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# Isotopic Anomalies from Neutron Reactions During Explosive Carbon Burning

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31 October 1978



The Ivan A. Getting Laboratories  
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## ABSTRACT

We examine the possibility that the newly discovered correlated isotopic anomalies for heavy elements in the Allende meteorite were synthesized in the secondary neutron capture episode during the explosive carbon burning--the possible source of the  $^{160}\text{Sm}$  and  $^{26}\text{Al}$  anomalies. Explosive carbon burning calculations under typical conditions were first performed to generate time profiles of temperature, density, and free particle concentrations. These quantities were inputted into a general neutron capture code which calculates the resulting isotopic pattern from exposing the pre-existing heavy "seed" nuclei to these free particles during the explosive carbon burning conditions. Comparison with published data shows that under certain conditions the anomalies can be explained by such a process, although the detailed fit is not perfect. This process may provide a viable alternative to the classical r-process which has also been proposed to explain the anomalies. Unlike the r-process, the present interpretation avoids the problem of the Sr isotopic data and may resolve the conflict between the time scales inferred from  $^{129}\text{I}$ ,  $^{244}\text{Pu}$ , and  $^{26}\text{Al}$ . Experimental studies of Zr and Ce isotopic composition can be used to test this model.

## CONTENTS

ACKNOWLEDGMENTS . . . . .	iv
ABSTRACT . . . . .	v
I. INTRODUCTION . . . . .	1
II. HEAVY FUN ANOMALIES . . . . .	3
III. NEUTRON CAPTURE REACTIONS DURING EXPLOSIVE CARBON BURNING . . . . .	5
IV. RESULTS AND DISCUSSION . . . . .	9
V. CONCLUSIONS . . . . .	14
REFERENCES . . . . .	15

## FIGURES

1. The time evolution of the neutron and proton mass fractions during explosive carbon burning are shown for the "cosmic seed" and the "He zone s-seed" cases . . . . .	8
2. The ratio $\epsilon$ (calculated)/ $\epsilon$ (observed) is plotted for the isotopes of Ba, Nd and Sm . . . . .	11

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## I. INTRODUCTION

The study of isotopic patterns in meteorites provides a "window" on the nucleosynthetic origin of the solar system. If material from different nucleosynthetic sources has not been completely homogenized before the formation of planetary objects then these objects may show isotopic abundance variations ("anomalies"). These anomalies can help in identifying the original sources. This approach is particularly powerful when correlated isotopic anomalies occur simultaneously for several elements in the same sample because, in this case, the observed pattern can be compared more directly with the predicted isotopic patterns from theoretical models of nucleosynthesis.

Correlated isotopic anomalies have recently been discovered in two Ca-Al rich inclusions of the Allende meteorite. These inclusions are characterized by extremely large mass fractionation effects in Mg (Wasserburg et al. 1977), Si (R. Clayton et al. 1978; Yeh and Epstein 1978), and O (R. Clayton and Mayeda 1977) as well as additional isotopic variations of unknown nuclear origin in Ca (Lee et al. 1978), Sr (Papanastassiou and Wasserburg 1978), Ba and Nd (McCulloch and Wasserburg 1978a), and Sm (Lugmair et al. 1978). After removal of fractionation, nuclear effects in Mg (Wasserburg et al. 1977) and Si (Yeh and Epstein 1978) also are apparent. This complicated set of anomalies has been collectively christened "FUN" (for Fractionation and Unknown Nuclear), and no comprehensive theoretical model is as yet available.

One feature of the FUN anomalies that has emerged is that the unshielded neutron-rich isotopes of Ba, Nd and Sm invariably show anomalous abundances. This has been interpreted as suggesting a nucleosynthesis



process that proceeds via neutron captures on the neutron rich side of the valley of beta stability, such as an r-process event (McCulloch and Wasserburg 1978a,b; D. Clayton 1978; Lugmair et al. 1978). However, the astrophysical site for r-process nucleosynthesis is unknown. Also, no theoretical model has so far been able to reproduce the solar system r-process abundance to the accuracy enjoyed by most other nucleosynthesis processes (cf. Norman and Schramm 1978). In addition, there are other episodes of neutron capture nucleosynthesis in a massive star (Howard et al. 1972; Arnett and Wefel 1978; Truran et al. 1978), which are not the "classical" r-process (Burbidge et al. 1957) but may be important contributors to heavy element nucleosynthesis.

Prior to the discovery of FUN, O isotopic anomalies due to the presence of  $^{16}\text{O}$  rich material had already been discovered in a wide variety of objects including Allende (R. Clayton et al. 1973, 1976, 1977). Furthermore, evidence for the presence of radioactive  $^{26}\text{Al}$  ( $t_{1/2} \sim 7 \times 10^5$  yr.) in the early solar system has also been found in Allende (Lee et al. 1977). Although the relation between  $^{16}\text{O}$  and  $^{26}\text{Al}$  anomalies remains an open question, various models have been proposed to produce them simultaneously. The production in the solar system by proton irradiation seems unlikely (D. Clayton et al. 1977; Schramm 1976) but cannot be completely ruled out (Lee 1978). However, it appears more plausible to assume that they were produced inside a nearby massive star, presumably by the carbon burning nucleosynthesis in either an explosive (Truran and Cameron 1978) or a high temperature "hydrostatic" (Arnett and Wefel 1978) environment. This star exploded as a supernova immediately before the formation of the solar system and injected into it the freshly synthesized nuclei to cause the anomalies (Cameron and

Truran 1977; Lattimer et al. 1978). Since carbon burning nucleosynthesis may be responsible for the  $^{26}\text{Al}$  and  $^{16}\text{O}$  anomalies, it is clearly important to investigate whether the neutron capture episode associated with this process can be the origin of the FUN anomalies. In this paper we examine neutron capture processes during explosive carbon burning (ECB) and compare the resulting isotopic pattern with the observed heavy element anomalies.

## II. HEAVY FUN ANOMALIES

The problem of interpreting isotopic anomalies is intimately related to the representation of the data. The data from isotopic ratio measurements are customarily reported as the fractional deviation (usually in units of parts in  $10^4$ ) for isotope  $i$  with respect to index isotope  $j$ , i.e.,  $\epsilon_j^i \equiv [(N^i/N^j)_s / (N^i/N^j)_o] - 1$ , where  $N$  is the abundance,  $s$  denotes the sample value, and  $o$  denotes the normal value. To obtain the "pure" nuclear effect a normalization isotope  $k$  has to be chosen such that the fractionation effect due to mass dependent processes in the laboratory and in nature can be "removed." Depending on the choice of the index isotope and the normalization isotope there are many possible representations of the data. If the sample is a mixture of normal solar system material and material from a peculiar source and if the index and normalization isotopes  $j$  and  $k$  are not produced in that source then the theoretical production ratio ( $p$ ) is related to the observed fractional deviations simply by

$$(\epsilon^\alpha / \epsilon^\beta) = (N^\alpha / N^\beta)_p / (N^\alpha / N^\beta)_o \quad (1)$$

Thus, with eq. (1), calculated  $\epsilon$ 's can be obtained from theoretical production ratios and compared to the observed  $\epsilon$ 's by forcing  $\epsilon^\beta(\text{calculated}) / \epsilon^\beta(\text{observed})$

to 1 for the reference isotope  $\beta$ . This representation facilitates the comparison between models and data but does depend on the above assumptions. In the discussion of nucleosynthesis in neutron rich environments the choice of  $j$  and  $k$  is obviously the shielded ( $s$ -process only) isotopes. Therefore, for Ba and Sm,  $^{134}\text{Ba}$ ,  $^{136}\text{Ba}$ ,  $^{148}\text{Sm}$ , and  $^{150}\text{Sm}$  are used. Nd has only one shielded isotope,  $^{142}\text{Nd}$ , and we adopt  $\epsilon(^{150}\text{Nd}) = 33.6$  for normalization. Using this representation and published data for the sample EK 1-4-1 (McCulloch and Wasserburg 1978a,b) the observed  $\epsilon$ 's for the isotope in parenthesis are: 13.2 ( $^{135}\text{Ba}$ ), 13.7 ( $^{137}\text{Ba}$ ), 1.6 ( $^{138}\text{Ba}$ ), 38.5 ( $^{147}\text{Sm}$ ), 36.5 ( $^{149}\text{Sm}$ ), 24.1 ( $^{152}\text{Sm}$ ), 31.6 ( $^{154}\text{Sm}$ ), 13.6 ( $^{143}\text{Nd}$ ), 18.0 ( $^{144}\text{Nd}$ ), 24.7 ( $^{145}\text{Nd}$ ), 13.8 ( $^{146}\text{Nd}$ ), 33.6 ( $^{148}\text{Nd}$ ). This set will be employed as the "data" against which the calculations are to be compared.

In the above representation the following three basic features of the heavy FUN anomalies become evident: (1) The unshielded isotopes have anomalous abundance suggesting that the anomalies are due to a peculiar component made in a neutron rich environment; (2) the data for EK 1-4-1 show large positive  $\epsilon$ 's indicating that this sample is enriched in the peculiar component; (3) C1, the other FUN sample, shows normal abundance for most isotopes except a small but negative  $\epsilon(^{135}\text{Ba})$  of -1.8 and consequently a deficiency relative to the normal. The last feature can be explained by a hold-up in  $^{135}\text{Cs}$  (McCulloch and Wasserburg 1978a). However, it may also be interpreted as implying that the normal is not a well-defined end component for the mixing but itself a mixture containing more of the peculiar material than C1. Note that this interpretation contradicts the assumption in deriving eq.(1).

Another feature of heavy FUN anomalies evident in this representation is that the abundance of the proton rich isotope  $^{144}\text{Sm}$  is also anomalous (Lugmair et al. 1978) and varies independently of that of

the neutron rich isotopes (McCulloch and Wasserburg 1978b). This implies that the heavy FUN anomalies require at least one more peculiar component involving the poorly understood p-process isotopes. The Sr isotopic results (Papanastassiou and Wasserburg 1978) also show that the data for EK 1-4-1 is not consistent with the addition of only the unshielded neutron rich isotopes. Therefore, the neutron rich component is not the sole cause of the FUN anomaly even when we consider only elements above the Fe peak. In this paper we will not tackle the production of the p-process component.

### III. NEUTRON CAPTURE REACTIONS DURING EXPLOSIVE CARBON BURNING

When a supernova explodes the shock wave traverses the carbon shell of the star. ECB occurs if the shock wave heats the shell to temperatures  $\sim 2.0 \times 10^9 \text{K}$  ( $T_9 \sim 2.0$ ) for typical initial densities of  $\sim 10^5 \text{g/cm}^3$ . Nucleosynthesis in this environment has been studied first by Arnett (1969) and then more extensively by Pardo et al. (1974).

During ECB neutrons are generated by reactions such as  $^{22}\text{Ne} (\alpha, n) ^{25}\text{Mg}$  and by the neutron channel of the  $^{12}\text{C} + ^{12}\text{C}$  reaction. For a hydrodynamic time scale, the entire neutron capture episode takes only 1-2 seconds, but the neutron density is never large enough to produce the conditions of the classical r-process (Howard et al. 1972). Therefore, a general formalism for neutron captures (n-process) (Blake and Schramm 1976) has been used to follow the neutron capture reactions. The n-process code calculates explicitly the  $(n, \gamma)$ ,  $(\gamma, n)$  and beta decay rates for all the nuclei in the network. It can go to the limits of either an s-process or an r-process and is an appropriate formalism for studying general neutron

capture nucleosynthesis including the intermediate neutron density environments during ECB. (For a more detailed description of the operation of the n-process code, see Blake et al. 1978.) Below  $A = 100$ , proton induced reactions become important, and (p,n) (n,p) (p, $\gamma$ ) reactions have also been included explicitly. Reaction rates, as a function of temperature, were obtained from a computer code developed by Woosley et al. (1975).

The n-process code requires as input a "seed" distribution and the temporal evolution of the neutron concentration, temperature, and density in ECB. For  $A < 100$  the evolution of proton concentration is also needed. We have used an ECB code, identical to that employed by Pardo et al. (1974), which contains a complete reaction network for the isotopes between  $^{12}\text{C}$  and  $^{38}\text{Ar}$ . In addition, a fictitious nuclide was included in the direct network. Temperatures in the range  $1.8 \leq T_9 \leq 2.8$  were assumed for an initial density of  $10^5\text{g/cm}^3$  and a hydrodynamic expansion time-scale. Initial abundances were those characteristic of the end of helium burning, both for stars that experience a limited s-process during core helium burning (Couch et al. 1974; Lamb et al. 1977) and for those that do not. This s-process alters the abundance of  $^{22}\text{Ne}$  and  $^{25,26}\text{Mg}$ , which affects the neutron generation, and modifies the initial distribution of heavy elements. For each set of conditions, the neutron and proton concentrations, temperature, and density were recorded as a function of time for input to the n-process calculation (Fig. 1).

The "seed" distribution, the relative abundances of the heavy elements which will capture neutrons, is determined by the evolution of the star prior to the explosion. Essentially this distribution is the "cosmic" or solar system abundance distribution (Cameron 1973)

augmented by any processing that occurs during core helium burning. Two types of seed distributions were studied: the solar system abundances and the modified yields following the s-process during helium burning (Lamb et al. 1977). The latter case enhances the relative abundance of "s-process" nuclei.

It is not our intention to carry out an exhaustive search for parameters which best fit the data. Instead we are interested in what is the resulting abundance pattern of heavy nuclei under typical ECB conditions. We also attempt to detect any qualitative change for different conditions in order to provide guidelines for more detailed work in the future. With this in mind, we have performed calculations for the following cases:

- (1) "Cosmic Seed" -  $T_9 = 2.1$  and solar system abundances as seed;
- (2) "He zone s-seed" -  $T_9 = 2.1$  and the seed distribution following an s-process during core helium burning;
- (3) "Hot" -  $T_9 = 2.8$  and the same seed distribution as in case 2.

Case 2 represents perhaps a more realistic set of conditions than the others since the temperature and density fall within the range identified by Pardo et al. (1974) and the seed distribution is appropriate to massive stars that are supposedly supernova progenitors. However, the degree of s-processing and shock heating can vary from stars of one mass range to the next.

In general, there are three stages to the evolution during ECB (Fig. 1). First there is a short burst of neutrons ( $\sim 10^{-2}$  sec) and a somewhat longer burst ( $\sim 10^{-1}$  sec) of protons which drive the neutron and proton reactions. Following this stage the free particle densities gradually decrease to zero in 1-2 seconds. However, the temperature

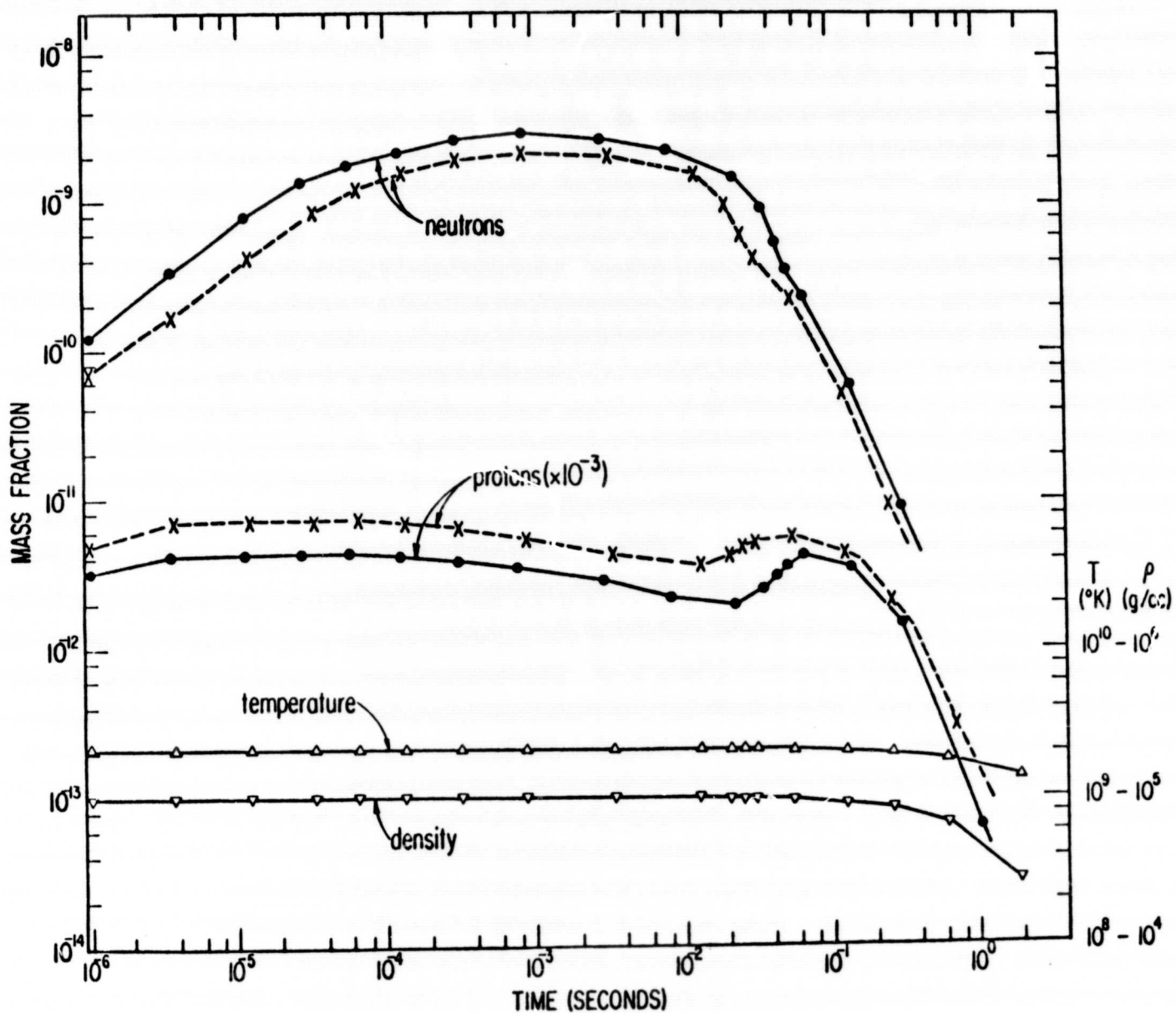


Figure 1: The time evolution of the neutron and proton mass fractions during explosive carbon burning are shown for the "cosmic seed" (●) and the "He zone s-seed" (x) cases. The temperature and density profiles are shown by the scales on the right.

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remains high so that photodisintegration reactions dominate. The final stage is the freeze-out during which the temperature decreases and only beta decays continue. During the initial neutron capture period the abundance of a given isotope is moved to the neutron-rich side of the valley of beta stability by about 3-6 neutrons, in contrast to the 10-15 neutron captures in the classical r-process. However, in the next stage of photodisintegration, the weakly bound species are eliminated by  $(\gamma, n)$  reactions. Finally, the abundances are altered by beta decay of the radioactive isotopes. The photodisintegration stage is a distinct feature of the ECB heavy element nucleosynthesis. This mechanism enhances the abundance of the tightly bound nuclei and reduces that of the weakly bound nuclei relative to the pattern dictated by simple  $(n, \gamma)$   $(\gamma, n)$  competition.

#### IV. RESULTS AND DISCUSSION

For the "cosmic seed" and "He zone s-seed" cases the isotopic abundance of a given element is usually moved to the unstable neutron rich isotopes and then replenished by the  $\beta$ -decays from the neutron rich unstable isotopes of the several elements immediately beneath it. Therefore the shielded isotopes are not produced. In the "hot case" considerably more neutrons are produced, but the photodisintegration rate increases even more. Therefore the neutron rich nuclei are destroyed and the  $\beta$ -decays are generally from the proton rich side. Thus, qualitatively, the neutron captures in the ECB zone fulfill the requirement for the source of the heavy FUN anomaly, since they can produce a component of heavy elements void of shielded isotopes under typical conditions. For quantitative comparison we have calculated



the values of  $\epsilon$  using eq. (1) for cases 1 and 2 and plotted the ratios to the experimental  $\epsilon$  in Fig. 2. Here a perfect agreement is signified by points along a line through 1. Within an uncertainty of a factor of five, roughly the accuracy of the theoretical r-process calculations, these results give a reasonable representation of the data except for the isotopes  $^{138}\text{Ba}$ ,  $^{144}\text{Nd}$  and  $^{149}\text{Sm}$ . The overabundance at  $^{138}\text{Ba}$  is due to the stability associated with neutron magic number of 82. The underabundance of  $^{149}\text{Sm}$  is because Pm has no stable isotopes while the low abundance of  $^{144}\text{Nd}$  is due to its progenitor  $^{144}\text{Pr}$  being a weakly bound odd-odd nuclide. Curiously, the presumably more realistic "He-zone s-seed" case gives a slightly worse fit than the "cosmic seed" results. In any case, these discrepancies seem to be a general result of the ECB heavy element nucleosynthesis and are distinct problems in this explanation of the FUN anomaly.

For comparison, results for the r-process are also shown in Fig. 2. The "cosmic" r-process curve is obtained from the solar system abundances by subtracting the s-process contributions following D. Clayton (1978) for Ba and Nd and using a constant  $\langle\sigma N\rangle$  of 6.4 for Sm. This distribution gives a remarkably good fit to the observed heavy element anomalies. However, it should be emphasized that the "cosmic" r-process abundance is obtained through a semi-empirical procedure and is not the result of a nucleosynthesis calculation. Fig. 2 also shows the  $\epsilon$ 's for a recent r-process calculation which is probably the most realistic attempt to reproduce the solar system r-process abundances (Norman and Schramm 1978). The fit for this case is not nearly as good as the "cosmic" r-process, and it is comparable to the fit of the present ECB results. This merely reflects the general lack of precision in the

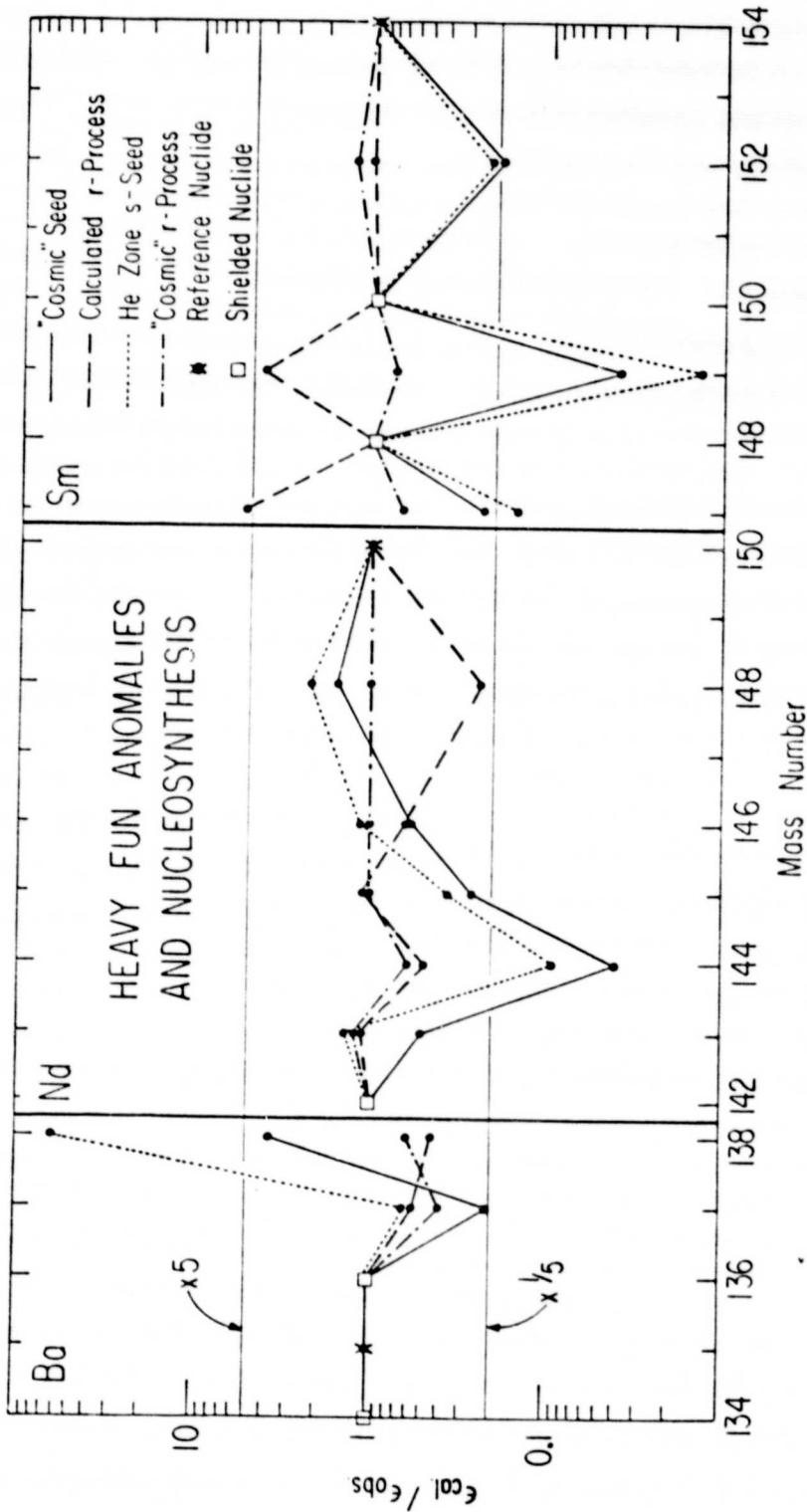


Figure 2: The ratio  $\epsilon(\text{calculated})/\epsilon(\text{observed})$  is plotted for the isotopes of Ba, Nd and Sm. The ECB n-process results for the "cosmic seed" and "He zone s-seed" cases are compared to the "cosmic" r-process and to the calculated r-process results. Agreement within a factor of five is indicated by horizontal lines. Perfect fit would be a horizontal line through 1.  $\epsilon's(\text{calculated})$  are obtained using eq. (1) with reference isotopes as indicated.

calculation of heavy element nucleosynthesis. It is well known that the superposition of r-process patterns calculated at slightly different conditions can result in a smoother pattern and thus a better fit (Seeger et al. 1965). It is conceivable that a range of explosive conditions for different portions of the carbon shell, when combined, may give a better fit to the experimental data.

There are other areas where ECB n-process and r-process explanations offer different predictions. In the case of Sr, the r-process predicts an enrichment in  $^{88}\text{Sr}$  only which is in direct conflict with the observation. The n-process produces virtually no Sr and this may be the reason why the other peculiar component required for FUN anomalies seems to dominate the Sr effect. However, this result depends on the estimate that the yet unmeasured neutron capture cross-section of  $^{86}\text{Kr}$  is smaller than that of  $^{88}\text{Sr}$ .

Another area of contrasting predictions is cosmochronology. It has been well established that the r-process produced isotopes  $^{244}\text{Pu}$  and  $^{129}\text{I}$  yield a consistent timescale of  $\sim 10^8$  years for the period between the last r-process event and the retention of xenon in meteorites (Schramm and Wasserburg 1970). Clearly, if  $^{26}\text{Al}$  is formed in the same event as  $^{244}\text{Pu}$  and  $^{129}\text{I}$ , there is a conflict in timescales. This difficulty is further accentuated if the FUN anomalies are attributed to the same r-process which produces Pu and I, unless one makes the ad hoc assumption that  $^{26}\text{Al}$ ,  $^{129}\text{I}$ , and  $^{244}\text{Pu}$  are each diluted by different factors.

Both  $^{129}\text{I}$  and  $^{244}\text{Pu}$  are weakly bound nuclei and therefore are under-produced in the ECB n-process relative to the classical r-process. In fact, essentially all of the actinides including Pu are destroyed. The

iodine region ( $^{127}\text{I}$  and  $^{129}\text{I}$ ) is similarly depleted. For example, the  $^{129}\text{I}/^{135}\text{Ba}$  production ratio in the ECB n-process is about factor 10 lower than that in the classical r-process. Therefore, the ECB n-process does not produce  $^{129}\text{I}$  and  $^{244}\text{Pu}$  and this result allows a more coherent scenario for the interpretation of the timescale between nucleosynthesis and solidification: The last r-process event took place  $\sim 10^8$  years before the solar system formed, perhaps associated with the last passage of the density wave, while the r-like material observed in FUN samples was produced along with  $^{26}\text{Al}$  in a supernova which exploded  $\sim 10^6$  years before solar system formation.

There are experimental tests that can distinguish the ECB n-process and r-process. The neutron flux in the n-process is much weaker than the r-process. Therefore, the abundance of an element, whose most neutron rich stable isotope has a magic number of neutrons, is pushed just barely beyond that isotope. For example, the abundance peak at  $^{138}\text{Ba}$  is pushed over to  $^{140}\text{Ce}$  and the  $^{88}\text{Sr}$  peak is moved to  $^{90}\text{Zr}$  and  $^{92}\text{Zr}$ . This is in sharp contrast with r-process where the corresponding abundance peaks are shifted much further ( $\gtrsim 10$  mass unit). For Ce the ECB n-process predicts an abundance ratio  $N(^{140}\text{Ce})/N(^{142}\text{Ce}) \sim 6$  or  $\epsilon(^{140}\text{Ce})/\epsilon(^{142}\text{Ce}) \sim 0.7$ , while r-process predicts  $\epsilon(^{140}\text{Ce})/\epsilon(^{142}\text{Ce}) \ll 1$ . For Zr the ECB n-process predicts the following pattern:  $N(^{90}\text{Zr}) : N(^{91}\text{Zr}) : N(^{92}\text{Zr}) : N(^{94}\text{Zr}) \sim 1 : 0.003 : 0.7 : 0.23$ ; while the cosmic r-process pattern probably shows a much gentler decrease from  $^{90}\text{Zr}$  to  $^{94}\text{Zr}$  although the precise abundances are difficult to estimate. The low predicted abundance of  $^{91}\text{Zr}$  should probably not be taken too seriously in view of the failure of the present model in similar nuclides such as  $^{149}\text{Sm}$ . However, we believe that the steep drop-off from  $^{90}\text{Zr}$  to  $^{94}\text{Zr}$  may be a distinct

feature of the ECB n-process. Therefore, the experimental studies of the Ce and Zr isotopic compositions may provide important tests for our model and, in general, supply information on the competition between neutron captures and photodisintegration reactions in the source zone of the heavy FUN isotopic anomalies. Unfortunately, the interpretation of the data will be hindered somewhat by appropriate isotopes for the removal of the fractionation effects and by the possible contribution of the poorly understood p-process component.

#### V. CONCLUSIONS

The heavy isotopic anomalies observed recently in the FUN inclusions from the Allende meteorite offer a challenge to theoretical explanation. Neutron reactions during ECB offer a reasonable fit to the positive anomalies in inclusion EK 1-4-1 and is thus a viable alternative to an r-process explanation of the anomalies. This model seems attractive because it produces heavy anomalies in the same zone where  $^{26}\text{Al}$  and  $^{16}\text{O}$  are produced and thus reduces the number of source zones required for the isotopic anomalies. In addition, the ECB n-process avoids the problem with the Sr anomaly and may resolve the problem of conflicting time scales between  $^{26}\text{Al}$  and the r-process isotopes  $^{129}\text{I}$  and  $^{244}\text{Pu}$ . Further studies are needed, both theoretically, into the details of the neutron captures in more realistic explosive environments, and experimentally, in the measurement of additional samples and elements, especially the isotopes of Zr and Ce.

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