Reports

Replication in Plastic of Three-Dimensional Fossils Preserved in Indurated Clastic Sedimentary Rocks

Abstract. *A new technique for replicating in plastic the fossils preserved in clastic rocks should now make available reliable morphologic and frequency data, comparable in quality to those derived from acid-prepared silicified faunas, for a major segment of the fossil record. The technique involves three steps: the dissolution of carbonate in fossiliferous rocks with hydrochloric acid, impregnation of resulting voids with liquid plastic, and dissolution of the rock matrix with hydrofluoric acid, leaving a concentrate of plastic-replaced fossils.*

Because it has been virtually impossible to use disaggregation and screening methods to recover three-dimensional fossils from well-indurated clastic sedimentary rocks, important morphological and statistical data on faunas from such facies have been unavailable to paleontologists.

We have developed a technique for extracting fossils from lithified clastics in a way analogous to that used for recovery of silicified fossils preserved in limestones. Silica is the natural replacing agent in limestones, and the rock matrix is dissolved with hydrochloric or acetic acid during preparation (*1*); in our method, fossils are artificially replaced with plastic, and the rock matrix dissolved with hydrofluoric acid. Our technique can be used for all three-dimensional fossils (*2*) that are acid-soluble and preserved in acid-resistant (not to HF) rock matrices as well as those that are preserved as natural molds. For description, we divide our technique into three stages: etching, impregnation, and matrix dissolution.

Etching. We immersed the HCl-resistant rock sample containing calcareous fossils or natural molds in a 25 percent HCl solution (Fig. 1, a and b) (*3–5*). Carbonates, if present, effervesced for as long as several days, depending on their volume in the sample. When effervescence stopped, the sample, still immersed in HCl, was placed in a vacuum chamber at 60 torr for 30 minutes (Fig. 1c). This was repeated three times a day until effervescence could no longer be reinitiated under vacuum (*6*). It was then assumed that all the carbonates had been dissolved. The HCl was decanted and the sample rinsed in slowly running water for 10 hours (Fig. 1d). The sample was next placed in an acetone bath under vacuum at 60 torr for 1 hour (Fig. 1e) and then allowed to dry at room temperature. To ensure thorough removal of moisture, the sample was dried in a microwave oven at 500 watts for 30 minutes or in a conventional oven at 130°C for 12 hours (Fig. 1f) (*7*).

Impregnation. Once thoroughly dried, the sample was impregnated with liquid plastic. For good results, the plastic must have a low viscosity, a slow rate of polymerization, and must be resistant to HF (*8*). We used ESCON CR64 diluted with 10 percent (by weight) methyl methacrylate monomer (acrylic monomer) or styrene (*9*); 0.25 percent cumene hydroperoxide and 0.05 percent methyl-ethyl ketone peroxide (by weight) were added together as catalysts. The appropriate viscosity was achieved by heating the plastic-catalyst mixture to 30°C. The container was then filled with the warmed, liquid plastic so that the sample was covered (*10*). The container and contents were then placed in vacuum at 60 torr for 10 minutes, returned to atmospheric pressure for several minutes, and then subjected to vacuum again (Fig. 1g); this procedure was repeated until bubbles were no longer exuded from the sample under vacuum. The container and contents were then placed in a fumehood for polymerization of the plastic, which normally required about 2 weeks (*11*). When the plastic had polymerized, final stability was achieved by heating in a conventional oven for 12 hours at 80°C (Fig. 1h). When the plastic block had cooled to room temperature, we removed the container and cut through the embedded rock with a diamond saw to expose a large surface (Fig. 1i) (*12*).

Matrix dissolution. The cut block was immersed in 40 percent HF, which attacked the rock matrix but not the plastic-replaced fossils (*13, 14*). The plastic replicas were gradually freed from the rock matrix by the acid, and they settled to the bottom of the container (Fig. 1j). They were sorted by gently washing

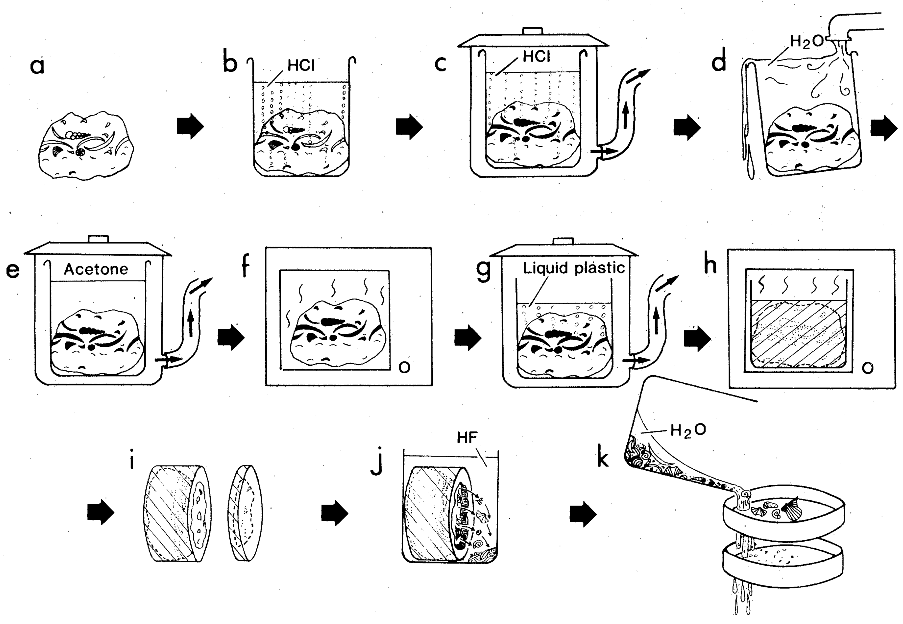

Fig. 1. Flow chart showing plastic replication technique: (a to f) etching stage; (g to i) impregnation stage; and (j and k) matrix dissolution stage.

them through a coarse and a fine screen (Fig. 1k) and allowed to air dry on the screens (*15*). Once dry, the fractions were tipped into trays for study under a binocular microscope. The replicas are, in general, less fragile than comparable silicified specimens.

We have tested our technique on a variety of fossiliferous rock samples and find it to be applicable for a broad range of fine-grained HCl-resistant clastic sedimentary rocks containing calcareous fossils or natural molds (Table 1). We emphasize that the surficial detail recorded in the plastic replicas is precise in all particulars (Fig. 2). For example, the replica of the crinoid ossicle (Fig. 2J) faithfully documents the honeycomb-like microstructure characteristic of the echinoderm skeleton, and the structurally complex and delicate fenestellid bryozoan pillar (Fig. 2H) has been reproduced in plastic with exactness. We have applied our technique to fossils of a wide range of sizes and affinities, from invertebrate microfossils such as ostracods (Fig. 2, F and K) to vertebrate macrofossils such as the dermal armor of primitive fishes (Fig. 2E), with a comparable degree of fidelity.

Our plastic-replication method has several major advantages over conventional methods of preparation. Preparation of fossils preserved in indurated clastic lithologies normally involves either the removal of the rock matrix from specimens by mechanical means or dissolution of exposed fossils with acid and casting of the resulting mold with latex rubber (fossils are sometimes dissolved during diagenesis leaving natural molds). The difficulty with mechanical methods

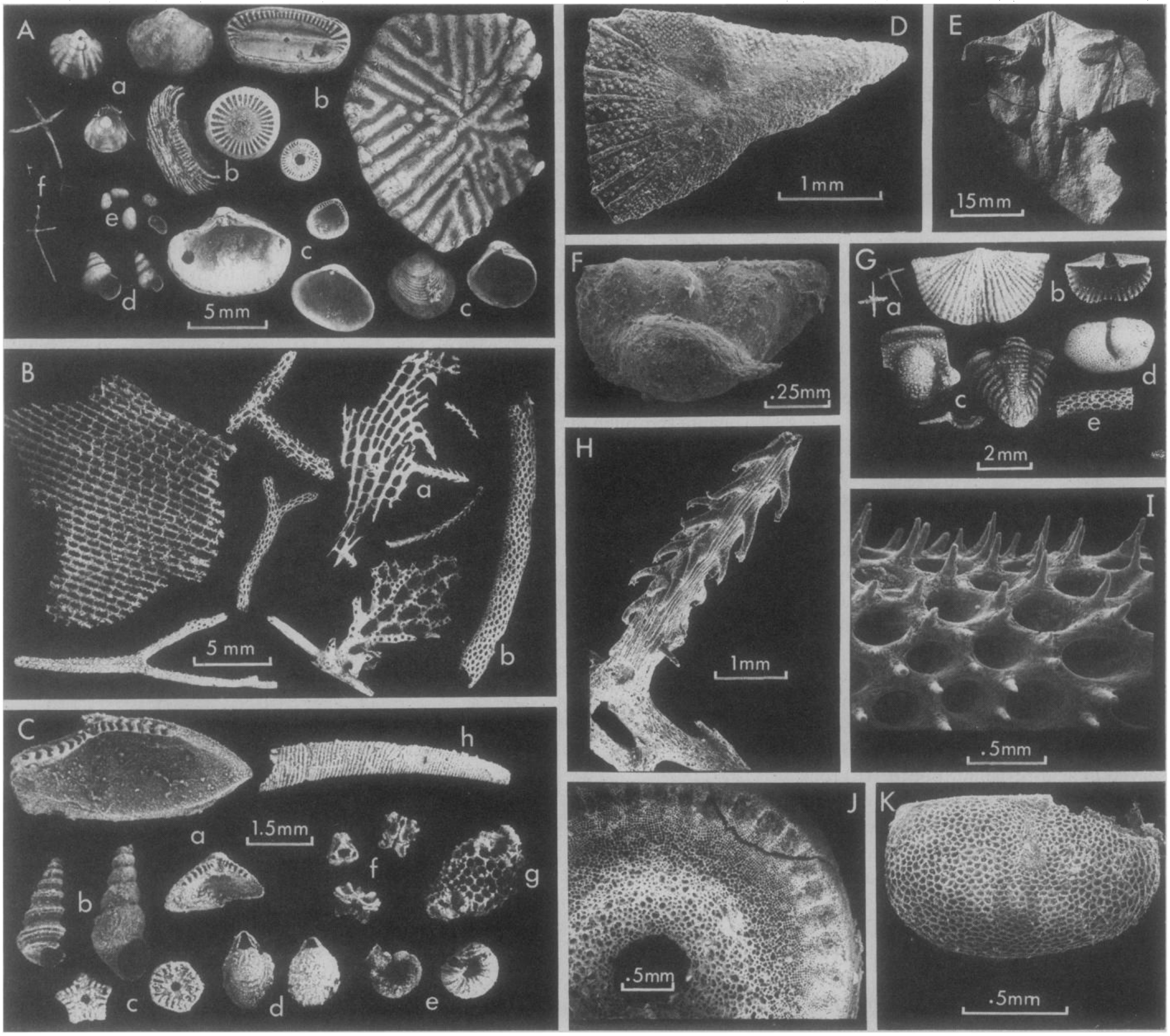

Fig. 2. Plastic-replaced fossils. Geologic unit and age for the fossils are given in Table 1. (A) (a) Brachiopods (note the cluster of sponge spicules around the pedicle foramen of the lowermost brachiopod; the spicules were probably originally held there by pedicle threads), (b) crinoid ossicles and plates, (c) bivalves, (d) gastropods, (e) ostracods, and (f) sponge spicules; (B) bryozoans; (C) (a) nuculoid bivalves, (b) gastropods, (c) crinoid ossicles, (d) terebratulid brachiopods, (e) spirorbid tubes, (f) fish vertebrae, (g) favositid coral, and (h) tube of uncertain affinities; (D) parablastoid plate; (E) ventral view of the posterodorsal bones of an arthrodire cranium showing the articulatory sockets (top) for the thoracic processes; (F) ostracod; (G) (a) sponge spicules, (b) brachiopods, (c) trilobite elements, (d) ostracod, and (e) bryozoan; (H) enlarged view of the fenestellid bryozoan pillar shown in (a) in (B); (I) enlarged view of a bryozoan of the type shown in (b) in (B); (J) crinoid ossicle showing microstructure of test; and (K) ostracod. (D, F, and H to K) Scanning electron micrographs. Specimens in (A to C and E and G) were whitened with ammonium chloride.

Table 1. Data for Australian rock units sampled by the plastic-replication technique.

Rock unit	Geologic age	Lithology	Main fossils	Mode of preservation	Figure
Ulladulla Mudstone	Early Permian	Siltstone; marine	Brachiopods, bivalves, bryozoans, crinoids	Natural molds	2, A, B, and H to K
		Black shale; marine	Brachiopods, bryozoans	Calcareous remains	
Wasp Head Formation	Early Permian	Lithic sandstone with calcareous cement; marine	Bivalves, rostroconchs,	Natural molds	
Clogham Shale	Late Devonian	Red siltstone and shale (red beds); nonmarine	Placoderm fishes	Calcareous remains	2E
Hunter Siltstone	Late Devonian	Siltstone; nonmarine	Placoderm fishes	Calcareous remains	
Cavan Bluff Formation	Early Devonian	Calcareous siltstone; marine	Bivalves, gastropods, crinoids, brachiopods, fish	Calcareous remains and natural molds	2C
Walker Volcanics	Late Silurian	Mudstone; marine	Bivalves, rostroconchs, parablastoids, ostracods	Calcareous remains and natural molds	2, D and F
		Siltstone; marine	Trilobites, brachiopods	Natural molds	2G

is that minute fossils or those with complex surfaces or delicate projections are almost impossible to prepare satisfactorily if the rock matrix is well cemented. Acid dissolution and latex casting are sometimes successful, but deep, narrow recesses on the surfaces of molds, such as those produced by elongate spines, resist latex penetration, and latex casts often tear when pulled from complex surfaces.

Mechanical methods and latex casting fail to provide reliable frequency data because they can be applied only to fossils that are visible on rock surfaces. Small organisms tend to be exposed on these surfaces less frequently than large organisms of comparable abundance; rare organisms, especially those of small size, are not likely to be exposed at all. Consequently, such organisms tend to be underrepresented or omitted from faunal lists and paleoecological analyses. The magnitude of these difficulties has been noted by Talent (*16*) who points out that faunas sampled by disaggregation and screening methods show a diversity 5 to 10 times greater than those sampled by splitting indurated lithologies with a hammer. The effect of this bias is magnified by the fact that clastics, the vast majority of which are too well indurated for disaggregation and screening methods, comprise about 80 percent of measured sedimentary rocks (*17*).

The significance of our method is that (i) all three-dimensional fossils enclosed in a given sample of rock are replaced and completely liberated from the rock matrix allowing their numbers to be calculated without the biases induced by conventional procedures; (ii) rare organisms, normally underrepresented or omitted from collections, are more likely to be recovered; and (iii) complex and delicate structures normally inaccessible for taxonomic analyses, can be recovered intact. Consequently, morphological and frequency data comparable in quality to those derived from acid-prepared silicified faunas can now be obtained from a large segment of the fossil record for which such data were not hitherto available.

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References and Notes

1. G. A. Cooper and H. B. Wittington, in *Handbook of Paleontological Techniques*, B. Kummel and D. Raup, Eds. (Freeman, London, 1965), p. 294.
2. It cannot be used with flattened fossils including most graptolites, natural molds that have collapsed during diagenesis, compressed plant megafossils, and so on.
3. Blocks of rock up to 15 by 15 by 8 cm gave the best results. Rocks that became soft or friable in HCl were suspended in nylon or plastic netting during etching and impregnation.
4. Theoretically, the type of acid could be varied, depending on the composition of the fossils, so long as the matrix is not dissolved. For example, thioglycolic acid might be used for fossils that have been replaced by ironstone. We have only used HCl, however.
5. We recommend that samples containing natural molds be subjected to the etching stage to ensure that any acid-soluble material adhering to the surface of the molds will be dissolved. If, however, the matrix cement of such samples is calcitic [for example, Wasp Head Formation (Table 1)], then proceed directly to the impregnation stage.
6. Renewed bubbling under vacuum represents the release of CO_2 bubbles that have been trapped in the sample. Presumably the CO_2 bubbles that form from the reaction of HCl and carbonate accumulate in microcracks and small cavities where they are held in place by surface tension. This impedes further advance of the HCl through these structures, and dissolution of carbonate ceases. When subjected to vacuum, the trapped CO_2 bubbles expand, become more bouyant, and are released from the sample, allowing further acid penetration and renewed carbonate dissolution. Through the repeated use of vacuum, we were able to get complete penetration of HCl to the core of roughly equidimensional dense blocks of mudstone and siltstone up to 3500 cm^3. To ensure complete penetration, one or two small holes can be drilled to the core of the sample before immersion in HCl.
7. Moisture in the sample must be completely removed so that polymerization of the plastic is not inhibited. We dried 31 samples in microwave ovens, and a small part of one nonmarine mudstone exploded forcibly; other samples of the same lithology did not. We subsequently wrapped samples in cloth to inhibit emission of rock fragments in the event of another explosion.
8. Slow polymerization provides time for penetration of very small spaces by capillary action. We obtained poor results with plastics that polymerized quickly.
9. ESCON CR64 is a preaccelerated, high clarity, low exotherm and low shrinkage polyester (Hatrick Chemicals, Botany, N.S.W., Australia).
10. The liquid plastic should be poured slowly to ensure upward displacement of air and to minimize trapping air pockets. The sample should be covered with 3 to 5 cm of liquid plastic since the level will drop through absorption by the sample and through evaporation of the acrylic monomer.
11. If contact with air keeps the top few millimeters of plastic from hardening, it can be placed under a sunlamp.
12. Cut parallel to bedding so that fossils do not etch out as ledges during matrix dissolution and break under their own weight.
13. The time required for complete dissolution varies with rock size and composition. A 500-g block of siltstone, for example, required about 5 days to dissolve. Usually acid must be replaced once or twice.
14. When subjected to HF, lithologies with a high organic content, such as Ulladulla Mudstone (Table 1), produced a black, tarlike substance that coated exposed surfaces, impeding the action of the HF on the rock matrix and obscuring details on the plastic fossil replicas. A light rinse of acetone dissolved the tar immediately. Prolonged soaking of the plastic in acetone should be avoided since this might cause warping. Calcium fluoride accumulated on the surface of some rock samples, such as Hunter Siltstone (Table 1), and impeded the action of the HF, but it was dissolved in warm HCl (about 30°C). Calcium fluoride probably accumulated because of the presence in some samples of small grains of calcite that were completely sealed off by noncalcareous phenoclasts and cement and hence were not dissolved by the HCl.
15. Pour the wet concentrate over a large area of the screens. If heaped, the concentrate tends to adhere together as it dries.
16. J. A. Talent, *Tr. Inst. Geol. Geofiz. Akad. Nauk SSR Sib. Otd.*, in press.
17. P. D. Krynine, *J. Geol.* **56**, 156 (1948).
18. We thank K. S. W. Campbell, R. C. Fox, and J. A. Talent for helpful comments, J. Cent for technical advice, especially concerning plastic impregnation, and R. Webster for taking some of the scanning-electron microscope photographs (Fig. 2, F, and I to K).

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