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Isotopic Evidence for Biological Origin of Shungite, Generation of Petroleum and ^{13}C Depleted Nature of Initial Biomass at 2.0 Ga ago

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Introduction

Intensive accumulation of organic material (OM) in Precambrian rocks is rare. The most remarkable occurrence of strongly matured OM is the Karelian shungite (SH) deposit from Karelia, NW Russia. The deposit may be classified as a giant carbon accumulation, one of the largest in the Palaeoproterozoic (C > 25 $x10^{10}$ tonnes, Galdobina, 1993). The explanation for such an exceptionally intensive accumulation of OM still remains rather controversial (Buseck et al., 1997; Melezhik et al., 1999a). This enigma is amplified by the fact that the accumulation of SH and synchronous pyrobitumens in N. America and Greenland postdate the Palaeoproterozoic positive excursion of $^{13}C/^{12}C$ in sedimentary carbonates, one of the largest in Earth history (Melezhik et al., 1999b).

Definitions

The term *'SH'* was introduced for a solid organic substance preserved in metamorphosed sedimentary rocks near the village of Shunga. SH was originally described as an amorphous, non-graphitised, organic substance consisting of C. SH occurs in both dispersed and concentrated forms in stratified rocks as well as in fracture veins. A series of optical, geochemical and genetic classifications of SH and SH-bearing rocks have been reviewed by Filippov (2000). The term *layer-SH* is used to describe shungite which occurs as either a layer or a lens which appear to be conformable with the bedding of the host rocks. The term *vein-SH* is used here to describe SH which occurs in veins cutting the host-rock bedding. The terms *layer-* and *vein-SH* substitute for the previously used *"shungite rocks I"*. Terms *SH rocks II* (35-75% C_{org}), *III* (20-35), *IV* (10-20) and *V* (<10) are used in this study in accordance with existing classifications.

Geological position

SH occurs in formations composing a ca. 2.0 Ga Palaeoproterozoic sequence which lies stratigraphically above the $\delta^{13}C$-high (+5 to +16‰) dolostones. These formations have a total thickness of 2500 m. The principal lithologies are greywacke and arkosic sandstones, basaltic tuffs and lavas, subordinate cherts and dolostones. The rocks were deformed and metamorphosed at greenschist facies (300-350ºC) during the Svecofennian orogeny.

The major volume of C-rich SH rocks occurs in the Zaonezhskaya Formation (ZF) where they form nine, 5-120 m-thick beds. The thickest known development of layer- and vein-SH has been found in the uppermost SH rock bed of the ZF, near the village of Shunga, where ten layer-SH seams, separated by dolostones, have been reported within a 4.5 m-thick succession.

Depositional environment, post-depositional alteration and petroleum generation

The ZF sediments were deposited in brackish water in a non-euxinic, lagoonal environment. The high C/S ratio (8-1000) with a zero intercept on the C-S cross-plot indicates that deposition occurred in S-deficient water. Intensive synchronous volcanism might have contributed to both the enhanced delivery of nutrients and an elevated sedimentation rate, and eventually to the high degree of preservation of OM.

The integrated data suggest that the OM has a biogenic origin, most likely algal or bacterial. The OM suffered complex catagenetic and metamorphic alteration which is reflected in: (i) four-modal distribution of C_{org} content (with maxima at 5, 30, 65 and 95%); (ii) highly variable $\delta^{13}C_{org}$ (-45‰ to -17‰); (iii) bimodal distribution of $\delta^{13}C_{org}$ (with maxima at -28 and -39‰); and (iv) low H/C ratios (0.005-0.2). Abundant diagenetic carbonates associated with SH ($\delta^{13}C_{carb}$ = -5 to -26‰) and the presence of pyrite ($\delta^{34}S$ -22 to +31‰) reflect a substantial loss of OM via bacterial reduction of sulphate during diagenesis. The SH rocks are characterised by a further substantial loss (> 50%) of biologically produced OM in the course of thermal maturation and by a depletion in ^{13}C (> 10‰). The isotopic composition of carbonate concretions does not indicate the involvement of fermentative diagenesis. Conservative estimates give a $\delta^{13}C_{org}$ of -34‰ as the best value of the initial biomass (Melezhik et al., 1999a). Vein- and layer-SH containing more than 80% C_{org} is considered to represent allochthonous, migrated bitumen (originally petroleum). The semi-lustrous and semimat layer-SH and SH rocks containing 55-75% C_{org} represent oil shales with both migrated bitumen and autochthonous kerogen residues.

It is likely that there is a relationship between the prior accumulation of ^{13}C-rich sedimentary carbonate and the ^{13}C-depleted nature of the initial biomass.

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