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Radio-Carbon Age Determination by Tandem Accelerator Mass Spectrometry Technique and Its Application To The Korean Sea

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탄뎀加速器에 의한 放射性炭素 年代測定과 韓國海에의 摘用

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ABSTRACT

^{14}C age dating by AMS (accelerator mass spectrometry) technique was performed on twenty five small sized fossil shells and one peat taken from the sixteen piston cores in the southern and southeastern Korean Sea. AMS technique is available to date only a few milligram of amorphous carbons compare than conventional dating technique. It is described in detail of sample pre-treatment and experimental, and applied to the reconstruction of the sea level changes since the late Pleistocene in the Korean Sea. Dated age ranges from 520±100 to older than 33,500 years. Sedimentary facies in the study area represents a different environmental set which is affected by sea level fluctuation since the late Pleistocene.

要 約

탄뎀加速器에 의한 放射性炭素 年代測定은 韓國南海 및 南東海의 海底로부터 25개의 化石種 조개와 1개의 피트를 對象으로 實施되었다. 특히 AMS技術은

종래의 C-14測定方法에 비해 量的으로 數밀리그램의 元素態炭素만으로써 충분히 測定가능하다. 本稿는 自體 製作한 分析라인에 의한 前處理方法 및 分析過程을 記述하고, 韓國南海 및 南東海에 대한 後期更新世의 海水面變動史에 대하여 考察한다. 測定結果年代는 -530±100年에서 -33,500年 以上의 範圍로 本海域의 堆積相은 後期更新世 以後의 海水準變動에 의한 複雜한 堆積環境을 시사한다.

INTRODUCTION

The technique of accelerator mass spectrometry (AMS) developed during the last decade, enables us to measure the low abundance nuclides such as ^{3}H, ^{10}Be, ^{14}C, ^{22}Na, ^{26}Al, ^{32}Si, ^{36}Cl, ^{41}Ca, ^{53}Mn, ^{129}I, ^{186}Os, ^{187}Os, etc. in natural samples (Nakamura et al., 1987). One aspect of this technique applied to ^{14}C dating is that samples can be dated which are much smaller in quantity than required for conventional ^{14}C dating. Conventional ^{14}C dating is based on the measurement of specific ^{14}C activity in the sample carbon. It can be carried out either by proportional gas counting (PGC), of CO_2, C_2H_2, CH_4 or C_2H_6, or by liquid scintillation counting (LSC) of C_6H_6. Radio-carbon age dating with AMS techniques is based on the

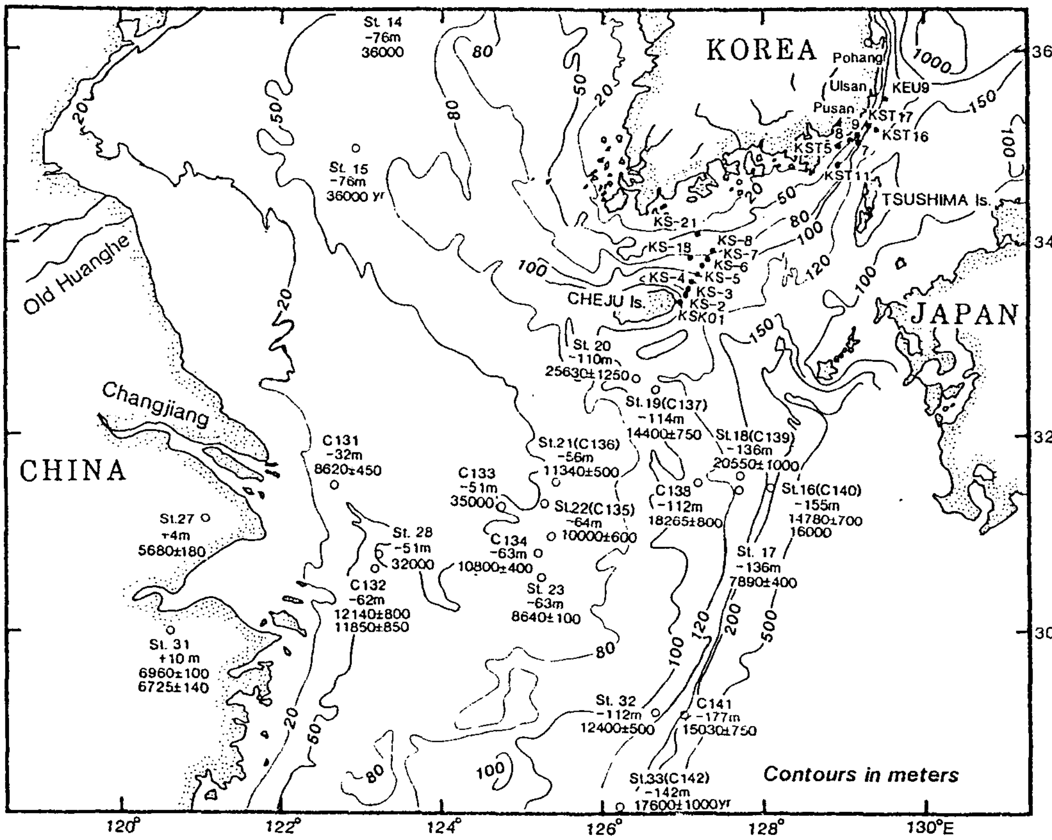


Fig. 1. Map showing sediment sampling locations and generalized submarine topography in the study area. Solid circles represent core sampling stations and open circles are compilation of the previous records of radiocarbon ages in the adjacent continental shelf. Water depths are in meters.

direct measurement of the $^{14}C/^{12}C$ abundance ratio (Nakai and Nakamura, 1984).

^{14}C age dating by AMS technique was applied to twenty five fossil shells and one peat taken from the sixteen piston cores in the southern and southeastern Korean Sea (Fig. 1). The purpose of this application was to reconstruct the sea level changes since the late Pleistocene in the Korean Sea and adjacent continental shelves. During late Pleistocene and early Holocene, the shelf area has experienced several sea level fluctuations, with the sedimentary records of these transgressive and regressive events being well preserved (Emery et al., 1971; Geng et al., 1987). The study area is good place to study sea level changes because of no glacial ice sheet covered on the continental shelf area and of relatively stable continental crust.

The purpose of this paper is to describe the nature of sample, experimental procedures and result. The detailed discussion of geological implications of the result will be presented elsewhere (Suk, 1989a, 1989b).

SAMPLING LOCALITY AND SEDIMENT FEATURES

^{14}C age dating by AMS technique was applied to twenty five fossil shells and one peat taken from the sixteen piston core in the southern and southeastern Korean Sea. A total of ten cores were selected for dating in the South Sea, and six cores from southeastern coast of the Korean Peninsula, respectively (Fig. 1).

In the studied area, there are many islands adjacent to the south and southeastern coast of the Korean Peninsula which is characterized by a ria type shoreline. Submarine topography of the South Sea lies very flat with the 0.04° to 0.07° slope gradient, and the bathymetric contour is mainly east-west direction up to the 90m water depth, however, it turns gradually towards northwest-southeast direction with increasing water depth. The Korea-Tsushima Strait is shallow with depths of about 30 to 230m and submarine contours to run parallel to the coast line of the Korean Peninsula. One submarine trough, the Korea Trough, extending northeast direction with a maximum depth of 230m was found in the central part of the Strait. The slope shows a relatively steep gradient of 3.8° in nearshore, gradual of 1.2° in inner shelf, and increasing rapidly up to 6.4° at the Korea Trough (Fig. 1).

Surficial sediments in the South Sea between Cheju Islands and Korean Peninsula appear to be variable. Relative thick muddy sediments are deposited nearshore of the south coast of Peninsula whereas sediments consisting mainly of sand, gravel including a lot of shell fragments (more than 30%) are dominant at the sea bottom deeper than about 80m in water depth. At the Korea-Tsushima Strait shows similar distribution pattern (Suk, 1989b). This sedimentary facies represents a different environmental set which is affected by sea level fluctuation since the late Pleistocene (Park and Song, 1971; Suk, 1981, 1986, 1989a, b).

EXPERIMENTAL PROCEDURES

Block diagram of sample preparation for the radio-carbon age dating is shown in Fig. 2, and the whole view of analysis system for sample preparation is shown in Fig. 3. The shell samples were cleaned with distilled water in an ultrasonic bath and then treated with 1.2N HCl for a short time (10 to 20 seconds) to eliminate surface contamination. The pretreated shell was powdered and then heated at 950°C for 25 minutes in a quartz test tube which is connected to an analytical line to produce CO_2. The evolv-

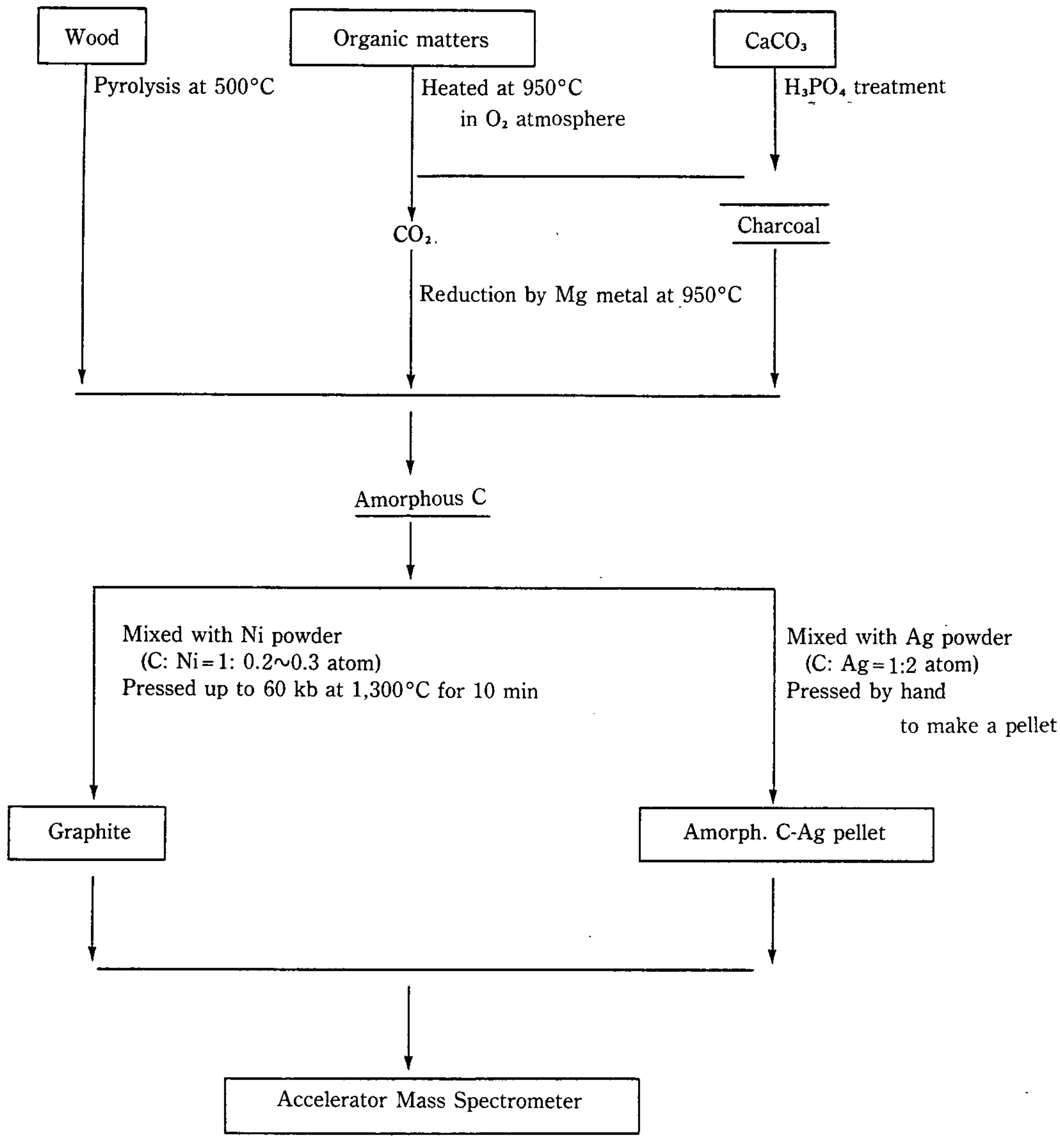


Fig. 2. Block diagram of the sample preparation process.

ed CO_2 was reduced to elemental carbon with Mg metal at 950°C for 3 hours. Peat sample was also cleaned in the same way as the shell sample and then treated with 4% NaOH and 1.2N HCl. The sample was pyrolyzed at 400°C in evacuated ampoules to poduce elemental carbon. Finally sample targets for AMS dating were prepared by mixing 2 to 5mg of amorphous carbon obtained with silver power (1:1 mol) and then pressing the mixture into a pellet of 3mm diameter.

ACCELERATOR MASS SPECTROMETRY OF ^{14}C

A Tandetron accelerator mass spectrometer (General Ionex, Model 4310) which has been installed at the Radioisotope Center, Nagoya

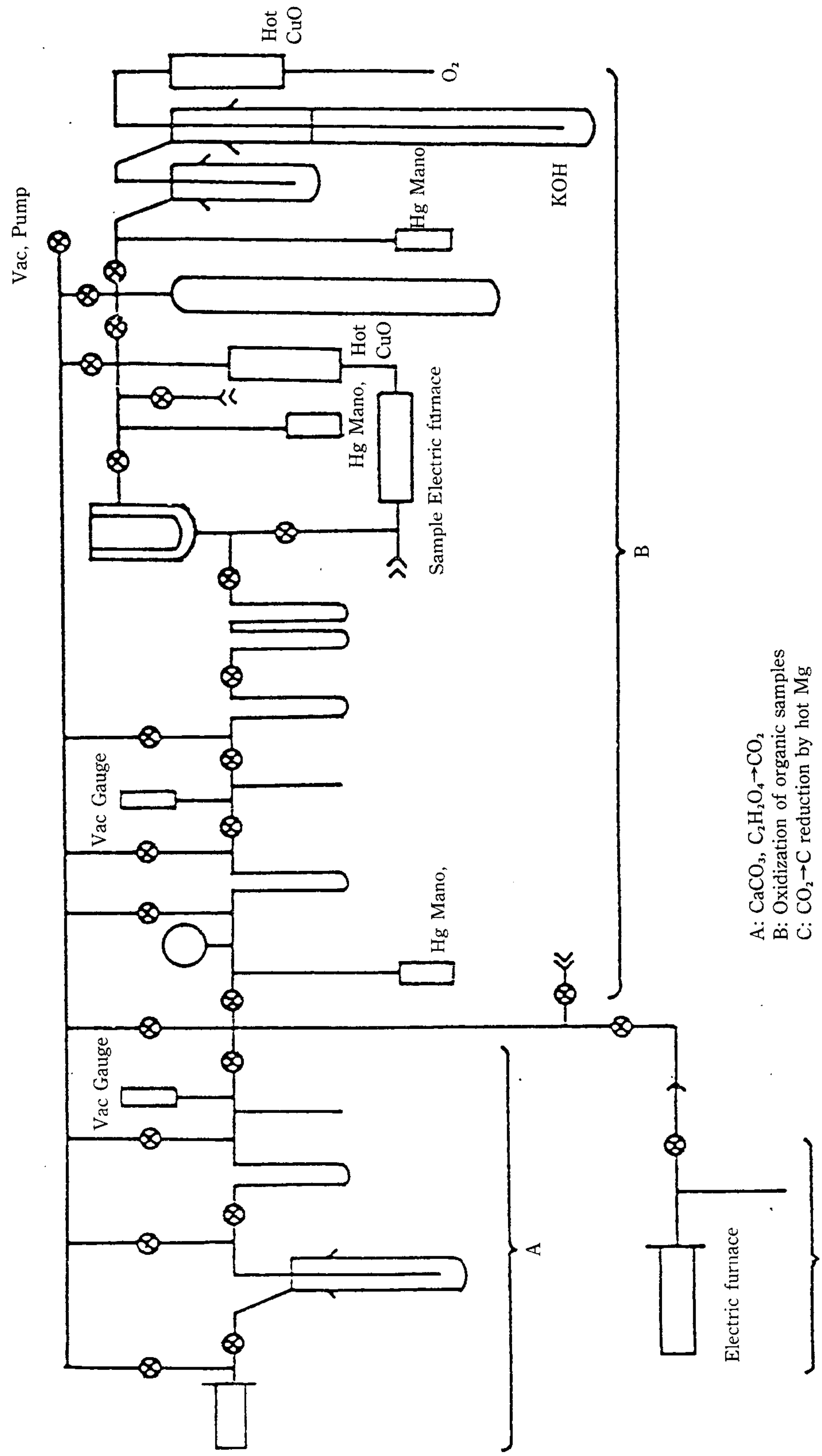


Fig. 3. The whole view of the sample preparation analytical line for Accelerator Mass Spectrometry technique. This line combined of three different stages: removal of organic matters, $CaCO_3$ to CO_2, and reduction of CO_2 stages.

University in Japan since 1982, is to detect and count ^{14}C atoms directly in natural carbon ($^{14}C/^{12}C = 10^{-12--15}$). The principal components and experimental layout of the system are illustrated schematically in Fig. 4. The accelerator is composed of the following major components: (1) a Cs sputter negative ion source to reject ^{14}N which is an isobar of ^{14}C, (2) a 3MV tandem accelerator with Cockcroft-Walton type voltage generator to break up molecular ions of Mass number 14 into atomic ions, (3) an electrostatic deflector and two sets of mass analyzing magnets to select ions correct mass, energy, and charge states, (4) a heavy ion detector system with an energy absorber of 2.54μm thick mylar foil, (5) an silicon surface barrier detector to isolate $^{14}C^{+3}$ from background ions.

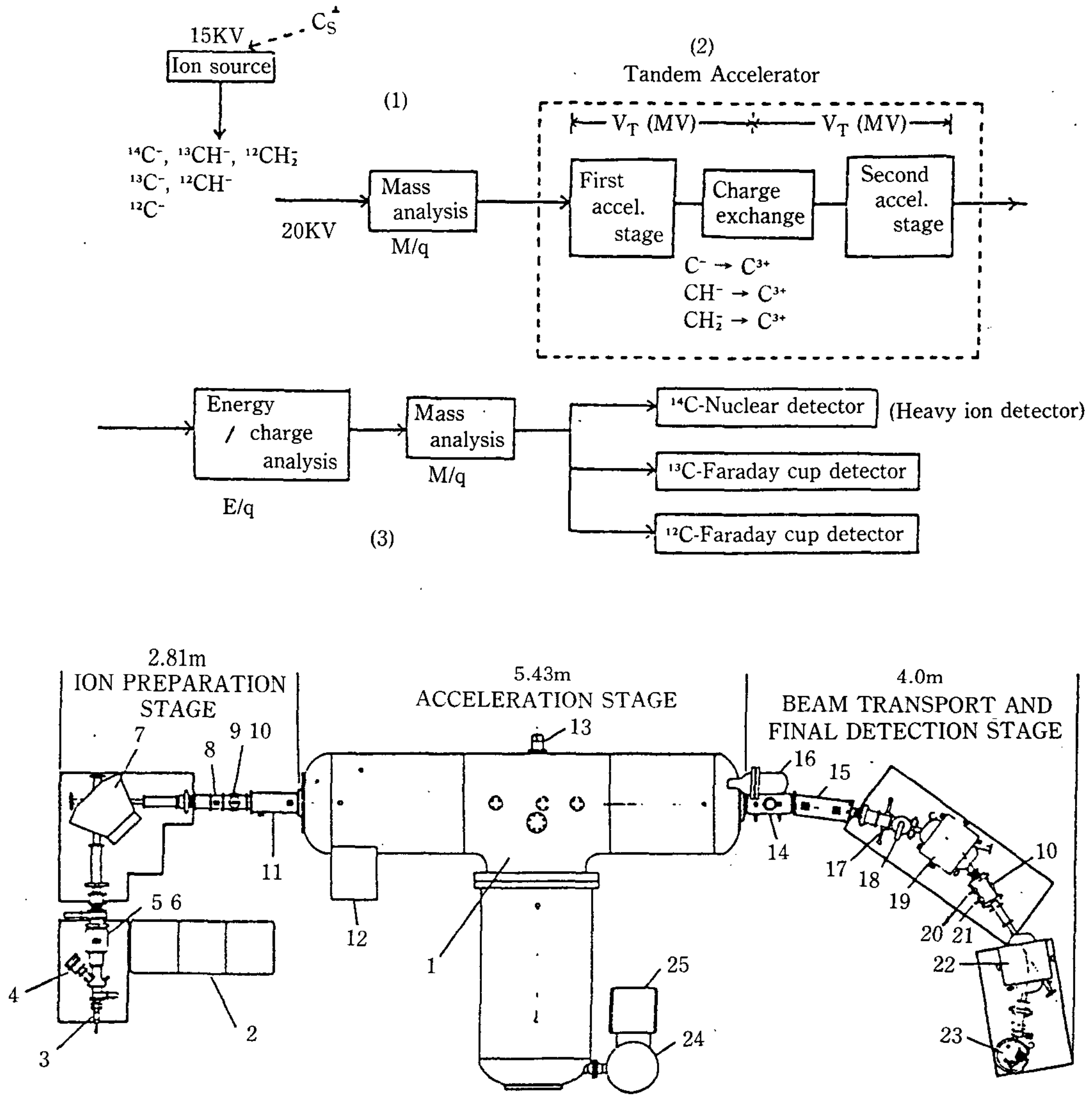
Negative ions are produced from solid target by Cs sputter negative ion source. Six targets can be mounted in the ion source at a time. The positive cesium ions of 12 KeV impinge on the surface of the target at an 45°. Then they sputter the target and produce negative carbon ions together with other atomic or molecular ions. The negative ion of nitrogen, an isobar of ^{14}C, is unstable and not produced. These negative ions are accelerated to an energy of 20KeV, focussed by an Einzel lens, and mass analyzed by high resolution 90° directional double focusing magnet. The beam intensity of the ions selected for mass can be monitored by a Faraday cup. The negative ions are then fed to the accelerator. After the acceleration at the first stage of the Tandem accelerator, negative ions are changed into positive ions by the collision with argon gas at the middle of the accelerator. In the charge exchange process, the molecular ions are decomposed to atomic ions, resulting in that no molecular ions with the same mass number as ^{14}C can survive.

The positive ions are accelerated further in the following second stage and then analyzed according to their values of energy/change ratio by the electrostatic deflector, which selects ions of only $^{+3}$ charge state for the ^{14}C measurement. The ion thus selected are mass-analyzed by the strong focusing magnet: the magnetic field is adjusted to let ^{14}C ions go through the central orbit of the analyzing magnet and $^{12}C^{3+}$ and $^{13}C^{3+}$ ions are focussed on two Faraday cups and their beam currents are measured independently. The ^{14}C ions are further mass-analyzed and directed to a silicon detector through the energy absorber of 2.54μm thick mylar. By the analysis of energy for ions arriving at the detector, the ^{14}C atoms can be identified from background atoms and counted.

The ^{14}C atoms are identified and counted using NIM electronics, together with $^{12}C^{3+}$ and $^{13}C^{3+}$ ion beam currents and these data are transferred to DEC LSI 11/23 computer via CAMAC interfacing. The switching for alternating measurements of mass 12, 13 and 14 ions is performed within a second by applying the relevant voltages to the magnetic chamber.

APPLICATION TO THE LOCAL HISTORY OF THE CHANGE OF SEA LEVEL

It is important to emphasize the lowering sea level during glacial time and the redistribution of sea bottom sediments in the transgressive phase of the post-glacial time. Several works have attempted to described the history of late Quaternary sea level fluctuations on the East China Sea continental shelf and Japan coasts (Emery, 1968; Fujii and Fuji, 1982; Fujii and Naruse, 1982; JAQR, 1987; Oshima, 1982; Wang and Wang, 1980). At the Korean coast, a few study have been done (Bloom and Park, 1985; Jo, 1980; Suk, 1986, 1989a, b; Youn et al., 1977). However, it still disputes the timing, rate of rise, and magnitude of vertical elevation



Names of Parts

1 3MV Tandetron
2 Control rack
3 Sample holder
4 Cs sputter ion source
5 Gridded Einzellens
6 500ℓ/s turbo molecular pump
7 Double focussing 90° magnet
8 X-Y steerer
9 Faraday cup
10 Cryopump
11 Matchig lens/X-Y steerer
12 Tube lens
13 Generating voltmeter
14 Electrostatic quadrupole lens
15 Electrostatic 15° deflector
16 Electric motor for terminal electric generator
17 X-Y slit
18 Faraday cup for total beam current
19 First double focussing 45° magnet
20 Faraday cup for ^{12}C beam current
21 Farday cup for ^{13}C beam current
22 Second double focussing 45° magnet
23 ^{14}C detector
24 Step up transformer for 3MV generator
25 Oscillator for 3MV generator

Fig. 4. General view of principal components (upper) and structure (below) of a Tandem Accelerator Mass Spectrometer installed at the Radioisotope Center in Nagoya University.

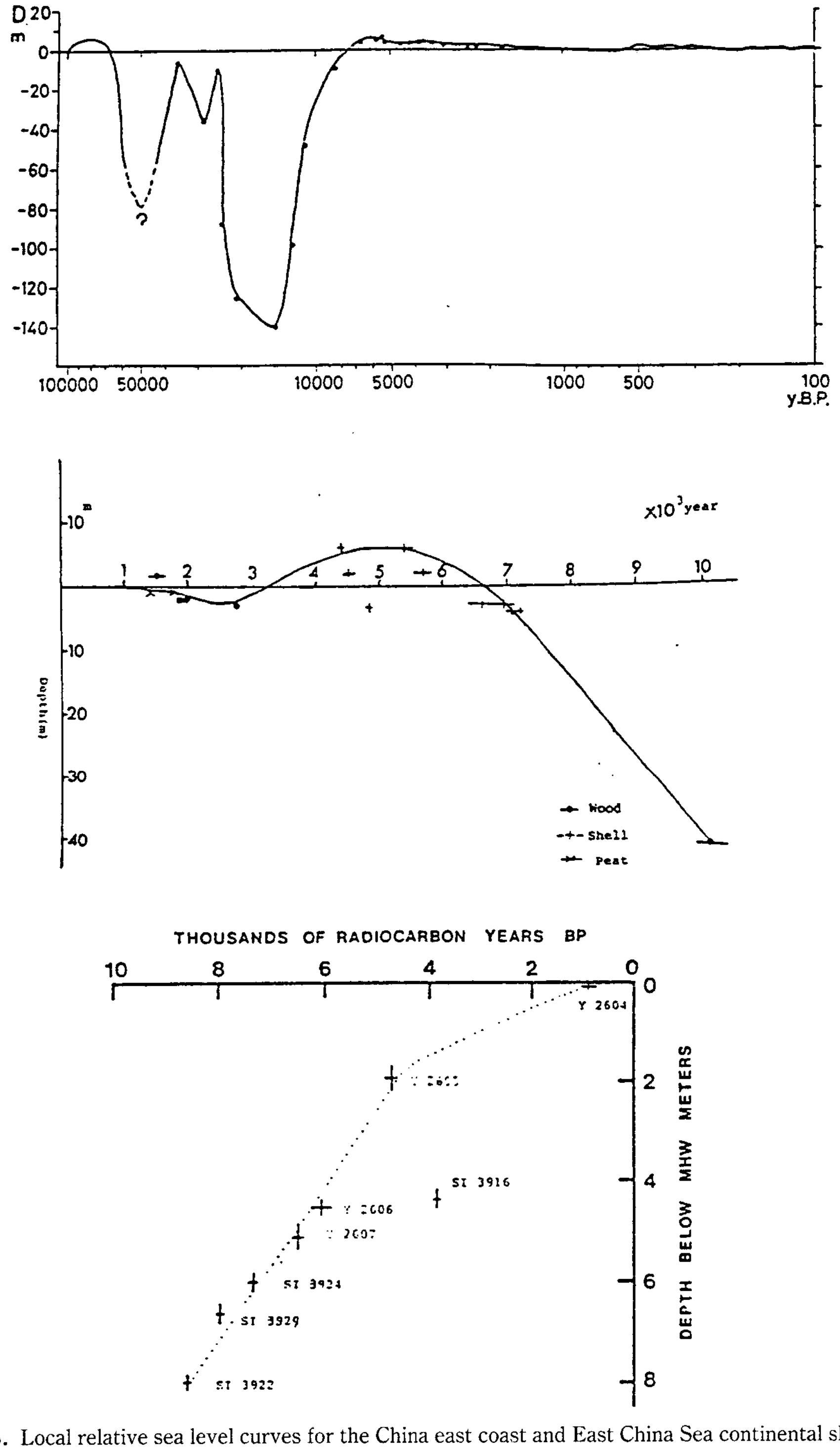


Fig. 5. Local relative sea level curves for the China east coast and East China Sea continental shelf (upper, after Wang and Wang, 1980), Toyama bay in Japan (middle, after Fujii and Fuji, 1982) and west coast of Korea (below, after Bloom and Park, 1985).

change of the sea level at the late Pleistocene and post-glacial time (Fig. 5).

The determined radiocarbon ages of all previous work followed conventional method such as gas proportional counter and liquid scintillation counter. No determination is small amount of samples for this method. The present study have been performed by AMS technique for radiocarbon age dating. The sampling location is shown in Table 1. Twenty-five samples contain mollusk shells from sixteen piston core sampling stations that were grouped into the following environments: brackish, intertidal flat, and shallow sea (Table 2). Water depth covers 36 to 135m, and are compensated by the piston core depth and living depth of mollusks. The ages determined range from 530 to older than 33,500 yrs. Each specimen of mollusca shells to be used age dating is shown in Plates 1 and 2. Nevertheless the care selection of autochthonous mollusca species, there are still contained some allochthonous species indicating complicate hydrodynamic condition for their deposition at that time.

The result showed that sea level fluctuation at Korean Sea is different that of the East China Sea and Japanese coastal area (Suk, 1989b). The sea level fluctuation pattern of the present study area, the southern and southeastern coast of the Korean Peninsula follows Shepard's curve pattern. It can be shown the locally different subsiding phenomena (Suk, 1989b). It inferred from the differences of regional eustasy, geothermal effect and local tectonics in addition to the global eustasy.

Table 1. Sampling locations for the radiocarbon age dating.

Station no.	Latitude (N)	Longitude (E)	Water depth (m)	Core length (cm)
KS-K01	33°25'25	126°57'56	95	90
KS-02	33°27'45	127°00'30	115	180
KS-03	33°32'15	127°03'45	135	172
KS-04	33°37'30	127°07'30	127	55
KS-05	33°41'10	127°11'20	102	160
KS-06	33°45'30	127°15'00	94	100
KS-07	33°50'10	127°18'45	88	160
KS-08	33°54'40	127°22'40	82	250
KS-18	33°50'40	127°08'20	90	60
KS-21	34°06'00	127°14'30	60	110
KST-03	34°36'38	128°51'30	35	180
KST-05	34°59'40	128°57'30	36	600
KST-07	35°02'50	129°09'50	81	74
KST-08	35°40'00	129°08'40	48	300
KST-09	35°05'50	129°11'10	63	290
KST-11	34°50'00	128°56'00	80	130
KST-16	35°08'20	129°30'00	110	40
KST-17	35°11'45	129°23'15	100	130
KEU-09	35°28'07	129°39'56	125	120

Table 2. Radio-carbon dated samples at Korean Sea.

St. No.	Water depth (m)	Sample depth (cm)	Species	Depth range (m)	Dated sea level (m)	C-14 age (yr)	Lab. No.
KS-K01	95	60-63	*Pitar sulfureum*	inter-tidal flat	95	7260±130	NUTA-530
KS-2	115	35-38	*Barbatia decussata*	ditto	115	9980±130	NUTA-536
KS-3	135	175-178	*Clinocardium buellowi*	10-20	122	10720±130	NUTA-534
			Barbatia desussata	inter-tidal flat	136	10430±130	NUTA-543
			Polamocorbula amurensis	brackish	136	19800±190	NUTA-544
KS-4	127	46-53	*Tellinella staturella*	inter-tidal flat	127	620± 95	NUTA-535
KS-5	102	85-90	*Modiolus difficilis*	0-10	93-103 (98)	11210±140	NUTA-541
		155-160	*Pitar sulfureum*	inter-tidal flat	103	12690±120	NUTA-540
KS-6	94	85-93	*Pitar sulfureum*	ditto	95	9450±170	NUTA-542
KS-7	88	150-157	*Anisocorbula venusta*	0-20	70- 90 (80)	10370±150	NUTR-537
KS-8	82	160-165	*Pitar sulfureum*	inter-tidal flat	83	7390±150	NUTA-538
KS-18	90	50-60	*Glycymeris albolineata*	5-20	70- 85 (75)	27020±240	NUTA-539
KS-21	60	35-38	*Glycymeris albolineata*	ditto	40- 55 (50)	30090±320	NUTA-533
		90-97	*Pitar sulfureum*	inter-tidal flat	60	5280±100	NUTA-531
KST-5	36	31	*Angulus vestalioides*	5-	0- 31 (10)	780± 95	NUTA-529
		337	*Pitar japonicum*	shallow water	10	700±125	NUTA-528
		542-545	lingnite		41	775± 85	NUTA-527
KST-7	81		*Arca arabica*	inter-tidal flat	81	9640±140	NUTA-575
			Acila divaricate	ditto	81	530±100	NUTA-550
KST-8	48	60	*Siphonalia trochulus*	20-50	0- 28 (13)	2240±140	NUTA-526
		116	*Mactrinula dolabrata*	30-	<20	5050±100	NUTA-525
		160	*Rapana thomasiana*	0-20	29-49	3450±130	NUTA-574
		203	*Pecten albicans*	10-30	20-40	2950±100	NUTA-522
KST-9	63	20	*Lucinoma annulata*	10-50	13-53	4100± 90	NUTA-521
KST-11	80	27	*Pitar sulfureum*	inter-tidal flat	80	>33500	NUTA-520
KST-16	110		*Balanus rostratus*	0-200	50 (?)	30400±500	TUGP-778
KST-17	100		*Rapana thomasiana*	0-20	80-100 (90)	10600± 90	TUGP-779
KEU-9	125	55-60	*Arca sp.*	shallow water	95	7240±110	NUTA-519

CONCLUSION

^{14}C age dating of a total of 26 specimens taken from the south and southeastern Korean sea was measured by AMS (accelerator mass spectrometry) technique. The detail of sample pre-treatment and analytical procedures by a self-made analytical line were described. By the AMS technique, dated age ranges from 520±100 years to older than 33500 years.

The sedimentary facies of the study area represents a different environmental set which is affected by sea level fluctuation since the late Pleistocene.

The sea level fluctuation pattern of the present study area, the southern and southeastern coast of the Korean Peninsula follows Shepard's curve pattern. It can be shown the locally different subsiding phenomena.

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REFERENCES

Bloom, A.L. and Y.A. Park, 1983. Holocene sea-level history and tectonic movements, Republic of Korea. The Quaternary Research, **24:** 77-84.

Emery, K.O., 1969. The continental shelves. Scientific America, **221:** 106-122.

Emery, K.O., H. Niino, B. Sullivan, 1971. Post-Pleistocene levels of the East China Sea. In: Late Cenozoic glacial ages, K.K. Turekian editor, Yale University Press, pp. 381-390.

Fujii, S. and N. Fuji, 1982. Postglacial sea-level changes in the Hokuriku region, Central Japan. The Quaternary Research, **21:** 183-193.

Fujii, S. and Y. Naruse, 1982. Sea level changes since the late Pleistocene in the Japanese Islands and related problems. The Quaternary Research, **21:** 283-287.

Geng, X., M. Fu, P. Li and X. Xiu, 1987. Quaterary sea-level changes and shoreline migrations in the People's Republic of China. In: Inventory of Quaternary Shorelines, T. YOSHIKAWA, editor, Nodai Research Institute, Tokyo Univ. Agriculture, Tokyo, pp. 94-101.

JAQR, 1987. Quaternary maps of Japan II Prehistoric Remains and Paleogeography, 1:4,000,000. Japan Association for Quaternary Research, Univ. of Tokyo Press.

Jo, W., 1980. Holocene sea level changes on the East coast of Korean Peninsula. Geographical Review Japan, **53**(5): 317-328.

Mook, W.G., 1984. Archaeological and geological interest in applying ^{14}C AMS to small samples. Nuclear Instruments and Methods in Physics Research, B5, 297-302.

Nakai, N. and T. Nakamura, 1984. Dating and measurements of radioactive isotopes with the Accelerator Mass Spectrometry. Radioisotopes, **32:** 645-655.

Nakamura, T., N. Nakai, S. Ohishi, S. Onoda and S. Tsubota, 1987. ^{14}C measurements with Accelerator Mass Spectrometry-Performance of the spectrometer and some applications to the earth sciences-, Application of Ion Beams in Materials Science, the 12th International Symposium of Hosei Univ., Tokyo, pp. 6.

Oshima, K., 1982. Lowest sea-level in the last glaciation of the Japanese Islands. The Quaternary Research, **21:** 211-222.

Park, Y.A. and M.Y. Song, 1971. Sediment of the continental shelf off the south coast of Korea. Journal Oceanolo. Soc. Korea, **6:** 16-25.

Suk, B.C., 1981. Depositional environment of Recent sediment on the continental shelf around the Jeju Island, Bulletin of Korea Ocean Research and Development Institute, **3:** 123-131.

Suk, B.C., 1986. Depositional environment of late Quaternary sediment and suspended particulate matter on the southeastern continental shelf, Korea. Journal of the Geological Society of Korea, **22:** 10-20.

Suk, B.C., 1989a. Sedimentology and history of sea level changes in the East China Sea and adjacent seas. In: A. TAIRA and H. MASUDA (editors), Sedimentary facies in the active continental margin, TERRAPUB. Co., 215-231.

Suk, B.C., 1989b. Quaternary sedimentation processes, structures, and sea level changes history in the East China Sea, the Yellow Sea and the Korea-Tsushima Strait regions, Dr. Sci. Thesis, University of Tokyo, Japan, 246pp.

Youn, O.K., B.K. Park and S.J. Han, 1977. Geomorphological evidences of post-glacial sea-level changes in Korea. Journal of the Geological Society Korea, **13:** 15-22.

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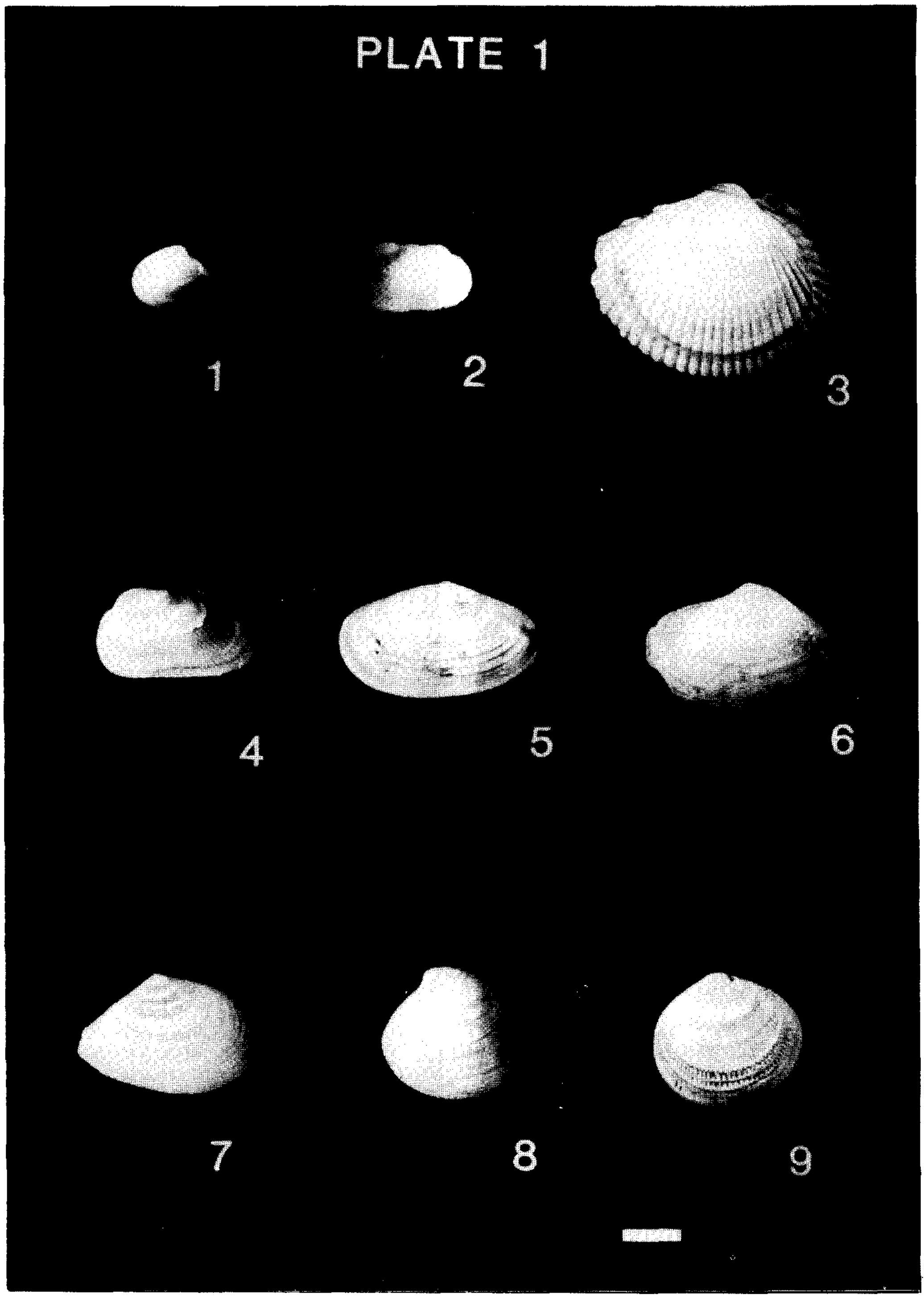
PLATE 1

Mollusca classification, living depth and radiocarbon age dates.
(Scale bar = 1cm)

Fig. 1. *Pitar sulfureum* Pilsbry: Sample KS-K01, intertidal flat, 7260±130yr.
Fig. 2. *Barbatia desussata* Sowerby: Sample KS-3, intertidal flat, 10430±130yr.
Fig. 3. *Clinocardium buellowi* Rolle: Sample KS-3, 10-20m, 10720±130yr.
Fig. 4. *Barbatia decussata* Sowerby: Sample KS-2, intertidal flat, 9980±130yr.
Fig. 5. *Angulus vestalioides* Yokoyama: Sample KST-5, >5m, 780±95yr.
Fig. 6. *Mactrinula dolabrata* Reeve: Sample KST-8, >30m, 5050±100yr.
Fig. 7. *Tellinella staturella* Ramarck: Sample KS-4, intertidal flat, 620±95yr.
Fig. 8. *Pitar sulfureum* Pilsbry: Sample KS-5, intertidal flat, 12690±120yr.
Fig. 9. *Arca sp.*: Sample KEU-9, shallow water, 7240±110yr.

PLATE 2

Mollusca classification, living depth and radiocarbon age dates.
(Scale bar = 1cm)
Fig. 1. *Glycymeris albolineata* Dunker: Sample KS-21, 5-20m, 30090±320yr.
Fig. 2. *Pitar sulfureum* Pilsbry: Sample KS-21, intertidal flat, 5280±100yr.
Fig. 3. *Pitar sulfureum* Pilsbry: Sample KS-8, inter-tidal flat, 7390±150yr.
Fig. 4. *Pitar japonicum* Kuroda et Kawamoto: Sample KST-5, shallow water, 700±125yr
Fig. 5. *Rapana thomasiana* Crosse: Sample KST-17, 0-20m, 10600±90yr.
Fig. 6. *Pecten albicans* Schroter: Sample KST-8, 10-30m, 2950±100yr.
Fig. 7. *Balanus rostratus* Hoek: Sample KST-16, 0-200m, 30400±500yr.
Fig. 8. *Lucinoma annulata* Reeve: Sample KST-9, 10-50, 4100±90yr.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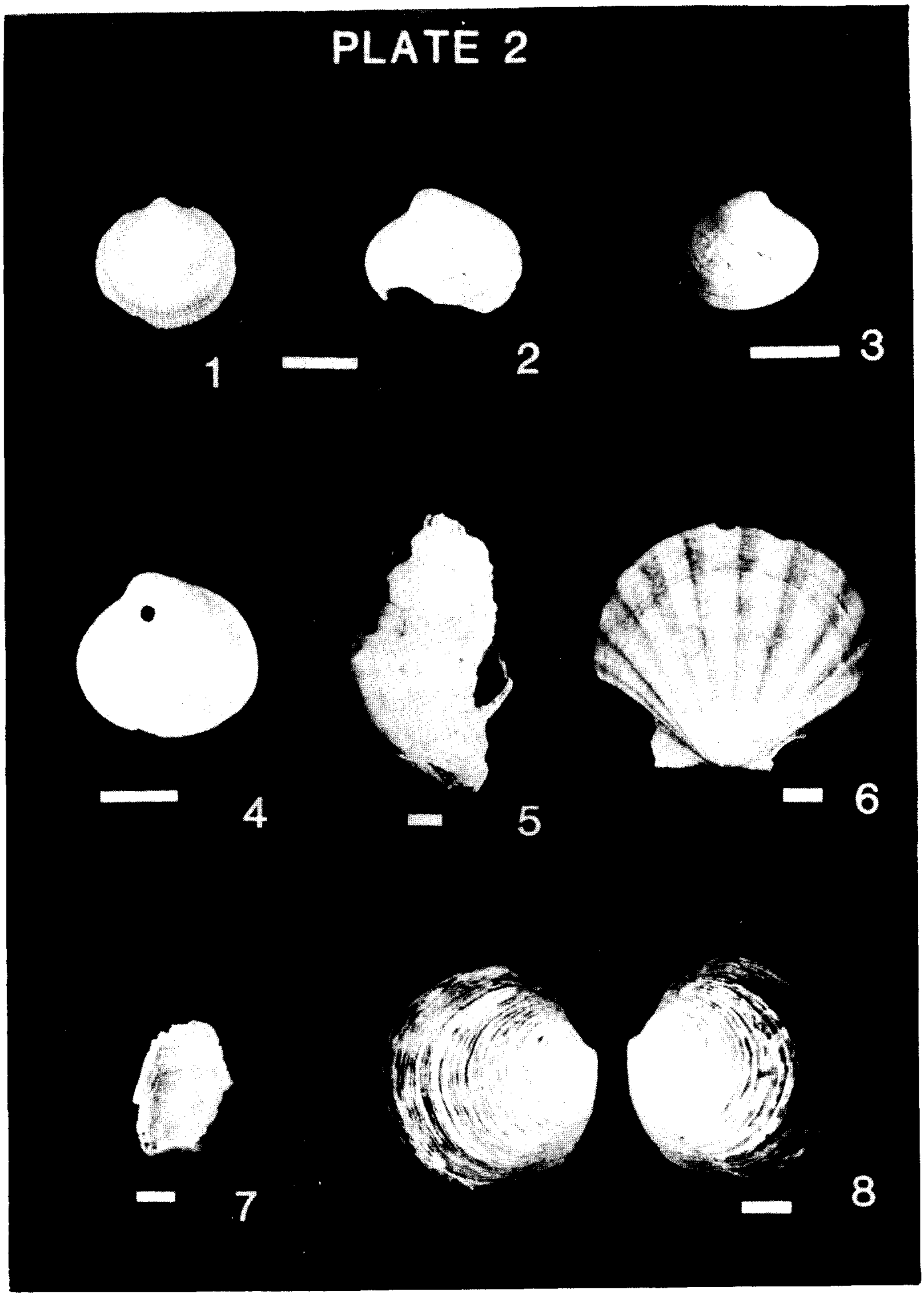
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