Figure 10. Interrelated geologic events/process, including ore formation, triggered by continental break-up.

planet. But, with available information, the rationale behind such preferential behaviour can only be outlined along its broadest contours. The space–time–process continuum that the earth's history represents can be perceived as an *n*-dimensional space where every single ore deposit obtains *n* specific coordinates. Only when all these coordinates are identified and rigorously determined for all known deposits can we expect to rationalize, fully, why ore deposits form *when* and *where* they do. The search for all conceivable coordinates is on, and it seems we have a long way to go.

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Design of crystal structure-specific surfactants based on molecular recognition at mineral surfaces

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The development of highly selective mineral processing reagents is urgently required in order to exploit the relatively difficult to process ore-deposits. Despite extensive research effort, there does not yet exist a comprehensive theory of reagent design. A critical review of past work suggests that new reagents should be not merely metal-ion-specific but also structure-specific. Both the choice of the functional group as well as the molecular architecture of the reagent is crucial to achieve the desired structure specificity. A systematic study of the separation systems where such structure specificity has already been observed and reported, would help delineate the appropriate interaction mechanisms required to accomplish it. A few examples are presented in this article to illustrate the concepts of structure specificity and molecular recognition, which are also of tremendous importance and of current research interest in the study of 'biomineralized' materials like oyster shells, corals, ivory and pearls.

Certain chemicals exhibit the characteristic property of enhanced adsorption at interfaces and hence possess the important attribute of being able to modify interfacial

interactions. Such chemicals known as surface-active agents or 'surfactants' are very widely used in a variety of modern industrial processes. Areas of application are as diverse as motor oils and other lubricants, laundry detergents, personal care products and cosmetics, food additives, petroleum recovery, specialized pharmaceutical formulations, leather processing, building materials, mineral and ceramic processing and effluent treatment[1-3].

The surfactants possess a characteristic chemical structure consisting of (a) molecular components that have little attraction for the solvent or bulk material, normally called the 'lyophobic' ('hydrophobic' or non-polar in aqueous systems) portion and (b) chemical units that have a strong attraction for the solvent called the 'lyophilic' (or 'hydrophilic' or polar) group[1]. The presence of hydrophilic group assists the solubilization of surfactants whereas the nature of the hydrophobic group largely governs its surface-activity, that is, its proclivity to concentrate at an interface, rather than remain in bulk. As the applications of the surfactants are becoming increasingly more complex and specific, the design of these surfactant molecules for particular application that is, the science and 'art' of making tailor-made surfactants has become, in recent years, an important area of interdisciplinary research. Since the desired properties of the surfactants differ from application to application, attempts are being made concurrently in many fields to establish the structure-property relationships for a variety of surfactants[3].

The success of surfactant-based separation processes, such as froth flotation and selective flocculation, adsorptive bubble separation including ion flotation, separation with liquid surfactant membranes, micellar-enhanced ultrafiltration, coacervate-based separation to remove slightly soluble organics and heavy metal ions from water and extraction with reverse micelles, depends on the availability of appropriate surfactants[2,3]. Certain important features of the surfactant design, particularly for mineral processing applications, are discussed briefly in this article.

Surfactants for mineral separation

With steady depletion of high grade, relatively easy-to-process ores, the mineral-processing industry is confronted with a challenging task of finding more efficient techniques to exploit low grade, complex and disseminated type of ore deposits. The development of novel, highly selective reagents is vital for this purpose[4-7].

The main distinguishing feature of mineral processing reagents is their efficiency in achieving selective separation. For instance, the flotation collectors are added to flotation pulp to adsorb selectively at the mineral/water interface to render the surface hydrophobic. Similarly selective adsorption of depressant helps to make the surface hydrophilic and/or to prevent collector adsorption. Frothers, added to the flotation pulp to assist the formation and stabilization of the froth phase, must adsorb selectively at the bubble/water interface. Selective dispersants and flocculants are typically polymeric surfactants which on adsorption selectively disperse or aggregate through bridging the desired components in a particulate suspension. Figure 1 is a schematic diagram illustrating the differences in the properties of commonly used mineral separation reagents. Both, the choice of the functional group (polar portion) essentially determining the selectivity and the design of appropriate molecular architecture (the nature of the nonpolar chain — aliphatic/aromatic, hydrocarbon/ polymeric, branched/linear, hydrophobic/hydrophilic,
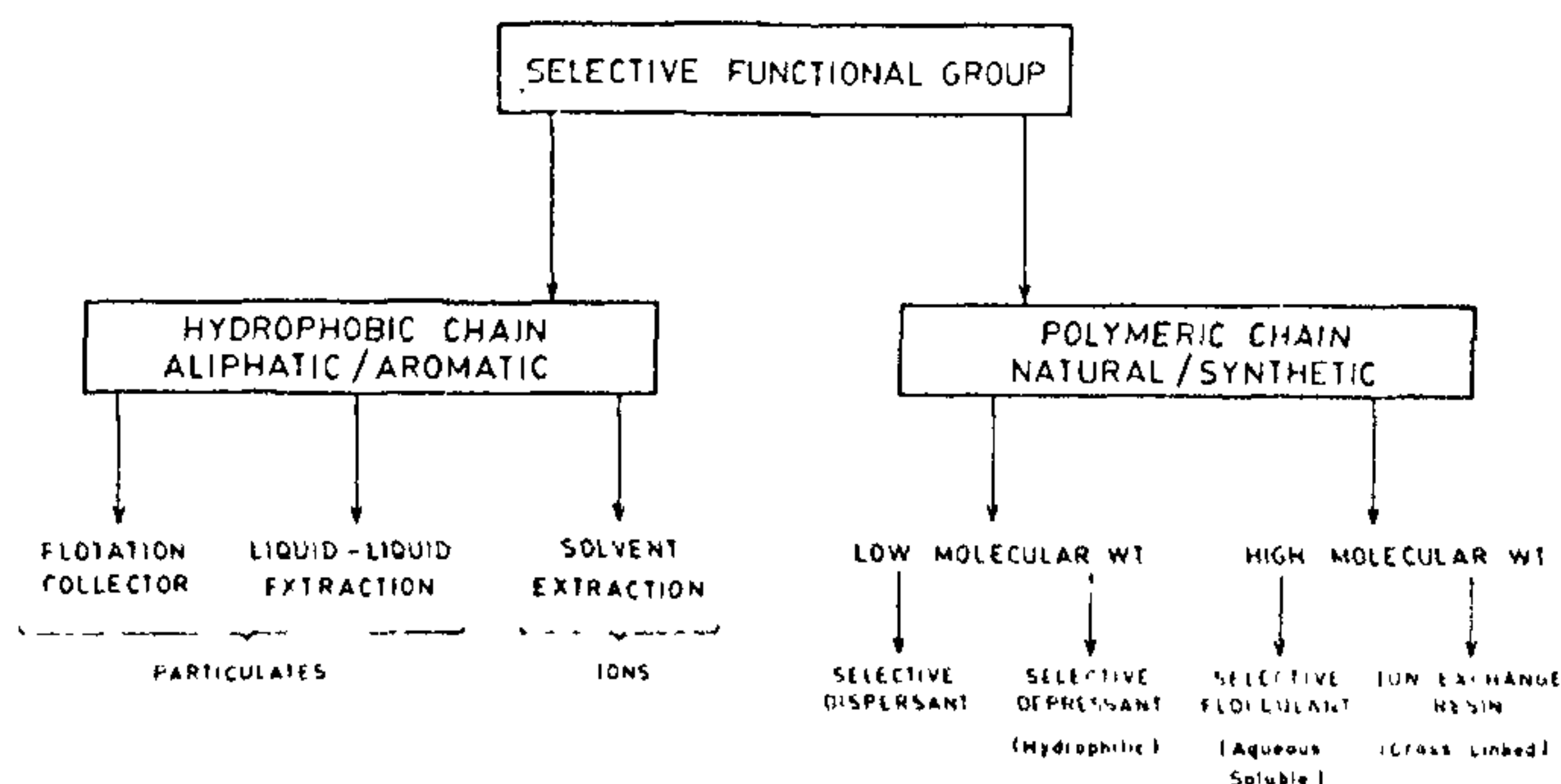

Figure 1. A schematic diagram showing commonly used mineral processing reagents comprising a selective functional group and a nonpolar portion depending upon the end use.

low high molecular weight, natural/synthetic origin, the positioning as well as the spacing between functional group in the chain) are thus crucial for achieving the required reagent performance in use.

It must be stressed that most of the commercial reagents currently being used in the minerals industry were discovered through trial and error methods. Despite extensive research efforts in the field, there does not yet exist a theoretical framework that can provide the practising engineers a scientific and rational basis to select and/or design reagents for a given separation problem at hand[6].

Design of selective reagents

A scientific approach to designing mineral separation reagents involves two important surface-chemical aspects, namely, (a) selection of the appropriate functional group, and (b) design of the corresponding molecular architecture depending on its end use. Presently the selection of functional group or groups is made on the basis of the knowledge available in analytical chemistry literature combined with trial and error experimentation. Several known chelating agents, for example, have been appropriately modified to make them act as selective flotation collectors, depressants, dispersants and flocculants[5,7]. This approach however suffers from a major drawback. Besides being unsatisfactory from a scientific viewpoint, it assumes that the metal ion specificity observed for a functional group in bulk aqueous systems would remain valid during surface chelation at the interface. Moreover, an approach based on only metal ion specificity is not applicable to those systems where separation is to be accomplished amongst minerals having the same constituent metal ion on the surface, for example, separation of fluorite (CaF_2) from calcite ($CaCO_3$), fluorapatite ($Ca_3(PO_4)_2F_2$) and dolomite $(Ca, Mg) CO_3$.

It is thus clear that a comprehensive theory of reagent design should be based on the reagent interactions not merely with the metal ion on the surface, but with the whole surface. Is it possible to design reagents which can recognize the molecular structure of the substrate? Should the reagent design not take into account the differences in the crystal structure of the mineral surfaces? If possible, it would be more appropriate to design reagents having functional groups so spaced that those are compatible with the relative positions of the metal ion sites available on the surface, that is, to design not just metal-specific but structure-specific reagents.

Molecular recognition

It is in this context that recent developments in our

understanding of molecular interactions, involved in the recognition of mineral surfaces by organic molecules present in the mineralization environment, are worth investigating. It is interesting to note that there exist a wide variety of biomineralized materials (engineered by living creatures), such as oyster shells, coral, ivory, pearls, sea urchin spines, magnetic crystals in bacteria, cuttlefish bone, limpet teeth, which consist of inorganic minerals intricately combined with and fashioned by various organic polymers[8]. One of the most striking features of biomineralization is the potential of organisms to synthesize single crystals of elaborate morphology which are species-specific and are replicated generation after generation[9]. Recent investigations have shown that the interaction of organic molecules with crystal surfaces during nucleation and crystal growth determines the crystallochemical specificity of bio-mineralized deposits[10,11]. Since the mechanisms of interaction in such systems are still unclear, several model systems are being studied to elucidate the nature of recognition processes occurring at inorganic/organic interfaces[12,13]. Design of tailor-made additives has been successfully accomplished for the control of morphology during crystallization[14,15]. For example, synthetic barite ($BaSO_4$) crystals prepared in the absence of additives exhibit simple rhombic plate-like morphology in which large (001) faces are bound by smaller (210) sides. The addition of certain stereochemically related diphosphonates (P–P distance of 5.7 Å in the additive molecules matches well with the nearest neighbour S–S distance of 5.6 Å between like-oriented ions on $BaSO_4[OH]$ surface) produces a characteristic morphological change leading to disc and elliptical morphologies as a consequence of incorporation of the additive molecules into the (011) surface which inhibit growth in the entire [001] zone[12]. Similar results have been obtained on crystal nucleation and growth of organic crystals in presence of 'tailor-made' surfactants which interact stereospecifically with crystal surfaces during growth[15].

Crystal structure specificity in mineral separation systems

A critical review of the published literature in mineral processing systems as well as certain observations made during our investigations indicate that there exist several separation systems where the above-mentioned structure specificity has already been reported. A systematic investigation of such systems is important to develop a comprehensive theory of molecular recognition in mineral–reagent interactions. One such system studied in our laboratories is discussed in the following section for illustration.

Iron oxide–alumina–kaolinite separation

The separation of iron oxide from associated alumina-containing minerals is an important industrial problem[16]. Extensive work carried out in our laboratories on this separation system[16-19] indicates that alumina and kaolinite, though both consist of aluminium as constituent metal ion, behave very differently with respect to the reagents investigated by us. These remarkable differences can be attributed to the differences in their crystal structure.

Table 1 lists the crystal structure and the cleavage planes for these minerals. While hematite and corundum have similar structure and cleavage planes, the constituent metal ion is different; corundum and kaolinite both contain a common metal ion (Al), but differ in crystal structure.

The selective flocculation results obtained on 1:1 synthetic mixtures of hematite–alumina and hematite–kaolinite with a modified polyacrylamide flocculant, PAMX, containing 19.3% hydroxamate groups in presence of sodium silicate dispersant, are presented in Figure 2 (ref. 19). The results are presented in the form of weight per cent settled as a function of pH. Perfect selectivity implies 100% settling of one component (Fe_2O_3) against no settling of the other component. While it was possible to separate iron oxide from kaolinite, alumina could not be separated from iron oxide using this reagent. Hydroxamate functional groups are known to chelate well with Fe^{3+} (compared to Al^{3+}) as illustrated by the bulk stability constant values presented in Table 2.

It is clear from the results that the differences in chelation ability of PAMX with Fe on hematite surface and Al on corundum surface are not adequate to achieve the desired selectivity. Selective separation was possible in hematite–kaolinite system because Al sites on the cleavage basal plane of kaolinite (surface) are not available as in the case of corundum. Kaolinite has broken Al–O bonds (Al sites required for adsorption) only on the edge surfaces which are relatively less predominant than the basal surface[20]. Therefore, PAMX does not chelate on kaolinite surface as effectively and hence no flocculation of kaolinite is observed in hematite–kaolinite mixtures. Furthermore, higher negative charge of kaolinite surface, compared to hematite and corundum, also prevents adsorption of
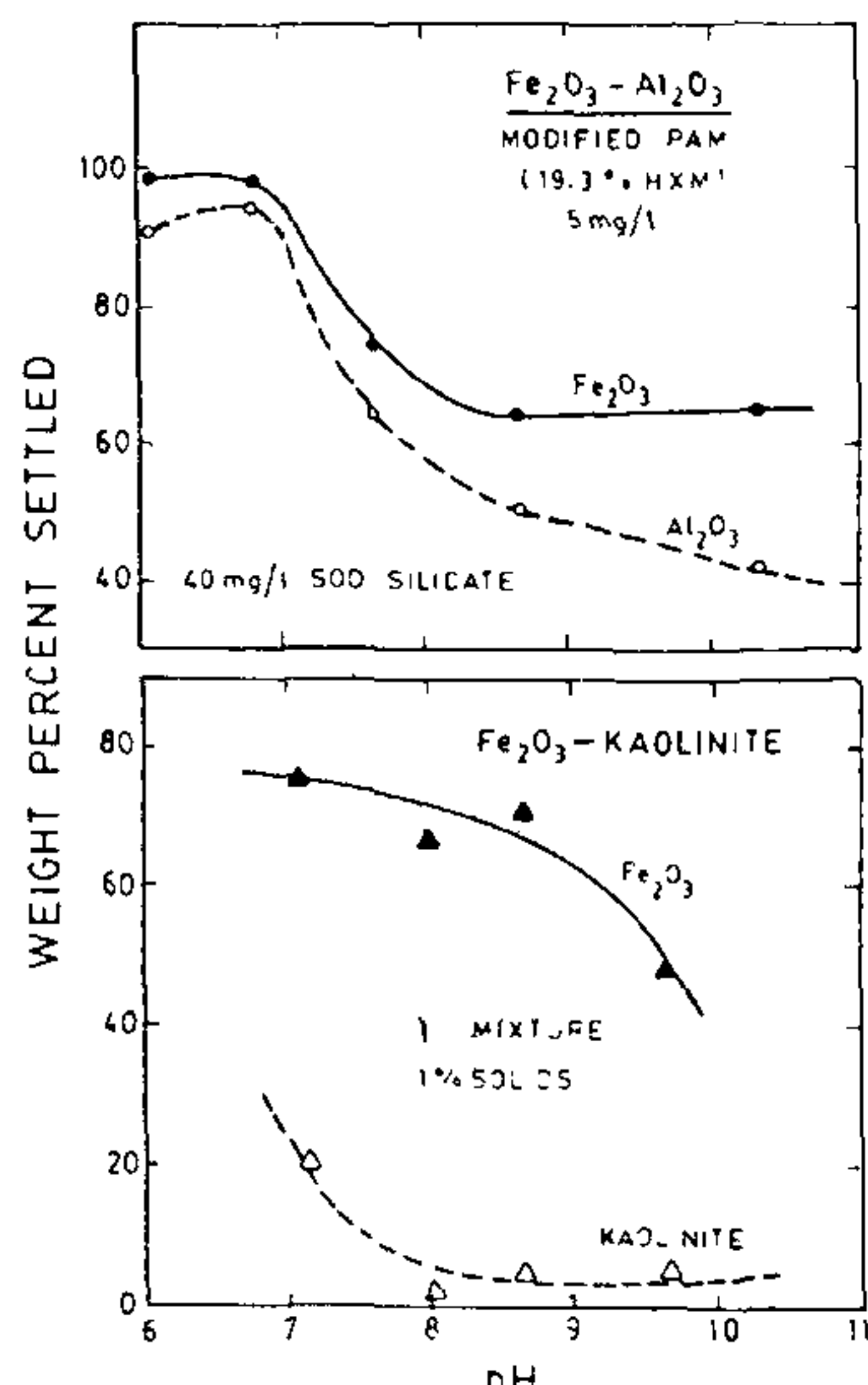

Figure 2. Selective flocculation of hematite (Fe_2O_3) from its synthetic mixtures with alumina and kaolinite respectively using a modified polyacrylamide (PAMX) flocculant containing 19.3% iron chelating hydroxamate functional groups (After ref. 19).

Table 2. Fe–Al hydroxamate system — stability constants

Cation	Log K_1	Log K_2	Log β_2	Log K_3	Log β_3
Fe^{3+}	11.42	9.68	21.1	7.23	28.33
Al^{3+}	7.95	7.34	15.3	6.18	21.47

where $K_n = \dfrac{[ML_n]}{[ML_{n-1}][L]}$ for reaction $ML_{n-1} + L \rightleftharpoons ML_n$

and $\beta_n = \dfrac{[ML_n]}{[M][L]^n}$ for reaction $M + nL \rightleftharpoons ML_n$.

highly negatively charged PAMX and hence less flocculation of kaolinite with PAMX[16]. The crystal structure, in addition to the metal ions, thus plays an important role in achieving selectivity.

The selective flocculation experiments carried out with starch, a nonionic polymer in this system, were found to be even more interesting[18]. Same 1:1 synthetic mixtures when flocculated with an essentially nonionic natural maize starch yielded remarkable results. Again, while there was no separation observed in hematite alumina system, almost perfect separation was possible in the case of iron oxide kaolinite (Figure 3). In this case since starch is a nonionic flocculant, the remarkable selectivity appears to be the consequence of different crystal structure only.
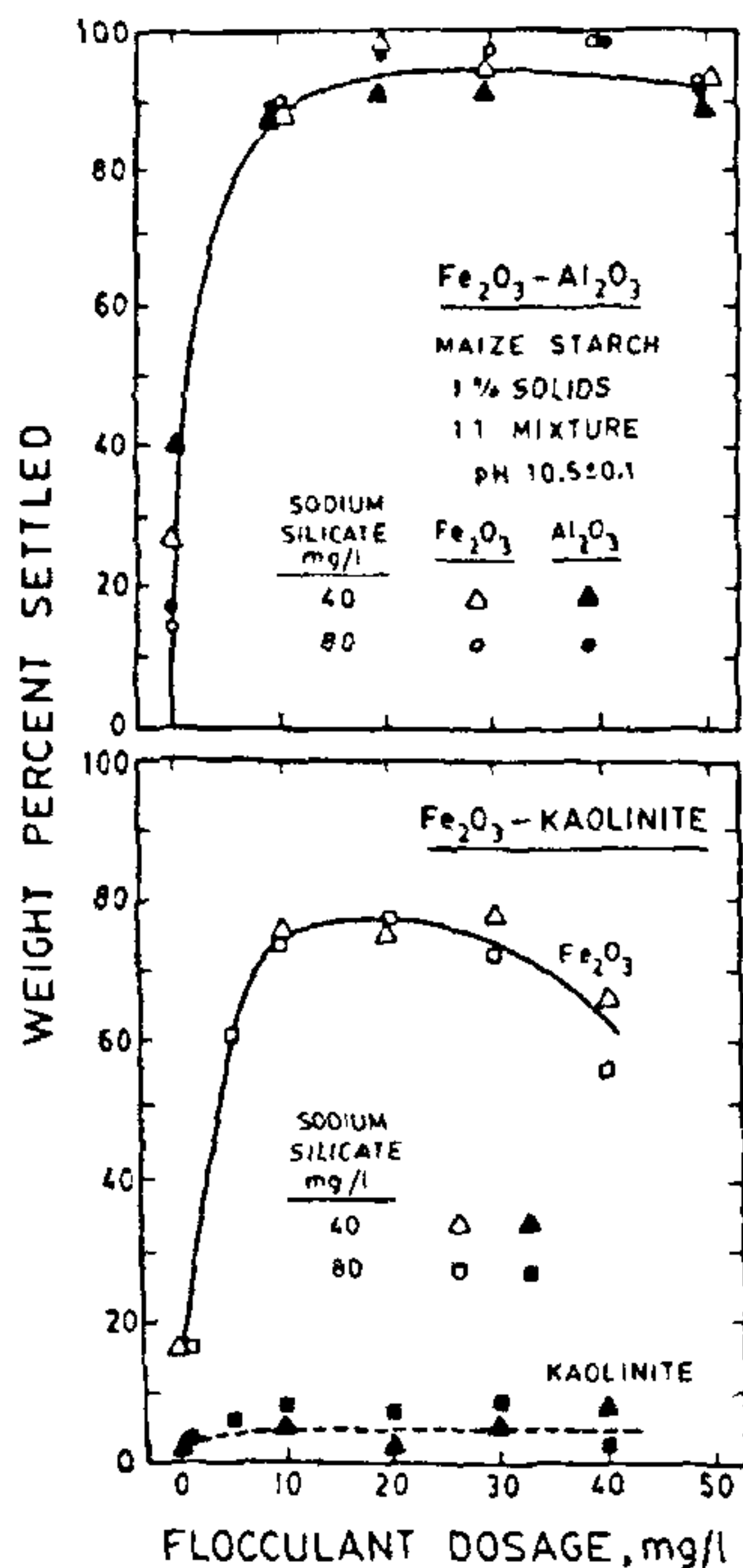
Table 1. Crystal structure and cleavage planes of minerals

Mineral	Crystal structure	Cleavage
Hematite, Fe_2O_3	Rhombohedral $c/a = 1.3656$	[0001] & [1011]
Corundum, Al_2O_3	Rhombohedral $c/a = 1.3630$	[0001] & [1011]
Kaolinite, $Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$	Monoclinic	Basal

Figure 3. Selective flocculation of hematite (Fe_2O_3) from its synthetic mixture with alumina and kaolinite respectively using a natural maize starch flocculant at pH 10.5 ± 0.1 in presence of sodium silicate dispersant (After ref. 19).
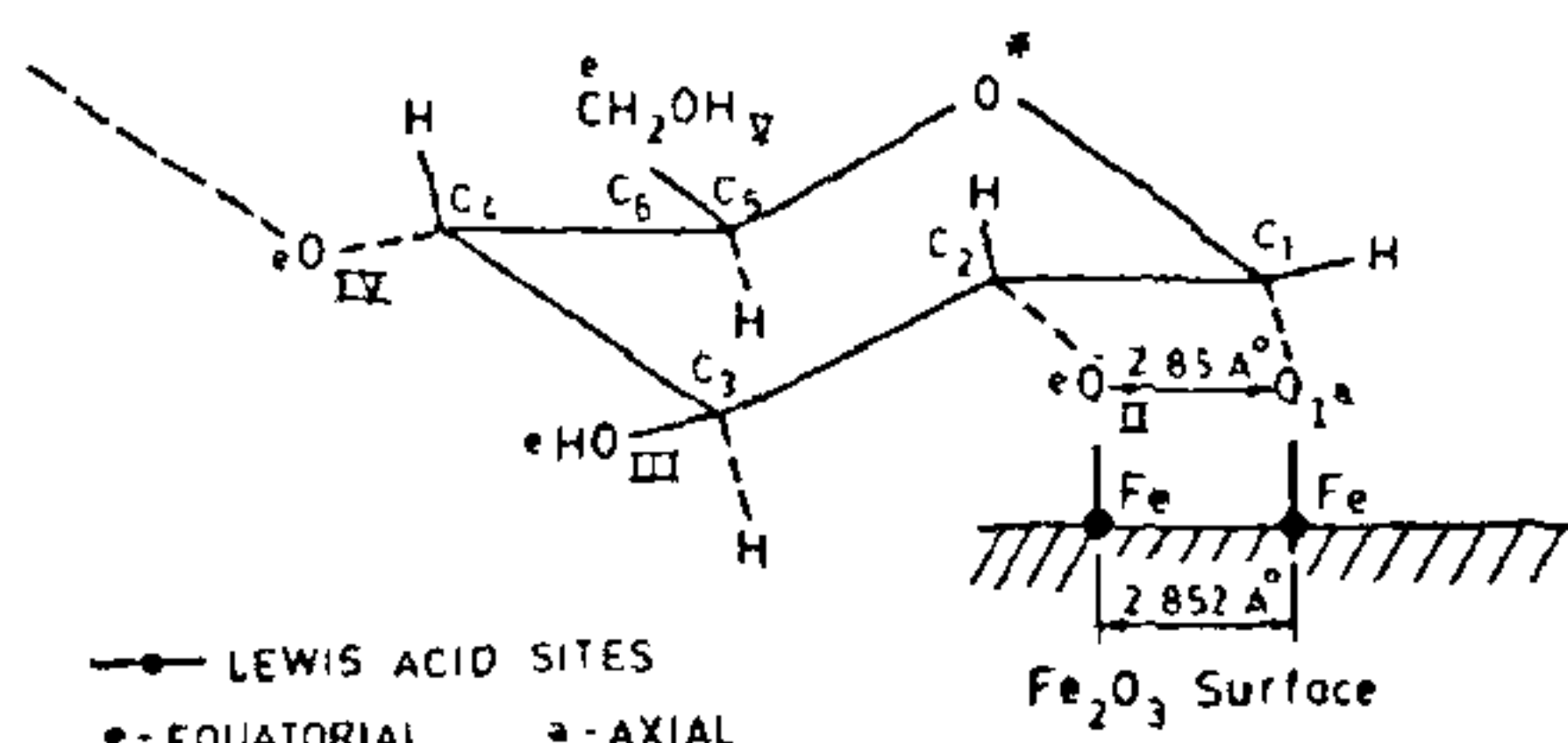

Figure 4. A schematic diagram illustrating the mechanism of starch adsorption through binuclear complexation with Fe sites on hematite (also corundum) surface (After ref. 18).

Ravishankar et al.[18] have further shown that starch adsorption on hematite and corundum surfaces appears to be highly structure-specific. Starch consists of amylopectin and amylose components. The relative content of these two components varies depending on the source of starch. The amylose having a low-molecular weight (10^5) contains 200–300 anhydroglucose units with one end group per molecule. Amylopectin is of higher molecular weight ($10^6–10^7$) and consists of more than 5000 cross-linked anhydroglucose units with, on an average, about one end group for every 20–30 glucose units. A typical end group of such starch molecules is shown in Figure 4. Amongst the four OH groups, $O_I H$ is the most polarizable due to the close proximity of the heterocyclic oxygen (denoted by O*). The distance between O_I and adjacent O_{II} can be calculated to be 2.85 Å (ref. 18). As shown in Figure 4, this O–O distance in adsorbing starch end groups is remarkably close to the average Fe–Fe (or Al–Al in

corundum) distance of 2.852 Å at the cleavage plane ($10\overline{1}1$) of hematite. Ravishankar et al. have proposed a mechanism of starch adsorption involving this binuclear complexation on hematite and corundum surfaces[18]. It is because of this structure-specific interaction of starch that we do not observe any selectivity between hematite and corundum. On the other hand, kaolinite is not flocculated well with starch since it does not have Al sites on its cleavage surface (basal plane). The separation of hematite from kaolinite is thus possible with starch-based flocculants.

There are a number of other mineral-reagent systems where similar observations, even though not explicitly stated, have been reported. For example, it is interesting that fluorite responds to oleate flotation at concentrations an order of magnitude lower than what is required for calcite flotation[21]. What is even more intriguing is the experimental observation that with hydroxamate reagent (a fatty acid derivative) this order is reversed. Calcite floats better than fluorite with hydroxamate collector[21]. Since the chelating cation is the same in calcite and fluorite, the differences in their flotation behaviour can only be attributed to the differences in their crystal structure. Even though not substantiated, Sorensen[21] has emphasized the role of fluoride anions in fluorite flotation vis-à-vis carbonate in case of calcite. Work is in progress in our laboratories to establish and then elucidate the role of crystal structure in determining the selectivity of reagent adsorption at mineral/water interfaces. This understanding would help us design structure-specific flotation collectors.

Role of molecular architecture

The structure specificity can also be achieved through an appropriate design of the adsorbing molecule, that is, by controlling the spacing between the chelating functional groups. In fact, in bulk chelation chemistry the importance of spacing between the chelating groups is well known. For example, hydroxamic acids have

been known to form stable complexes with heavy metals such as Fe(III). Of particular interest among the natural products containing hydroxamic acids are those which consist of tri-hydroxamic acids called siderophores. A distinguishing feature of these natural compounds (e.g. ferrioxamine-B) is the fact that the three hydroxamic acid groups are spaced along the chain at just the right intervals to favour the formation of intra-molecular octahedral iron (III) complex, and hence their unusually high affinity for Fe(III) as compared to monohydroxamic acids.

Winston and coworkers[22-24] synthesized a series of polymers having side chains terminated by hydroxamic acids. The influence of spacing between the hydroxamic acid groups on the stability constants with iron was thus studied. With side chains of sufficient length, three hydroxamic acids from neighbouring side chains could easily fit the octahedral coordination sphere of iron, and therefore the stability constant with iron was observed to be appropriately high. With less distance between hydroxamic acids (shorter side chains), the complex stability was observed to be lower, reflecting the decreasing ability of hydroxamic acids to coordinate iron. With very short side chains, however, the iron complexes of polymers precipitated immediately on addition of iron. With too short side chains, the intramolecular complexation was not possible and extensive crosslinking due to intermolecular complexation made the polymer insoluble and hence the precipitation[24]. Thus, through the control of these subtle changes in the structure and arrangement of the chelating functional groups, the authors demonstrated the possibility of enhancing the selectivity and the effectiveness of a new iron-chelating drug[23].

The role of molecular architecture in the design of flotation reagents was demonstrated through an excellent study made by Nagaraj and Somasundaran[25] on the flotation of chrysocolla using certain hydroxyoxime derivatives. Certain compounds were found more efficient than others as illustrated in Figure 5. It is important to note here that the collecting action (flotation recovery) is a combined effect of both the complexing power of the reagent as well as its hydrophobicity. Hydroxyoximes chelate on the surface through two types of bonds — an ionic bond between copper ion and the phenolic oxygen and the bond between the same copper ion but with the oximic nitrogen. EHMO calculations reported by Aliaga and Somasundaran[26] showed that due to higher electron density on the oximic nitrogen, OHAPO and OHBUPO could form stronger complexes with copper compared to SALO and OHNPO and therefore those were found to be better collectors. Better flotation with OHBUPO than OHAPO was attributed to the more hydrophobic nature of the substituent in the former.

It is thus clear that both the choice of the functional

Figure 5. Flotation of chrysocolla using hydro-oxime derivatives: Effect of molecular architecture (After ref. 25).

group as well as the molecular architecture of the molecule determine the selectivity and the structure specificity achieved during adsorption on the surface. An enhanced understanding of the origin of crystal-structure specificity in mineral-reagent systems already reported in the literature, would help researchers develop the scientific theory of reagents design.

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Conjugal gene transfer in filamentous cyanobacteria

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Though cyanobacterial gene manipulation is very difficult and time consuming than bacteria due to constraints like longer generation time, cryptic genetic material, lack of colony formation (filamentous cyanobacteria). But, the time has come when genetic improvement has become inevitable for the better utilization of biotechnologically important cyanobacteria like *Anabaena, Nostoc, Oscillatoria, Calothrix, Westiellopsis, Spirulina*. Cyanobacteria are prokaryotes with gram-negative bacteria like cell wall and eukaryotes like aerobic photosynthetic apparatus. Cyanobacteria have a high market value as natural therapeutic and colouring substance besides, its conventional use as protein suppliment and nitrogen fixers. To bring down the product cost, extensive cyanobacterial genetic manipulations are badly needed. Cyanobacterial microbiologists were handicapped due to lack of reproducible gene-transfer system in filamentous cyanobacteria till Wolk *et al.* for the first time reported successful gene transfer in *Anabaena* using shuttle vector pVW_1 and its derivatives, broad host range conjugal plasmid RP_4 of incompatibility group (IncP) and helper plasmid pGS101/pGJ28. The use of these plasmids made the conjugation possible in cyanobacteria. Indeed conjugation is the only technique now available for gene transfer in filamentous cyanobacteria, viz. *Anabaena, Nostoc, Fremyella, Fischerella*. It has opened the way for gene manipulation studies in other biotechnologically important filamentous cyanobacteria like *Spirulina* and *Westiellopsis*, provided one takes for surety of conjugal contact, degradation of transferred DNA from host's (recipients) restriction enzyme digestion, and the transferred gene (plasmid) is capable of replication or integration into the recipient cell.

CYANOBACTERIA are a unique group of prokaryotes with both oxygenic photosynthesis and the capacity to fix elemental nitrogen. An understanding of the coexistence of these two primary biosynthetic processes thus promises a closer application in higher plants than other nitrogen-fixing microbes, which either have anoxygenic photosynthesis or are heterotrophic. Besides, cyanobacteria have a high market value as a source for natural therapeutic and colouring substances[1–3]. To bring down the product cost, extensive cyanobacterial gene manipulations are needed.

Investigations on gene organization and function in these microbes (cyanobacteria) are limited because of their slow growth, in many cases multicellular nature of their colony forming units, the spreading tendency of many of them on agar plates and nonavailability of simple and efficient gene transfer systems. The identification and characterization of a 'classic' mechanism for genetic recombination in cyanobacteria lags behind tremendously in comparison to other prokaryotes. For