GEOLOGY



Manuscript received 24 May 2022 Revised manuscript received 15 July 2022 Manuscript accepted 4 August 2022

Published online 14 October 2022

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Novel age constraints for the onset of the Steptoean Positive Isotopic Carbon Excursion (SPICE) and the late Cambrian time scale using high-precision U-Pb detrital zircon ages

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ABSTRACT

The Steptoean Positive Isotopic Carbon Excursion (SPICE) is a prominent +4–5‰ shift in the Cambrian $\delta^{13}C$ record used for global chronostratigraphic correlation. The onset of this excursion is traditionally placed at the base of the Pterocephaliid trilobite biomere (base of the Furongian Series). Recent studies have documented local controls on the expression of the SPICE and emphasize the need for chronostratigraphic standards for these complex biogeochemical signals. We build upon prior work in western Laurentia by integrating $\delta^{13}C$ and biostratigraphy with high-precision isotope dilution U-Pb detrital zircon maximum depositional ages that are coincident with the onset, peak, and falling limb of the SPICE. Our study provides the first useful numerical age constraint for the onset of the SPICE and the Laurentian trilobite biozones and requires revision of the late Cambrian geologic time scale boundaries by several million years.

INTRODUCTION

The marine carbonate δ^{13} C record of the Cambrian is characterized by protracted intervals (>1 m.y.) of relatively stable δ^{13} C values punctuated by short-lived (1 m.y.), likely global, δ^{13} C excursion events (Zhu et al., 2006). This Cambrian δ^{13} C record has emerged as a powerful correlation tool, especially where key age-diagnostic trilobite markers are missing (Saltzman et al., 2000). However, all Cambrian time-scale Global Stratotype Sections and Points (GSSPs) are defined by cosmopolitan agnostids, which leaves the endemic trilobitebearing shelfal facies difficult to correlate across discrete biogeographic realms without chemostratigraphic features. The later Miaolingian and Furongian epochs of the Cambrian are particularly poorly constrained by radioisotopic ages, such that global interpretation of Cambrian Earth-system dynamics relies upon a time scale constructed through weak assumptions of proportional scaling of trilobite biozone duration (Peng et al., 2020).

The Steptoean Positive Isotopic Carbon Excursion (SPICE) is a globally preserved geochemical event characterized by a prominent +4-5% and +2-3% shift in the $8^{13}C_{carb}$ and $8^{13}C_{org}$ records, respectively. First described from western Laurentia, the SPICE is coincident with the Marjumiid-Pterocephaliid (M-P) biomere boundary and biotic crisis (Brasier, 1993; Saltzman et al., 1998). This biomere boundary corresponds with the base of the *Aphelaspis* Biozone, which defines the base of the Laurentian Steptoean Stage (Ludvigsen and Westrop, 1985). Because the base of the *Aphelaspis* Biozone contains the cosmopolitan trilobite *Glyptagnostus reticulatus*, the SPICE is correlative to the base of the international Furongian Series and Paibian Stage (Palmer, 1962; Peng et al., 2020).

The SPICE is commonly ascribed to an increased burial rate of organic carbon, anoxic or euxinic conditions, and/or increased continental weathering associated with sea-level lowstand at the Sauk II–III transition (Saltzman et al., 1998, 2000; Gill et al., 2011; Dahl et al., 2014; Rooney et al., 2022). The SPICE is coincident with notable bioevents, such as the restructuring of polymerid trilobites globally, with the extinction of marjumiid and radiation of pterocephaliid groups in Laurentia, and the

replacement of the dameseliid group with leiostegiid and ceratopygid trilobites in Gondwana (Palmer, 1965a; Peng et al., 2004). Turnover in agnostids is also recognized by the transition from *G. stolidus* to *G. reticulatus* fauna. Shoaling of anoxic deep water onto the continental shelves has been proposed as the causal link between these events (Gill et al., 2011).

Oceanic mixing models suggest that the ocean is well-circulated, even in times of poor ventilation, on timeframes of 103 yr. Excursions in short-residence time (105 yr) geochemical tracers such as carbon are thus fairly rapid and likely a more precise correlation tool than biostratigraphy, and it has been suggested that more GSSPs should be defined by geochemical events (Kiehl and Shields, 2005; Cramer and Jarvis, 2020; Gradstein, 2020). However, recent work has challenged the reliability of the SPICE as a global correlation tool specifically because the onset of the SPICE is not always clearly linked with the M-P biomere nor the G. stolidus-G. reticulatus boundary (Schiffbauer et al., 2017; Pulsipher et al., 2021; Ren et al., 2021). The expression of the SPICE signal may be more controlled by local conditions than was previously understood, and it may vary in stratigraphic thickness and magnitude as a function of paleogeography, water depth, facies, and unconformities (Wang et al., 2020; Pulsipher et al., 2021).

Concerns over the SPICE signal's chronostratigraphic fidelity are challenging to assess without well-constrained, integrated chemostratigraphic, biostratigraphic, and geochronologic data. Here, we present a robust, tripartite data set over the SPICE interval from western Laurentia and include direct radioisotopic constraints on its timing and duration. This

CITATION: Cothren, H.R., Farrell, T.P., Sundberg, F.A., Dehler, C.M., and Schmitz, M.D., 2022, Novel age constraints for the onset of the Steptoean Positive Isotopic Carbon Excursion (SPICE) and the late Cambrian time scale using high-precision U-Pb detrital zircon ages: Geology, v. 50, p. 1415–1420, https://doi.org/10.1130/G50434.1

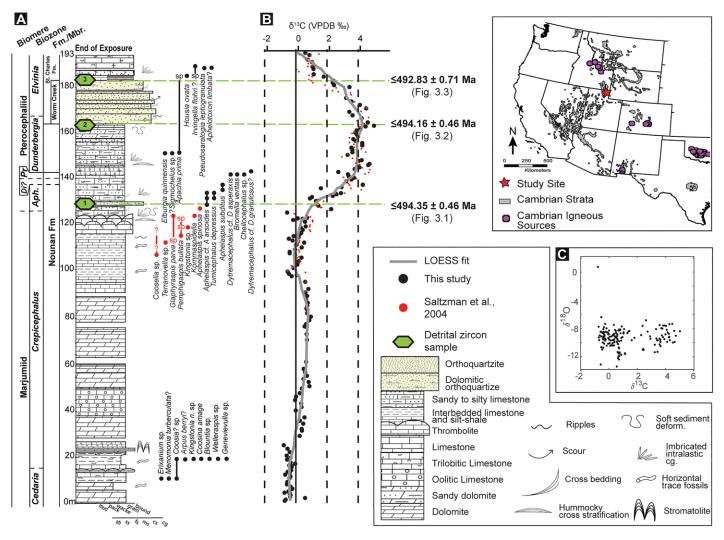


Figure 1. (A) Lithostratigraphy of the Smithfield Canyon Section (northern Utah, USA) with integrated biostratigraphy of biozones and biomeres. Solid lines indicate well-defined boundaries (e.g., first occurrence and last occurrence of biozone fauna are <1 m), and dashed lines indicate estimated biozone boundary position. (B) δ^{13} C stratigraphy; black points are from this study, and red points are from Saltzman et al. (2004) (see Table S2 [see footnote 1]). Locality map inset shows Cambrian strata exposed in the western United States in gray (polygons have a 5 km buffer); Smithfield Canyon section is marked with an orange star; and Cambrian-age igneous bodies are indicated by purple points. (C) δ^{13} C and δ^{18} O cross plot. *Aph—Aphelaspis*; *Di—Dicanthopyge*; *Pr—Prehousia*; Fm.—Formation; Mbr.—Member; VPBD—Vienna PeeDee Belemnite; cg.—conglomerate; deform.—deformation.

chronostratigraphic reference section of the SPICE is used to solidify biostratigraphic and chemostratigraphic correlations and refine the upper Cambrian geologic time scale.

GEOLOGIC SETTING

The SPICE in northern Utah, USA, was first reported by Saltzman et al. (2004) and is recorded in the upper Nounan Formation (Cedaria to Dunderbergia biozones) and overlying lower St. Charles Formation (Dunderbergia to Taenicephalus biozones), including the basal Worm Creek Member (Fig. 1A). Strata embrace the typical pentad trilobite zonation of the Pterocephaliid biomere in the central Great Basin: Aphelaspis, Dicanthopyge, Prehousia, Dunderbergia, and Elvinia (Palmer, 1984; Peng, 2020). The upper Nounan Formation straddles the M-P biomere and Marjuman-Steptoean Stage boundaries. These shelfal carbonate strata are charac-

terized by prominent microbialite boundstone, trilobite grainstone, imbricated intraformational conglomerate, and sandy-silty fossiliferous grainstone that are interrupted regionally by the 20–300-m-thick, orthoquartzite-dominant Worm Creek Member (Link et al., 2017). These orthoquartzites are interpreted to represent the influx of siliciclastic sediment at the Sauk II-III transition maximum regression. Strata were deposited during a hothouse climate at equatorial latitudes on the western (modern-day orientation) Laurentian margin (Bond et al., 1984, Müller et al., 2018, Goldberg et al., 2021).

METHODS

Field work was conducted in Smithfield Canyon near Smithfield, Utah (Fig. 1). Fossiliferous carbonates were targeted for integrated trilobite biostratigraphy and stable carbon- and oxygenisotope analyses every 0.5–1 m (see the Supplemental Material¹). $\delta^{13}C$ data were smoothed using the "loess" function in the MATLAB and Statistics Toolbox Release 2022a (The MathWorks, Inc., Natick, Massachusetts, USA). Biostratigraphic data at the M-P boundary from Saltzman et al. (2004) were integrated into our section using the thrombolite marker horizon (115–122 m). Quartz-rich units were targeted for U-Pb detrital zircon (DZ) geochronology. Zircon grains were separated from a calcareous sandstone bed in the upper Nounan Formation and from orthoquartzites at the base and top of the Worm Creek Member. Zircons were dated using laser ablation—

¹Supplemental Material. Biostratigraphic data (S1, S2, S7); chemostratigraphic data (S3); detailed methodology (S4, S8, S9); geochronological data (S5, S6); and location details (S10). Please visit https://doi.org/10.1130/GEOL.S.21183523 to access the supplemental material, and contact editing@geosociety.org with any questions.

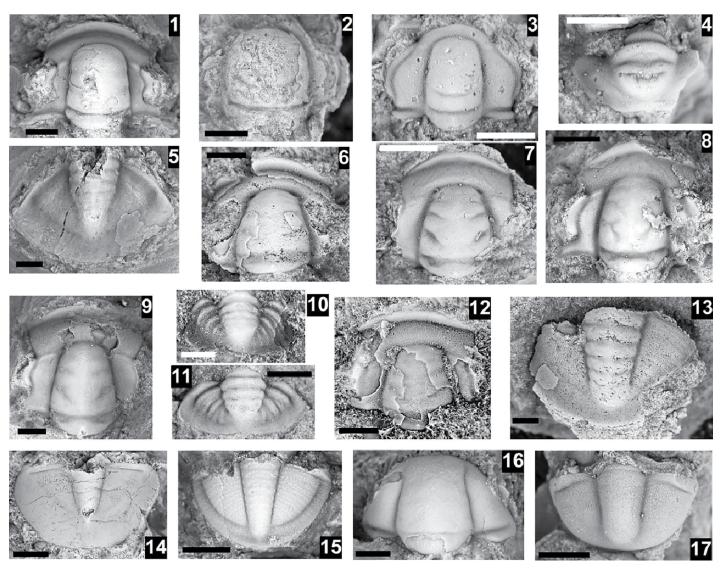


Figure 2. Examples of trilobites from the Crepicephalus (14–17), Aphelaspis (9 and 11), Prehousia (6, 10, 12, and 13), Dunderbergia (7 and 8), and Elvina (1–4) biozones of Smithfield Canyon (northern Utah, USA). Black scale bars = 2 mm, white scale bars = 1 mm. 1: Pseudosaratogia leptogranulata cranidium. 2–4: Irvingella flohri? cranidium; pygidium. 5: Housia ovata mostly pygidium. 6 and 10: Bromella veritas mostly exfoliated cranidium; pygidium. Elburgia quinnensis cranidium. 8: Apachia prima cranidium. 9 and 11: Aphelaspis subditus cranidium; pygidium. 12: Dytremacephalus cf. D. asperaxia cranidium. 13: Cheilocephalus sp. pygidium. 14: Coosia sp. pygidium. 15: Blountia sp. Pygidium. 16 and 17: Kingstonia sp., cranidium; pygidium. See Table S1 (see footnote 1) for taxonomic references and locality details.

inductively coupled plasma—mass spectrometry (LA-ICP-MS) to screen for the young Cambrian DZ population, and a randomly selected subset of this young population was analyzed via chemical abrasion—isotope dilution—thermal ionization mass spectrometry (CA-ID-TIMS) to obtain a precise and accurate maximum depositional age (MDA) from each sample (for detailed methods, see the Supplemental Material). We report our newly interpreted MDAs from the three samples as the mode and 95% confidence interval of the youngest mode from the probability density plot of all CA-ID-TIMS ²⁰⁶Pb/²³⁸U zircon dates.

RESULTS

Paleontology and Chemostratigraphy

The Smithfield Canyon section contains diagnostic fauna of the Cedaria, Crepicepha-

lus, Aphelaspis, Prehousia, Dunderbergia, and Elvinia biozones (Fig. 2; see the Supplemental Material and Table S1 therein). The observed biostratigraphy is in general agreement with that of Saltzman et al. (2004) (Fig. 1A), but with the addition of taxa attributable to the *Cedaria* and *Prehousia* biozones.

The SPICE occurs over \sim 70 m of the upper Nounan and the lower St. Charles formations (Figs. 1A and 1B). In the upper *Cedaria* and *Crepicephalus* biozones, δ^{13} C values range from -0.5% to +1%, which is generally consistent with background δ^{13} C_{carb} values for the Phanerozoic (Kump and Arthur, 1999). The excursion rises to peak values of +5% in the *Aphelaspis*, *Prehousia*, and *Dunderbergia* biozones and exhibits a plateau over 35 m (136–171 m) within the *Dunderbergia* and *Elvinia* biozones before

returning to near pre-SPICE background values within the *Elvinia* Biozone. δ^{18} O trends do not co-vary with δ^{13} C trends (Fig. 1C). We define the onset of the δ^{13} C excursion as the greatest rate of change in the LOESS fit of δ^{13} C values (*sensu* Cramer et al., 2010). From our analysis, the SPICE begins at 127.9 m, which is above the first occurrence (FO) of the *Aphelaspis* Biozone of Saltzman et al. (2004) at 127 m, but below the FO of *Aphelaspis* observed in our section at 129.5 m (Fig. 1A).

Geochronology

Sample 1 (TF21-SC2), from a 0.25-m-thick calcareous sandstone bed in the upper Nounan Formation at 129 m, yielded a CA-ID-TIMS MDA of 494.35 \pm 0.46 Ma (n = 6) determined from the mode and 95% confidence interval of the

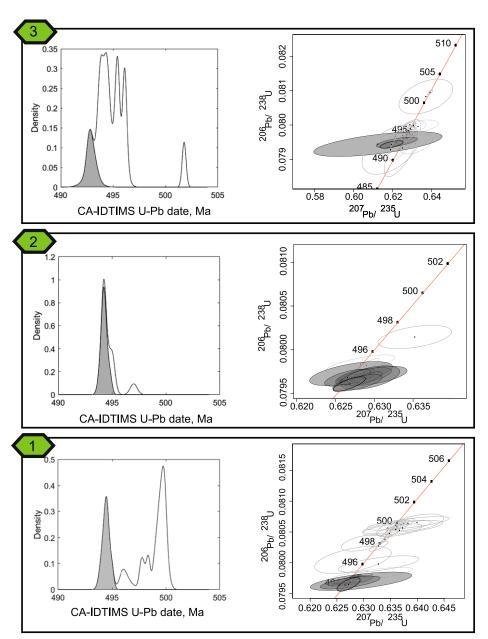


Figure 3. U-Pb chemical abrasion–isotope dilution–thermal ionization mass spectrometry (CA-ID-TIMS) geochronology of samples 1, 2, and 3. (Left) Probability density plots (PDPs) of CA-ID-TIMS detrital zircon data, (Right) U-Pb concordia plots. Shaded areas in PDPs indicate interpreted maximum depositional ages (MDAs) and their uncertainties as determined from the mode and 95% confidence interval of the youngest statistical peak of all CA-ID-TIMS dates. Shaded ellipses denote analysis used in interpreting the MDA. Stratigraphic position of samples is noted in Figure 1A.

youngest peak of CA-ID-TIMS DZ dates (Fig. 3). Sample 2 (TF21-SC1), from an orthoquartzite in the basal Worm Creek Member at 163 m, yielded a CA-ID-TIMS MDA of 494.16 \pm 0.46 Ma (n = 10; Fig. 3). Sample 3 (HC-20–77) is from an orthoquartzite at the top of the Worm Creek Member at 182 m and yielded a CA-ID-TIMS MDA of 492.84 \pm 0.73 Ma (n = 3; Fig. 3).

DISCUSSION Chronology of the SPICE

As a result of our U-Pb CA-ID-TIMS DZ MDA near the base of the *Aphelaspis* Biozone,

the base of the Furongian Epoch and initiation of the SPICE at Smithfield Canyon is \leq 494.4 \pm 0.5 Ma. This is the first numeric age control on the onset of the SPICE and is congruent with recent numerical depositional age constraint for the peak SPICE values in Baltica of 494.6 \pm 2.9 Ma (Rooney et al., 2022), suggesting that the SPICE may be globally synchronous at the 1 Ma timescale but substantially younger than estimated in previous time-scale calibrations. Additionally, the base of the Jiangshanian Stage is defined by the first appearance datum (FAD) of *Agnostotes orientalis* and co-

occurs with the trilobite *Irvingella angustilimbus* (Palmer, 1965b; Peng et al., 2012a). The FO of *Irvingella* observed in our section at 183 m, 1 m above DZ Sample 3, suggests the base of the Jiangshanian Stage is \leq 492.83 \pm 0.71 Ma, which again is substantially younger than current estimates of ca. 494.2 Ma (Peng et al., 2020).

When defined as at the greatest rate of change in the LOESS fit of $\delta^{13}C$ values and integrated with biostratigraphic data sets in Smithfield Canvon, the onset of the SPICE event and the M-P biomere turnover are collocated at a scale of less than 1 m, which we estimate as having been deposited in <100 k.y. We cannot falsify the hypothesis that the onset of the SPICE is correlated to the M-P biomere, especially if the M-P biomere boundary is regarded as an interval rather than a discrete time horizon. This interval can arise from sampling and preservational biases that are often underestimated in work exploring casual links between the SPICE and associated faunal turnover (Schiffbauer et al., 2017). If an excursion is correlated based on a biostratigraphic horizon, then its apparent timing is contingent on the stratigraphic position of the indicator taxon's sampled first or last occurrence datum, which must be nested within the true interval of the FAD to last appearance datum (LAD). This interval may also be exemplified by the crisis or crucial interval represented by the Coosella perplexa subzone (Palmer, 1984; Taylor, 2006). In this framework, it is probable that the true FAD of Aphelaspis precedes the true LAD of Crepicephalus, and it has been suggested that the biomeres themselves are "leaky" such that in basinal settings the "exotic" Aphelaspis taxa may appear within the Marjumiid biomere, prior to the crisis interval, and likewise "Lazarus" Crepicephalus taxa appear in the Pterocephaliid biomere following the crisis interval (Palmer, 1965a, Taylor, 2006). Future age modeling of the SPICE interval should refine the numerical resolution of the synchrony between the carbon isotope and biotic signals.

Biostratigraphic and Geologic Time Scale Implications

In recent global Geologic Time Scale compilations (Peng et al., 2012b, 2020), the conterminous bases of the international Furongian Series, Paibian Stage and Laurentian Steptoean Stage are correlated based on the FAD of the cosmopolitan G. reticulatus in Paibi, China, and the co-occurrence of Aphelaspis fauna and G. reticulatus in Laurentia (Palmer, 1962; Peng et al., 2004). The estimated age of the appearance of G. reticulatus was calculated assuming that agnostid trilobite biozones represent an equal length of time of ~ 1 m.y. (sensu Peng et al., 2020), such that six agnostid trilobite biozones from the closest radioisotopic age

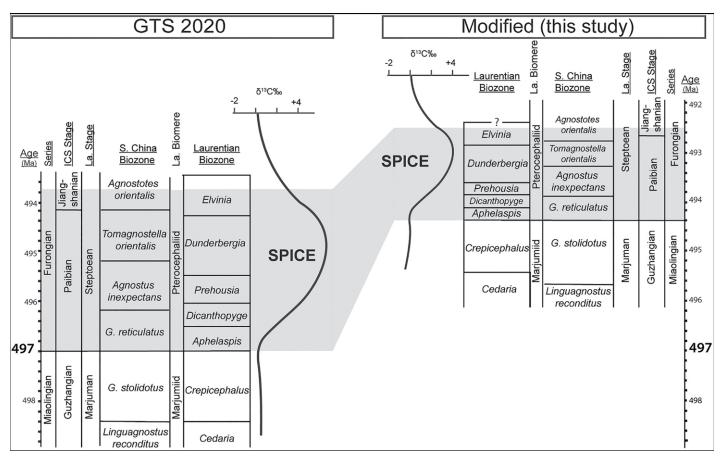


Figure 4. (Left) Chronostratigraphic diagram of the Smithfield Canyon Section (northern Utah, USA) (ca. 499–494.4 Ma), modified from Peng et al. (2020) and Karlstrom et al. (2020, their figure DR2); the Laurentian biostratigraphy (i.e., biomere and biozone); and the South China biostratigraphy. (Right) Modified chronostratigraphic diagram with hypothesized calibration of trilobite biozones, Steptoean Positive Isotopic Carbon Excursion (SPICE) chemostratigraphy, and associated Geologic Time Scale boundaries as a result of integrated chemical abrasion-isotope dilution—thermal ionization mass spectrometry (CA-ID-TIMS) detrital zircon maximum depositional ages. Note that the age (in Ma) at far left and right are at the same scale, such that the interval studied now spans ca. 496–492 Ma. GTS—Geologic Time Scale; ICS—International Commission on Stratigraphy; La.—Laurentia; S.—South; G.—Glyptagnostus.

of 502.2 ± 3.5 Ma (Encarnación et al., 1999) yielded an estimated age of 497 Ma (Fig. 4). Our new MDA within the *Aphelaspis* Biozone in Smithfield Canyon indicates that the Furongian Epoch, Paibian Age, and Steptoean Age must be \leq 494.4 \pm 0.5 Ma. Likewise, the base of the *Aphelaspis* Biozone and Pterocephaliid biomere must young accordingly. Similarly, the uppermost MDA of \leq 492.8 \pm 0.7 Ma within the *Elvinia* Biozone requires the base of the biozone to become younger than its previously estimated boundary (cf. 494.3 Ma; Peng et al., 2020). Biozones within the now compressed *Aphelaspis-Elvinia* interval must become younger and shorter in duration (Fig. 4).

We note that our new radioisotopic constraints on the late Cambrian numerical time scale are at odds with recent astrochronological interpretations of the Alum Shale Formation of Scandinavia (Zhao et al., 2022), likely due to incorrect tuning assumptions applied to the cyclostratigraphic signal. By contrast, the 3 Ma compression of the Furongian Epoch demanded by our work is fully consistent with a similar

conclusion for the base of the Miaolingian Epoch based on new MDAs in the Tapeats Sandstone in southwestern Laurentia (Karlstrom et al., 2020; Sundberg et al., 2020). Taken together, 3 m.y., or $\sim 13.5\%$ of the former duration of the traditional middle and upper Cambrian, must be reapportioned to the latter part of the early Cambrian, with implications for estimates of biodiversity and evolutionary rates based upon numerical time scales (Paterson et al., 2019).

ACKNOWLEDGMENTS

Funding for this work was provided in part by U.S. National Science Foundation (NSF) grant EAR-1337887 to C. Dehler and M. Schmitz; NSF grant EAR-1954583 to M. Schmitz; U.S. Geological Survey EDMAP award G20AC00235 to C. Dehler; Utah State University (USU) J. Stewart Williams Award to H. Cothren; and Boise State University (BSU) W.L. Burnham Research Award to T. Farrell. Special thanks go to the USU and BSU laboratory staff. We thank R. Kubina, T. Nielson, M. Pulsipher, and Silty for field assistance; Steve Westrop for palaeontologic support; and Justin Wilkins (U.S. Forest Service) for assistance in obtaining permits. Constructive reviews from J. Schiffbauer, S. Pruss, and B. Gill improved this manuscript.

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