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# CLASSIFICATION OF HIGH-TEMPERATURE SUPERCONDUCTING *YBCO* THIN FILMS BY FUZZY CLUSTERING

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The present paper deals with the classification of high-temperature superconducting thin films (HTS) by the use of fuzzy clustering. A dataset from 84 YBCO thin films obtained by chemical vapour deposition technology was treated by the fuzzy approach. The films described were summarized by their input (oxygen pressure, chemical precursors, substrate parameters) and output (critical temperature, critical current) physical characteristics. The aim of the study was to confirm a previous classification of the films by crisp cluster analysis, and to improve the film separation into similar clusters by the more advanced and reliable fuzzy clustering method. It has been shown that fuzzy clustering allows a better classification, ascribing a membership function to each HTS thin film.

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## 1. Introduction

The fabrication of high-temperature superconducting (HTS) materials still presents a serious challenge to many research laboratories. It is well known, however, that the approaches to the fabrication procedures are, more or less, trial-and-error methods. The reason for this is the multivariate nature of the process of fabrication, for which many factors which are quite different in nature should be taken into account. Thus, the substrate properties, precursor chemistry, chemical vapour deposition parameters etc. have to be carefully selected in order to achieve reasonable critical temperature values. In a previous paper [1] an attempt was made to classify data available from the experience of many international laboratories, concerning the input factors of HTS thin films and the respective outputs (critical temperature $T_c$ or critical current $J_c$). It has been shown by the use of cluster analysis (CA), principal components analysis (PCA) and multiple linear regression analysis (MLRA) that a kind of pattern recognition among the various thin films could be constructed and interpreted [1].

It is the aim of the present communication to offer another point of view in the classification already carried out, by applying a new and promising chemometric approach called fuzzy cluster analysis (FCA). The paper indicates the advantages of the new method and compares the classification obtained with the previous cluster analysis procedure for the HTS thin films.

## 2. Experimental details

Data for the classification procedure were taken from the review paper of Leskela *et al.* [2]. The authors' attention was concentrated mainly on Y-Ba-Cu-O thin films obtained by chemical vapour deposition. Altogether, 84 cases were presented in the review, comprising experimental evidence about the physical input variables (substrate lattice parameter LP, substrate thermal

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expansion coefficient TE, substrate temperature $T_s$, oxidizer partial pressure P, precursor chemical nature Y, Ba, Cu,) and physical output parameters (critical temperature $T_c$ for all cases and critical current $J_c$ for some of them).

In essence, fuzzy theory is an exact mathematical theory which has matured into a wide-ranging collection of concepts and techniques, as well as applications in almost all branches of science [3]. Its substantial concept is a membership of a set (the concept of fuzzy sets) but not in the sense of probability. For instance, a spectroscopic line has to be identified in order to specify a functional group in IR spectroscopy, or to decide on the presence of an element in atomic spectroscopy. This is done by comparing the line position with those appearing in a library of reference lines. Since the experimentally-obtained line will surely not exactly match the library line, an interval around the reference (library) line is usually defined in order to decide whether or not the library line coincides with the experiment one. A value of 1 is assigned to a line that matches the interval and the value 0 is assigned to lines outside this interval. So only a yes/no answer is possible in the traditional way of grouping experimental results. With fuzzy theory, this situation could be described in much more detail. For a line that come closest to the library line, a value near to 1 is assigned, whereas the more the line position deviates from the exact match the lower the value assigned. The outcome of the comparison does not only reveal information on whether or not the match has been successful (a 1/0 answer), but it also gives a rating of the quality of the match, graduated between 0-1.

The main idea of founding fuzzy set theory was the generalization of the common set notion. When defining a common set, say $M_c$, we are given a collection of elements, the universe $X$, being of interest in our context. Then, $M_c$ is defined by specifying, for each element of the universe, whether it belongs to $M_c$ or not. This can be expressed mathematically by a characteristic function, say $m_c$ , that assigns an element $x$ of the universe $X$ a value of 1 if it belongs to $M_c$ and of 0 otherwise:

$$m_c\,(x) = 1 \text{ if } x \,\varepsilon\, M_c \cap \mathrm{X} \tag{1}$$

$$m_c\,(x) = 0 \text{ if } x \,/\varepsilon\, M_c \cap \mathrm{X} \tag{2}$$

If we also allow values between 0 and 1 to be grades of membership with respect to $X$, we obtain a fuzzy set $M$. Thus, the transition between membership and non-membership may be described gradually, rather than abruptly. The function $m$ valuing the elements $x \,\varepsilon\, X$ with numbers in the closed interval [0, 1]. and thus defining the fuzzy set $M$, is called the membership function of $M$. A common set, for which the membership function yields only two values, 0 and 1, is called crisp in this context.

For interpretation, there can be elements $x \,\varepsilon\, X$ for which we do not state whether they belong to $M$ or not. The value $m(x)$ can be explained as to which grade or degree the element $x$ belongs to $M$.

When we consider more than one fuzzy set, say $M$ and $N$ simultaneously, we shall add the name of the set as an index to the corresponding membership function, e.g. $m_M$, $m_N$.

In all cases of application, we have to start by specifying universes and fuzzy sets in them. Remembering the example mentioned above – comparison of a crisp spectroscopic line $x$ to a fuzzy candidate reference line. The universe $X$ which is to be specified would be the energy or wavelength axis, and the uncertainty of the appearance of the line could be modeled by a fuzzy set M having the shape of a bell or of a triangle. Thus, the membership value $m\,(x) = 1$ would be assigned only to that element $x$ of the universe that meets the maximum of the membership function, i.e. the experimental line taken as being crisp matches the reference line. Elements around the reference line are characterized by the degree to which the experimental line is comparable with the reference one.

A special case of a fuzzy set is a fuzzy measurement or fuzzy observation. Here, $m(x)$ indicates the degree to which $x$ is to be considered as a result of our measurements at hand.

It makes sense to explain the values of $m$ as shades on a scale [0, 1], where 0 means white and 1 black. In this scheme, fuzzy sets correspond to gray – tone pictures and could be manipulated by image processing equipment.

For specifying the membership function, one must exploit the special knowledge of the expert, together with the preliminary information on the problem at hand. Frequently, the specification of a membership function can in the broadest sense be inspired by statistical material. Fortunately, the choice of the specific mathematical form of the membership function has, as a rule, only a small influence on the conclusions to be drawn. In general, the guideline is dictated by mathematical convenience. We mention here the following types for the one-dimensional case: $(1 + c|x\text{-}a|^p)^{-1}$ or $\exp(-c|x\text{-}a|^p)$ or $1 - \exp(-c|x\text{-}a|^p)$ for $x \geq a$ with suitably chosen constants *a*, *c*, *p*, where *c* and *p* have to be positive numbers. A simple generalization of the multidimensional case is given by replacing the difference $|x\text{-}a|$ between the numbers *x* and *a* by a distance between the points *x* and *a*.

If we have to take into account asymmetry in specifying *m* we can, at *a*, connect branches of the chosen type with different parameters, and even branches of different types.

When using image processing equipment, the choice of a suitable type becomes less essential; the membership function may be constructed locally, e.g. by splines.

In order to handle fuzzy sets efficiently, notions known from common set theory have to be applied. Besides these, however, other notions will occur. They include the support of a fuzzy set with a positive grade of membership; the cardinality of a fuzzy set (the number of its elements for a finite set or its suitably defined contents for an infinite set); the grade of containment etc. Furthermore, fuzzy numbers have to be determined when some conditions are satisfied (convex membership function, only one mean value); fuzzy points, fuzzy observations represented by fuzzy points, a fuzzified function, which is a family of fuzzy numbers, fuzzy relations, etc.

The notions introduced form the basis for more complicated problems and their solution. For example, we may consider the problem: to which grade is “near to 20” “essentially larger” than “near to 10”? Although this could be treated with conventional mathematical tools, the best solution is given by the fuzzy approach as described in [4, 5].

It is interesting to mention that the notion “linguistic variable” is also important for fuzzy operations. This variable can assume several variables, which are verbal units, words, or sentences, in a natural or artificial language. A simple example is given by the variable “stature” as used in warrant of apprehension. The values are certain labels, e.g. very tall, tall, medium, short, very short. Each of these variables corresponds to a fuzzy set on the universe “height”: (0, 3) when measured in meters.

Fuzzy data sets are observed and described by the specific membership function (MF). In the case of fuzzy cluster analysis, an iterative procedure called generalized fuzzy n – means (GFNM) is used for detecting clusters within a collection of objects (universe) by MF. The iterative process starts with an arbitrary initialization of the partition, and ends when two successive partitions are close enough. It is then possible to have the distance between two partitions. In such a way, step by step, the membership of each object to the separate classes (clusters) is found [6].

## 3. Results and discussion

In the previous study, hierarchical crisp clustering of Y-based HTS thin films indicated that the universe of objects (84 altogether) can be separated into 5 clusters. A more careful consideration of the clustering shows that, in principle, the majority of the objects (thin films) are quite similar: at the first level of significance (33.3 % of $D_{max}$). Only 4 objects are linked above this level. Some subclusters were found and discussed. Within them, the linkage between the objects was due mainly to a “national” indicator – similar scientific schools, similar technological procedures, similar instrumentation. The outliers were attributed to some specific technological reasons, or to the more different physicochemical and morphological properties assuring better output. In Table 1, results from the fuzzy clustering of HTS thin films (YBCO) are summarized.

In this clustering procedure, one obtains three well-defined classes or clusters. The first two of them indicate a typical fuzzy structure, since the objects included in them could be ascribed to a certain extent to the next group. Indeed, the membership percentage is quite high, this being a sign for a stable structure of the classes. However, on the other hand, it might be assumed that class 1 and class 2 are quite similar to each other. In this aspect, classes 1 and 2 resemble the subclusters 1, 2 and 3 in the hierarchical dendrogram in [1]. However, the possibility of merging classes 1 and 2 into one bigger class is not acceptable, since the objects classified within each of them possesses high values of the membership function. Therefore, one could accept that classes 1 and 2 are divided for some reason.

Table 1. Final clustering of 84 objects with fuzzy algorithm

| Class | Objects and membership to class (%) |
|---|---|
| 1 | **2**(89), **5**(81), **8**(94), **11**(87), **15**(86), **21**(85), **22**(89), **23**(90), **24**(89), **29**(87), **34**(79), **36**(89), **37**(92), **42**(88), **47**(84), **55**(85), **56**(88), **59**(90), **62**(90), **63**(87), **65**(89), **67**(83)**, 68**(87), **74**(90), **77**(91), **78**(89), **79**(90), **83** (90) |
| 2 | **4**(88)**, 6**(89)**, 9**(90)**, 10**(86)**, 12**(84)**, 13**(83)**, 14**(79)**, 16**(87)**, 18**(86)**, 19**(90)**, 20**(79)**, 25**(88)**, 28**(80)**, 30**(85)**, 31**(90)**, 32**(91)**, 33**(87)**, 35**(82)**, 38**(89)**, 39** (89), **40**(88)**, 45**(86)**, 46**(85)**, 53**(86)**, 54**(83)**, 57**(88), **58**(89)**, 60**(88)**, 61**(87)**, 64**(86)**, 69**(86), **70**(84)**, 71**(89)**, 72**(88)**, 75**(85)**, 76**(88), **80**(90)**, 81**(89)**, 82**(88) |
| 3 | **1, 3, 7, 17, 26, 27, 41, 43, 44, 48, 49, 50, 51, 52, 66, 73, 84** (For all objects the membership percentage is 100). |

The third cluster in the fuzzy procedure is well defined and separated from the other two (none of the objects indicate membership of the other classes). This means that the third group of objects differs from the other two in the most significant way.

## 4. Conclusions

A very important conclusion from this study is that by the use of fuzzy clustering, one could get a better separation of groups of similarity and even explain the reason for fuzzy behaviour. In this particular case, the separation of the 84 YBCO thin films into three main classes could occur for the following reasons:

- Class 1 – dependence on the lattice parameter and the thermal expansion coefficient (both parameters possess the lowest possible values); relatively high critical temperature values are reached (above $85^{o}$ K);
- Class 2 – this is the cluster of similar objects realized by the use of higher values of the substrate parameters. These, however, do not lead to a significant increase in the critical temperature values. On the contrary, in many cases a decrease of the critical temperature is observed.
- Class 3 – in this cluster of objects, one could find some with the behaviour of outliers (e.g. a very low critical temperature or very low oxidizer partial pressure etc.). It might be stated that this is the group of objects which are difficult to classify with the traditional agglomerative clustering procedures.

As a final conclusion, it may be suggested that the class 1 objects should be considered as the optimal “pattern” for YBCO HTS thin films.

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