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Amphibole asbestos from Libby, Montana: Aspects of nomenclature

ANN G. WYLIE[1] AND JENNIFER R. VERKOUTEREN[2,*]

[1]Laboratory for Mineral Deposits Research, Department of Geology, University of Maryland, College Park, Maryland, 20742, U.S.A.
[2]Chemical Science and Technology Laboratory, National Institute of Standards and Technology, Gaithersburg, Maryland, 20899, U.S.A.

ABSTRACT

Richterite-asbestos and winchite-asbestos are not listed in the federal regulations governing asbestos. However, asbestiform winchite is found in the gangue at the Libby, Montana, vermiculite deposit, where asbestos-related diseases have been reported among the miners and millers. Changing amphibole nomenclature, uncertainties in Fe^{2+}/Fe^{3+}, and natural compositional variability result in samples of the asbestiform amphibole from Libby being variably classified as soda tremolite, richterite, sub-calcic actinolite, and winchite. A classification of winchite-asbestos is assigned for two samples of Libby asbestos analyzed for this report, consistent with the most recent International Mineralogical Association classification system. Although some of the unit-cell parameters and optical properties reported here are distinctive, others are very similar to the tremolite-actinolite series.

INTRODUCTION

The U.S. Occupational Safety and Health Administration (OSHA) and the U.S. Environmental Protection Agency (EPA) have regulated asbestos since the early 1970s (summarized by Vu 1993). The current regulations specify chrysotile and the asbestiform habit of five amphiboles: tremolite, actinolite, anthophyllite, riebeckite (listed as crocidolite) and cummingtonite-grunerite (listed as amosite) (Title 40, Code of Federal Regulations, Part 61 and Part 763; Title 29, Code of Federal Regulations, Part 1910 and Part 1926[1]). These minerals were known at the time the regulations were first written to have been mined commercially as asbestos. Although there have been modifications to the regulations since they were first promulgated, most notably to clarify that cleavage fragments are not asbestos (Federal Register 1992), the minerals that are regulated have not changed. In particular, the sodic-calcic amphiboles winchite and richterite are not regulated.

An asbestiform amphibole occurs as a gangue mineral in the Zonolite® vermiculite ore body in Libby, Montana, which was mined from 1923 to 1990. Estimates of the abundance of the amphibole in the unprocessed ore range from 0 to ≈ 5 wt% (Atkinson et al. 1982). An elevated incidence of mesothelioma, the hallmark of asbestos exposure, has been reported among the miners and millers of Zonolite® in several studies that were summarized by Ross et al. (1993). In late 1999 and early 2000, many deaths alleged to be due to asbestos exposure in Libby were reported in the popular press, stimulating Congressional oversight (106th Congress 2000). Of particular significance for the regulatory community is the identity of the asbestiform amphibole.

The asbestiform amphibole at Libby has been referred to under a variety of names, including tremolite, actinolite, soda tremolite, richterite, and winchite. The current nomenclature used in the popular press and by the residents of Libby is tremolite, or tremolite/actinolite. Deer et al. (1963) give an analysis of an amphibole from Libby (taken from Larson 1942) that they identify as "richterite (soda tremolite)."

In the amphibole classification system of Deer et al. (1963), Miyashiro's (1957) classification of the alkali amphiboles was generally adopted. However, of particular significance to the Libby amphibole, Deer et al. (1963) used the name richterite in place of soda tremolite, dividing tremolite from richterite at $NaCa_{1.5}$, (they considered winchite to be a subset of richterite). The International Mineralogical Association (IMA) classification (Leake 1978) continued the use of richterite in place of soda tremolite and added specific chemical parameters for distinguishing the actinolite series from richterite and for applying the name winchite. The parameters for richterite were $^{B}(Ca + Na) \geq 1.34$ atoms per formula unit (apfu) and $0.67 < {}^{B}Na < 1.34$ apfu (classifying the amphibole as a member of the sodic-calcic group) and $Si > 7.5$ apfu and $^{A}(Na+K) \geq 0.5$ apfu. By the IMA 1978 classification scheme, winchite is also a member of the sodic-calcic group and is distinguished from richterite by $^{A}(Na + K) < 0.5$ apfu. Members of the actinolite series belong to the calcic group and have $^{B}(Ca + Na) \geq 1.34$ apfu and $^{B}Na < 0.67$ apfu. Another relevant evolution in the nomenclature was the division between tremolite and actinolite; according to Deer et al. (1963), tremolite contained between 0 and 20% ferro–actinolite while according to the IMA, tremolite contained no more than 10% ferro–actinolite. The most recent nomenclature changes in Deer et al. (1997) and the revised IMA classification (Leake et al. 1997) changed the positions of the subdivisions to fit a 50% rule. Under these changes, ^{B}(Na +

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[1] Regulations dealing with asbestos can be obtained through the websites maintained by OSHA (www.osha.gov) and the EPA (www.epa.gov).

* E-mail: jennifer.verkouteren@nist.gov

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Ca) ≥ 1.0 apfu is now used to define the calcic and sodic-calcic groups, and the calcic group has $^{B}Na < 0.50$ apfu.

RESULTS

Two samples of asbestiform amphibole from Libby were analyzed following the experimental procedures detailed in Verkouteren and Wylie (2000). Chemical compositions, cell parameters, and optical properties are given in Table 1. One of the samples had been in our collection for several years; the other was obtained recently. Sample 1 is relatively pure, loose fiber and sample 2 was collected from the mine dump and is composed primarily of asbestiform amphibole. In both cases, the fibers are light green and asbestiform. The wt% Fe (analyzed as FeO) was converted to formula proportions of cations assuming first all Fe^{2+} and then all Fe^{3+}. It seems likely that at least some portion of the iron is trivalent, as more than 8.0 apfu Si cannot be accommodated in the tetrahedral sites. Hence, the "true" formulae must lie somewhere between these two extremes.

Because $^{B}(Na + Ca) \geq 1.0$ apfu this amphibole is either a sodic-calcic or a calcic amphibole. If all the iron is Fe^{2+}, then $^{B}Na = 0.63$ or 0.61 apfu, and according to Miyashiro (1957) the amphibole should be called soda tremolite. According to Deer et al. (1963), it would probably be richterite. According to Leake (1978), it would be sub-calcic actinolite, and according to Deer et al. (1997) and Leake et al. (1997), it would be winchite. If all the iron is Fe^{3+}, ^{B}Na increases to 0.75 or 0.67 apfu and according to Leake (1978), Deer et al. (1997) and Leake et al. (1997), it would be winchite. The chemical composition of the Libby amphibole as reported by Larson (1942) corresponds to a current classification of richterite.

Deer et al. (1963) chose $NaCa_{1.5}$ as the dividing line between richterite and tremolite because it was consistent with a "relatively sudden" change in optical properties, specifically a decrease in birefringence, stronger pleochroism, lower indices of refraction, and smaller optic axial angle. The refractive indices given in Table 1 are different for the two samples, consistent with the change in 1 – Mg/(Mg + Fe + Mn). Comparison of the optical properties to those of the actinolite series (Verkouteren and Wylie 2000) indicates that, for both samples, n_α is high and n_γ is low, although not statistically outside the population of actinolite samples. The birefringence given in Table 1 is much lower than any actinolite sample in Verkouteren and Wylie (2000) and is a clear outlier; this is also true for the birefringence of the Libby amphibole given by Larson (1942). No difference exists between the optic axial angle given in Table 1 and the actinolite series; however, the optic axial angle for the Libby amphibole reported by Larson (1942) is distinctly smaller than that of corresponding actinolites.

Comparison of the lattice parameters to those of the actinolite series (Verkouteren and Wylie 2000) indicates that, for both samples, *a* and *c* are at or within the 95% prediction limits for actinolite, but *b* is outside the lower 95% prediction limit by more than 0.025 Å. The values of *a* are high given a Ca value of 1.3 apfu; these samples would fall into an anomalous region in the actinolite series where *a* and Ca are positively correlated (Verkouteren and Wylie 2000), and we would predict an *a* of 9.83 Å or lower. The values of β for the Libby samples are consistent with the actinolite series and the positive correlation of β and Ca. The potassian winchite-asbestos described by Wylie and Huggins (1980) has a larger *a* dimension, a smaller *b* dimension, and the same *c* dimension when compared with the actinolite series. Similarly, the three non-Ti bearing richterites in Oberti et al. (1992) have larger *a* dimensions, smaller *b* dimensions, and the same *c* dimensions when compared to the actinolite series.

TABLE 1. Chemical composition, optical properties, and cell parameters of 2 samples of winchite-asbestos, Libby, Montana. (1σ errors in parentheses)

Oxide	wt%			apfu§, all Fe^{2+}		apfu§, all Fe^{3+}	
	Sample 1*	Sample 2†		Sample 1	Sample 2	Sample 1	Sample 2
SiO_2	56.6(4)	56.1(2)	Si	8.04	8.01	7.92	7.92
TiO_2	n.d.	n.d.	^{IV}Al	–	–	0.08	0.07
Cr_2O_3	n.d.	n.d.	ΣT	8.04	8.01	8.00	8.00
Al_2O_3	0.5(1)	0.4(2)	^{VI}Al	0.09	0.07	0.01	–
FeO	6.0(6)	4.2(4)	Mg	4.28	4.45	4.21	4.41
MnO	0.1(0)	0.3(3)	Fe	0.72	0.50	0.71	0.50
MgO	20.2(5)	21.0(4)	Mn	0.01	0.03	0.01	0.03
CaO	8.3(10)	8.8(2)	ΣC	5.10	5.05	4.94	4.94
Na_2O	3.2(8)	3.4(2)	ex. C	0.10	0.05	–	–
K_2O	0.7(1)	0.8(2)	Ca	1.27	1.34	1.25	1.33
Total	95.6	95.0	^{B}Na	0.63	0.61	0.75	0.67
			ΣB	2.00	2.00	2.00	2.00
			^{A}Na	0.25	0.33	0.12	0.26
			^{A}K	0.13	0.15	0.13	0.15
			ΣA	0.38	0.48	0.25	0.41
			1-Mg/(Mg+Fe+Mn)	0.15	0.11		

Notes: optical properties: Sample 1: $n_\alpha = 1.621(1)$, $n_\beta = 1.631(1)$, $n_\gamma = 1.637(1)$, $c \wedge Z = 15.8(0.5)°$. Sample 2: $n_\alpha = 1.618(1)$, $n_\beta = 1.628(1)$, $n_\gamma = 1.634(1)$, $c \wedge Z = 15.8(0.5)°$ $\delta‡ = 0.016$, $2V_Z‡ = 104.9$.

Cell dimensions: Sample 1: *a* = 9.855(1) Å, *b* = 18.032(1) Å, *c* = 5.288(3) Å, β = 104.54(2)°. Sample 2: *a* = 9.861(2) Å, *b* = 18.003(5) Å, *c* = 5.276(6) Å, β = 104.37(4)°.

* Average of 6 analyses.

† Average of 3 analyses.

§ Calculated on the basis of 23 O atoms.

‡ Calculated from the measured refractive indices.

DISCUSSION

The composition of the Libby asbestiform amphibole as given in Table 1 is consistent with an identification of winchite-asbestos, based on Leake et al. (1997). The samples can be identified as winchites despite the uncertainty in site occupancies resulting from the unknown oxidation state of Fe. The *b* lattice dimension and the birefringence are consistent with what is known about winchite (and richterite) and are distinct from actinolite. Ross et al. (1993) report that both tremolite and richterite asbestos fibers were found in a specimen of Libby vermiculite. Our two samples were collected approximately ten years apart, and probably from different areas in the mine, and both are winchites, although our sample 2 is close to richterite in composition ($\Sigma A = 0.48$ to 0.41 apfu). Given the fact that the Libby amphibole reported by Larson (1942) is a richterite, it is possible that the amphibole composition ranges from winchite to richterite, and possibly to actinolite, throughout the vermiculite deposit. Asbestiform winchite and richterite are also known from other localities, where they are similarly associated with the alteration of alkali igneous rocks (Wylie and Huggins 1980; Deer et al. 1997).

It is unfortunate that a regulatory decision could hinge on such details as the amount of ^{B}Na and the choice of classification scheme. While the distinctions among amphiboles are important from a scientific standpoint, they do not add significantly to the regulatory terminology unless they are correlated with risk assessment. There are data that show differences in disease potential among different minerals with similar morphology, such as between talc and tremolite (Guthrie and Mossman 1993), but it is clear that the asbestiform winchite in Libby, Montana poses a health threat (106th Congress 2000). From an analytical standpoint, the identification of the specific asbestiform mineral is necessary for complete characterization of the asbestos component in any sample. The regulatory requirement to identify the mineral can be addressed by providing reference values for known asbestiform amphiboles, which was, in part, the impetus behind the study described in Verkouteren and Wylie (2000) and the current note. It would be reasonable for the regulations to be revised to provide a broader description of asbestiform amphiboles to avoid similar hair-splitting problems in the future.

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