# DIAMONDS AND THEIR INCLUSIONS:

# ARE THE CRITERIA FOR SYNGENESIS VALID?

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## INTRODUCTION

The general axiom that mineral inclusions in diamonds (DIs) are syngenetic with their host diamonds (Ds) is herein seriously questioned. Virtually all papers on DIs start with the major premise that the Ds and their DIs are *syngenetic* -- i.e., formed simultaneously under the same P-T-X conditions and from the same genesis. However, supporting evidence for this assumption is rarely presented. Even if there are valid evidences for syngenesis, such criteria are seldom checked. We have already presented evidence (Taylor et al., 2003) negating the possible syngenesis for some types of DIs, considered by most to be syngenetic. In this paper, we will address the development of thought that led to the present-day criteria for establishing the syngenesis of diamond inclusions. These criteria will be addressed as to their validity, since the syn-chronous, syn-genetic nature of diamond inclusions is at the very foundation of inclusion studies. Inherent in many studies is *the requirement for co-crystallization of diamonds with their mineral inclusions*.

Mineral inclusions in diamonds were originally divided into two groups: *syngenetic* and *epigenetic.* Epigenetic phases (literally, of later origin) are secondary minerals, typically associated with crustal processes. These minerals are atypical to the primary minerals in mantle xenoliths. All other DIs was considered as *syngenetic*. Inherent in this group is the requirement of formation of the inclusion and its host diamond at exactly the same instant and by the same genetic process. Later, Henry Meyer (1987) suggested the addition of a third class of DIs, *protogenetic*, for inclusions that had formed *before* the encapsulation by the host diamond. However, *the distinction between the syngenetic and protogenetic DIs is difficult*. This is at the heart of this paper. We contend that the thesis, presented by Pearson and Shirey (1999), that the diamond age-dating studies performed to date have been conducted on syngenetic inclusions needs serious reconsideration.

## SYNGENETIC CRITERIA

Several *reviews* of the criteria for 'syngenesis' of diamonds and their mineral inclusions have been written over the years, starting with Sobolev (1977), followed by Harris (1979), and most recently by Pearson and Shirey (1999). Much of the following discussion has been culled from these publications.

Concerning syngenetic DIs, Harris (1968, 1979) proposed, based on optical criteria, that diamond has imposed its growth pattern on the silicate inclusions. Reportedly, this occurred during simultaneous growth of both host and inclusion, and the inclusion nucleated on the octahedral or cubic plane of the diamond substrate (e.g., Harris and Gurney, 1979). For example, cubo-octahedral morphology is a common feature of olivine and enstatite DIs that have orthorhombic crystal structures. In fact, olivine with its own orthorhombic morphology is rare as a DI. Even cubic garnet morphologies, that are distinctly different from those of diamond, are seldom present. Harris (1968) stated that specific alignments of the crystallographic orientations between inclusions and their host diamonds were not common phenomenon, yet it had been observed in several instances. Sobolev et al., (1972) thought that the octrahedral morphology of the pyrope was imposed by the diamond as a result of the growth of garnet in etched pits [negative crystals] on diamond {111} growth surfaces. However, Harris (1979) felt that this encapsulation process did not adequately explain the complete diamond morphology of the garnet. It is thought that the inclusion morphology developed as a result of the greater 'form energy' of diamond, thereby imposing its morphology upon the inclusion, *during mutual growth.* The morphology of the DI was regarded as indicative of an approach to textural equilibrium at the time of encapsulation. *The most common criterion for syngenesis is the imposition of the morphology of the host diamond on the inclusion* (Harris, 1968; Meyer, 1985, 1987). Pearson and Shirey (1999) stated that the synchronous growth hypothesis seems the simplest and best explanation for the production of oriented, faceted inclusions in diamonds. Indeed, where DI morphologies are noted, such as by Harte et al. (1999), it is stated the imposition of dominant host crystallographic faces upon inclusions is common with diamonds. It is thought to be not only a reflection of the crystalloblastic force of diamond faces, but conclusively demonstrates that the *growth of both diamond and inclusion occurred simultaneously* (Harris and Gurney, 1979).

X-ray diffraction studies of diamonds and their mineral inclusions, dating back to the 1950s, demonstrated that, in many cases, the principal surfaces of the inclusions are closely related to {100} and/or {111} planes of the dia-

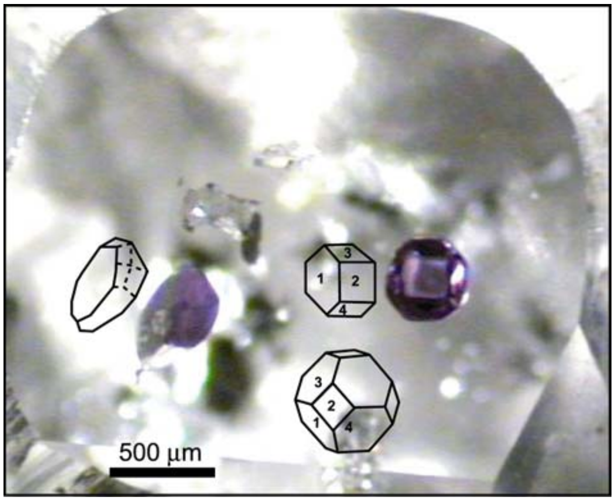


Figure 1. A P-type diamond with a "window" cut parallel to (100) showing the cubo-octrahedral morphology of the harzburgitic garnet inclusions.

diamonds. Mitchell and Giardini (1953) examined olivine inclusions in diamonds and found that the olivines had a specific crystallographic orientation. That is, the (101) and (010) planes of the olivine are aligned approximately parallel to the (10$\bar{1}$) and (111) of the diamond. This similarity between crystallographic orientations of the inclusion and its host demonstrates an epitactic relationship, and is well reviewed by Harris (1979). Pearson and Shirey (1999) summarily stated that *the epitaxial relationship between inclusions and host diamond, as revealed by X-ray studies, is considered as 'powerful' evidence in favor of the syngenetic origin of many inclusions in diamonds.* In summary of their review of the various criteria for recognition of syngenetic DIs, Pearson and Shirey (1999) stated that substantial weight of evidence is in favor of the syngenetic growth of most DIs.

This leaves open the evaluation criteria for the recognition of protogenetic inclusions; however, little has been published on this subject. Meyer (1987) felt that the inclusions that existed prior to their encapsulation in the diamonds typically should have either irregular morphologies or euhedral morphologies that are entirely consistent with the crystal structure of the mineral. Thus, *a syngenetic garnet should have an octrahedral diamond morphology, whereas a protogenic garnet inclusion should have a dodecahedral form.* An octrahedral habit has never been observed for 'normal' garnet.

The above discussion has emphasized that the most commonly used criteria for the identification of syngenetic DIs [e.g., Harris (1968); Pearson and Shirey (1999)] is the imposition of the morphology of the host diamond on the DI, with the possible epitactic relationship of the two. The remaining criteria for syngenesis are all concerned with the chemistry of the DIs relative to each other and to their host rocks (Sobolev, 1977; Bulanova, 1995). In summation of these criteria using mineral chemistry, they are largely based on the hypotheses that the DIs should resemble the other minerals that are of mantle origin, and that if multiple inclusions do occur in a single diamond, their compositions should indicate progressive crystallization sequence from the diamond core to the rim, a logic that has been repeatedly contradicted by several of our studies (e.g., Taylor et al., 1998, 2000).

One additional criterion has been put forth by Bulanova (1995). Based upon detailed examination with CL of polished surfaces of diamonds, Bulanova felt that diamond growth zones are always interrupted (i.e., cut) by the diamond/inclusion contact; supposedly, there are no cases of the growth zones wrapping around the inclusion, which would signify a protogenetic inclusion. Such interrupted zonations are supposedly evidence for synchronous growth of diamonds and their syngenetic inclusions. Our experience, albeit limited, has shown that most DIs are surrounded by CL "dead zones." Such interrupted zones may exist, but we do not feel that this is an unambiguous criterion for syngenesis.

## HARZBURGITIC DIAMOND INCLUSIONS

The majority of all garnets found as DIs are of the harzburgitic variety (see Fig. 1). It is based upon the morphologies of harzburgitic DIs, as well as their chemistry, that we address this paradigm of syngenesis.

As part of our investigation of DIs in Yakutian diamonds, we routinely use the *in-situ* technique of examination of the

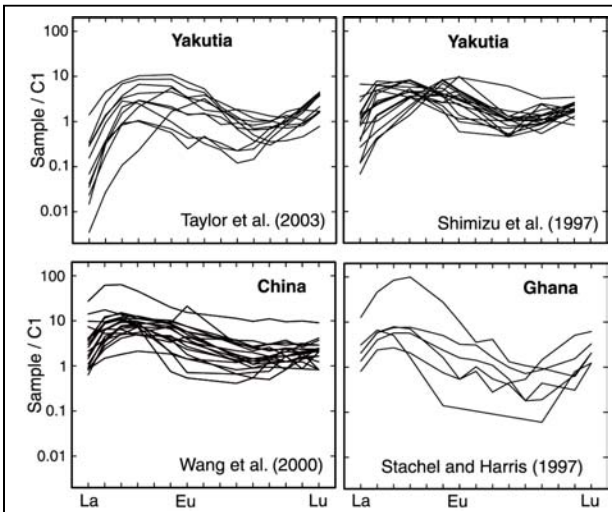


Figure 2. Representative REE patterns of harzburgitic garnet DIs from worldwide locales. Note the typical sinusoidal REE patterns.

inclusions inside of and on polished surfaces of the diamonds (Bulanova, 1995), with substantial photographic documentation. These diamonds are examined in detail by CL on an EMP for possible cracks, possibly indicative of "open-system behavior." Figure 1 shows a P-type diamond from Komsomolskaya (Sobolev et al., this volume) with a "window" cut into it for observation, and in preparation for polishing to expose the inclusions simultaneously on one plane. This cut is approximately parallel to the (100) plane of the diamond. Note the purple color typical of harzburgitic garnets. Also note the cubo-octrahedral morphologies of the two garnets, which have obviously been imposed by the diamond. ***This morphological control of the habit by the diamond is evidence that these DIs are definitely "syngenetic"*** (Harris, 1968; Sobolev, 1977; Harris, 1979; Meyer, 1987; Pearson and Shirey, 1999). But, are they really?

The most striking chemical feature of harzburgitic garnets, both in rocks and as DIs, is the sinusoidal REE pattern observed worldwide. Figure 2 shows typical REE patterns for harzburgitic garnets from several significant studies that cover various locales worldwide. Figure 3 illustrates further that such patterns are not restricted to particular garnet compositions, instead are present across the entire harzburgitic range. What process in the formation of the harzburgites has produced this unusual, albeit typical, sinusoidal REE pattern? Each change of slope may be conveying a genetic message.

The complicated genesis and evolution of the harzburgitic garnet DIs are difficult to explain. Although the proposed schemes for the formation of these REE patterns vary somewhat, all investigators agree that the sinusoidal-REE-patterned garnets have experienced complex evolutionary histories (e.g., see ref in Taylor et al., 2003). Figure 4 is a cartoon, modified from Taylor et al. (2003), that segments the overall harzburgitic REE pattern into three portions, thereby permitting an evolutionary reconstruction of the complete pattern. An ancient melting event resulted in the initial depletion signature of the garnet [A in Fig. 4], followed by mantle metasomatism [B of Fig. 6], culminating in a possible late-stage depletion episode [C of Fig. 6].

It has been suggested, based on chemical and radiometric data of both diamond inclusions and mantle xenoliths from

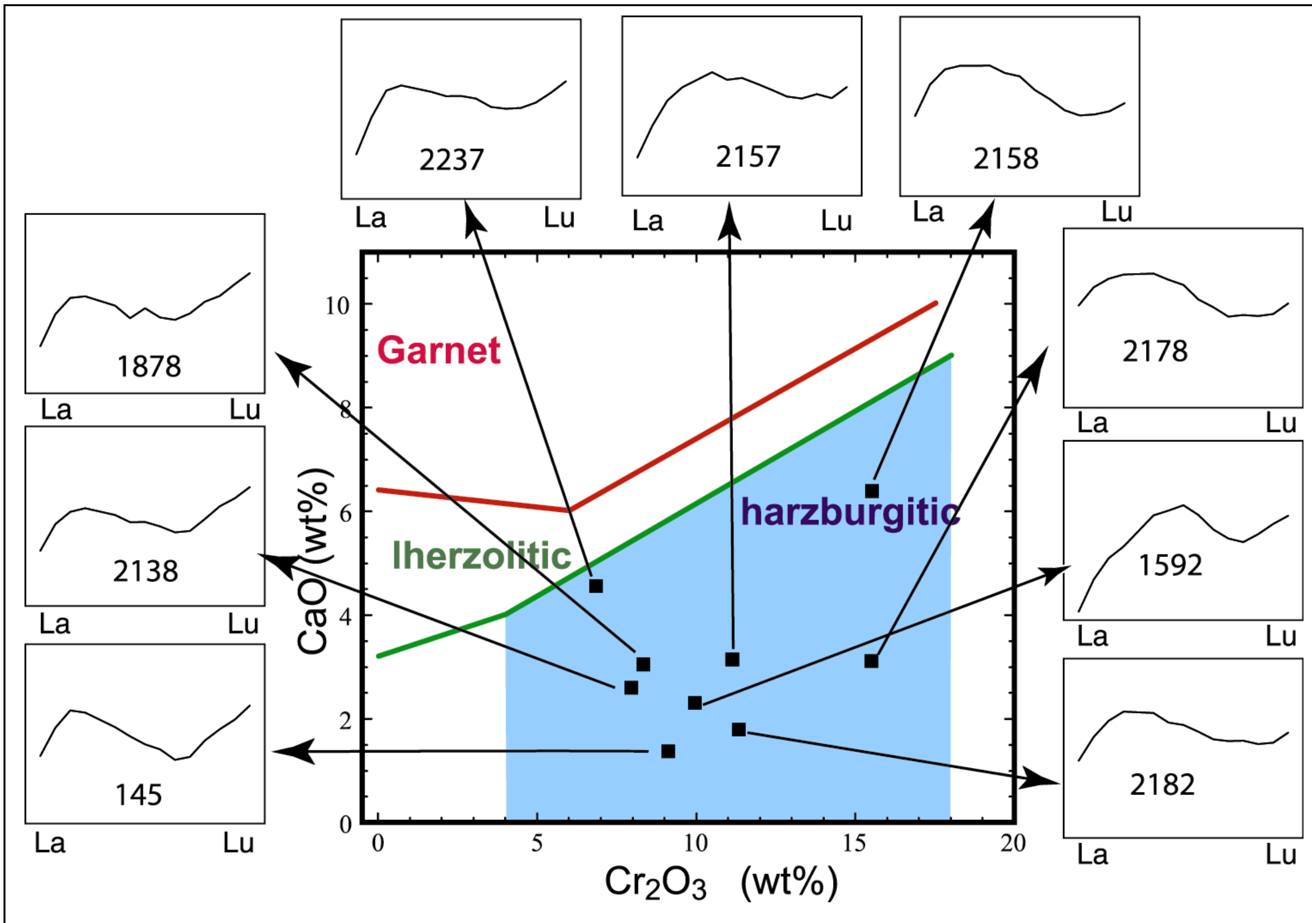


Figure 3. Sinusoidal REE patterns for harzburgitic garnet DIs illustrating that garnet major-element composition has no affect the REE patterns significantly (data from Taylor et al., 2003).

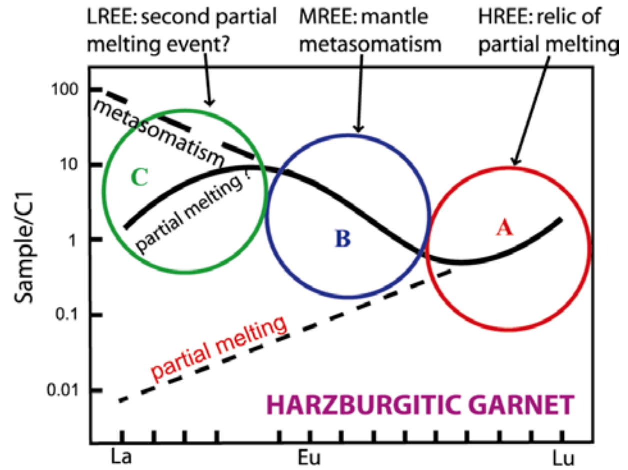


Figure 4. Schematic diagram of the sinusoidal REE pattern, typical in harzburgitic-garnet diamond inclusions. The REE pattern suggests multiple processes that begin with a partial melting event (A), followed by metasomatic enrichment (B), and with a possible late-stage episode of partial melting (C). Modified after Taylor et al. (2003).

cratonic lithospheres, that this initial depletion episode corresponds with komatiite magmatism as a major crust-forming event in the Archean (Boyd and Gurney, 1986; Boyd et al., 1997, Pearson et al., 1999). That it was metasomatism [B in Fig. 6] that modified the initially depleted REE patterns in harzburgitic, and certain lherzolitic, garnets is agreed upon by all. However, the details of the process (es) to form the MREE [B] and LREE [C in Fig. 5] portions of the sinusoidal shapes in response to metasomatic enrichment are only qualitatively understood (reviewed in Taylor et al., 2003). Such typical positive slopes of REEs with negative-sloped MREE patterns are usually interpreted as the result of a depletion episode, such as one that accompanies a partial-melting event. If this is the case here, it can be said that the event that last occurred before encapsulation of the garnets in the diamonds was this partial melting. This would appear to remove the earlier metasomatism from the diamond formation, since the diamond formed only afterward.

An important implication from the complex petrogenetic history of the harzburgitic garnet DIs is the requirement that the garnets underwent extensive processing *prior* to diamond encapsulation, proof of the non-syngenetic formation of diamond and their DIs. Granted, a scenario can be imagined whereby the garnets may been been metasomatized by the same fluids that precipitated the diamonds. However, the entire chemistry of the garnets is not modified at this time, making for a 'new' garnet, especially since they retain a signature of the initial early partial-melting event. This is evidence for the non-syngenetic (i.e., protogenetic) formation of diamonds and their harzburgitic DIs.

## CONUNDRUM

The widely accepted criterion of diamond-imposed morphology indicates "syngenesis" for the harzburgitic garnets. Yet, the sinusoidal REE pattern definitely indicate that these harzburgitic garnets are all "protogenetic" – i.e., formed before their encapsulation by the diamonds. The bottom line is that the morphologic/crystallographic criteria have been established based mainly on sound scientific logic, but have not been demonstrated to be unambiguous. A new paradigm states that ***most, if not all, harzburgitic garnet inclusions, worldwide, are not syngenetic with their diamond hosts.***

## DATING DIAMONDS

The chemistry of diamond, consisting of 99+% carbon, has not permitted, to date, the absolute determination of its age. Instead, diamonds have been dated based upon the Rb-Sr and Sm-Nd radiogenic systematics of garnet and pyroxene inclusions (e.g., Richardson et al., 1984, 1997). Effectively, these inclusions are recovered from literally hundreds of diamonds from a locale, combined to give sufficient mass for analyses, and subjected to standard radiometric treatment. Inherent in this approach is the major assumption that the DIs are: 1) synchronous with the diamond formation; 2) all similar in chemical composition; and 3) all formed at about the same time.

Inclusion compositions can be diverse within the same diamond (e.g., Sobolev et al., 1998; Taylor et al., 2000) and between different diamonds from the same xenolith (Keller et al., 1999; Taylor et al., 2000). Initially, this would seem to conflict with the isotopic dating studies of inclusions from numerous diamonds that have yielded unique, consistent model and even 'isochron' ages. This apparent enigma may be explained if diamonds commonly form by metasomatic processes, such that changes in mantle composition during diamond growth, now seen as variable inclusion compositions, occurred over periods of time so short that they are within the analytical error of isotopic dating techniques.

## CONCLUDING REMARKS

It is now generally agreed that diamonds do not crystallize from a silicate melt, rather they form from a metasomatic C-O-H-N-S or carbonatitic fluid. Based upon studies of peridotitic diamond inclusions, it has been suggested that peridotitic diamonds possibly grew under subsolidus conditions (Boyd and Finnerty, 1980; Hervig et al., 1980), rather than in igneous melts. Why, then, should we suppose that the igneous minerals in the peridotites formed synchronous and syngenetically with their host diamonds? Indeed, the compositions of the harzburgitic garnet DIs are indicative of formation before their encapsulation in diamond -- a protogenetic origin.

***Diamonds are time capsules, but just as the coins in a person's pocket do not relate to a his/her age, so the inclusions in the diamonds may not tell us the age of the diamond.*** The general axiom in the diamond community that DIs are syngenetic with their diamonds, therefore, is seriously questioned. The genetic relationship between diamonds and their mineral inclusions is pivotal to diamond research, especially for dating of diamonds.

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