boundary at the beginning of the infiltration process.
Solving Eq.(4b) with respect to *z* and substituting the result into Eq.(4a) after uncomplicated transformations one obtains:

$$\frac{dx}{dt} = \frac{\alpha f}{\eta} \frac{P}{x - z_0}$$

The integration of this equation yields:

$$x = z_0 + \sqrt{\frac{2\alpha f P}{\eta}}\, t^{1/2}$$

Taking into account that at uniform distribution of the resin between the powder particles the co-ordinate of the farthest distance reached by the resin is $x_{end} = \frac{z_0}{1-f}$, the total time required to infiltrate a powder layer with thickness $x_{end} = \frac{z_0}{1-f}$ and fractional density *f* is:

$$t_{tot} = \Phi_{plane}(f) \frac{\eta z_0^2}{\alpha P} \tag{5}$$

with $\Phi_{plane}(f) = \frac{1}{2} \frac{f}{(1-f)^{1/2}}$.

b. Cylindrical geometry of the resin deposit

In this case it is assumed that the resin is deposited in the form of an infinite cylinder with radius R_0 surrounded with powder. The Darcy's equation and the mass balance equation for the resin are in this case:

$$\frac{dr}{dt}=\frac{\alpha}{\eta}\frac{P}{r-R} \quad (6a)$$

$$(1-f)(r^2-R^2)+R^2=R_0^2, \quad (6b)$$

with r the co-ordinate of the farthest distance reached by the resin at time t, R the co-ordinate of the resin-powder boundary and R_0 the resin-powder boundary at the beginning of the infiltration process. The substitution of Eq.(6b) into Eq.(6a) leads to the following differential equation:

$$\frac{dr}{dt}=\frac{\alpha P}{\eta}\frac{1}{r-\left[\frac{R_0^2-(1-f)r^2}{f}\right]^{1/2}}.$$

The solution of this equation yields for the total time of infiltration:

$$t_{tot}=\Phi_{cylinder}(f)\frac{\eta R_0^2}{\alpha P} \quad (7)$$

with $\Phi_{cylinder}(f)=\frac{f}{2(1-f)}+\frac{1}{2}\left\{1-\frac{1}{[f(1-f)]^{1/2}}\left[\frac{\pi}{2}-\arcsin\left[(1-f)^{1/2}\right]\right]\right\}$.

c. Spherical geometry of the resin deposit (droplets)

In this case it is assumed that the resin is deposited in the form of sphere (droplet) with radius R_0 surrounded with powder. The Darcy's equation and the mass balance equation for the resin in this case read as:

$$\frac{dr}{dt}=\frac{\alpha}{\eta}\frac{P}{r-R} \quad (8a)$$

$$(1-f)(r^3-R^3)+R^3=R_0^3 \quad (8b)$$

with r the co-ordinate of the farthest distance reached by the resin at time t, R the co-ordinate of the resin-powder boundary and R_0 the resin-powder boundary at the beginning of the infiltration process. The substitution of Eq.(8b) into Eq.(8a) leads to the following differential equation:

$$\frac{dr}{dt}=\frac{\alpha P}{\eta}\frac{1}{r-\left[\frac{R_0^3-(1-f)r^3}{f}\right]^{1/3}}.$$

Unfortunately this equation cannot be solved analytically. In this case we solved it numerically. The obtained solution has been treated with the aid of non-linear regression analysis. The algebraic function with highest correlation coefficient has been chosen to describe the solution of the above equation. The total time of infiltration in this case is given by:

$$t_{tot}=\Phi_{sphere}(f)\frac{\eta R_0^2}{\alpha P} \quad (9)$$

with $\Phi_{sphere}(f)=\frac{-9.1*10^{-5}+0.084f-0.102f^2+0.01967f^3}{1-2.39f+1.803f^2-0.413f^3}$ as algebraic function obtained by the non-linear regression.

As mentioned above the value of the permeability coefficient α is of crucial importance to the applicability of the model. In the next section we tested some of the most frequently used permeation coefficients and their applicability to the discussed here case of resin infiltration.

EXPERIMENTAL RESULTS AND DISCUSSION

An experiment was carried out in order to check the validity of the obtained above theoretical results. Three specimens have been prepared. The specimen were with rectangular form (100x15x10 mm) and have green fractional density of about f=0.6. Spherical droplets with different size (diameters of 3, 5 and 7 mm) have been put in the centre of the green powder compact. The so prepared specimens have been kept under pressure (750 MPa) for 30 s each. It was estimated that the mean fracional density of the compact during the pressing process should be $\bar{f}$=0.9. The obtained results are shown in Figs. 2, 3 and 4. The photos have been taken at the farthest boundary reached by the resin, at the centre of the droplet and at half the distance between the centre and the boundary reached by the resin. For the smallest droplet (the one with diameter 3mm) photos have been taken only at the centre and the farthest boundary. The infiltration of the resin (black) through the powder compact (white) is clearly visible. One easily notes the structure difference between the droplets with the different diameters and the structure difference along the specimens. The obtained results can be summarised as follows:

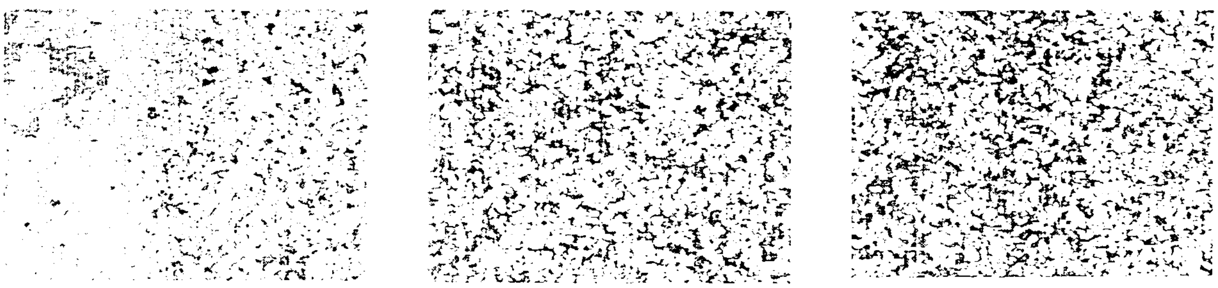

Fig.2a Fig.2b Fig.2c

Fig.2 SEM micro-graphs of the resin distribution after pressing with 750 MPa for 30s. The initial diameter of the deposit was 7mm. The photos have been taken at the farthest boundary reached by the resin (Fig.2a), at half the distance between the centre and the boundary reached by the resin (Fig.2b) and at the centre of the droplet (Fig.2c).

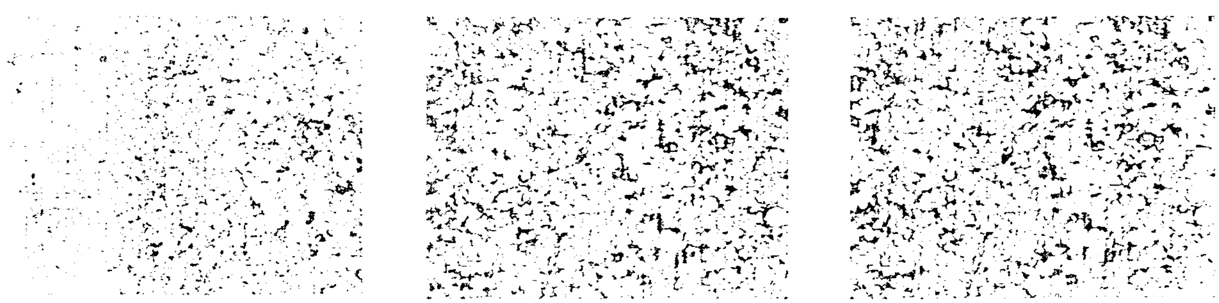

Fig.3a Fig.3b Fig.3c

Fig.3 SEM micro-graphs of the resin distribution after pressing with 750 MPa for 30s. The initial diameter of the deposit was 5mm. The photos have been taken at the farthest boundary reached by the resin (Fig.3a), at half the distance between the centre and the boundary reached by the resin (Fig.3b) and at the centre of the droplet (Fig.3c).

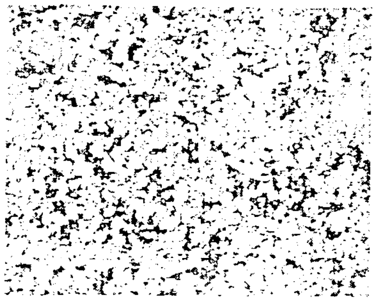

Fig.4a

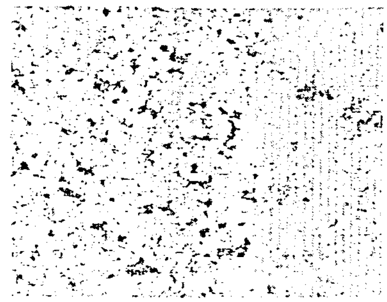

Fig.4b

Fig.4 SEM micro-graphs of the resin distribution after pressing with 750 MPa for 30s. The initial diameter of the deposit was 3mm. The photos have been taken at the farthest boundary reached by the resin (Fig.4b), and at the centre of the droplet (Fig.4a).

-The fractional density of the resin in the centre of the specimens is lower. This is due to the fact that the pressurisation time was not long enough to lead to uniform distribution of the resin among the powder particles. Another explanation of this behaviour is that the fractional density of the powder at the resin-powder boundary becomes too high due to the high pressure. This can lead to closure of the capillaries between the powder particles and thus according to Eqs. (5), (7) and (9) to a high flow resistivity.
-It is clearly seen that the initial radius of the deposited droplet is of significant importance for the uniform infiltration of the resin. This is also predicted by our model (see Eq.(9)). The fractional density at the centre of the droplet with larger initial radius (7mm) is lower than at the centre of the ones with smaller radiuses. Therefore, in order to obtain uniform infiltration of the resin among the powder particles it can be recommended the radius of the deposited droplets to be kept as small as possible. This fact is confirmed from the obtained by us results, which predict that the infiltration time is proportional to the square of the deposit thickness (see Eqs.(5), (7) and (9)).
-The value of the permeability coefficient α is of crucial importance to the applicability of the model. In Table 1 are compared the total infiltration times required for evenly distribution of the resin between the powder particles predicted by different permeability models. It is evident that all of the permeability dependencies used underestimate the infiltration time. As seen from the micrographs in Figs. 2,3 and 4 the resin is not evenly distributed for the pressing time of 30s used in the present research. This is probably due to the fact that the used here dependencies for the permeability coefficient have been obtained empirically for media's different from the one studied by us (metallic powder). Thus, more investigations are needed in order to obtain proper permeability dependence. However the above permeation dependencies, especially the first one, can be successfully used for the estimation of the infiltration time.

Table 1 Comparison of the total infiltration times t_{tot} [s] according to different permeation models [7-9]. The following values for the involved physical parameters have been used: D=60μm, η=0.6 Pas, f=0.9, R_0=1.5mm and P=750 MPa.

α	$\alpha(0.9)$	t_{tot} [s]	Reference
$4.6\,10^{-11}\,D^{0.73}\,(1-f)^{5.8}$	$1.448\,10^{-16}$	0.11	German [8]
$4.8\,10^{-13}\,D^{1.3}\,(1-f)^{4.8}$	$1.556\,10^{-15}$	0.01	Smith and Marth [9]
$10^{-14}\,D^{2}\,(1-f)^{4}f^{-2}$	$4.44\,10^{-14}$	$3.4\,10^{-4}$	German [7]

-As mentioned above the geometry of the deposit leads to crucial difference of the with different geometries are related as follows: $t_{plane}:t_{cylinder}:t_{sphere}$= 22:7:0.1s. It is seen that depending on the deposit's geometry, the infiltration time may vary up to two orders of magnitude.

CONCLUSIONS

The results can be summarised as follows:

- A model of the infiltration kinetics during production of SMC is proposed, based on the classical Darcy's equation. The equation combines most important physical parameters of the process.
- According to the obtained criterion the influence of the surface tension of the resin on the infiltration kinetics is negligible.
- The permeability coefficients used here considerably underestimate the infiltration time. This is probably due to the fact that they are obtained for media's different from the one studied here. In order to better match the predicted and the real infiltration time, additional research is needed in this direction.
- The proposed model predicts that the most optimal way of resin deposition is to disperse the latter as small droplets into the powder. This considerably reduces the infiltration time and leads to a more uniform distribution of the resin into the compact.

In the discussed case here, $\frac{l^2}{\sigma}\frac{dP}{dx} >> 1$, the infiltration process is driven by the pressure gradient. Thus, the omission of the terms with higher order in the Taylor expansion in Eq. (1a) is not fully correct. In this case one should also keep the terms with a higher order, depending on the value of the expansion coefficients in front of the different expansion members. Unfortunately, this would lead to additional coefficients in the kinetic equation. Therefore, additional experimental work is needed in order to further improve of the description of kinetics of resin infiltration into powders.

Acknowledgement

The authors would like to thank FWO (Fonds voor Wetenschappelyk Onderzoek Vlaanderen), project No. 9.0420.99, for the financial support of the present research.

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