Systematic Violation of Loewenstein's Rule Established

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Since its formulation 50 years ago, Loewenstein's rule [1] has become a standard argument in the crystal chemistry of, notably, layer and framework silicates and zeolites. The validity of the rule is usually taken for granted, and it is used rather schematically, sometimes even without making reference to the original paper. The rule is normally used as an argument to account for two experimental observations: i) In alumosilicates, AlO_4-tetrahedra tend to avoid each other, leading to alternating AlO_4- and SiO_4-tetrahedra for a Si : Al ratio of 1:1; ii) Except in rare cases, alumosilicates have an Al : Si ratio smaller than or equal to 1. Observation i) could be rationalized theoretically to be due to the expenditure in elastic energy which is necessary to deform the structure when the bigger Al^{3+} replaces Si^{4+}[2]. Observation ii) is not easily explained, in fact, it does not seem that fully convincing arguments have been put forward for its explanation.

We have prepared series of compounds where Loewenstein's rule is breached systematically, and continuously. These belong either to the melilite, or to the sodalite structure type. Crystallographic, structural, chemical and physical features and properties have been determined and will be reported.

[1] Loewenstein W., *Amer. Miner.*, 1954, **39**, 92. [2] Bosenick A., Dove M.T., Myers E.R., Palin E.J., Sainz-Diaz C.I., Guiton B.S., Warren M.C., Craig M.S., Redfern S.A.T., *Mineralogical Magazine,* 2001, **65**, 193.

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