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Conditions and New Extrapolation Method for ESR Dating of Aragonitic Mollusk Shells in Taiwan

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ESR dating of aragonitic shells using different magnetic field amplitude modulation and microwave power levels has been studied to establish appropriate conditions of ESR measurement. The ESR ages obtained using the signal at $g=2.0007$ (rather than using the signal at $g=2.0014$) and a microwave power of 100 mW, are consistent with those of the biological methods such as the pollen and nannofossil data regardless of the species. The two step extrapolation method assuming a theoretical growth curve in nature and including the additive dose method gave ages around 140 ka while the conventional additive dose method alone gave ages 90 ka younger than expected from other geologic data.

Keywords
ESRDD, ESR condition, defect lifetime, Szekou formation, aragonitic mollusk

1. Introduction

There is some controversy over the appropriate ESR dating signal and measurement conditions for mollusk fossils (Ikeya & Ohmura, 1981; Radtke et al., 1985; Molodkov, 1988; Katzenberger and Willems, 1988; Katzenberger et al., 1989; Skinner, 1989; 1992; 1993; Ikeya, 1993). Skinner used three different conditions to test the appropriate methodology (Skinner, 1989), including the so called $g=2.0014$ signal for dating (Skinner & Weicker, 1992) which Katzenberger insisted should not be used (Katzenberger et al., 1989). Similar experimental comparison has been done in this paper in order to check the appropriate conditions. The conditions suggested by Radtke et al. and Molodkov are used in this study while Skinner used slightly different conditions from theirs. It seems that the dating signal $g=2.0007$ at high microwave power (100 mW) is better than the signal at $g=2.0014$ at low microwave power. Furthermore, the intensity of the $g=2.0057$ signal might be dependant on mollusk species. The *Gerastoderma sp.*, which is claimed by Radtke etc not to be used for ESR dating, become useful specimen for ESR dating.

Figure 1 shows the sample localities. All shells were obtained from the Szekou Formation, in the Hengchun area, at the southern tip of Taiwan. The Hengchun platform occupies the west area of Hengchun peninsula. The Szekou Formation, about 40 m thick, forms part of the basement of this uplift platform. The altitude of Szekou Formation ranges from 10 - 80 m due to the eastward tilting of the platform. Here, the altitude (<100 m) and latitude (22°) do not cause significant differences to the cosmic ray dose compared with that at sea level (Prescott & Stephan, 1982; Ikeya 1993). This formation is composed of unconsolidated silty mud with abundant mollusk fossils comprising 257 mollusk species (Cheng, 1991). The sedimentary environment of Szekou Formation was a Pleistocene lagoon.

Ten species were used here for dating purposes. Four species and five complete bivalve shells (*Eucrassalella nana*) from loc.S2 (bottom of this formation) were excavated within a 1 m×1 m×50 cm area to test the concordance of the ESR ages and the possibility of individual variations within one species, respectively. Loc.S2 and S5 are at the bottom of this formation, 1m above the

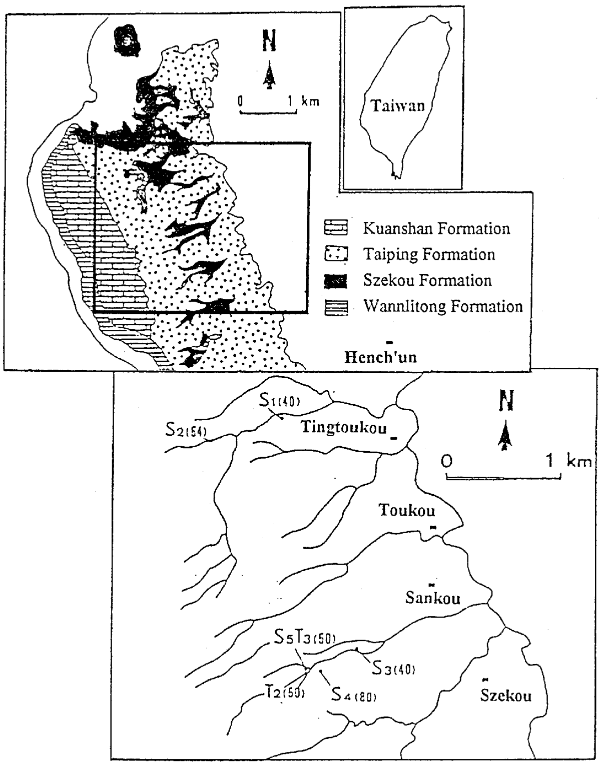


Fig. 1: ESR sample localities. All specimens were sampled from the Szekou Formation. S2 and S5 are at the bottom of the Szekou Formation, 1 m above the Wannilitoung Formation. S1 and S4 are at the upper part. S3 is in the middle part. The Arabic numerals enclosed in parentheses denote the altitude. (Geological map after Chen, 1991)

Hengchun Limestone. Loc. S1 and S4 are at the top and upper part of this formation, respectively. Loc.S3 is in the middle part. All shells were excavated from at least 30 cm deep within the surface of the outcrop (several to tens of meters from the top surface). Only shells from loc.S1 were picked up directly at the surface in order to test the possibility of the bleaching effect of sunshine. The maximum thickness of shells was less than 2 mm except some shells (5 mm) from loc.1. Thus considering the thickness of the sample, the external β–dosage should be included.

There are still many problems in ESR shell dating (Skinner & Weicker, 1992). One problem is the determination of external dose. This is why they chose shells which were in situ and imbedded in a uniform carbonate site. However, it is a special case and cannot be used widely for dating purposes. Furthermore, specimens must be sampled and cleaned with difficulty from associated semi-consolidated carbonates. Another approach for obtaining a reliable ESR age is to use shells not only in situ but also excavated from a very small area. The smaller area we choose, the higher the chance of uniform external environment. This has been already mentioned by Molodkov (1988) and is also what we did here. Whether the surrounding matrix is uniform or not should still be checked from the results.

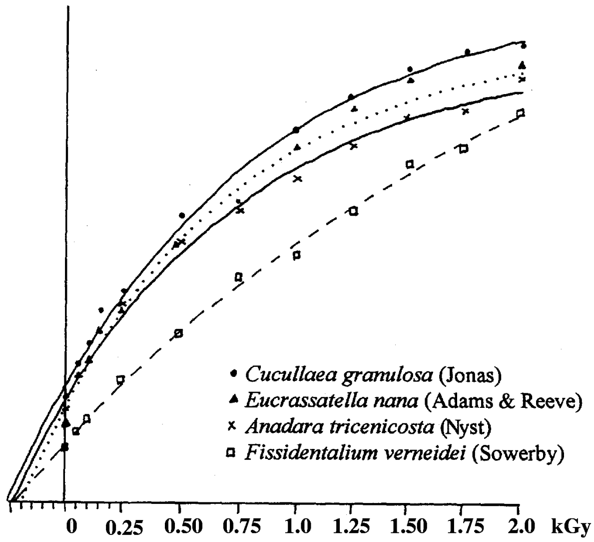


Fig. 2: ED obtained by ESR curve fitting. All shells were excavated within 1 m × 1 m × 50 cm at Loc. 2. *Cucullaea granulosa* (Jonas), *Anadara tricenicosta* (Nyst) and *Eucrassatella nana* (Adams & Reeve) belong to type 1. *Fissidentalium verneidei* (Sowerby) belongs to type 2. Both type 1 and type 2 get the same ED when using the conditions of case 2, (i.e. both can be used for dating purposes). But in case 1 and case 3, the ED of type 2 shells will be much younger than type 1 shells.

2. Experimental procedures

Samples were washed and cleaned ultrasonically, following by etching them with 5% HCl for 5 minutes to remove the external dosage effect, and powdered in a hand vise. The fraction 250-350 μm was selected. Each sample was divided into 9-13 aliquots, approximately 250 mg each. One aliquot of each sample was for INAA analysis and the other aliquot was unirradiated. The other 7-11 aliquots were then irradiated, by a cobalt-60 source which delivered 102 Gy/h. The artificial dosages were given to levels of 0.05, 0.10, 0.15, 0.25, 0.5, 0.75, 1.00, 1.25, 1.50, 1.75 and 2.00 kGy. (The intensity of maximum dosage, 2000 Gy is about 7-9 times the natural one in case 1, 5-6 times in case 2 and 6-7 times in case 3, as stated in the following).

After storage for at least one week, samples of precisely 250.0 ± 0.5 mg were put into a quartz tube (inner diameter : 2 mm) with 2.5 cm height which was equal to the width of the microwave gate. All the samples were investigated at room temperature using a Bruker esp-300E spectrometer (using X-band: 9.77GHz) with a ST4102 cavity. Three conditions have been tested and compared.

Case 1 used 0.5 Gpp, 2 mW, $g = 2.0014$ (Radtke et al., 1985); case 2 0.5 Gpp, 100 mW, $g = 2.0007$ and case 3 10 Gpp, 50 mW, $g = 2.0012$ (Molodkov, 1988). Total doses were derived from the additional dose method with

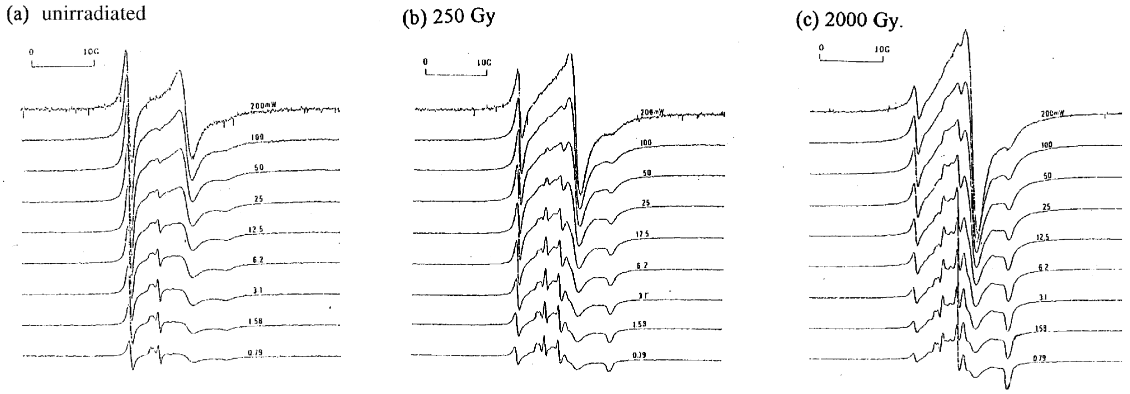


Fig. 3: ESR spectrum of *Eucrassatella nana* (Adams & Reeve) showing changes with different microwave power: (a) unirradiated (b) 250 Gy (c) 2000 Gy.

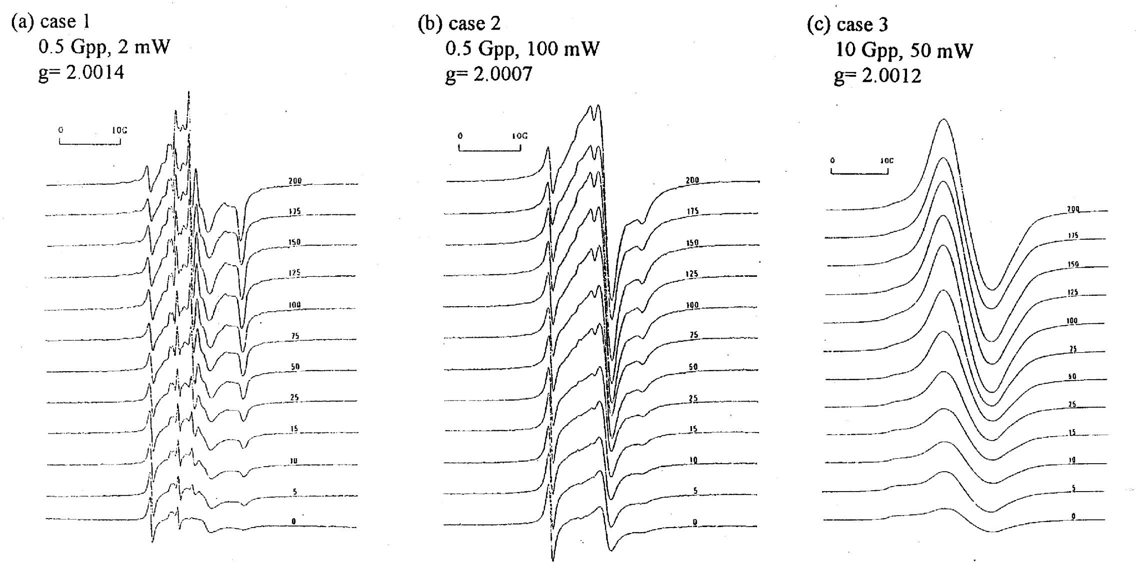


Fig. 4: The growth of the ESR spectrum of *Eucrassatella nana* (Adams & Reeve) as radiation dose increases in three cases: (a) case 1 uses 0.5 Gpp, 2 mW, $g = 2.0014$ (Radtke et al., 1985), (b) case 2 uses 0.5 Gpp, 100 mW, $g = 2.0007$ and (c) case 3 uses 10 Gpp, 50 mW, $g = 2.0012$ (Molodkov).

exponential curve fitting. The external dosages of γ-rays and cosmic rays were measured directly by TLD at loc.S5. No calcite could be detected by X-ray diffraction except in *Turritella filliola* from loc.S5. which had 0.5% calcite after 3 minutes etching by 5% HCL as detailed in Table 1.

3. Results and Discussion

3.1. Is the signal $g = 2.0014$ at low microwave power the appropriate dating signal?

Table 1 shows the results of three different experimental conditions, with the ED and the apparent ESR age. Case 1 ($g = 2.0014$, 0.5 Gpp, 1 mW) has the smallest ED while case 2 ($g = 2.0007$, 0.5 Gpp, 100 mW) has the largest one. And as Radtke pointed out, there did exist two types of shell (actually there may be more than two types) in case 1. Therefore, *Fissidentalium verneidei* (belong to the *Gerastoderma sp.* type) , which can not be used for dating, does exhibit a very different ED from the others. The same situation holds in case 3. However, in case 2, after saturating many signals using high microwave power (100 mW), there seems to be no differences between type I and type II. Similar ED's at loc.2 (Fig. 2) were obtained and both can be used for ESR dating.

The age of the Szekou Formation is reliably controlled by the nannofossil *Emiliania huxleyi* (start of NN21) of which the FAD (First Appearance Datum) is known to be 260 ka, so the age must be younger than 260 ka (Wang, 1984). An age ranging from 100-200 ka is suggested. A cool interval within Stage 5 (70-140 ka) is also suggested by pollen data (Liew, 1983; Cheng, 1986). ESR dating of "corals" from the Kuanshan Limestone which is either above the Szekou Formation (Shih, 1989) or contemporaneous with it according to Wu (1990) are around 90 ka and are supposed to be around 128 ka. A Carbon-14 age on wood from the upper part of this formation gave >50 ka. Two Carbon-14 ages derived from two shells by AMS are also over 50 ka. Therefore, the apparent ESR age in case 1 is too young to be accepted.

Table 1. A synoptic picture of ESR dating on aragonitic mollusk shells of Szekou Formation in Taiwan

locality	mollusk species	excavated depth (cm)	used number	etching time (5%HCl) (min)	grain size (μ)	aragonite (%)	radiation rate (Gy/h)	Radtke (1985) type	Case 1 0.5Gpp, 2mW *g*=2.0014 ED (Gy)	Case 1 age (ka)	Case 2 0.5Gpp, 100mW *g*=2.0007 ED (Gy)	Case 2 age (ka)	Case 3 10Gpp, 50mW *g*=2.0012 ED (Gy)	Case 3 age (ka)
S1	*Anadara granosa* (Linne)	ground	1	60	250-350	100	102.5	I	97	40.6-64.2				
S1	*Cyclina sinesis* (Gmelin)	ground	1	60	250-350	100	102.5	I	100	43.8-71.4	143	62.7-102	107	46.9-76.4
S1	*Gelonia papua* (Losson)	ground	1	60	250-350	100	102.5	I	46	19.4-30.9	86.0	36.2-57.8	60	25.3-40.7
S2	*Anadara tricenicosta* (Nyst)	>30	1	5	250-350	100	102.5	I	123	53.0	178	76.7	144	62.1
S2	*Anadara tricenicosta* (Nyst)	0 - 10	1	5	250-350	100	102.5	I	128	54.7	179	76.5	174	74.3
S2	*Cucullaea granulosa* (Jonas)	10 - 50	1	5	250-350	100	103.6	I	132	57.9	216	94.7	166	72.8
S2	*Eucrassatella nana* (Adams &Reeve)	10- 50	6	5	250-350	100	103.6	I	134	58.8	198	86.8	124	58.8
S2	*Fissidentalium verneidei* (Sowerby)	> 30	4	5	250-350	100	103.6	I	87	38.0	212	92.6	73	31.9
S2	*Fissidentalium verneidei* (Sowerby)	0 - 15	4	5	250-350	100	103.6	II	84	36.8	211	92.5	97	42.5
S3	*Fusinus gracillinus* (Adams & Reeve)	> 30	1	5	250-350	100	102.5	?	111	47.6				
S4	*Paphia (neotapes) undulatus*(Born)	> 30	3	5	250-350	100	103.6	I	137	53.5	195	76.2	165	64.5
S4	*Paphia (neotapes) undulatus* (Born)	> 30	3	5	0-250	100	103.6	I	138					
S5T3	*Turritella filiola* (Yokoyama)	> 30	200	2	250-350	99	103.6	I	129	56.6	221	96.9	180	78.9

locality	
S1	ground soil attached on the shell
S2	mudstone
S3	mudstone
S4	mudstone
S5	mudstone

Note : For 1 ppm U, $D'\beta / D'\gamma$=1.32; for 3 ppm U + 10 ppm Th + 1% K, $D'\beta / D'\gamma$=1.35; for 3 ppm U + 10 ppm Th + 2.5% K, $D'\beta / D'\gamma$=1.81. Here, external β dose rate is calculated from (Dex-1/2Dcosmic)×($D'\beta / D'\gamma$)×β attenuation factor, where Dex=1.40 mGy/a (from TLD), Dcosmic=0.15 (assume 1m from top surface (Prescott & Stephan, 1982)). The ambiguity caused from Dcosmic which ranges from 0-0.15 (taking the depth from 50m-1m (Ikeya,1993)) is only 3% in ESR ages in this paper., βattenuation factor = 0.5 (considering an infinite plane with shell thickness of 1mm (Mejdahl,1979)). Thus, here, the external β dose rate equals (1.40-0.07)×1.32×0.5 = 0.88 (mGy/a). (If use $D'\beta / D'\gamma$=1.81, here, ESR age is younger by 15%.)

(Continued)

locality	mollusk species	Th (ppm)	U (ppm)	α ray (k=0.05) (inner) (mGy/a)	β ray (inner) (mGy/a)	γ-ray + cosmic ray(TLD) (external) (mGy/a)	β ray (external) (mGy/a)	total annual dose rate (mGy/a)	ESR age (ka)
S1	*Anadara granosa* (Linne)	0	0.46	0.053	0.056	(1.40)	(0-0.88)	2.39-1.51	
S1	*Cyclina sinesis* (Gmelin)	0	0	0	0			2.28-1.44	62.7-102
S1	*Gelonia papua* (Losson)	0	0.38	0.44	0.046			2.37-1.49	40.4-66.9
S2	*Anadara tricenicosta* (Nyst)	0	0.17	0.019	0.021	(1.40)	(0.88)	2.32	76.7
S2	*Anadara tricenicosta* (Nyst)	0	0.26	0.030	0.032			2.34	76.5
S2	*Cucullaea granulosa* (Jonas)	0	0	0	0			2.28	94.7
S2	*Eucrassatella nana* (Adams & Reeve)	0	0	0	0			2.28	86.8
S2	*Fissidentalium verneidei* (Sowerby)	0	0.05	0.006	0.006			2.29	92.6
S2	*Fissidentalium verneidei* (Sowerby)	0	0	0	0			2.28	92.5
S3	*Fusinus gracillinus* (Adams & Reeve)	0	0.19	0.022	0.023	(1.40)	(0.88)	2.33	
S4	*Paphia (neotapes) undulates* (Born)	0	1.17	0.135	0.143	(1.40)	(0.88)	2.56	93.8
S4	*Paphia (neotapes) undulatus* (Born)	0	1.28	0.148	0.156				
S5T3	*Turritella filiola* (Yokoyama)	0	0	0	0	1.40	(0.88)	2.28	116
S1	ground soil attached on the shell	4.40	1.16		assume 1% K			assume 2.5% K	
S2	mudstone	9.73	3.13		D'β / D'γ ≒ 1.34			D'β / D'γ ≒ 1.93	
S3	mudstone	11.05	2.94		D'β / D'γ ≒ 1.29			D'β / D'γ ≒ 1.76	
S4	mudstone	12.54	3.32		D'β / D'γ ≒ 1.24			D'β / D'γ ≒ 1.68	
S5	mudstone	9.38	2.45		D'β / D'γ ≒ 1.36			D'β / D'γ ≒ 1.86	

The reason why Radtke et al. got a good match in case 1 is probably because they used linear fitting. However, we should use the growth line to fit and the exponential fitting discussed by Apers (1981) is closer to the growth line. The ED of modern shells will shift too far from the original point if we use the linear fit. This has already been demonstrated by Ninagawa et al (1985).

Skinner & Weicker (1992) excluded a condition similar to case 2 stating that "even with high power and high modulation amplitude there remain at least two peaks in the spectrum of the natural sample, and the shape and location of the peak shift as the radiation dose increases. This makes it inappropriate to use these conditions in this case." Almost at the same time, Barabas, et al. (1992) had shown a clear signal (2.0006) at high microwave power (200 mW) and suggested the importance of using high microwave power. They stated that " In order to enhance the dating signal it is recommended to use high microwave power because it is the only signal in this region that does not saturate and, hence, at high microwave power this signal typically dominates the ESR spectrum." Our data support part of both opinions and challenge part of both.

Fig. 3 shows the ESR spectrum changing with different microwave power at (a) unirradiated (b) 250 Gy and (c) 2000 Gy, respectively. As you can see, using high microwave power can saturate several peaks and make the dating peak $g = 2.0007$ (the same as found in Molodkov, 1988, $g = 2.0012$, in Barabas, 1992, $g = 2.0006$) easier to read. For Case 2 $g = 2.0007$ and $g = 2.0019$ (?) did fuse into one signal at high microwave power (200 mW) but when radiation dose increases, they are still discrete ((c) and Fig. 4(b)). On the contrary, using a low microwave power, the spectrum is complex (more than two peaks) and the signal at $g = 2.0014$ is a compound signal and hard to read. Therefore, we should not exclude the high microwave power condition because compound signals exist in both low and high microwave conditions, especially in low microwave conditions. Second, we did not see the peak shifting as the radiation increased.

Fig. 4 shows the signal increase in the three cases. (In case 3, 10 Gpp is not always large enough, especially for shells with a large signal A ($g = 2.0057$)) It seems that all signals increase without shifting their positions. Therefore, Case 1 is an inappropriate condition and Case 2 may be an appropriate one since it is consistent with the geological data and more convenient for dating purposes.

3.2. ESR dating using signal C ($g = 2.0007$)

Table 1 shows the species, ED obtained by the additive dose method, γ-ray and cosmic dose rate with TLD in this study (here $D_{ex} = D_\gamma + D_c$, where D_{ex} is the external dose rate, D the gamma dose rate and D_c the cosmic ray dose rate). The external β-ray dose rate D was partly calculated by estimating the sedimentary β-ray dose rate assuming an infinite plane with a shell thickness of 1 mm. Here, the external beta dose rate was estimated by subtracting the cosmic component ($1/2 D_c$ due to the cliff geometry) from the D_{ex} and then multiplying by the factor 1.32 (D_β / D_γ for 1 ppm of U)(also see the note in Table1)

$D_\beta = (D_{ex} - 1/2\ D_c) \times 1.32 \times$ (β attenuation factor)

Hence the total dose rate D we should use is

$D = D_{in} + D_{ex} + D_\beta$

D_{in} was calculated considering the content of U, Th and K of shells. All shells here contain no Th as measured by INAA and no K (two species chosen for a two-species test by ICP and PIXE). The U content measured by INAA is usually below 0.5 ppm except in *Paphia (neotapes) undulatus* (1.2 ppm). Therefore, in this area, D_{in} can affect usually only 5% of the total dose and 10% is the maximum. The ambiguity caused from the cosmic dose rate $1/2 D_c$ which ranges from 0.07-0.15 mGy/a is only 3% of the ESR age. The problem here is that the shape and thickness of the shells (<2 mm) play an important role in estimating the attenuation factor of each shell. D_β plays a significant role in D and therefore, affects the ESR age.

3.3. Intensity of $g = 2.0057$: dependency on species

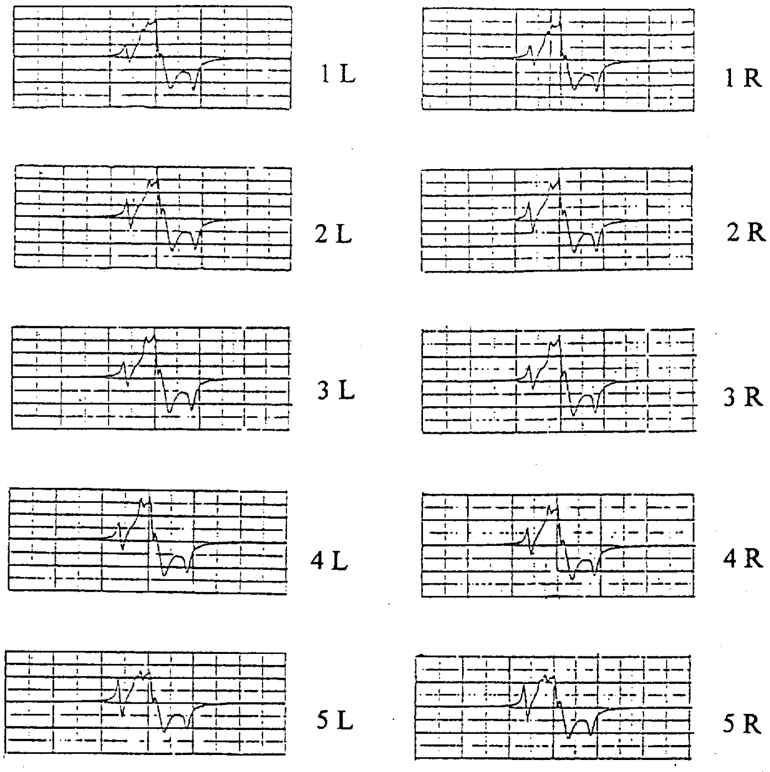
The signal intensity of A ($g = 2.0057$) was ascribed to SO_2^- within the shell since it appears after thermal annealing of SO_3^-. Some shells show an intense signal and some others show a weak signal. Resultant discussion proposes that there are two types of shells (Radtke et al., 1985). Our results indicate that three species in loc.2 (1 m × 1 m × 50 cm) show weak signals while one species shows an intense signal. This indicates that the signal intensity depends on the species rather than the thermal history. If the signal is really associated with SO_2^-, SO_2^- might arise from decomposition of sulfate-containing proteins like conchiolin in the shell. Further studies on different of species are needed in order to clarify the origin of the signal A.

3.4. Individual variation within one species

10 valves from 5 complete bivalve shells (*Eucrassalella nana*, a kind of pelecypod) from loc.2 (1m x 1m x 50cm) were irradiated at the same time and then analyzed. Figure 5 shows the shapes and intensities of ESR spectra from these 10 valves. It was found that

(i) Any shell of a pair is more similar to the other pair member rather than to a member of another pair, and this is not only in the intensity but also in the signal shape.

(ii) The standard deviation is 7.3% for 10 valves, 6.2% for right valves, 6.2% for left valves and only 2.9% for the differences between one valve and its pair.



Shell	No.	1	2	3	4	5
ESR intensity (a.u.)	left valve: L	4.90	5.30	5.45	4.65	4.65
	right valve:R	5.00	5.15	5.40	4.40	4.70
average :	4.96	standard deviation : 0.36 (7%)				

Fig. 5: ESR spectrum of 10 valves of 5 complete bivalve shells (*Eucrassalella nana*). The right (left) valve is similar to its left (right) valve rather than right (left) valves of other shells not only in intensity but also in signal shape.

Therefore, the standard deviation cannot be totally explained by the error from artificial radiation or measurement since it conflicts with statements (i) and (ii). It should be explained as tiny variations in dose in individual shells.

3.5. Revised age with two step growth

ESR ages were calculated from the equivalent doses obtained by the conventional additive dose method by assessing the annual dose rate within the sediment. However, the ESR ages were consistently about 90 ka younger than expected. The age was revised taking the lifetime of defects into account with a new method of "Two-step extrapolation" (Ikeya, 1992; 1993), the software of which was developed by Takaki (1999) and is open-source for use in ESR dating. One can download this directly from the home page or from the Appendix of the Proceedings of 2001-ESRDD-Osaka. The revised ages are listed in Table 2.

As mentioned above (see 3.1), the age of Szekou Formation is reliably controlled within the range, 50 ka~260 ka by C-14. and the nannofossil (NN21) method. Thus, all the ages in Table 2 could possibly be candidates. However, a cool stage (at the lower part of Szekou Formation) was suggested from pollens, shells and also from oxygen isotopes of shells. Besides, a transgression lag at the bottom/lower part of Szekou Formation has been recognized by molluscan assemblages (Chen, 1991) and we know the Isotope Stage 5e (last interglacial:128 ka) is the highest sea level at that time. Although some fluctuations exist (relative high sea level: Stage5a (80 ka), Stage5c (100 ka)), Stage 5 is a warm period without problems. Therefore, a transgression from Stage 6 (130 ka~190 ka) to Stage 5 can well explain the geology here. Now the good news is that the ESR-age obtained by the two-step growth curve method can support this well. Using $T = 22°C$, $\tau = 1.5 \times 10^5$, we obtain an age of around 140 ka for the bottom/lower part (S2) of Szekou Formation which is impossible to obtain by only using exponential fitting without considering the lifetime of the centers. Considering the lifetime of the defects instead of assuming "a forever center", we obtain ESR ages on the older side. Although we can not make a definitive answer on the age of the Szekou Formation due to uncertain assumptions about temperature of the Szekou formation and lacking annealing experiments,

Table 2: Correction of the shell age at Taiwan using the lifetime of the signal

locality	mollusk species	ED (Gy)	Age(ka) τ >> Age	τ at T_{amb} = 22 °C Age (ka) $\tau = 1.2 \times 10^5$	 Age (ka) $\tau = 1.5 \times 10^5$	 Age (ka) $\tau = 3.0 \times 10^5$
S1	*Geloina papua* (Losson)	86.0	36.2 ~ 57.8	45.7 ~ 84.0	43.7 ~ 77.1	40.4 ~ 66.9
S2	*Anadara tricenicosta* (Nyst)	178	76.7	135	113	90
S2	*Anadara tricenicosta* (Nyst)	179	76.5	123	108	88
S2	*Cucullaea granulosa* (Jonas)	216	94.7	234	164	116
S2	*Eucrassatella nana* (Adams & Reeve)	198	86.8	174	137	104
S2	*Fissidentalium verneidei* (Sowerby)	212	92.6	191	149	112
S2	*Fissidentalium verneidei* (Sowerby)	211	92.5	187	148	111
S4	*Paphia (neotapes) undulates* (Born)	195	76.2	131	111	89
S5T3	*Turritella filiola* (Yokohama)	221	96.9	244	169	119
S2	AVE and S.D. of shells at S2			174 }40 (23%)	137 }22 (16%)	104 }12 (11%)

our ESR data on aragonitic shells very significantly support the conclusion that Szekou Formation should have been formed during the transgression from period Stage 6 to Stage 5 and the surface of the West Hengchun Hill/top surface of Kuanshan Formation (Henchun Limestone) could be the S surface (Stage 5e).

4. Conclusion

Fitting a growth curve using the laboratory additive dose method and constructing a theoretical growth curve in the environment have been carried out for dating aragonitic shells using the recently created program and an additional parameter, the lifetime of the signal. The ESR ages of shells in the Szekou Formation, southern Taiwan using the signal at g = 2.0007 and microwave power of 100 mW (case 2) were less than 100 ka, but shifted to about 140 ka with the two-step growth curve. The obtained ESR ages are consistent with the other geologic data. This method should be used in both TL and ESR dating of materials whose signal lifetime is comparable with or older than the age.

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