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TECHNICAL REPORT NO. 2

Chemical Applications of Topology and Group Theory. 18. Polyhedral Isomerizations of Seven-Coordinate Complexes¹

bу

R.B. King

Prepared for Publication

in

Inorganic Chemistry

University of Georgia Department of Chemistry Athens, Georgia

January 2, 1985

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Abstract

This paper presents all possible isomerizations involving the 34 combinatorially distinct seven-vertex polyhedra. Such isomerizations are expressed in the form $P_1 \neq P_2 \uparrow P_3$ in which P_1 and P_3 have the same number of edges and the intermediate polyhedron P2 has fewer edges than P_1 or P_3 . Isomerizations of this type are regarded as degenerate if P_1 is combinatorially equivalent to P3 and planar if P2 is a planar polygon. Non-planar isomerizations can be classified as n-pyramidal processes where n is the number of edges on the new face of P2 generated by edge removal from P1; 4-pyramidal processes also known as diamond-square-diamond processes favorable energetically, Degenerate are the most diamond-square-diamond isomerization processes involving the chemically significant pentagonal bipyramid and capped octahedron must involve an intermediate polyhedron P_2 having two fewer edges than the starting and finishing polyhedra P1 and P3, respectively. The chemically significant 4-capped trigonal prism is a possible intermediate polyhedron for such degenerate double diamond-square-diamond isomerizations of either the pentagonal bipyramid or the capped octahedron.

1. Introduction

A topic of considerable interest to inorganic chemists is the stereochemical nonrigidity in ML_n coordination complexes (M = central atom, generally a metal; L = ligands surrounding M). Several theoretical approaches have been used to study such stereochemical nonrigidity. Thus selected types of possible isomerizations of ML_n polyhedra [n = 4 (ref. 2), 5 (ref. 3), 6 (ref. 4), and 8 (ref. 5)] have been represented topologically^{2,6} as graphs in which the vertices represent different polyhedral isomers and the edges represent possible one-step isomerizations. Selected individual polyhedral isomerizations have been described in terms of processes originally called diamond-square-diamond (dsd) processes⁷ but which can be more systematically called 4-pyramidal processes.⁸ \hat{A} recent paper of this series⁸ used Gale transformations⁹ to find all possible non-planar isomerization processes for polyhedra having five and six vertices. Using this approach all degenerate non-planar isomerizations of five-vertex polyhedra were shown to be formulated as sequences of Berry pseudorotation processes, the prototypical dsd or 4-pyramidal process. Furthermore, study of the Gale transformations reveals that degenerate nonplanar isomerizations of the seven combinatorially distinct six-vertex polyhedra can be expressed in terms of six distinct types of 4-pyramidal processes and the two prototypical 5-pyramidal processes.

The success of the Gale transformation approach for the study of polyhedral isomerizations lies in the ability of Gale transformations to reduce the dimensionality of the problem. Thus the Gale transform of a (three-dimensional) polyhedron having v vertices can be imbedded into (v-4)-dimensional space.⁸ The Gale transform of a tetrahedron is thus a single point, the Gale transform of a five-vertex polyhedron (trigonal bipyramid or square pyramid) consists of points on a line, and the Gale transform of any of the seven six-vertex polyhedra consists of points in a plane. This dimensionality reduction is the key to the value of Gale transforms in the study of isomerizations of polyhedra with "few" vertices⁹ (i.e., < 6 vertices).

Polyhedra having more than six vertices are also of interest to inorganic chemists. However, Gale transformations offer no advantage for the study of the isomerizations of such polyhedra since they no longer

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provide dimensionality reduction (i.e., for v > 6, $v-4 \ge 3$). However, the experience provided by the Gale transformation study of five-and six-vertex polyhedra⁸ coupled with the still manageable number of combinatorially distinct seven-vertex polyhedra, namely 34 (ref. 10), allows an exhaustive study of isomerizations of seven-vertex polyhedra. This paper summarizes the results of such a study. Polyhedra having seven vertices are of interest in representing the coordination polyhedra of the large variety of known ML7 complexes.¹¹

2. Background

A polyhedral isomerization step may be defined⁶ as a deformation of a specific polyhedron P₁ to the point that the vertices define a new polyhedron P₂. Of particular interest in the context of this work are sequences of two polyhedral isomerization steps $P_1 \rightarrow P_2 \rightarrow P_3$ with the following properties:

(a) P_3 has the same number of edges as P_1 .

(b) P_2 has one less edge than P_1 (or P_3).

Such a polyhedral isomerization can be considered to result from removal of one edge from P_1 to give P_2 followed by adding the edge back to P_2 in a different way to give P_3 . Note that by definition all polyhedra involved in a sequence of isomerization steps must have the same number of vertices. If P_1 and P_3 are combinatorically equivalent, the polyhedral isomerization $P_1 + P_2 + P_3$ is called a <u>degenerate</u> polyhedral isomerization.

Let us now consider the effects of edge removal from P_1 on the face structure of the resulting polyhedron P_2 . Removal of a single edge will convert two faces into a single larger face, which can be called the <u>pivot</u> face. If the pivot face has n edges, then the process $P_1 \rightarrow P_2 \rightarrow P_3$ defined as above will be an n-pyramidal process. Note that for rearrangements of polyhedra having v vertices, the admissible values for n are $4 \leq n$ $\leq v$. A v-pyramidal polyhedral isomerization of a v-vertex system is a <u>planar</u> polyhedral isomerization since the intermediate "polyhedron" P_2 is actually a planar polygon with v vertices.

These concepts may be illustrated more concretely. A 4-pyramidal process involves removal and subsequent addition of an edge in the following

-2-

way so that P_1 and P_3 have the same number of edges:



This, of course, is the familiar dsd process.^{7,12} A 5-pyramidal process merges adjacent triangle and quadrilateral faces into a pentagonal face in the intermediate polyhedron P_2 as follows:



An example of this type of process is described in the Gale diagram paper.⁸ The 4-pyramidal and 5-pyramidal processes (1) and (2) can be combined to give the following process:



In this process (3) <u>two</u> edges are removed from P_1 to give P_2 and those two edges are added back to P_2 in a different way to give P_3 . Such a combination of an m-pyramidal and an n-pyramidal process where n>m is conveniently classified as an n-pyramidal process since the intermediate polyhedron P_2 has a pivot face having n edges.

This paper also considers parallel polyhedral isomerization processes. In a parallel process two or more <u>equivalent</u> edges are removed from P_1 to give P_2 and the same number of edges are added in a different way to equivalent sites of P_2 to give P_3 . Examples of parallel processes include the double dsd interconversion of two eight-vertex D_{2d} -dodecahedra through a square antiprismatic intermediate⁵ and the triple dsd interconversion

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of two six-vertex octahedra through a trigonal prismatic intermediate (the "Bailar twist").¹³

The total number of possible rearrangements for polyhedra having v vertices can rapidly become unmanageably large with increasing v even for cases where v is still of chemical interest. Energetic and symmetry considerations can be used to select rearrangements of specific interest in a chemical context. Thus rearrangements $P_1 \rightarrow P_2 \rightarrow P_3$ involving a pivot face in P_2 having a minimum number of edges are energetically favored since a minimum number of ligands need to approach coplanarity in the intermediate polyhedron P_2 . A large number of coplanar ligands is definitely unfavorable in terms of interligand repulsion and in extreme cases (> 6 coplanar ligands) may require impossible atomic orbital hybridizations. Such energetic considerations make polyhedral isomerizations involving dsd processes the most energetically favorable in accord with the remarkable insight of Lipscomb nearly 20 years ago. Similar considerations lead to exclusion of planar polyhedral isomerizations for energetic reasons. Use of such energetic considerations appears to be sufficient to reduce the number of interesting rearrangements of seven-vertex polyhedra to a manageable number. However, use of a similar approach to study rearrangements of eight-vertex polyhedra will undoubtedly require not only such energetic considerations but also restriction to polyhedral systems having certain symmetry since otherwise the total number of combinatorially distinct eight-vertex polyhedra is an unmanageable 257 (ref. 10).

In exploring relatively large numbers of seven-vertex polyhedral isomerizations, it has proven most convenient to consider first the intermediate polyhedra P₂ in rearrangements of the type $P_1 + P_2 + P_3$. Such polyhedra must necessarily have at least one non-triangular face and can conveniently be called non-deltahedra. The five seven-vertex deltahedra thus cannot be intermediate polyhedra P_2 in polyhedral rearrangements. This reduces the number of possible seven-vertex intermediate polyhedra the 29 seven-vertex non-deltahedra. Furthermore, one of to these non-deltahedra, namely the hexagonal pyramid, can be the intermediate polyhedron only in energetically unfavorable 6-pyramidal processes. For the remaining 28 seven-vertex non-deltahedra it is feasible to draw diagonals across the non-triangular faces in all possible ways thereby generating all possible transformations $P_1 + P_2$ and $P_2 + P_3$ involving the non-deltahedron in question as P_2 . If two different ways of drawing diagonals (conveniently called <u>diagonalization</u>) across the non-triangular faces of a given non-deltahedron lead to combinatorially equivalent polyhedra P_1 and P_3 , then the non-deltahedron in question can serve as the intermediate polyhedron P_2 in a <u>degenerate</u> isomerization $P_1 + P_2 + P_3$.

3. Results

Federico¹⁰ has described the properties of the 34 combinatorially distinct polyhedra having seven faces. These polyhedra P_i can be converted to their duals⁹ P_i^* by the following process:

(1) The vertices of P_i^{\star} are located at the midpoints of the faces of P_i .

(2) Two vertices of P_i^* are connected by an edge if and only if the corresponding faces of P_i share an edge.

The duals of the 34 polyhedra having seven faces are the 34 possible combinatorially distinct polyhedra having seven vertices.

The properties of these 34 seven-vertex polyhedra are listed in Table 1 including the following for each polyhedron:

The number of the dual in Federico's paper¹⁰ so that readers can relate material in this paper to that earlier work. Federico's paper contains the Schlegel diagrams⁹ of all of the polyhedra having seven faces.
 The degrees of the vertices (vertex index) expressed as a four-digit

number $v_6v_5v_4v_3$ where v_n is the number of vertices of degree n.

(3) The sizes of the faces (face index) expressed as a four-digit number $f_6f_5f_4f_3$ where f_n is the number of faces having n edges (and T is used as a digit for ten).

(4) The symmetry point group.¹⁴

(5) Other distinctive features such as the number of edges of a particular type where $e_m e_n$ means an edge connecting a vertex of degree m with one of degree n.

All of the non-deltahedra in Table 1 have been investigated as intermediate polyhedra P_2 for polyhedral rearrangements of the type $P_1 \rightarrow P_2 \rightarrow P_3$. This has been done by the diagonalization process described in the previous section, i.e., all possible diagonals have been drawn across vertices of non-triangular faces and the resulting polyhedra have

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been determined (and in Table 1 are indexed according to the Federico number). For non-deltahedra having only triangular and quadrilateral faces the number of different ways of drawing such diagonals is 2f4 where f4 is the number of quadrilateral faces. For non-deltahedra having pentagonal faces (E in Table 1) only diagonalization of the pentagonal face has been considered in Table 1 since diagonalization of a pentagonal face is assumed to be energetically more favorable than diagonalization of a quadrilateral face. There are five different ways of drawing a diagonal across a pentagonal face to split the pentagonal face into a triangular and a quadrilateral face.

A non-deltahedron P_2 can be the intermediate polyhedron in a degenerate polyhedral isomerization $P_1 \rightarrow P_2 \rightarrow P_3$ (P₁ and P₃ are combinatorically equivalent; P_2 has one less edge than P_1 or P_3) if and only if two or more different ways of diagonalizing a non-triangular face of P_2 lead to the same polyhedron corresponding to P1 in one case and P3 in the other case. If the non-triangular face of P1 being diagonalized is quadrilateral, isomerization is a single dsd process. Such degenerate such an isomerizations are not particularly common and therefore are starred in Interestingly only one of the five seven-vertex deltahedra, Table 1. namely the low symmetry (C_2) #13, can undergo degenerate isomerization through a single dsd process. The other seven-vertex deltahedra require multiple dsd processes or energetically relatively unfavorable 5-pyramidal or 6-pyramidal processes for their degenerate polyhedral iscmerizations.

These considerations allow one to define a <u>dsd rigidity index</u> of a polyhedron P_1 as the number of edges that must be removed from P_1 in a dsd manner (i.e., converting two adjacent triangular faces into a single quadrilateral face) in order to give an intermediate polyhedron P_2 , which upon adding back the same number of edges gives a polyhedron P_3 combinatorially equivalent to P_1 , i.e., $P_1 \rightarrow P_2 \rightarrow P_3$ is a degenerate isomerization involving only dsd provesses. Four of the five seven-vertex deltahedra including the chemically significant¹¹ pentagonal bipyramid and capped octahedron have a dsd rigidity index of 2 which means that a

-6-

degenerate isomerization must involve loss of two edges to give an intermediate polyhedron having two quadrilateral faces. Four of the 28 seven-vertex polyhedra having only triangular and quadrilateral faces cannot undergo degenerate isomerizations involving only dsd (4-pyramidal) processes and therefore have a dsd rigidity index of 0. The degenerate isomerization of such polyhedra must involve less energetically favorable n-pyramidal ($n\geq 5$) processes. None of the seven-vertex polyhedra having a dsd rigidity index of 0 has been found to be chemically significant.

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The relationships between dsd processes involving seven-vertex deltahedra are depicted in the dsd-graphs shown in Figure 1. The vertices of the dsd graphs represent seven-vertex polyhedra designated by the The edges of the Federico numbers of their duals as given in Table 1. dsd graph represent a "diamond-square" $(P_1 + P_2 \text{ in } (1) \text{ above)}$ or a "square-diamond" ($P_2 \rightarrow P_3$ in (1) above) isomerization step in a process depending upon the direction the edge is traversed. Vertice on the left of these dsd graphs represent seven-vertex deltahedra having ten ngular faces (i.e., Federico numbers #11, #12, #13, #20, and #23), vertices in the center of these dsd graphs represent seven-vertex polyhedra having eight triangular faces and one quadrilateral face (i.e., Federico numbers #14, #15, #16, #21, #22, #24, #25, and #28), and vertices on the right of these dsd graphs represent seven-vertex polyhedra having six triangular and two quadrilateral faces (i.e., Federico numbers #18, #26, #27, #30, #31, #32, #33, #36, and #37). Degenerate single dsd processes (i.e., those corresponding to starred polyhedra in the diagonalization column of Table 1) appear as double edges in the dsd graphs. Degenerate double dsd processes are circuits of length 4 including a left vertex corresponding to the starting deltahedron. In forming these circuits only a double edge can be traversed twice; a single edge can only be traversed once.

The dsd graphs in Figure 1 clearly indicate the dsd rigidity index of 1 for the C₂ deltahedron #13 (i.e., through #16 or #21 as an intermediate) and the dsd rigidity indices of 2 for the other four seven-vertex deltahedra including the chemically significant pentagonal bipyramid (#23) and the capped octahedron (#20). Of particular interest are the degenerate double dsd isomerizations of these two polyhedra through the route #20 or #23 + #28 + #36 + #28 + #20 or #23 since the intermediate polyhedron #36 is the capped trigonal prism which is a chemically

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significant non-deltahedral seven-vertex coordination polyhedron¹¹ with six triangular and two quadrilateral (rectangular) faces. In the case of degenerate isomerization of the pentagonal bipyramid the subtraction of two edges to form the capped trigonal prism intermediate and the subsequent addition of two edges to reform a combinatorially equivalent pentagonal bipyramid can each be viewed as concerted processes so that the symmetry of this system never falls below the C_{2v} symmetry of the capped trigonal prism. This is a good example of two parallel dsd processes and is feasible since the C_{2v} point group of the capped trigonal prism is a subgroup of the D5h point group of the pentagonal bipyramid. A similar concerted view of the degenerate double dsd isomerization of a capped octahedron through a capped trigonal prismatic intermediate is not feasible since the two edges of the capped octahedron which are removed to form a capped trigonal prism are non-equivalent. This is a consequence of the fact that the C_{2v} point group of the capped trigonal prism is not a subgroup of the Cov point group of the capped octahedron. Despite the attractiveness of the degenerate isomerizations of the pentagonal bipyramid and the capped octahedron through capped trigonal prismatic intermediates because of the relatively high symmetry of the key polyhedra and the chemical significance of the intermediate polyhedron, Figure 1 indicates a variety of other possible dsd processes for the degenerate isomerization of the pentagonal bipyramid and the capped octahedron. Distinguishing between these diverse dsd processes is likely at best to be extremely difficult experimentally, although differences in symmetry might be exploited in very carefully designed low temperature n.m.r. experiments on specially constructed systems in order to obtain some relevant information.

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TABLE 1

7

SOME IMPORTANT PROPERTIES OF THE 34 SEVEN-VERTEX POLYHEDRA

Distinctive Features ^f		2v6	$1v_6 + C_3$ axis	$1v_6 + C_2$ axis	capped octahedron	pentagonal bipyramid			0 e33	1 e33		3 e45	2 e45	0 e55	1 e55	
Polyhedra formed by Diagonalization ^e					-				#11, #12	#11, #13	#13* [;]	#13*	#11, #23	#11, #23	#13, #20	#20, #23
dsd Rigidity Index ^d		2	2	1	2	2			2	l	2	2	I	2	1	l
Symmetry		^C 2v	c ₃ v	c ₂	c _{3v}	D _{5h}		ateral Face	c ^s	C1	C ₂ v	cs S	ດ _s	C _s	сı	С _В
Face Index ^c		000T	000T	000T	000T	000T		One Quadril	0018	0018	0018	0018	0018	0018	0018	0018
Vertex Index ^b	hedra	2032	1303	1222	0331	0250		edra with i	1123	1123	1042	0313	0313	0232	0232	0151
Federico Number of Dual ^a	A) Delfa	#11	#12	#13	#20	#23	•	B) Polyh	#14	#15	#16	#21	#22	#24	#25	#28

	Distinctive Features ^f			2 connected e33	2 disconnected e33	1 e44	2 e44	2 e44	3 e44	4-capped trigonal prism (l e33)	0 e33			3-capped trigonal prism, self-dual	self-dual	self-dual	lv3 murrounded by 3v4, melf-dual	4 quadrilateral faces
tinued)	Polyhedra formed by Diagonalization ^e		#15*	#14,#15,#22,#25	#15*。#22*	#22*。#28×	#14,#22,#24,#28	#16,#21,#25*	#15,#24,#25,#28	#28*	#25* , #28*		#18,#26,#30,#33	#32*	#30* , #36*	#27,#32*,#33*,#37	#26,#30,#31,#33, #36,#37	#35*,#40*,#42*
TABLE 1 (Con	dad Rigidity Index ^d		2	2	0	1	2	1	l	1	2	eral Faces	1	0	1	0	1	0
	Symmetry	teral Faces	c2v	сı	c_2	c_2	с ^в	C1	сı	c ₂ v	c ₂	ir Quadrilat	c _s	c ₃ v	c ₃ v	c_2	c1	C2v
	Face Index ^c	wo Quadrila	0026	0026	0026	0026	0026	0026	0026	0026	0026	hree or Fou	0034	0034	0034	0034	0034	0042
	Vertex Index ^b	edra with T	1024	. 0214	0214	0133	0133	0133	0133	0052	0052	edra with T	0115	0034	0034	0034	0034	0016
	Federico Number of Dual ^a	C) Polyh	#18	# 26	# 27	#30	#31	#32	#33	#36	#37	D) Polyh	#35	# 39	\$40	1 4 1	# 42	****

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TABLE 1 (Continued)

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Federico Number of Dual ^a	Vertex Index ^b	Face Index ^C	Symmetry	dsd Rigidity Index ^d	Polyhedra formed by Diagonalization ^e	Distinctive Features ^f
E) Polyh	edra with	Pentagonal o	or llexagonal	Faces		
#17	1024	0107	C _B	1	#14*,#15*,#16	3-capped pentagonal pyramid
#29	0133	0107	C _B	1	#22,#25*,#28*	
#34 .	0115	0115	c1	1	#26,#27,#30,#31*	self-dual
#38	0034	0115	8 0	1	#32*,#36,#37*	
#43	0016	0123	C ₈		#39,#41×,#42×	
# 19	1006	1006	c ₆ v		#11*,#12*,#13*,#14*, #17*,#18*	hexagonal pyramid

These numbers are the same as those given in Table 1 of Federico, P.J. Geom. Ded. 1975, 3, 469. (a)

- The vertex index corresponds to v₆v₅v₄v₃ where v_n is the number of vertices of degree n (see text). <u>م</u>
- H The face index corresponds to $f_6f_5f_4f_3$ where f_n is the number of faces with n edges and the letter is used as a digit for "ten." () ()
- (d) The dsd rigidity index is defined in the text.
- These correspond to degenerate pyramidal isomerization Federico numbers of their duals (note a and text). Combinatorially equivalent polyhedra formed by two The polyhedra formed by drawing all possible diagonals on non-triangular faces are indicated using the different diagonalization processes are starred. processes. (e)
- or edges (e_me_n) of distinctive types, and/or self-duality. The edge designation e_me_n refers to an edge Distinctive features include special names for the polyhedra, the presence or absence of vertices (v_n) connecting a vertex of degree m with one of degree n. E

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(pentagonal bipyramid)

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