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Application of the Fuzzy-C-Means Algorithm to the Study of Relationships Between Composition and Properties: Physical Properties of Glasses

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Summary. The opportunities of applying the Fuzzy-C-Means (FCM) algorithm for cluster analysis to study composition-property relations are discussed. Physical properties of 98 aluminosilicate glasses serve as an example. The influences of the initial partition and of the number of clusters chosen are discussed in detail. The FCM algorithm allows to find characteristic members of clusters (these form the "nuclei" of the clusters) and to find outliers of the data set under consideration. Thus the results of the cluster analysis form a basis for a deeper understanding of relations between the composition and properties of the glasses studied.

Keywords. Fuzzy-C-Means method; Cluster analysis; Composition-property relations; Glasses.

Anwendung des Fuzzy-C-Means-Algorithmus bei der Untersuchung von Beziehungen zwischen Zusammensetzung und Eigenschaften: Physikalische Eigenschaften von Gläsern

Zusammenfassung. Es werden die Möglichkeiten des Fuzzy-C-Means-(FCM)-Algorithmus für Cluster-Analysen zur Untersuchung von Zusammensetzung-Eigenschafts-Relationen diskutiert. Die physikalischen Eigenschaften von 98 Aluminosilikat-Gläsern dienen als Beispiel. Es wird der Einfluß von Anfangsverteilung und Clusteranzahl untersucht. Der FCM-Algorithmus erlaubt das Auffinden von charakteristischen Teilen von Clustern (diese bilden den „Kern" des Clusters) und auch von Außenseitern des betrachteten Datensatzes. Somit liefern die Ergebnisse der Clusteranalyse die Grundlage für ein tieferes Verständnis der Beziehungen zwischen Zusammensetzung und Eigenschaften von Gläsern.

Introduction

The presence of a large set of experimental data which have (i) to be evaluated concerning their reliability, (ii) to be systematized with respect to the problem under consideration and, finally (iii) to be interpreted, is the common situation in a scientist's daily work. Classification methods, like e.g. cluster analysis [1–3] offer

the opportunity to support evaluation, systematization and interpretation of data sets. Cluster analysis is a tool to subdivide a given set of objects characterized by definite properties into groups (clusters) of similar objects. This subdivision forms a basis to test or to develop hypotheses about the source for similarities or differences of the properties of the objects under study [1].

In chemistry today application of cluster analysis is well established, e.g. in analytical chemistry [5, 6], NMR [7–9], EPR [10, 11], Raman [12] spectroscopies, and also used to systematize further chemical and physical properties [13, 14].

Since cluster analysis may be used to test or to develop hypotheses it may also support the study of relations between chemical composition, structure and properties of "objects" [15–18]. In this field non-hierarchical methods have proofed to be valuable tools for the data treatment [1, 3, 19]. One can decide between (i) methods which lead to a so called "hard partition" (e.g., *k*-Means [1]) and (ii) methods which use the theory of fuzzy sets [20–24]. In the framework of the last method one object may belong to more than one cluster, i.e., each object belongs to each cluster with a certain "degree of membership". Thus the result of a cluster analysis based on the theory of fuzzy sets is a table which contains the degrees of membership for all objects under consideration.

It is advantageous to have the chance to assign an object to more than one cluster since such an assignment can given evidence for outliers or hybrids, i.e., objects which should be excluded from an interpretation first. According to Bratchell this advantage may be one reason that fuzzy cluster analysis "will receive more attention in the future" [4].

It is the objective of this work to demonstrate, from a chemist's point of view, the problems and advantages resulting from the application of the FCM method to the study of composition-property relations. As an example a set of physical property data of glasses is used. This set offers the advantages, that (i) chemical composition of the objects could be varied stepwise and in a wide range, thus forming a comparably large data set and (ii) results of the application of *k*-Means to a respective set of objects are already known [18] so it will be possible to compare expense and profit of the different algorithms.

Objects and Methods

The objects chosen are 98 glasses of the $CaO–Al_2O_3–SiO_2$ system characterized by the values of the physical properties molar refraction, molar volume of oxygen, transformation temperature and coefficient of linear thermal expansion. The distribution of these glasses in the composition diagram can be taken from Fig. 1.

Autoscaling (e.g. [25]) was applied to the experimentally determined values of the physical properties under consideration before performing the cluster analysis. The normalization of the data was sensible because no arguments could support any weighting of the properties.

The cluster algorithm applied here is the Fuzzy-C-Means (FCM) method. The ideas of the fuzzy theory are described in [20–22], in a short form quite understandable for chemists in [24], and the basic principles of FCM are described in detail in Refs. [9, 23, 26, 27]. Our calculations are based on the computer program listed in [27] and essentially we follow the notations and abbreviations used there.

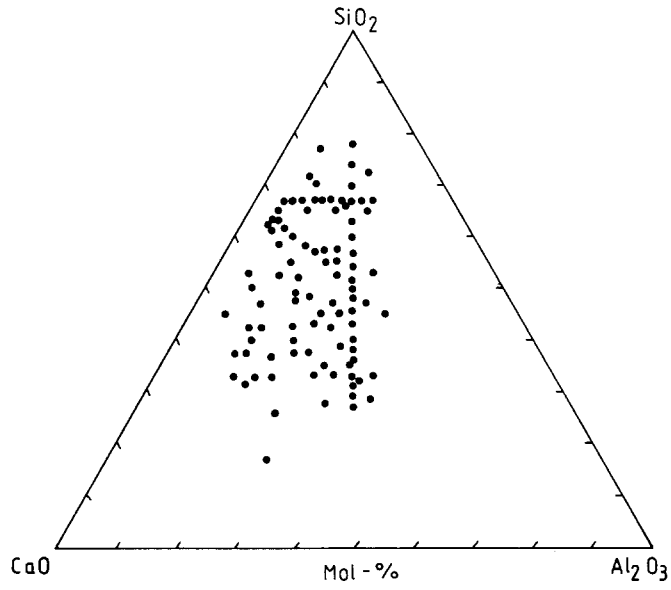

Fig. 1. Graphical display of composition (mol%) of glasses under consideration

The results will be compared with those of the k-Means (KM) method (cf. [18, 28]). Both of these algorithms, FCM and KM, are nonhierarchical ones. Within FCM one object may partially belong to more than one cluster, whereas in the result of k-Means, being a special case of FCM, each object belongs to only one cluster. The membership of the objects y_k of the clusters u_i is described by a degree $u_i(y_k) = u_{ik}$ ($i = 1, \ldots, c$; $k = 1, \ldots, n$), with c being the number of clusters and n being the number of objects. The membership degrees u_{ik} form the matrix U. For the further treatment the degree of membership has to be regarded as normalized in the range between 0 and 1; so for FCM $u_{ik} \in [0, 1]$ is valid and for k-Means $u_{ik} = \begin{cases} 1; & y_k \in Y_i \\ 0; & \text{otherwise} \end{cases}$ holds, Y_i is the hard partition of the sample set Y, (compare [27]). Both methods minimize the sum over intra class distances (optimization parameter).

For FCM the functional

$$J_m(U, v) = \sum_{i=1}^{c} \sum_{k=1}^{n} (u_{ik})^m \| y_k - v_i \|^2 \tag{1}$$

has been proposed by Bezdek et al. [27] as a metric, analogous to the functional of k-Means $\sum_{i=1}^{c} \sum_{k \in Y_i} \| y_k - v_i \|^2$.

Here v_i is the center of gravity of the cluster i, v is the vector of the centers and m is a weighting exponent, $m \in [1, \infty)$. As a distance we have applied the Euclidean distance

$$\sqrt{\| y_k - v_i \|^2} = \sqrt{(y_k - v_i)^{\mathrm{T}} (y_k - v_i)}.$$

The application to NMR data of glassy aluminosilicates is described in [12, 29].

Obviously the formation of clusters is not only influenced by the data structure but also by some other quantities. For k-Means these quantities are the predestined number of clusters, the normalization of the data as well as the initial distribution of the objects within the clusters [9, 11, 18], and FCM additionally is affected by the weighting exponent m [9, 27, 29]. As formula 1 shows, the optimization parameter depends on the input value m, which controlls the delocalization of the objects over the clusters. If m approaches 1, then the results of FCM tend to be identical to those of the k-Means algorithm, i.e., for $m = 1$ k-Means is a special case of FCM. For large values of m, exactly if m tends to infinity, then the objects are homogeneously distributed over all clusters. This leads to a result which contradicts to the aim of cluster analysis to bring out the structure of an unknown data matrix and to show objects with common and similar properties. The parameter m has not any substantial meaning in our case. It is an empirical quantity, reasonable selection of which has to be decided by a sensible interpretability of the results. Examination of Bezdek et al. [27] has pointed out that a sensible range lies between $1.5 \leqslant m \leqslant 3$. If $m = 3$ holds, then our calculation with the data set of physical properties of alumosilicate glasses and with artificial data [30] have shown that the clusters have to establish a clearly distinguishable structure, i.e. the sums of the intra cluster distances have to be small in comparison with the inter cluster distances, otherwise the results are hardly interpretable. For our data set the weighting exponent $m = 2$ caused a diminution of the membership degree to a value of about 0.7 of many objects. These were to many objects as to be useful for our purposes to extract the core of the cluster from its surrounding. If $m = 1.5$ was chosen, then the results led to a plausible interpretability.

It is the focus of this work to investigate the influence of the initial partition on the results of cluster analysis. The assertions with regard to the sensibility of the normalization and the distance measure were already given above. The argumentation for the selection of a reasonable cluster number from the point of view of interpretability has been performed in a previous work [12]. There was additionally shown, that the dependence of the entropy $H_c(U) = -\sum_{k=1}^{n} \sum_{i=1}^{c} \frac{1}{n} u_{ik} \log(u_{ik})$ on the cluster number c, recommended in [27] for the identification of the "true" cluster number with some limitations, was not helpful for the interpretation of our complicated data structure. The dependence of the end partition on the initial partition for the cluster algorithms applied by us is caused by the property of many optimization procedures to find a local minimum which is not necessarily the global one. From a first theoretical point of view it should be possible to find the global minimum by total enumeration when applying conventional cluster procedures to a finite number of objects. This fails because of the combinatorial augmentation of the initial partitions with the number of objects.

For procedures like k-Means there exist investigations about the dependency of the result on the initial partition. A common recommendation suggests to calculate the end partitions from a sufficiently large number of initial partitions (100 or more) and to regard that end partition as the right one which exhibits the smallest optimization parameter. Another method of verification consists in applying the results of hierarchical cluster strategies as an input for the nonhierarchical ones. At third it has been shown in [18] how to diminish the ambiguities of interpretation

by constructing generalized partitions based on a collective treatment of multiple end partitions. By this means some of the objects under consideration in the present work were already successfully classified as not clearly assignable, especially those that where scattered in the transition region of two or more clusters. For objects being situated in the property space rather distant from cluster centers, their detection as delocalized ones was less successful. In our investigation we have led similar investigations on the influence of the initial partition for the FCM and have compared the results with those got by k-Means [18]. For the latter purpose it was sometimes necessary to adjust the results from FCM with those from k-Means, that's why we had partially to withdraw the basic principles of FCM – the assignment of one object to more than one cluster – in the course of the interpretation of the results.

The calculations presented here are based on the FORTRAN program given in [27], which was implemented on a PDP-like computer. The procedure was extended permitting us to create up to 100 initial partitions and to preselect the end partitions, being practically "identical" – really very similar. By doing so the expense to explain the results of the FCM procedure was greatly diminished. Two partitions have been regarded as "identical" if the differences of all the membership degrees Δu_{ik} were not greater than 0.05. So, in the worst case, end partitions with $\Delta u_{ik} = 0.1$ were accepted as identical, what is sensible and allowable for our purposes. Additionally the algorithm of generating the random initial partitions was modified, i.e.

$$u_{ik} = \frac{R_{ik}}{\sum_{i=1}^{c} R_{ik}}.$$

For every object and cluster a [0,1]-distributed random number (R_{ik}) is assigned and divided by the sum of the random numbers over the clusters to guarantee the normalization condition $\sum_{i=1}^{c} u_{ik} = 1$.

Results and Discussion

Table 1 summarizes the data optimization parameter, entropy and frequency characterizing the end partitions obtained for a partition of the 98 objects into two to five clusters and gives the numbers of different end partitions found by applying the k-Means method to the same data set.

A visual analysis of the presorted results of FCM-analysis of 100 initial partitions has reduced the number of end partitions to be regarded as different as follows: 3 clusters – 4 end partitions, 4 clusters – 2 end partitions and 5 clusters – 1 end partition. This visual analysis should be explained for cluster 4 in more detail. In Figs. 2 a–c three partitions are visualized for that cluster number by a plot of the first and second principal components (for a description of principal component analysis see e.g. [3]) of the autoscaled data. Objects having $u_{ik} \geqslant 0.9$ are characterized by its cluster index, $0.8 \leqslant u_{ik} < 0.9$ by capital letters, and $0.6 \leqslant u_{ik} < 0.8$ by small letters, respectively. The relations 1-A-a, 2-B-b and so on hold. All other objects are signed by the asterisk "*". The plots 2 a–c are in accordance to the sequence of the partitions of Table 1. The difference between the assignments in Figs. 2 a and 2 b

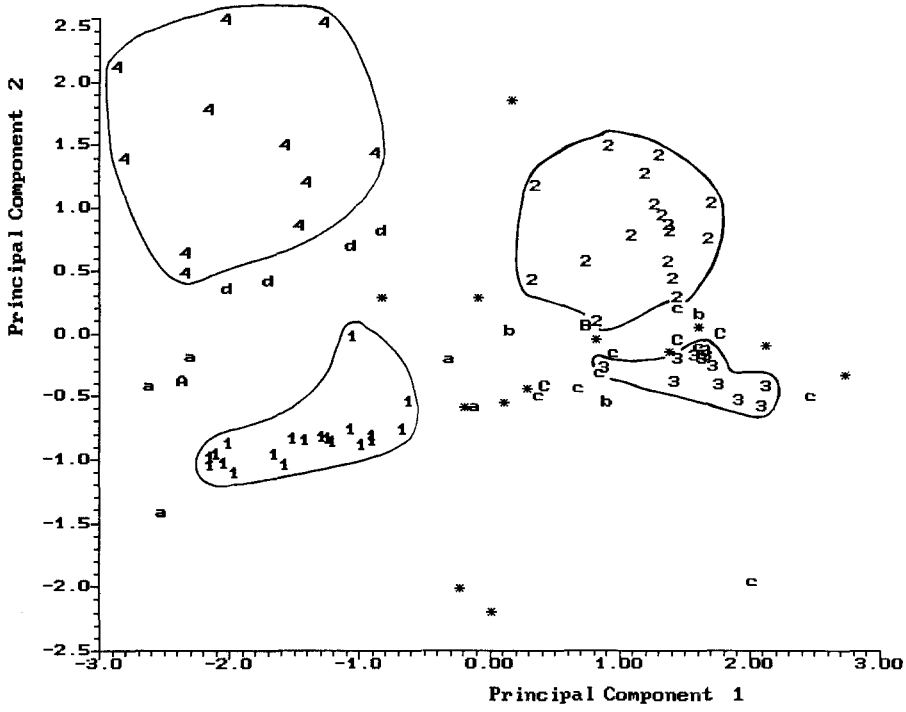
Principal Component 2
Principal Component 1
2.5
2.0
1.5
1.0
0.5
0.0
-0.5
-1.0
-1.5
-2.0
-2.5
-3.0
-2.0
-1.0
0.00
1.00
2.00
3.00

Fig. 2a

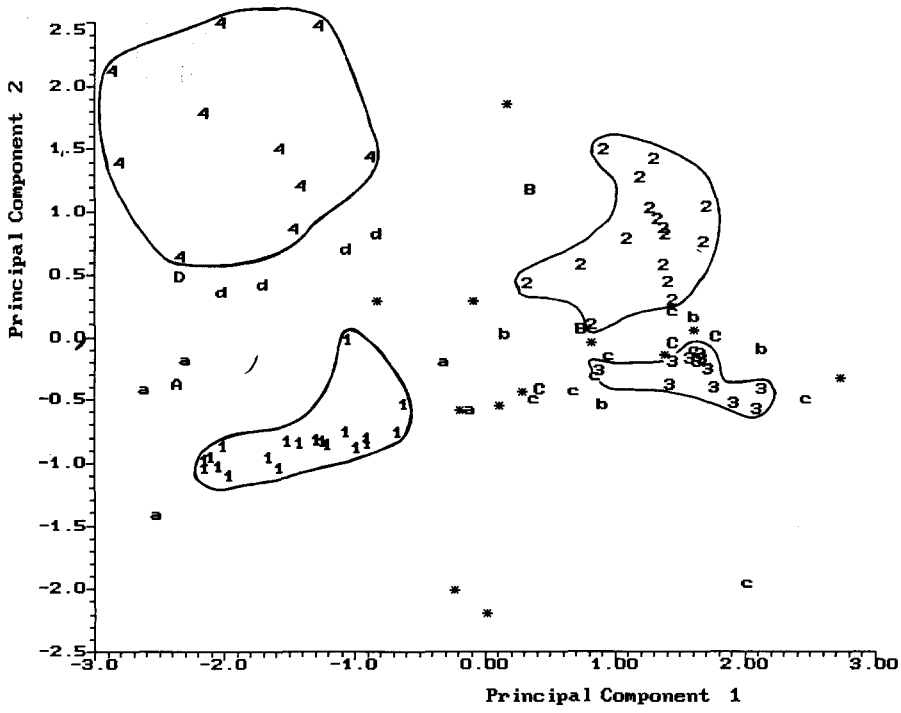
Principal Component 2
Principal Component 1
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Fig. 2b

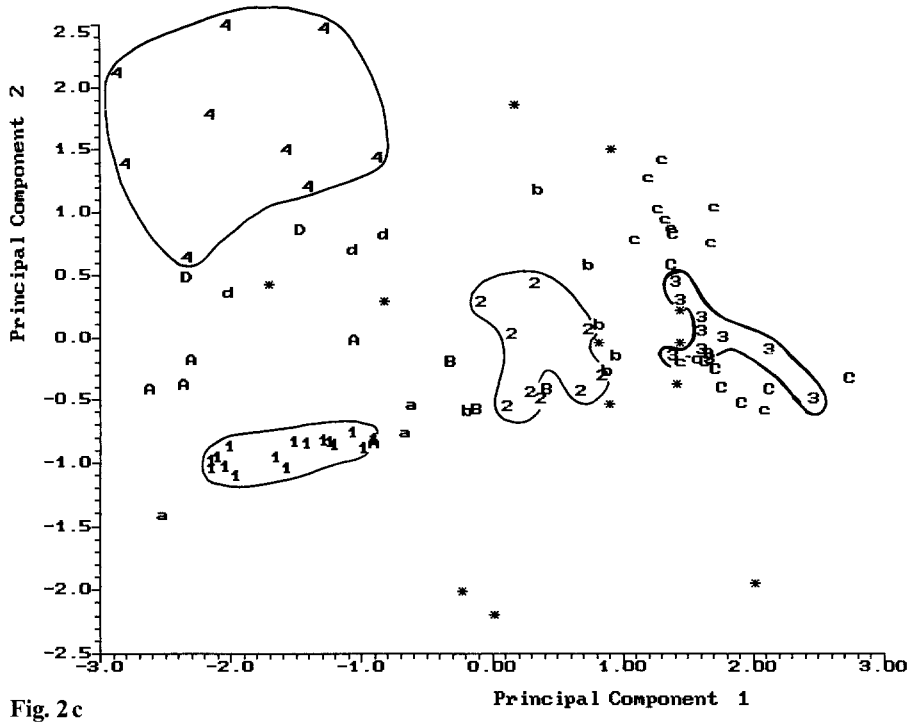

Fig. 2c

Fig. 2. Principal component plots (**a**–**c**) of preselected partitions got for 4 clusters. Numbers characterize the nucleus of the cluster where $u_{ik} \geqslant 0.9$, capital letters the range $0.8 \leqslant u_{ik} < 0.9$, small letters the range $0.6 \leqslant u_{ik} < 0.8$, according to the relation 1-A-a; all other objects are signed by "*". The end partitions **a**–**c** are in accordance with the sequence of Table 1

stems from two objects (50 and 62), which are translated only from one range Δu_{ik} into an adjacent one. The membership degrees u_{ik} of object 62 with respect to the appropriate partitions are (0.02, 0.904, 0.04, 0.03)* and (0.02, 0.898, 0.05, 0.04)*, respectively, which means that both partitions are practically identical; only the rather arbitrary assignment of the boundary of 0.9 led to the discrimination.

An analogous conclusion is valid for object 50. The numerical values (not given here) have also shown that neither object 50 nor 62 was responsible for the distinction during the preselection, but one of the objects where $u_{ik} < 0.6$ holds. The visual analysis has shown that both partitions are equivalent with respect to their interpretability.

On the other hand the situation is very different with Fig. 2 c. Whilst the differences between clusters 1 and 4 with regard to the appropriate clusters in Fig. 2 a, b are characterized by small transitions within the adjacent Δu_{ik}-ranges and are negligible from the interpretive point of view, clusters 2 and 3, however, show a completely different structure. The further partitioning (examination of 5 clusters) acts on those clusters 2 and 3 as may be seen from Fig. 3. Here the first partition with respect to Table 1 is presented. From a similar analysis performed for 5 clusters, according to

* Nonfulfillment of normalization because of round off errors

Table 1. Parameters of the different end partitions got starting from 100 different initial partitions within Fuzzy-C-Means and k-Means clustering of the objects under consideration

Number of clusters	Optimization parameter	Entropy	Frequency [%]	Number of end partitions/k-Means [18]
2	188	0.193	100	–
3	139	0.299	78	4
	146	0.377*	12	
	146	0.377	8	
	151	0.442	1	
	151	0.438	1	
4	104	0.408*	90	8
	104	0.408	8	
	112	0.468	2	
5	86	0.458	49	8
	86	0.460	33	
	86	0.457	14	
	86	0.459	4	

* Though the same optimization parameters and entropies were obtained for two end partitions for cluster numbers 3 and 4 each, these are different concerning the criterion ($\Delta u_{ik} < 0.05$) used here (see text)

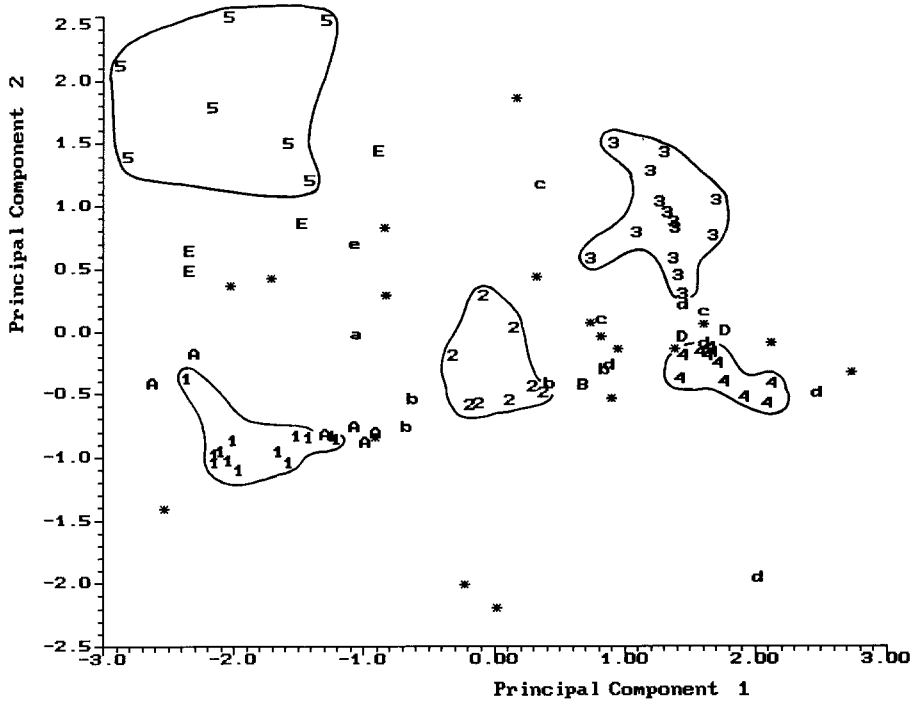

Fig. 3. Principal component plot of preselected partitions obtained for 5 clusters of the first partition of Table 1; the meaning of the symbols is the same as in Fig. 2

that one described above, is to be concluded that all of the four preselected end partitions are practically identical. From Figs. 2 a–c and 3 it can also be concluded, that the principal component plots already express the cluster structure in rough outlines. For certain objects the higher principal components are significant and hence in the plots of only two principal components these objects, though they are fuzzy ones, are projected into the core (cores of the clusters are outlined by an unbroken line).

Thus, after having got a first insight into the differences between the end partitions for a subdivision of the objects into four and five clusters by using the principal components plots (Figs. 2 a–c and 3) a more detailed discussion shall be based on the assignments of the objects to the clusters according to the membership functions (FCM) including a comparison to the results of k-Means cluster analysis [18]. Figure 4 again shows the differences between the end partitions for four clusters obtained by FCM and also the respective end partition got by applying the k-Means method. Each box represents one cluster and the four clusters (denoted I to IV) belonging to one partition are arranged in one row. The FCM cluster boxes are subdivided by broken lines according to certain ranges for the membership functions. All objects were assigned to that cluster where they have the largest value of membership function, e.g. in the most frequent partition object 87 has the following values of membership function: Cluster I: 0.347; cluster II: 0.262; cluster III: 0.354; cluster IV: 0.036. Thus it actually lies between clusters I to III but was according to our "rule" assigned to cluster III.

Comparing these three different FCM end partitions the following is to be observed: (i) The two partitions with the optimization parameter of 104 are similar to each other in that the "compositions" of the clusters are similar, i.e., the same objects are always together in analogous clusters. (ii) The composition of the clusters forming the partition with the highest optimization parameter differs from the two other ones, despite cluster IV. (iii) For clusters of same composition belonging to different end partitions the values of the membership functions of the objects are different, e.g. for object 62 $1 \geqslant u_{ik} > 0.9$ holds in the first, $0.9 \geqslant u_{ik} > 0.8$ in the second and $0.8 \geqslant u_{ik} > 0.6$ in the third partition. (Remember, that the discussion above showed that the differences between the first and second end partitions are practically negligible, but the differences between these two and the third end partition are significant.) The appropriate end partition with the lowest optimization parameter obtained by applying the k-Means algorithm [18] shows many similarities to the FCM end partitions but the compositions of the clusters are not identical.

Figure 5 shows the results for a partition into five clusters (I to V) obtained by applying the FCM method, and the k-Means generalized partition taken from [18]. The comparison of these end partitions yields the following peculiarities: (i) The comparison of the first, second ... clusters respectively belonging to the different end partitions shows, that the respective clusters are always comprizing the same objects. The differences of the entropies belonging to each individual end partition indicate, that the entropies are obviously more sensitive to the changes in membership degrees than the criterion applied here. However, the discussion showed that the criterion choosen to differentiate the end partitions is sufficient. The scaling of the membership function arbitrarily chosen makes the differences between the different

	I	II	III	IV
$1 \geq u > 0.9$	1-4 12-15 41-45 66 76-78 83 88 89	7-11 33-40 62-64 80	18-21 24-29 75 86	57-61 72 79 85 92 96 97
$0.9 \geq u > 0.8$	84	54	17 23 30	
$0.8 \geq u > 0.6$	5 46 67 74	6 32 71	22 48 52 53 68 81 82 95	56 70 90 93
$0.6 \geq u$	16 65 69 91	31 49-51 55 94 98	47 87	
$1 \geq u > 0.9$	no change	7-11 33-40 63 64 80	no change	57-61 72 79 85 92 97
$0.9 \geq u > 0.8$	84	54 62	17 23 30	96
$0.8 \geq u > 0.6$	5 67 73 74	6 32 50 71	22 48 52 53 68 81 82 95	56 70 90 93
$0.6 \geq u$	16 46 65 69 91	31 49 51 55 94 98	47 87	
$1 \geq u > 0.9$	1-3 12-14 41-45 66 76	6 7 47 54 81 82 87 95	28-34 49 50 52 53	57-61 72 79 85 92
$0.9 \geq u > 0.8$	73 74 78 84 89	5 17 46	20 21 26 27 35 36 51 75	96 97
$0.8 \geq u > 0.6$	4 15 67	8 16 18 48 62 80	9 10 11 19 25 37-39 63 86	56 90 93
$0.6 \geq u$	65 69 91	71 94	22-24 40 68 98	70
k-Means GP [18]	1-5 12-16 41-46 65-67 69 73 74 76-78 83 84 87-89	6-11 31-40 47-55 62-64 71 80 81 94 98	17-30 52 68 75 82 86	56-61 70 72 79 85 90-93 96 97

Fig. 4. End partitions obtained by FCM and k-Means methods for partition of the 98 glasses into four clusters

end partitions transparent, for $0.9 > u_{ik}$ differences between analogous clusters occur, cf. e.g. the data of object 76 (cluster I). (ii) In the range $1 \geqslant u_{ik} > 0.9$ the clusters I to V belonging to different end partitions are identical, i.e., these 53 objects should serve as a starting point for the further interpretation of the partitions. Construction of a generalized partition from the k-Means cluster analysis led to the clusters given in Fig. 5 and four objects (78, 81, 89, 91) were called "difficult to classify" [18], i.e.,

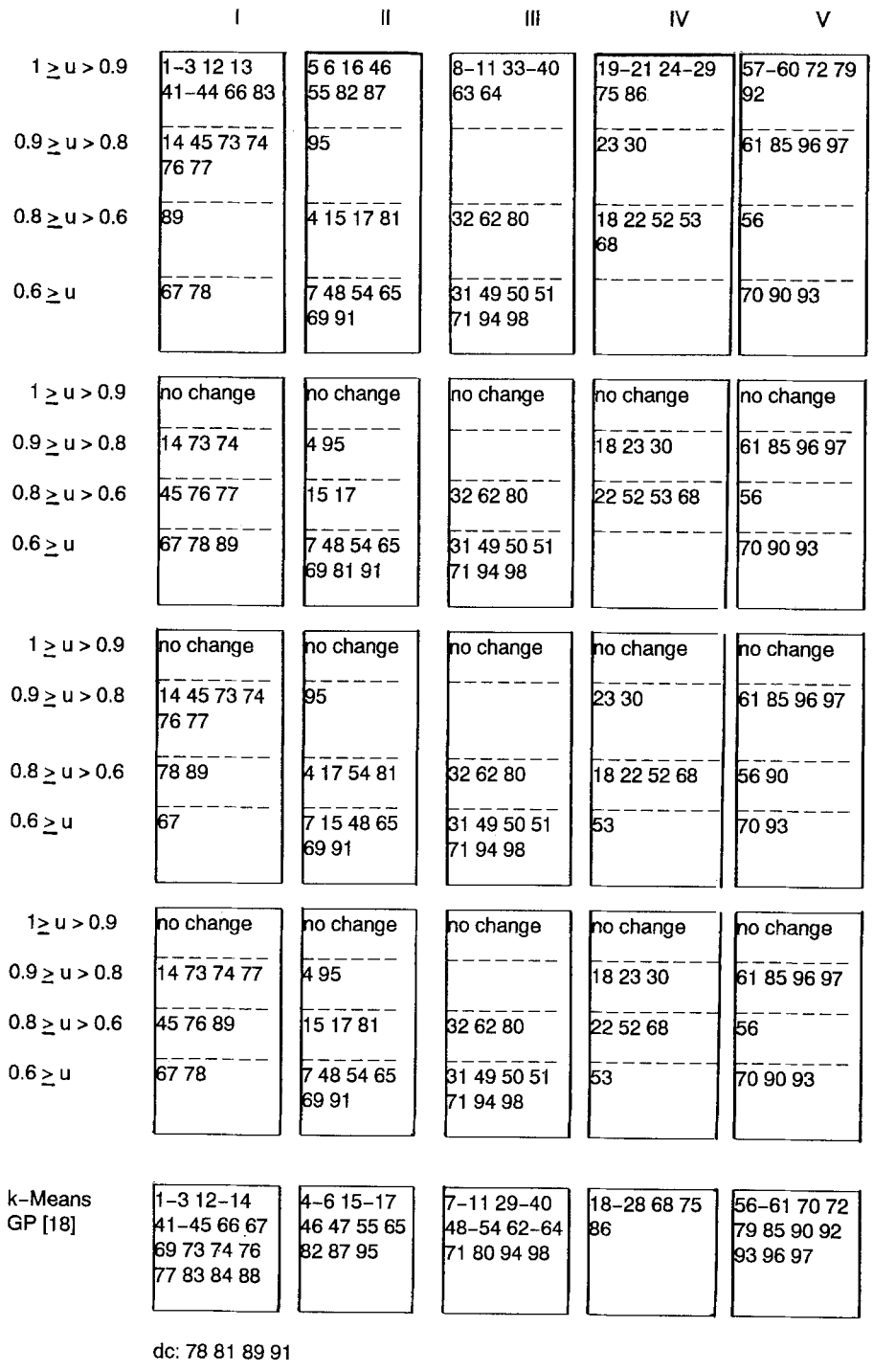

Fig. 5. End partitions obtained by FCM cluster analysis for partition of the 98 glasses into four clusters and "generalized partition" obtained by using the *k*-Means method

they did not fit to any of the five clusters of the generalized partition. For all of these "dc" objects $u_{ik} < 0.8$ holds. The generalized k-Means partition differs partially from the FCM partitions.

The basis for further discussion will be the partition into five clusters depicted in Fig. 6. These clusters were constructed as follows: (i) Objects with values of $u_{ik} \geqslant 0.9$ form the core of the clusters I to V, and (ii) objects with $u_{ik} < 0.9$ are

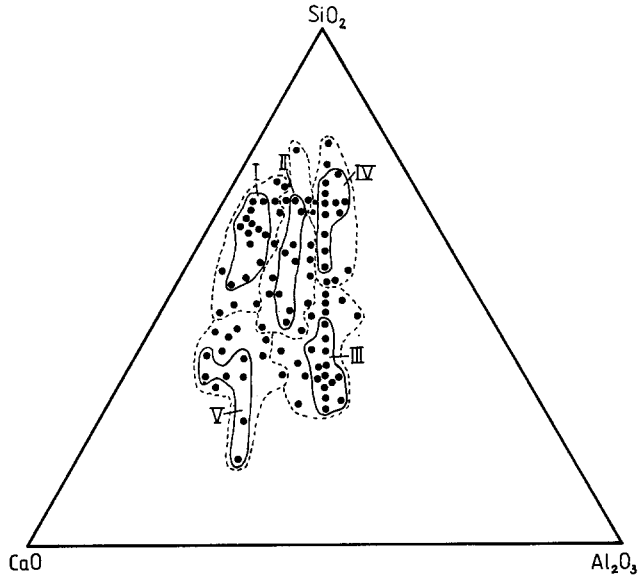

Fig. 6. Graphical display of the end partition obtained by FCM cluster analysis for partition of the 98 glasses into five clusters and differentiating between objects with membership functions being greater than/equal to (—— nuclei) or smaller than 0.9 (– – – – members)

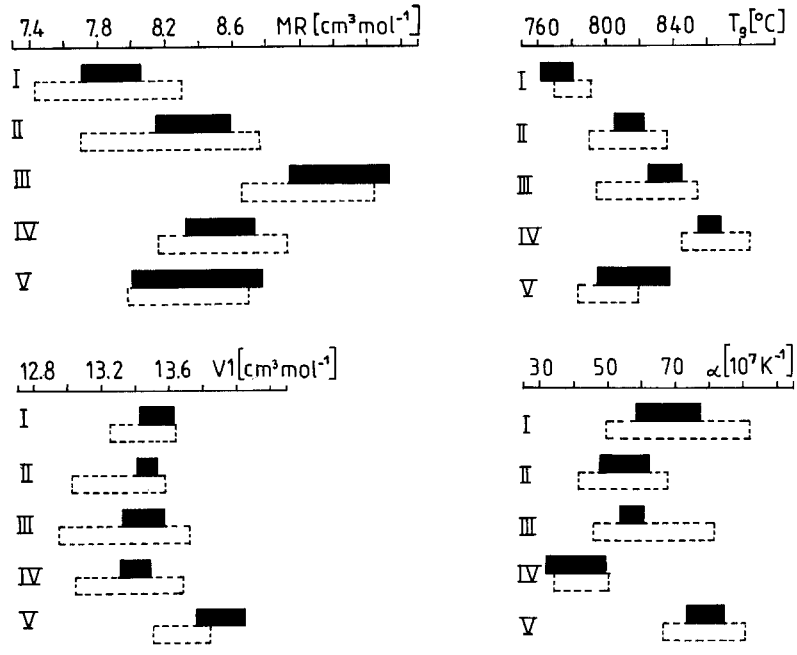

Fig. 7. Plot of ranges of property data molecular refraction (MR), transformation temperature (Tg), molar volume of oxygen ($V1$) and coefficient of linear thermal expansion (α) for nuclei and members of clusters I to V

"members" of the clusters I to V. This arbitrary definition chosen here has the advantage, that the small differences between the different end partitions do not influence the "simplifyed" end partition got by discriminating between objects with membership functions being either $\geqslant 0.9$ or < 0.9. Figure 6 shows that the cores of the clusters are situated in closed compositional ranges and the members are arranged around the cores. Though this arrangement of the objects in the composition diagram is an interesting and valuable result for the glass chemist, it is necessary to remind that the basis for the cluster analysis was not formed by the composition but by physical property data. Figure 7 shows the ranges of property values belonging to the objects which are forming centers and members of clusters I to V. The following peculiarities are to be observed: (i) The ranges of the properties of objects forming the centers of clusters are not necessarily smaller than the ranges for the objects being members of the respective clusters, (ii) the ranges of property data belonging to centers of different clusters may overlap. The degree of pair-wise overlapping between these property ranges depends on the properties, e.g., the centers of clusters I to IV overlap for *V1* but are clearly different in case of T_g.

The results discussed above allow the glass chemist to draw conclusions of the following kind: Within the glass forming range of a special system, e.g. $CaO–Al_2O_3–SiO_2$, by means of cluster analysis certain compositional ranges may be deduced were the glasses are similar to each other regarding the properties under consideration. Such a result of a cluster analysis does allow to draw conclusions about possible combinations of property data for certain compositional ranges.

Conclusions

Summarizing the results, it has to be stated that the cluster procedure of the FCM-algorithm is influenced by the initial partition too. By choosing $u_{ik} < 0.5$ as a criterion to differentiate between "different" end partitions and constructing 100 different initial partitions, up to five different end partitions could be found for a given number of clusters. The different end partitions got for one cluster number have exhibited a clearly different cluster structure. Such a behavior was the origin for a further partitioning of the data up to 5 clusters. For the partition into five clusters an unequivocal partition could be found. An effect of k-Means, the fluctuation of single objects between different clusters, did not take place, which is understandable because those fluctuations occurred with the objects belonging to the surrounding, and here FCM comes to its full advantage identifying those objects as fuzzy classified. Partitions may appear, whose surrounding objects differ in membership degrees of about $\Delta u_{ik} \cong 0.3$. However, the principal conclusion that those objects are fuzzy classified will not be changed.

Using the membership functions to discriminate cores and members of clusters the results of a FCM cluster analysis may be systematized. The obtained partitions support the solution of practical problems, e.g. the investigation and description of relationships between composition and properties of glasses. The definition of centers and members of clusters, taking the value of the membership function as a discriminating criterion, is advantageous especially for large numbers of objects, since the interpretation of the results may start from the cores, formed by a smaller number of objects.

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