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DEVELOPMENT AND EVALUATION OF DENSIFICATION MODELS FOR Al-Pb ALLOYS PRODUCED BY POWDER METALLURGY

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ABSTRACT

The recent research into the alternate sliding bearing materials unequivocally point to the beneficial role of lead (Pb) in aluminum (Al). But, these alloys offer a manufacturing challenge, due to wide immiscibility gap. For such applications, apart from homogeneous microstructure, porosity control is also equally important, as it influences the tribological performance through spreading of lead in aluminum matrix. These objectives can be achieved together by mechanical alloying through powder metallurgy (P/M). However, die compaction in P/M results in inhomogeneous density distributions leading to distortion during sintering process. In order to control the shape of final P/M parts, the appropriate models are necessary for densification of composite powders to simulate cold compaction responses. In the present work, the authors studied the effect of alloy composition and ball to charge ratio on densification behaviour of conventional ball milled Al-Pb alloys, with a view to develop non-linear regression models that can best describe the densification behaviour of ball milled Al-Pb alloys from compressibility test data. Based on their validation, the study concludes that Richards model effectively predicts densification behaviour of P/M processed Al-Pb alloys. Such modeling will be of great significance in simulating the cold compaction response, especially in case of alloys where no historical data exists regarding effect of processing conditions on densification behaviour.

Key words: *Al-Pb Alloys, Compressibility Test, Densification Modeling, Powder Metallurgy.*

1. Introduction

In the manufacturing technologies of P/M products, die compaction is widely used. However, P/M parts formed by die compaction have inhomogeneous density distributions due to the friction between the powder and the die well. Inhomogeneity in density leads to non-uniform shrinkage or distortion during sintering process, and thus makes it difficult to control the shape of final P/M parts [1]. In order to control the shape of final P/M parts, the appropriate models are necessary for densification of composite powders to simulate cold compaction responses. Theories describing the deformation of porous material are enumerated and the changes in density in powder forming process are analyzed with the help of a theoretical and numerical analysis by Bruhns and Sluzalec [2] to model the densification behaviour of porous materials.

A number of yield functions have been developed for densification behaviour of metal powder, so far. Kuhn and Downey [3], Shima and Oyane [4], and Doraivelue et al [5] proposed yield functions based on uniaxial tests on sintered powder compacts. Brown and Weber [6], however, showed that densification behaviour of loose powder is different from that of sintered powder in uniaxial tests. The discrepancy in densification behaviour is more important, in the early stage of compaction, because the inter-particle cohesion of loose powder compact is much lower than that of a sintered powder compact. Thus, these models [3-5] may not be appropriate for the early stage of compaction response. A macroscopic constitutive model from particle deformations was proposed by Fleck et al. [7]. However, this yield function did not agree well with experimental data of soft metal powder during die compaction [8].

A number of researchers also adopted models, for densification of metal powder from soil mechanics. These models include Drucker-prager model [9], cam-clay model [10], and cap models. Out of them, the cap model may be useful to predict densification behaviour of loose soft metal powder in all stages of compaction, because this model based on experimental data of lose powder incorporates densification due to inter-particle movements as well as particle's plastic deformation.

The hyperbolic cap model proposed by Lee and Kim [1] agreed well with the experimental data in the overall density region under cold compaction of Aluminum alloy powder. Whereas the finite element

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based models such as Shima –Oyane model and other models underestimated the experimental data in the low density region and in the high density region respectively. In view of the limitations in the existing densification models [2-10] an attempt is made in this work, to apply the techniques of non-linear regression models, for the experimental data generated on Al-Pb alloys.

2. Experimental Details

The Al and Pb powders of various compositions, shown in Table 1 are used in the present work. Their particle distributions are presented in Figure 1(a) and Figure 1 (b). These metal powders are mixed in conventional ball mill operated at 200 RPM.

Table 1: Composition of Metal Powders

Sl. No.	Al (wt. %)	Pb (wt. %)
1	95	5
2	90	10
3	85	15
4	80	20
5	75	25

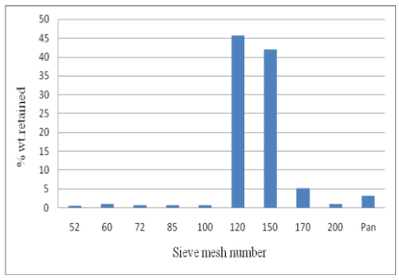

Fig. 1(a) Particle Size Distribution of Al Powder

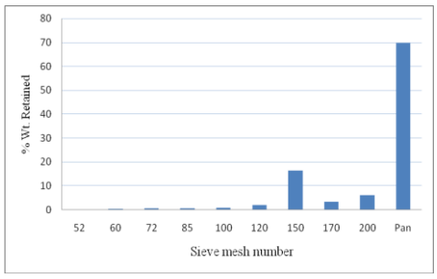

Fig. 1(b) Particle Size Distribution of Pb Powder

Samples are withdrawn from the ball mill after 45 min. of continuous run and subjected to compressibility test. The compressibility test is carried out on a hydraulic press using one set of die and punches that conform to cylindrical shape of test sample with 1 inch diameter. For the purpose of compressibility test, the lower punch is inserted into the die and the assembly is placed on the lower platen of the hydraulic press. Using supporting spacers which may be wooden blocks, the die is raised so that the depth of powder fill will be proper to give a green compact height equal to the compact diameter + or – 10%.The powder depth varies with the particular powder used, and was determined empirically. Sufficient quantity of alloy powder is poured into the die cavity to fill it to overflowing. Then, the powder is leveled off flush with the top of the die by passing the straight edge of the spatula horizontally across the die top. The die is tapped a little to make the powder level settle slightly below the die top. The upper punch is positioned so that it rests in the die and on top of the powder. A preliminary force of 0.2 Kgf is applied and then removed, after which the spacers are removed. Then, a force of 500Kgf is applied, building up to this amount at a constant rate of loading. The applied force is gradually withdrawn and the green compact is ejected by use of a die push – down spacer placed between the die top and the bottom of the platen above it. Subsequently, the height of the green compact is measured, to calculate the volume of the green compact. The weight of the compact is determined to the nearest 0.001 gram, to calculate the green density of the compact. These steps are repeated, using appropriately larger amounts of powder and final compacting forces of 1,000, 1,500, 2,000, 2,500, 3,000, 3,500, 4,000, 4500 kgf etc. The compressibility test results of Al-Pb alloys are shown in Table 2.

3. Modeling of Densification Behavior

3.1 Model selection

Given a set of data points, from the compressibility test, the data is fitted to a model in the form of a parametric equation. One important consideration in selecting an appropriate model is the underlying law that the data represents. Optimally, the model should be chosen to reflect that law so that the parameters in the curve fit have physical interpretation and meaning. In the work presented here, the nonlinear models known as the sigmoidal family models are selected based on the nature of the experimental data.

The sigmoidal or "S-shaped" growth curves (namely Gompertz Model, Logistic Model, Richards Model, MMF Model and Weibull Model) start at a fixed point and increase their growth rate monotonically to reach an inflection point. After this, the growth rate approaches a final value asymptotically. These five models are fitted to the experimental data pertaining to various compositions of Al-Pb alloys made by

conventional ball milling, at two different ball to charge ratios. The ball to charge ratios used in the experiments are selected based on general engineering practice for such applications as well as the volume capacity of the attrition mill.

3.2 Solution methodology

This program uses the Levenberg-Marquardt method to solve nonlinear regression. This method combines the steepest-descent method and a Taylor series based method to obtain a fast, reliable technique for nonlinear optimization. The Levenberg-Marquardt (LM) algorithm allows for a smooth transition between these two methods as the iteration proceeds. In general, the data modeling equation (with one independent variable) can be written as follows:

$$Y=Y(x, \bar{a}) \quad (1)$$

The above expression simply states that the dependent variable y can be expressed as a function of the independent variable x and vector of parameters a of arbitrary length. Note that using the ML method, any nonlinear equation with an arbitrary number of parameters can be used as the data modeling equation. Then, the "merit function" we are trying to minimize is

$$\chi^2(\bar{a}) = \sum_{i=1}^{N} [(Y_i - Y(X_i, \bar{a})) / \sigma_i]^2 \quad (2)$$

Where N is the number of data points, xi denotes the x data points, yi denotes the y data points, σ_i is the standard deviation (uncertainty) at point i, and y(xi,a) is an arbitrary nonlinear model evaluated at the i_{th} data point. This merit function simply measures the agreement between the data points and the parametric model; a smaller value for the merit function denotes better agreement. This merit function is also called as the chi-square. The Taylor series method states that sufficiently close to the minimum, the function can be approximated as a quadratic. Without detailed explanation of that method, a step from the current parameters $\bar{a}_{cur}$ to the best parameters $\bar{a}_{min}$ can be written as

$$\bar{a}_{min} = \bar{a}_{cur} + H^{-1} [-\nabla \chi^2 (\bar{a}_{cur})] \quad (3)$$

Where H is the Hessian matrix (a matrix of second derivatives). If the approximation of the function as a quadratic is a poor one, then we might instead use the steepest-descent method, where a step to the best parameters from the current parameters is

$$\bar{a}_{min} = \bar{a}_{cur} - C \nabla \chi^2 (\bar{a}_{cur}) \quad (4)$$

This equation simply states that the next guess for the parameters is a step down the gradient of the merit function. The constant C is forced to be small enough that a small step is taken and the gradient is accurate in the region that the step is taken.

Since we know the chi-square function, we can directly differentiate to obtain the gradient vector and the Hessian matrix. Taking the partial derivatives of the merit function with respect to a gives

$$\frac{\partial \chi 2}{\partial a_k} = -2 \sum_{i=1}^{N} \left[\frac{Y_i - Y(x_i ; \bar{a})}{\sigma_i^2} \cdot \frac{\partial Y(x_i ; \bar{a})}{\partial a_k} \right] \quad (5)$$

To obtain the Hessian matrix, take the gradient of the gradient above (so that we have a matrix of partial second derivatives)

$$\frac{\partial \chi 2}{\partial a_k \partial a_l} = -2 \sum_{i=1}^{N} \left[\frac{1}{\sigma_i^2} \cdot \frac{\partial Y(x_i ; \bar{a})}{\partial a_k} \cdot \frac{\partial Y(x_i ; \bar{a})}{\partial a_l} - \frac{Y_i - Y(x_i ; \bar{a})}{\sigma_i^2} \cdot \frac{\partial^2 Y(x_i ; \bar{a})}{\partial a_k \partial a_l} \right] \quad (6)$$

Now, for convenience, define the gradient vector and the curvature matrix as

$$G_K = -\frac{1}{2} \frac{\partial \chi 2}{\partial a_k} = \sum_{i=1}^{N} \left[\frac{Y_i - Y(x_i ; \bar{a})}{\sigma_i^2} \cdot \frac{\partial Y(x_i ; \bar{a})}{\partial a_k} \right] \quad (7)$$

$$C_{kl} = \frac{\partial^2 \chi 2}{\partial a_k \partial a_l} = \sum_{i=1}^{N} \left[\frac{1}{\sigma_i^2} \cdot \frac{\partial Y(x_i ; \bar{a})}{\partial a_k} \cdot \frac{\partial Y(x_i ; \bar{a})}{\partial a_l} \right] \quad (8)$$

Table 2: Compressibility test results of conventional ball milled Al-Pb alloys

Pressure Mpa	Density (Gr/Cm^3)									
	95Al-5Pb alloys		90Al-10Pb alloys		85Al-15Pb alloys		80Al-20Pb alloys		75Al-25Pb alloys	
	BCR = 5:1	BCR =10:1	BCR = 5:1	BCR =10:1	BCR = 5:1	BCR =10:1	BCR = 5:1	BCR =10:1	BCR = 5:1	BCR = 10:1
10	1.312	1.575	1.669	1.673	1.752	1.735	1.865	1.804	1.808	1.988
20	1.389	1.60	1.713	1.730	1.807	1.779	1.932	1.886	1.839	2.054
30	1.425	1.629	1.754	1.784	1.847	1.851	1.990	1.964	1.979	2.103
40	1.498	1.677	1.819	1.856	1.945	1.928	2.046	2.028	2.015	2.162
50	1.538	1.738	1.872	1.918	2.011	2.003	2.099	2.097	2.103	2.259
60	1.605	1.798	1.947	1.992	2.052	2.048	2.168	2.204	2.174	2.335
70	1.649	1.851	1.993	2.037	2.106	2.135	2.226	2.273	2.25	2.406
80	1.699	1.89	2.05	2.103	2.179	2.203	2.293	2.327	2.307	2.472
90	1.715	1.949	2.105	2.152	2.237	2.25	2.348	2.386	2.361	2.541
100	1.765	2.034	2.155	2.146	2.287	2.323	2.401	2.455	2.429	2.589
110	1.809	2.039	2.189	2.269	2.319	2.363	2.489	2.491	2.492	2.637
120	1.867	2.059	2.264	2.292	2.372	2.413	2.559	2.541	2.571	2.677

Table 3: A Comparison of Densification Models for Ball milled Al-Pb Alloys

Sl.No.	Alloy composition	Densification Model	Coefficients	Standard error (S)	Coefficient of correlation (r)	Comments
1.	95Al-5Pb (ball to charge ratio=5:1)	Gompertz Relation: y=a*exp(-exp(b-cx))	a = 2.384449, b = -0.446123, c = 0.00774622	0.01196878	0.99809979	The fit converged to a tolerance of 1e-006 in 30 iterations. No weighting used
		Logistic Model: y=a/(1+b*exp(-cx))	a = 2.2470045, b = 0.7827956, c = 0.0108170	0.0122433	0.9980115	The fit converged to a tolerance of 1e-006 in 12 iterations. No weighting used
		MMF Model: y=(a*b+c*x^d)/(b+x^d)	a = 1.233188, b = 411.95285, c = 5.8028301.	0.0121162	0.9982692	The fit converged to a tolerance of 1e-006 in 26 iterations. No weighting used.
		Weibull Model: y=a-b*exp(-c*x^d)	a = 3.0046159, b = 1.7624897, c = 0.0049914, d = 0.9286785	0.0122362,	0.9982347,	The fit converged to a tolerance of 1e-006 in 100 iterations. No weighting used.
2.	95Al-5Pb (ball to charge ratio=10:1)	Logistic Model: y=a/(1+b*exp(-cx))	a = 3.4394767, b = 1.2909257, c = 0.0057404523	0.0215148	0.9939606	The fit converged to a tolerance of 1e-006 in 55 iterations. No weighting used.
		Richards Model: y=a/(1+exp(b-cx)^(1/d))	a = 2.052706, b = 39.213857, c = 0.3657327, d = 126.89219	0.0153676	0.9972656	The fit converged to a tolerance of 1e-006 in 23 iterations. No weighting used.
3.	90Al-10Pb (ball to charge ratio=5:1)	Logistic Model: y=a/(1+b*exp(-cx))	a = 3.5803115, b = 1.2340121, c = 0.00621108	0.0103343	0.9988791 .	The fit converged to a tolerance of 1e-006 in 51 iterations. No weighting used.
		Richards Model: y=a/(1+exp(b-cx)^(1/d))	a = 2.502839, b = 4.1727808, c = 0.0303377, d = 9.4789474	0.0104810,	0.9989752,	The fit converged to a tolerance of 1e-006 in 18 iterations. No weighting used
4.	90Al-10Pb (ball to charge ratio=10:1)	Gompertz Relation: y=a*exp(-exp(b-cx))	a = 3.1580973, b = -0.3842547, c = 0.0062701	0.0201725	0.9960780	The fit converged to a tolerance of 1e-006 in 48 iterations. No weighting used.

		Logistic Model: y=a/(1+b*exp(-cx))	a = 2.9138955, b = 0.82027625, c = 0.009190747	0.0201681	0.9960797	The fit converged to a tolerance of 1e-006 in 22 iterations. No weighting used.
		MMF Model: y=(a*b+c*x^d)/(b+x^d)	a = 1.6156402, b = 582.20585, c = 3.7683579, d = 1.165871	0.0211842	0.9961554	The fit converged to a tolerance of 1e-006 in 87 iterations. No weighting used.
		Weibull Model: y=a-b*exp(-c*x^d)	a = 3.0148363, b = 1.4017246, c = 0.0030244, d = 1.1247359	0.0212466	0.9961327	The fit converged to a tolerance of 1e-006 in 86 iterations. No weighting used.
5.	85Al-15Pb (ball to charge ratio = 5:1)	Gompertz Relation: y=a*exp(-exp(b-cx))	a = 3.2529345, b = -0.406826, c = 0.00625533	0.0129936	0.9984171	The fit converged to a tolerance of 1e-006 in 50 iterations. No weighting used.
		Logistic Model: y=a/(1+b*exp(-cx))	a = 3.0062812, b = 0.7961878, c = 0.00913700	0.0127534	0.9984751	The fit converged to a tolerance of 1e-006 in 22 iterations. No weighting used.
		Richards Model: y=a/(1+exp(b-cx)^(1/d))	a = 2.4995539, b = 3.9725305, c = 0.0355696, d = 10.166325	0.0127354	0.9986485,	The fit converged to a tolerance of 1e-006 in 58 iterations. No weighting used.
		MMF Model: y=(a*b+c*x^d)/(b+x^d)	a = 1.7178972, b = 1165.7997, c = 3.0707762, d = 1.4597335	0.0122039	0.9987590	The fit converged to a tolerance of 1e-006 in 55 iterations. No weighting used.
		Weibull Model: y=a-b*exp(-c*x^d)	a = 2.6826822, b = 0.9699176, c = 0.0017231, d = 1.3545622.	0.0122700	0.9987455	The fit converged to a tolerance of 1e-006 in 97 iterations. No weighting used.
6.	85Al-15Pb (ball to charge ratio = 10:1)	Gompertz Relation: y=a*exp(-exp(b-cx))	a = 3.5461106, b = -0.2639019, c = 0.0058341195.	0.0117797	0.9989389	The fit converged to a tolerance of 1e-006 in 58 iterations. No weighting used.
		Logistic Model: y=a/(1+b*exp(-cx))	a = 3.187259, b = 0.93490289, c = 0.009019316.	0.0113370	0.9990172	The fit converged to a tolerance of in 26 iterations. No weighting used.
		Richards Model: y=a/(1+exp(b-cx)^(1/d))	a =2.5348328, b = 4.7052507, c = 0.041548986, d = 11.198612	0.0097946	0.9993480	The fit converged to a tolerance of in 12 iterations. No weighting used.
		MMF Model: y=(a*b+c*x^d)/(b+x^d)	a = 1.7023093, b = 1656.6775, c = 3.1262541, d =1.5479796.	0.0089810	0.9994519	The fit converged to a tolerance of in 57 iterations. No weighting used
7.	80Al-20Pb (ball to charge ratio= 5:1)	Richards Model: y=a/(1+exp(b-cx)^(1/d))	a = 3.0297479, b = 8.0406403, c = 0.0450205, d = 15.827954	0.0086652	0.9994509	The fit converged to a tolerance of 1e-006 in 25 iterations. No weighting used
8.	80Al-20Pb (ball to charge ratio = 10:1)	Gompertz Relation: y=a*exp(-exp(b-cx))	a = 3.2166758, b = -0.45728495, c = 0.0083337461	0.0124989	0.9989550	The fit converged to a tolerance of 1e-006 in 26 iterations. No weighting used.
		Logistic Model: y=a/(1+b*exp(-cx))	a = 3.0373077, b = 0.77418514, c = 0.011594177	0.0118826	0.9990556	The fit converged to a tolerance of 1e-006 in 12 iterations. No weighting used.
		Richards Model: y=a/(1+exp(b-cx)^(1/d))	a = 2.655366, b = 3.572541, c = 0.035945342, d = 8.4353705	0.0105830	0.9993342	The fit converged to a tolerance of in 38 iterations. No weighting used
		MMF Model: y=(a*b+c*x^d)/(b+x^d)	a = 1.7659829, b = 955.60305, c = 3.2247146, d = 1.4610421	0.0111686	0.9992584	The fit converged to a tolerance of in 70 iterations. No weighting used.
9.	75Al-25Pb (ball to charge ratio = 5:1)	Richards Model: y=a/(1+exp(b-cx)^(1/d))	a =4.9266647, b = 11.310197, c =0.03941985,	0.0467050	0.9906603	The fit converged to a tolerance of 1e-006 in 84 iterations. No weighting used.

			d =10.866504			
10.	75Al-25Pb (ball to charge ratio = 10:1)	Logistic Model: y=a/(1+b*exp(-cx))	a =3.4197151, b = 0.8057931, c =0.00911015	0.0161802	0.9981114	The fit converged to a tolerance of 1e-006 in 24 iterations. No weighting used.
		Richards Model: y=a/(1+exp(b-cx)^(1/d))	a =2.7211645, b = 6.9937057, c = 0.0653764, d =19.944137	0.0105317	0.9992892	The fit converged to a tolerance of 1e-006 in 4 iterations. No weighting used
		MMF Model: y=(a*b+c*x^d)/(b+x^d)	a =1.9794184, b = 5436.2893, c = 3.0548515, d = 1.9267836	0.0082975	0.9995588	The fit converged to a tolerance of 1e-006 in 76 iterations. No weighting used.
		Gompertz Relation: y=a*exp(-exp(b-cx))	a =3.7220418, b = -0.3906493, c = 0.0061553974,	0.0167863	0.9979672	The fit converged to a tolerance of 1e-006 in 50 iterations. No weighting used.

Table 4: A Comparison between Best Fit Densification Models for Al-Pb Alloys

Sl.No.	Composition of Alloys	Particulars of best fit Densification Models	
		By min. value of Std. error (S)	By max. value of Coeff.of correlation (r)
1	95Al-5Pb (ball to charge ratio=5:1)	Gompertz Relation: S=0.01196878	MMF Model: r = 0.9982692
2	95Al-5Pb (ball to charge ratio=10:1)	Richards Model: S = 0.0153676	Richards Model: r = 0.9972656
3	90Al-10Pb (ball to charge ratio=5:1)	Logistic Model: S= 0.0215148	Richards Model: r = 0.9989752
4	90Al-10Pb (ball to charge ratio=10:1)	Logistic Model: S= 0.0201681	MMF Model: r =0.9961554
5	85Al-15Pb (ball to charge ratio=5:1)	MMF Model: S= 0.0122039	Weibull Model: r = 0.998745
6	85Al-15Pb (ball to charge ratio=10:1)	MMF Model: S =0.0089810	Logistic Model: r = 0.9990172
7	80Al-20Pb (ball to charge ratio=5:1)	Richards Model: S = 0.0086652	Richards Model: r = 0.9994509
8	80Al-20Pb (ball to charge ratio=10:1)	Richards Model: S = 0.0105830	Richards Model: r = 0.9993342
9	75Al-25Pb (ball to charge ratio=5:1)	Richards Model: S=0.0467050	Richards Model: r =0.9906603
10	75Al-25Pb (ball to charge ratio=10:1)	MMF Model: S = 0.0082975	MMF Model: r =0.9995588

Table 5: Validation of Richards Model-Stage-I

(a) Conventional Ball Milled 95Al-5Pb Alloys

Compaction pressure(MPa)	Density (Gr/ Cm^3)		
	Ball to charge ratio =5:1		
	Experimental value	Model predicted value	Deviation(%)
130	2.29450	2.29685	0.102419
140	2.32395	2.33054	0.521027
150	2.36632	2.36875	0.102691
160	2.39467	2.39703	0.098552
170	2.41795	2.42042	0.102153

(b) Conventional Ball Milled 90Al-10Pb Alloys

Compaction pressure (MPa)	Density (Gr/ Cm^3)		
	Ball to charge ratio = 10:1		
	Experimental Value	Model Predicted Value	Deviation (%)
130	2.04709	2.05270	0.274048
140	2.04720	2.05271	0.269148
150	2.04720	2.05271	0.269148
160	2.04720	2.05271	0.269148
170	2.04720	2.05271	0.269148

(c) Conventional Ball Milled 80Al-20Pb Alloys

Compaction pressure (MPa)	Density (Gr/ Cm^3)					
	Ball to charge ratio =5:1			Ball to charge ratio = 10:1		
	Experimental value	Model predicted value	% Deviation	Experimental value	Model predicted value	% Deviation
130	2.60949	2.62093	0.43840	2.55947	2.56647	0.273494
140	2.66559	2.68707	0.80582	2.60870	2.59042	-0.700730
150	2.72886	2.75037	0.78824	2.62008	2.60848	-0.442730
160	2.80743	2.80907	0.05842	2.62674	2.62183	-0.186920
170	2.85934	2.86131	0.06889	2.62979	2.63154	0.066545

(d) Conventional Ball Milled 75Al-25Pb Alloys

Compaction Pressure (MPa)	Density (Gr/ Cm^3)		
	Ball to charge ratio =5:1		
	Experimental value	Model predicted value	Deviation (%)
130	2.77171	2.78775	0.578704
140	2.88348	2.89048	0.242762
150	2.98876	2.99685	0.270681
160	3.09792	3.10694	0.291163
170	3.21065	3.22076	0.314890

Table 6: Validation of Richards Model -Stage-II

Compaction Pressure(MPa)	Density (Gr/Cm^3)				
	Experimental value	Value predicted by Hyperbolic cap model by Lee and Kim	% Deviation	Value predicted by Richards model	% Deviation
20	1.674	1.661	-0.77658	1.674	0.00000
50	1.809	1.809	0.00000	1.807	-0.11056
70	1.890	1.890	0.00000	1.891	0.05291
100	1.998	2.025	1.35135	1.997	-0.05005

It can be noted that the second derivative term in C will be ignored because of two reasons: it tends to be small because it is multiplied by (y-yi), and it tends to destabilize the algorithm for badly fitting models or data sets contaminated with outliers. This action in no way affects the minimum found by the algorithm; it only affects the route in getting there. So, the Taylor series method (inverse Hessian method) can be written as the following set of linear equations:

$$\sum_{k=1}^{N_P} C_{kl}.\,\delta_{al} = G_k \qquad (9)$$

Where, N_P is the number of parameters in the model that is being optimized. This linear matrix will be our workhorse for this method after some modification; it can be solved for the increments δ_a that, when added to the current approximation for the parameters, gives the next approximation. Likewise, we can substitute our "convenient" definitions into the steepest descent formula to obtain

$$\delta_{al} = c.G_{al} \qquad (10)$$

The steepest descent method works best far away from the minimum and the Taylor series method works best close to the minimum. The Levenberg-Marquardt (LM) algorithm allows for a smooth transition between these two methods as the iteration proceeds. The first issue in deriving the LM method is to attach some sort of scale to the constant c in the steepest-gradient method (equation 10). Typically, there is no obvious way to determine this number, even within an order of magnitude. However, in this case, we have access to the Hessian matrix; examining its members, we see that the scale on this constant must be 1/Cll. But, that still may be too large, so let's divide that scale by a non-dimensional factor (l) and plan on setting this much larger than one so that the step will be reduced (for safety and stability). The second issue to formulate the LM method is noting that the steepest-descent and Taylor series methods may be combined if we define a new matrix M_{ij} by the following

$$\mathbf{M_{ii} = C_{ii}\,(1+\lambda)}$$
$$\mathbf{M_{ij} = C_{ij}\,,\quad i \neq j}$$

This matrix combines equations (9) and (10) into a convenient and compact form. So finally, we have a means of calculating the step δ_a in the parameters by the following system of linear equations

$$\mathbf{\sum_{K=1}^{Np} M_{kl}.\,\delta_{al} = G_k} \qquad (11)$$

Note that when $\boldsymbol{\lambda}$ is large, the matrix M is forced to be diagonally dominant; consequently, the above equation is equivalent to the steepest descent method (equation 10). Conversely, when the parameter $\boldsymbol{\lambda}$ goes to zero, the above equation is equivalent to the Taylor series method (equation 9). So, $\boldsymbol{\lambda}$ is varied to switch between the two methods, continually calculating a parameter correction da that we apply to our most recent guess for the parameter vector. The steps that are taken in the LM algorithm are as follows

1. Compute $\chi^2(a)$
2. Pick a conservative value for λ (0.001)
3. Solve the linear equations (equation 3) for δa
4. Evaluate $\chi^2(a+\delta a)$
5. If $\chi 2(a+\delta a) >= \chi 2(a)$, increase λ by a factor (10) and go back to step [3].
6. If $\chi 2(a+\delta a) < \chi 2(a)$, decrease λ by a factor (10), correct the parameter vector by $a=a+\delta a$, and go back to step [3].

Iteration is stopped when $| \chi 2(a+\delta a) - \chi 2(a)| <$ tolerance. This tolerance is the value specified for regression convergence. The non-linear algorithm uses this number to determine whether or not it has converged on the correct modal parameters. The difference between the standard errors in two consecutive iterations must be smaller than this tolerance for the computation to terminate. The smaller the tolerance, the more accurate the parameters will be,

but the algorithm will take more time to converge. In this work, the value of tolerance considered is 1μm.

For the models under consideration, the Coefficients, Covariance, Residuals, Coefficient of correlation and Standard error of estimate are computed. The performance of a curve fit is judged by two quantities - the correlation coefficient and the standard error of the estimate. In general, the correlation coefficient will range from 0 to 1, with a value of 1 being the best. For the alloys under consideration, the converging mathematical models are tabulated, for ball milled Al-Pb alloys in Table 3**.** The best fitting mathematical models are compared for five compositions of Al-Pb alloys and at two ball to charge ratios (BCR), in Table 4, based on maximum value of coefficient of correlation and minimum value of standard error of estimate.

From the results presented in Table 4, it can be observed that Richards model emerged as the best fit model, for a good number of the ball milled alloys, satisfying maximum value of coefficient of correlation and minimum value of standard error of estimate.

4. Validation of Densification Model

The best fitting densification model (i.e. Richards Model) is validated in two stages. In the first stage, validation is taken up by considering the data outside the range of experimentation, in the present study. The results of first stage validation are presented in Table 5(a) to (d). It can be observed from these tables, that in case of ball milled alloys, the deviation between model predicted values and experimental values of density varied from – 0.7% to 0.8%.

In the second stage, validation of Richards model is taken up by considering the experimental data generated from the presentations by Lee and Kim [1] on Aluminum alloys in cold compaction, under similar test conditions as the ones used in this work. The results obtained in this validation of Richards model are compared with the values predicted by the hyperbolic cap model developed by Lee and Kim. On the basis of the comparison shown in Table 6, it can be observed that the deviation between experimental values considered and predicted values due to Lee and Kim is in the range of -0.776% to 1.351%. Whereas the deviation observed with Richards model is in the range of -0.11% to 0.05%. This clearly demonstrates that for the composition of Al-Pb alloys and particle distribution considered in the present work, Richards model effectively predicts the density values as a function of applied compaction pressure.

5. Discussion

Powder metallurgical processing quite often yields materials containing a substantial amount of residual porosity along particle boundaries. Successful utilization of the process, therefore, needs careful control of the deformation, elimination of flash formation and, perhaps the most important, proper densification.

Theories describing the deformation of porous material are enumerated and the changes in density in powder forming process are analyzed with the help of a theoretical and numerical analysis by Bruhns and Sluzalec [2] to model the densification behaviour of porous materials.

These models [3-5] may not be appropriate for the early stage of compaction response, in view of the fact that the inter-particle cohesion of loose powder compact is much less. Out of several models developed in the past, the hyperbolic cap model proposed by Lee and Kim [1] agreed well with the experimental data in the overall density region under cold compaction of Aluminum alloy powder. Since, the processing parameters such as composition, particle distribution, ball to charge ratio, compaction pressure etc., will exhibit significant influence on densification behaviour, it is very important to develop densification models for the alloys under consideration, validate them and then use them to predict appropriate compaction pressure, so as to successfully implement P/M processing.

In this work, the modeling of compressibility test data is carried out by using non-linear regression algorithm, fitting five different sigmoidal models, for various compositions of ball milled and attrition milled alloys separately. Based on the results presented in Table-6, it can be concluded that Richards model emerged as the best fit model, for a good number of ball milled alloys satisfying maximum value of coefficient of correlation and minimum value of standard error of estimate. Further, this model is validated considering the data outside the range of experimentation and found to predict density, within ± 1 % for ball milled alloys. This is also confirmed by validating Richards model with the data generated from the presentations by Lee and Kim (2002) on Aluminum alloys in cold compaction. This approach is found quite helpful in deciding the compaction pressure for powder metallurgical processing of Al-Pb alloys of various compositions, where there are a number of variants influencing densification behaviour.

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