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A predictable glass forming ability expression by statistical learning and evolutionary intelligence

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ABSTRACT

This paper demonstrates how principal component analysis of multivariate BMG alloy data and the genetic programming of the extracted features in the form of principal components can be used to develop a meta-modeling scheme for GFA expression. The proposed GFA model can estimate the glass forming potential of an alloy from its composition data, unlike the characteristic temperature based glass forming ability expressions, consisting of T_g, $T_x$and T_l. The BMG alloys have been described by means of generic attributes of the constituent elements and corresponding composition of the alloy yielding a multi-dimensional descriptor space for a 594 BMGs compiled from literature. The PCA model of the data base plausibly reduced the dimensionality into a two dimension in terms of two extracted features by first two principle components capturing the 82% of the data knowledge. Successively, these principle components are used to develop a constitutive model for glass forming ability using genetic programming. The combinatorial analysis of the meta-model for GFA expression is applied to the prediction of potential compositional zone in five different experimentally explored ternary systems. The predicted composition zones are discussed in the context of available experimental data in literature and the energy of formation of the stable phases in respective alloy systems.

1. Introduction

The bulk metallic glasses (BMGs) offer attractive properties to consider them as potential material for many advanced technological applications [1]. However, the limited section thickness, which can be produced as glass, without significant crystallization, limits such possibilities. Many compositional design methodologies have been proposed and used to design the composition of the bulk metallic glasses, with marginal success. For instance, the compositions explored and established for high glass forming ability generally bear costly elements like Pd and rare earths [1]. Therefore, it is essential to search for low cost BMGs with high glass forming ability (GFA) to utilize the technological advantages offered by these glassy materials. The experimental exploration for the compositions with high glass forming ability is an expensive and tedious method. Therefore, as an alternative, it is proposed to predict the glass forming ability of an alloy composition, before its actual physical preparation.

The glass forming ability indicator expressions may be instrumental to formulate a better compositional design strategy. Nevertheless, different glass forming ability indicator expressions have been reported in terms of characteristic temperatures(T_g, T_x and T_l,whereT_g stands for glass transition temperature, T_x-onset temperature of crystallization and T_l is offset temperature of melting)of the metallic glass [2–21]. These characteristic temperatures based GFA indicator expressions are useful to estimate the glass forming ability once the metallic glass is physically prepared. These GFA expressions lack the predictive capability from the generic data of the alloy, *i.e.* they cannot be used to estimate the glass forming ability of a composition before actual preparation of alloy [12]. Despite of the intensive efforts from the metallic glass workers, quantifying as well as predicting the glass forming ability still remains an open question [12].

Also few alternative approaches have been reported in the literature. The Inoue's empirical rule [3], deep eutectic based compositional design [22,23], structure and topologybased approach [24–26], data training of artificial neural network [27,28] are to name a few effort made to develop a strategy for compositional design.Also the thermodynamics based approach [29–33], cluster line based approach [34–36], molecular dynamics simulation based approach [37,38] and a golden mean analysis based approach [39] has been attempted to design the BMG compositions.However, these attempts have led to limited

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success due to one or other constraints.

Nevertheless, over the past few years the data driven approachesare gaining reasonable attention of the metallic glass workers to derive more plausible solution of the problem [40–45].Therefore, development of data driven model-capable to predict glass forming ability of a composition may providebetter opportunity for combinatorial analysis to audit a wide range of species of BMG composition.In this work, a meta-modeling approach using principle component analysis to statistically model the new features,*i.e.* principal components, subsequently used as model input, to model the GFA expression using evolutionary intelligence of genetic programming has been presented.The prediction results of the proposed meta-model of GFA expression has been discussed as case study in few ternary alloy systems.

2. The meta-modeling methodology

The primary interest of this work is to employ the statistical learning ability of principle component analysis and the evolutionary intelligence of genetic programming in the meta-modeling of BMG data knowledge for evolution of GFA expression. It has been attempted to explore the relation between GFA of the alloy with the generic attribute of the constituting elements and their quantitative presence in the alloy. Eventually an eleven dimensional descriptors space has been described by means of weighted sum of the generic attribute which numerically represents the alloy [46]. The paper extends the previous work of the authors where the PCA has been applied to BMG data for developing the classification model of the BMG alloys with the atomic fraction weighted sum of the genericattributes, which includes atomic radius, electro-negativity, electron density at the boundary Wigner-Seitz cell, work function, bulk modulus, melting point, linear expansion coefficient, heat of fusion, molar heat capacity, density and thermal diffusivity [46]. The same attribute has been adapted in this work to describe these descriptors which actually take into account the thermodynamic, electronic and physical characteristics of the alloy in the form of latent variables. The principal component analysis is necessarily appropriate to this computational methodology for meta-modeling as it significantly reduced the data dimensionality thorough feature extraction without much loss of data knowledge. In this meta-modeling exercise PCA predicts the model inputs for the GFA model to develop,*i.e.*, the principal components, which latently adapts the physical, electronic and thermodynamic notion of the BMG alloy from the data knowledge. In successive progress of the computation, the first two predicted principle components of the alloy has been used to model the GFA expression which collectively relate to the generic attribute of the compositional elements of the alloys seems to be responsible for formation of glassy metallic alloys.Fig. 1 schematically illustrate the details of the computational methods for meta-modeling approach of evolution of predictable glass forming ability expression using statistical learning by PCA and evolutionary intelligence of genetic programming.

The computer implementation of the proposed meta-modeling approach is of two fold. First, a MATLAB routine has been developed in MATLAB version R2010a in order to perform the principal component analysis [47]. For the second part of the computation, a in-house computer code in FOTRAN platform has been developed for applying genetic programming [48,49] on the principal component data of the alloys to search optimal expression for estimation of GFA, in the terms of principal componentsPC_1 and PC_2. The details about the methodology of development of the algorithm for genetic programming has been explained in detail by Tripathi*et al.* [21].The process of evolution attempts to maximize the correlation of the expression under consideration in terms of PC_1 and PC_2 with the D_{max} for the 594 BMGs [46].

3. Results and discussion

3.1. Glass formation ability expression development

The proposed meta-modeling approach has been implemented for the analysis of the experimentally reported critical diameter, D_{max}, for 594 synthesized BMGs alloys. In the initial stage, the model computes the scores of the principal components (PC_1 and PC_2) which describe the alloys in two dimension at the cost of marginal loss of knowledge present in the original eleven dimensional data space as the atomic fraction weighted sum of the eleven generic attribute of the composition data of the alloys. It is worth to note here that the PC_1 and PC_2 are mutually orthogonal in multi-dimensional principle component space. Therefore, it is plausible to consider them as independent parameter to describe the data, however, their physical significance from the subject domain i.e., the principle of glass formation in the multicomponent alloy system remains unknown [46]. The predictive model for the score of the PC_1 and PC_2 evolved in principal component analysis are shown in terms of atomic fraction weighted sum of the generic attributes [46], in Equation (1) and Equation (2).

$$PC_1 = (0.1396\overline{R} - 0.3452\overline{E} - 0.3399\overline{\phi} - 0.3628\overline{n}_{WS} - 0.3595\overline{B} - 0.3459\overline{T}_m + 0.1533\overline{\alpha} - 0.3553\overline{H}_m + 0.2771\overline{C}_p - 0.2471\overline{\rho} + 0.2788\overline{k}) \quad (1)$$

$$PC_2 = (-0.6632\overline{R} + 0.1875\overline{E} - 0.1261\overline{\phi} - 0.0154\overline{n}_{WS} + 0.1127\overline{B} - 0.1232\overline{T}_m + 0.2463\overline{\alpha} + 0.0301\overline{H}_m - 0.3908\overline{C}_p - 0.4169\overline{\rho} + 0.3048\overline{k}) \quad (2)$$

where, $\overline{R}$, $\overline{E}$, $\overline{\varphi}$, $\overline{n}_{WS}$, $\overline{B}$, $\overline{T}_m$, $\overline{\alpha}$, $\overline{H}_m$, $\overline{C}_p$, $\overline{\rho}$ and $\overline{k}$ represents the atomic fraction weighted sum of the atomic radius (Å), electro negativity $(eV)^{-½}$, work function (J), electron density at the boundary of Wigner–Seitz cell (C/m^3), bulk modulus (Pa), melting point (K), linear expansion coefficient (K^{-1}), heat of fusion (J/mol), molar heat capacity (J/mol/K), density (kg/m^3) and thermal diffusivity (m^2/s) respectively.

In the latter stage the model develops the GFA expression using genetic programming with the objective to successfully describe the critical diameter, D_{max}, data for 594 synthesized BMGs alloys. The evolutionary intelligence of the genetic programming computation is performed such that it generates different expressions usingPC_1 and PC_2as input variables and calculates the coefficient of the correlation between the developed expression using input variables and the critical diameter, D_{max}, data for 594 alloys. The optimal expression of GFA in terms of PC_1 and PC_2, is presented in Equation (3) and referred as PG_p.

$$PG_p = \left[-\left\{ \frac{(PC_2)^2}{1 + PC_1 * PC_2)} \right\} * \left\{ \frac{PC_2 + 2}{PC_1} \right\} \right] \quad (3)$$

Fig. 2 (a) shows the variation of *PGp* parameter against maximum diameter (D_{max}), where as Fig. 2(b) shows the variation of prediction residue with respect to experimentally observed maximum diameter. It has been demonstrated that PG_p shows good correlation with the D_{max} data as shown in Fig. 2 (a) where the larger magnitude of negative value of PG_p corresponds to higher experimental diameter of BMGs. In Fig. 2 (b) shows the clear trend that the residue in prediction by PG_p parameter increases with higher experimentally reported diameters, i.e., in high GFA regions. It is worthy to note that correlation coefficient (R = **0.5769**) has been recorded for the PG_p (refer Equation (3)) for 594 BMGs. It is apparent that the correlation coefficient for the PG_p is not too high, however, its comparable to other characteristic temperature based GFA expression such as G_p (R = 0.6692), new- β (R = 0.5718) etc. This lower value of correlation coefficient in characteristic temperature based GFA expression can be attributed to the critical diameter data compiled from literature where the BMGs are synthesized by different techniques. The PG_p is not only efficient to estimate the glass forming ability of metallic glass compositions, but it