OXIDATION OF 19β,28-EPOXY-18α-OLEANAN-3-ONE AND -1-ONE WITH PERACIDS*

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The reaction of 3-oxo derivative *I* and 1-oxo derivative *XII* of 19β,28-epoxy-18α-oleanane with peracids leads to corresponding lactones *II* or *XIII*, respectively, which were converted to A-seco acids *III*, *V*, and *XIV* and esters *IV*, *VI*, *VII*, and *XV*. Under acid catalysis oxidation of ketone *I* results in the degradation of one methyl group in position 4 and formation of compounds *VIII* and *IX*. On reaction with 3-chloroperbenzoic acid the α,β-unsaturated ketone *XVI* gives epoxyketone *XVII*.

Some triterpenoid A-seco acids which contain a further oxygen-containing functional group or a double bond in the vicinity of the carboxyl group, display antibacterial activity (see ref.[1,2]). They are formed by second order Beckmann rearrangement of ketoximes and subsequent hydrolysis of the unsaturated nitriles formed[1,3]. A further possible way for the preparation of triterpenoid A-seco acids and their derivatives is the Baeyer–Villiger oxidation of ketones with peracids; this reaction was used in the case of some triterpenoid and 4,4-dimethylsteroid 3-ketones, leading to lactones of 4-hydroxy-3,4-seco-3-oic acids[4–6]. In this paper we discuss the reaction of oxo derivatives *I*, *XII* and *XVI* (derived from 19β,28-epoxy-18α-oleanane) with peracids and the conversion of the products formed to seco acids. The preparation of the starting ketones *I*, *XII*, and *XVI* is described in literature[7,8].

Oxidation of 3-oxo derivative *I* with 3-chloroperbenzoic acid in dichloromethane gave lactone *II* in a 80–86% yield. Lactone *II* is also formed in good yield (81–85%) when peracetic or performic acids are used. In the case of performic acid the reaction is carried out in a two-phase system (chloroform–formic acid and 30% hydrogen peroxide) and the oxidation reagent must be exchanged several times in order to bring the reaction to completion.

Hydroxy acid *III* was prepared by alkaline hydrolysis of lactone *II*. The acid was converted to methyl ester *IV* with diazomethane. The same methyl ester was also obtained in 78% yield directly from lactone *II* on reaction with methanol in dichloromethane, under catalysis with a small amount of sulfuric acid (0·03%). When a larger

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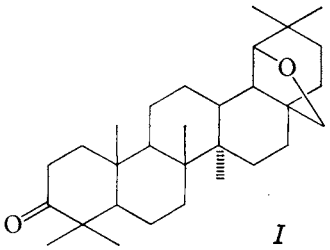
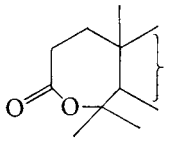
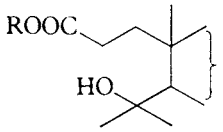
amount of sulfuric acid is used (1%) elimination takes place simultaneously and the unsaturated methyl ester *VI* is formed, identical with the ester described in ref.[3]. Similarly, unsaturated ethyl ester *VII* was obtained from lactone *II* under the effect of ethanol or ethyl acetate and sulfuric acid. Methyl ester *VI* was also prepared on dehydration of hydroxy ester *IV* with phosphorus oxychloride in pyridine or



I



II

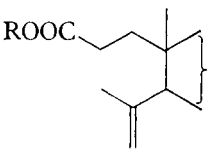


III, R = H
IV, R = CH_3

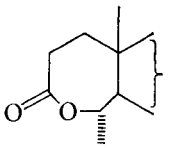
on heating lactone *II* at 250°C and subsequent reaction of the pyrolysate with diazomethane. A similar pyrolysis of a lactone has been described in the 4,4-dimethyl-5α-cholestane series[4]. When reacting lactone *II* with sulphuric acid in a mixture of dichloromethane (or acetone) and ethylene glycol, the known[3] acid *V* is obtained. The conversion of lactone *II* to unsaturated acid *V* and its esters *VI* and *VII* is practically quantitative and the yields after crystallization range between 83 and 95%. For the preparation of these compounds crude lactone *II* may be used, obtained by oxidation of ketone *I*. This procedure is a suitable method for the preparation of unsaturated 3,4-secotriterpenoids in cases when no further functional group is present in the molecule of the starting ketone, which could react with peracids.

If the oxidation of ketone *I* with 3-chloroperbenzoic acid is carried out in the presence of acid catalysts, one carbon atom splits off. Oxidation in a mixture of dichloromethane, acetic acid and sulphuric acid gives rise to the known[2] 24-norlactone *VIII* (73%). Oxidation in dichloromethane in the presence of boron trifluoride etherate gave 4-formyloxy-24-noracid *IX* (37%) which was characterized as methyl ester *X*. Since methyl ester *X* gives on partial hydrolysis on alumina 4-hydroxy-24-nor-methyl ester *XI*, identical with the ester for which configuration 4*S* has been derived[2], the configuration of acid *IX* and ester *X* on $C_{(4)}$ must also be *S*. Methyl ester of 4-formyloxy-24-noracid *X* is also described by Hase[9], but the melting point and optical rotation differ considerably from our values. These differences can be caused by a different configuration on $C_{(4)}$. The loss of one methyl group from position 4 during the oxidation of 3-ketones with peracids under similar conditions has already been observed[5,9].

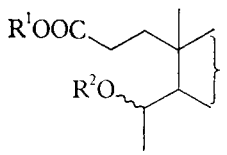
Oxidation of 1-oxo derivative *XII* with 3-chloroperbenzoic acid in chloroform gives lactone *XIII*. Under the effect of sulfuric acid in acetone the lactone is converted to unsaturated 1,10-seco acid *XIV*. On acid catalysed reaction of lactone *XIII* with



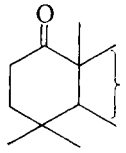
V, R = H
VI, R = CH_3
VII, R = CH_2CH_3



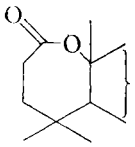
VIII



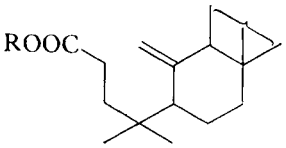
IX, R^1 = H, R^2 = CHO
X, R^1 = CH_3, R^2 = CHO
XI, R^1 = CH_3, R^2 = H



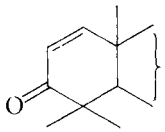
XII



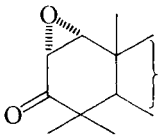
XIII



XIV, R = H
XV, R = CH_3



XVI



XVII

methanol unsaturated methyl ester *XV* is formed, identical with the ester obtained from acid *XIV* on reaction with diazomethane. In contrast to saturated ketones *I* and *XII*, the α,β-unsaturated ketone *XVI* does not give a lactone on reaction with 3-chloroperbenzoic acid. Epoxidation of the double bond takes place and the known[10] 1α,2α-epoxy ketone *XVII* is obtained as the main product.

The structures of all the compounds prepared were confirmed by infrared and ^{1}H NMR spectra. The antibacterial activity of some A-seco acids prepared in this study has already been published[1].

EXPERIMENTAL

The melting points were measured on a Kofler block and they are not corrected. Optical rotations were measured in chloroform on an automatic polarimeter, ETL-NPL (Bendix–Ericsson) with a ± 2° accuracy (at *c* 0·5 to 1·0). The infrared spectra were measured in chloroform on a UR-20 spectrometer (Zeiss, Jena); the ^{1}H NMR spectra on a Tesla BS 487A instrument (80 MHz) in deuteriochloroform, using hexamethyldisiloxane as internal reference, the chemical shifts are referred to tetramethylsilane and given in ppm (δ-scale). For thin-layer chromatography silica gel according to Stahl (Merck) was used. When working up reaction mixtures the procedure *A* consisted of washing with water, saturated sodium hydrogen carbonate and water, drying over sodium sulfate and evaporation of the solvent to dryness by distillation. Procedure *B* consisted of washing with water, 5% potassium iodide solution, water, 5% sodium sulphite solution and continuing as in procedure *A*. The identity of the substances was confirmed by mixture melting point determination, infrared spectra and thin-layer chromatography. The samples for analysis were dried under reduced pressure at 100°C, over phosphorus pentoxide.

Oxidation of 19β,28-Epoxy-18α-oleanan-3-one (*I*) with Peracids

a) A solution of ketone *I* (5 g, see ref.[7]) and 3-chloroperbenzoic acid (8 g) in dichloromethane (120 ml) was allowed to stand at room temperature in the dark for 7 days. The solution was diluted with chloroform and worked up by procedure *B*. Crystallization of the residue from chloroform and methanol gave 4·30 g (83%) of 19β,28-epoxy-4-hydroxy-3,4-seco-18α-oleanan-3-oic acid lactone (*II*), m.p. 246–248°C, $[\alpha]_D$ +101°. IR spectrum: 1 714 (C=O), 1 030 cm^{-1} (COC). ^1H NMR spectrum: 0·80, 0·91, 0·93, 1·02, 1·08, 1·39 and 1·47 (7 × CH_3), 2·30–2·90 m ($C_{(2)}H_2$), 3·54 s ($C_{(19)}H$), 3·45 d and 3·78 d (J = 8 Hz, $C_{(28)}H_2$). For $C_{30}H_{48}O_3$ (456·7) calculated: 78·89% C, 10·59% H; found: 78·59% C, 10·43% H.

The reaction was repeated under the same conditions, using various ratios of peracid and ketone *I*. The time necessary for the termination of the reaction was sought using thin-layer chromatography on silica gel. The molar ratio of 3-chloroperbenzoic acid and ketone *I* and the reaction time (in brackets) were as follows: 10·3 (16 h), 7·7 (2 days), 4·1 (7 days), 3·8 (9 days), 1·3 (the reaction is not terminated even after 30 days). The yields of lactone *II* after crystallization were 80–86%.

b) Hydrogen peroxide (30%, 10 ml) was added under stirring to acetic anhydride (48 ml) at 40°C and the mixture was stirred for another 15 min and then cooled to room temperature. A solution of ketone *I* (0·60 g) in acetic acid (40 ml) was then added and the mixture allowed to stand at room temperature for 12 days. After dilution with water the precipitate formed was filtered under suction, washed with water and dissolved in chloroform. The solution was then filtered through a layer of alumina and the chloroform distilled off. Crystallization of the residue from ethanol gave 0·53 g (85%) of lactone *II*, identical with the preparation described under *a*). M.p. 245–248°C, $[\alpha]_D$ +102°.

c) A mixture of ketone *I* (0·50 g), chloroform (10 ml), 98% formic acid (20 ml) and 30% hydrogen peroxide (10 ml) was stirred at room temperature for 17 h. The chloroform layer was separated and the aqueous layer washed with chloroform. Formic acid (20 ml) and hydrogen peroxide (10 ml) were added to the combined chloroform layers and the mixture was stirred for 12 h. Peroxide and formic acid were again added to the chloroform layer as above, and the mixture was stirred for another 15 h and then worked up using procedure *A*. Crystallization of the residue from a mixture of chloroform and methanol gave 0·42 g (81%) of lactone *II*, m.p. 245 to 247°C, $[\alpha]_D$ +100°.

d) 10% Sulfuric acid in acetic acid (3 ml) was added to a solution of ketone *I* (0·50 g) and 3-chloroperbenzoic acid (1·0 g) in dichloromethane (15 ml) and the mixture was allowed to stand at room temperature for 4 days. After dilution with ether it was processed by procedure *B*. Crystallization of the residue from methanol gave 365 mg (73%) of the lactone of (4*S*)-19β,28-epoxy-4-hydroxy-3,4-seco-24-nor-18α-oleanan-3-oic acid (*VIII*), identical with the lactone described in literature[2]. M.p. 292–295°C (decomposition), $[\alpha]_D$ +43°. Ref.[2] gives m.p. 290 to 293°C, $[\alpha]_D$ +46°. ^1H NMR spectrum: 0·79 and 0·89 (2 × CH_3), 0·92 (2 × CH_3), 0·98 (CH_3), 1·26 d (J = 6·5 Hz, $C_{(4)}CH_3$), 2·20–3·00 m ($C_{(2)}H_2$), 3·52 s ($C_{(19)}H$), 3·44 d and 3·77 d (J = 8 Hz, $C_{(28)}H_2$), 4·48 m ($W_{1/2}$ = 25 Hz, $C_{(4)}H$).

e) Boron trifluoride etherate (0·2 ml) was added to a solution of ketone *I* (80 mg) and 3-chloroperbenzoic acid (150 mg) in dichloromethane (8 ml) and the mixture was allowed to stand at room temperature for 12 h. It was then diluted with chloroform and worked up using procedure *B*. Crystallization of the residue from acetone and from ether gave 33 mg (37%) of (4*S*)-19β,28-epoxy-4-formyloxy-24-nor-3,4-seco-18α-oleanan-3-oic acid (*IX*), m.p. 230–234°C. IR spectrum: 2 400–3 500, 1 723 (COOH and CHO), 1 199 (OCHO), 1 038 cm^{-1} (COC). On reaction with

diazomethane methyl ester *X* was obtained, m.p. 194–197°C (chloroform–methanol), $[\alpha]_D$ +22°. Ref.[9] gives m.p. 223–224°C, $[\alpha]_D$ +48°. IR spectrum: 1 730 (C=O), 1 204 (OCHO), 1 038 cm^{-1} (COC). ^1H NMR spectrum: 0·79, 0·85, 0·90, 0·93, and 0·98 (5 × CH_3), 1·20 d (J = 6·6 Hz, $C_{(4)}CH_3$), 1·90–2·50 m ($C_{(2)}H_2$), 3·50 s ($C_{(19)}H$), 3·43 d and 3·76 d (J = 8 Hz, $C_{(28)}H_2$), 3·65 s (OCH_3), 5·26 m ($W_{1/2}$ = 20 Hz, $C_{(4)}H$), 7·96 s (OCHO). For $C_{31}H_{50}O_5$ (502·7) calculated: 74·06% C, 10·03% H; found: 74·32% C, 9·89% H.

A solution of ester *X* (20 mg) in benzene (3 ml) was adsorbed on a column of alumina (2 g). After two days elution with ether gave methyl (4*S*)-19β,28-epoxy-4-hydroxy-24-nor-3,4-seco-18α-oleanan-3-oate (*XI*), identical with the ester described in ref.[2]. M.p. 196–199°C (acetone). Ref.[2] gives m.p. 198–200°C.

19β,28-Epoxy-4-hydroxy-3,4-seco-18α-oleanan-3-oic Acid (*III*)

A 5% potassium hydroxide solution in ethanol (30 ml) was added to a solution of lactone *II* (0·30 g) in benzene (2 ml) and the mixture was allowed to stand at room temperature for 3 days. After dilution with water, neutralization with dilute hydrochloric acid and treatment with ether, it was submitted to procedure *A*. Crystallization of the residue from benzene–ethanol mixture gave acid *III* (246 mg, 82%), m.p. 206–208°C, $[\alpha]_D$ +58°. IR spectrum: 3 612, 3 500 (OH), 3 200–2 700, 1 740, 1 703 (COOH), 1 028 cm^{-1} (COC). For $C_{30}H_{50}O_4$ (474·7) calculated: 75·90% C, 10·62% H; found: 75·65% C, 10·52% H.

Methyl ester IV: A solution of lactone *II* (105 mg) in a mixture of methanol (5 ml), dichloromethane (2 ml) and sulfuric acid (0·002 ml) was allowed to stand at room temperature for 30 min and then diluted with chloroform and worked up using procedure *A*. Yield, 85 mg (78%) of methyl ester *IV*. M.p. 193–196°C (ether), $[\alpha]_D$ +55°. IR spectrum: 3 615(OH), 1 727, 1 436, 1 170 ($COOCH_3$), 1 028 cm^{-1} (COC). ^1H NMR spectrum: 0·80, 0·91, 0·93, 0·99, 1·01, 1·24, and 1·28 (7 × CH_3), 2·0–2·6 m ($C_{(2)}H_2$), 3·53 s ($C_{(19)}H$), 3·43 d and 3·76 d (J = 8 Hz, $C_{(28)}H_2$), 3·65 s (OCH_3). For $C_{31}H_{52}O_4$ (488·7) calculated: 76·18% C, 10·72% H; found: 76·32% C, 11·01% H. The same methyl ester was also obtained on reaction of acid *III* with diazomethane.

19β,28-Epoxy-3,4-seco-18α-olean-4(23)-en-3-oic Acid (*V*)

A mixture of lactone *II* (50 mg), ethylene glycol (0·50 ml), dichloromethane (3 ml) and sulfuric acid (0·02 ml) was allowed to stand at room temperature for 2 h, then diluted with chloroform and processed according to procedure *A*. Yield, 45 mg (90%) of acid *V*, m.p. 236–238°C, identical with a sample described in literature[3] (m.p. 238–240°C). IR spectrum: 2 400–3 600, 1 717, 1 421 (COOH), 1 037 (COC), 1 643 and 900 cm^{-1} (C=CH_2). Acid *V* was obtained in 91% yield when the reaction was carried out in a mixture of ethylene glycol with acetone.

Methyl 19β,28-Epoxy-3,4-seco-18α-olean-4(23)-en-3-oate (*VI*)

a) A solution of lactone *II* (41 mg) in a mixture of methanol (1 ml), dichloromethane (1 ml) and sulfuric acid (0·02 ml) was allowed to stand at room temperature for 5 h, then diluted with chloroform and worked up using procedure *A*. Yield, 35 mg (83%) of methyl ester *VI*, identical with a preparation described in literature[3]. M.p. 158–160°C (chloroform–methanol), $[\alpha]_D$ +61°. Ref.[3] gives m.p. 156–158°C, $[\alpha]_D$ +59·5°. IR spectrum: 1 740 (C=O), 1 647 and 901 (C=CH_2), 1 038 cm^{-1} (COC). ^1H NMR spectrum: 0·80 and 0·85 (2 × CH_3), 0·93 (2 × CH_3), 1·02 (CH_3), 1·72 bs (CH_3–C=), 1·7–2·5 m ($C_{(2)}H_2$), 3·52 s ($C_{(19)}H$), 3·42 d and 3·78 d (J = 8 Hz, $C_{(28)}H_2$), 3·64 s (OCH_3), 4·65 bs and 4·84 bs (C=CH_2).

b) A solution of ester *IV* (0·30 g) and phosphorus oxychloride (9 ml) in pyridine (45 ml) was refluxed for 2 h. After cooling it was poured onto ice and extracted with ether. The ethereal solution was washed with dilute hydrochloric acid and further worked up as in procedure *A*. Chromatography of the residue on alumina (activity II, 15 g, elution with benzene) and crystallization from a dichloromethane–methanol mixture and methanol gave methyl ester *VI* (120 mg, 34%), identical with the preparation under *a*).

c) Lactone *II* (30 mg) was heated at 250°C for 15 min and the product was methylated with diazomethane. Crystallization from methanol gave methyl ester *VI* (16 mg, 52%), identical with the preparation described under *a*).

Ethyl 19β,28-epoxy-3,4-seco-18α-olean-4(23)-en-3-oate (*VII*)

A solution of lactone *II* (94 mg) in a mixture of ethanol (2 ml), dichloromethane (2 ml) and sulfuric acid (0·05 ml) was allowed to stand at room temperature for 6 h and then diluted with chloroform and worked up using procedure *A*. Yield, 86 mg (86%) of ethyl ester *VII*, m.p. 146–148°C (chloroform–ethanol), $[\alpha]_D$ +62°. IR spectrum: 1 738 (C=O), 1 648 and 902 (C=CH_2), 1 038 cm^{-1} (COC). ^{1}H NMR spectrum: 0·80 and 0·85 (2 × CH_3), 0·93 (2 × CH_3), 1·02 (CH_3), 1·24 t and 4·10 q (J = 7·2 Hz, CH_3CH_2O), 1·71 bs (CH_3—C=), 3·52 s ($C_{(19)}H$), 3·42 and 3·78 d (J = 8 Hz, $C_{(28)}H_2$), 4·65 bs and 4·84 bs (C=CH_2). For $C_{32}H_{52}O_3$ (484·4) calculated: 79·28% C, 10·81% H; found: 79·46% C, 10·83% H. The same ethyl ester was also obtained in a 95% yield, when the reaction was carried out in ethyl acetate with 1% sulfuric acid and refluxing for 4 h.

Lactone of 19β,28-Epoxy-10α-hydroxy-1,10-seco-18α-oleanan-1-oic Acid (*XIII*)

A solution of ketone *XII* (150 mg, see ref.[8]) and 3-chloroperbenzoic acid (225 mg) in chloroform (3 ml) was allowed to stand at room temperature for 14 days, then diluted with chloroform and worked up using procedure *A*. Crystallization of the residue from a chloroform–methanol mixture gave lactone *XIII* (83 mg, 53%), m.p. 187–189°C, $[\alpha]_D$ +65°. IR spectrum: 1 708 (C=O), 1 030 cm^{-1} (COC). For $C_{30}H_{48}O_3$ (456·7) calculated: 78·89% C, 10·59% H; found: 78·72% C, 10·47% H.

19β,28-Epoxy-1,10-seco-18α-olean-10(25)-en-1-oic Acid (*XIV*)

A drop of sulfuric acid was added to a solution of lactone *XIII* (150 mg) in acetone (10 ml) and the solution was allowed to stand at room temperature for 2 h. After dilution with chloroform the mixture was worked up according to procedure *A*. Crystallization from methanol gave acid *XIV* (130 mg, 87%), m.p. 190–194°C, $[\alpha]_D$ −25°. IR spectrum: 3 200–2 600, 1 742, 1 707 (COOH), 1 640 and 891 (C=CH_2), 1 030 cm^{-1} (COC). ^{1}H NMR spectrum: 0·79, 0·82, 0·94, 0·97, 1·00 and 1·04 (6 × CH_3), 3·59 s ($C_{(19)}H$), 3·45 d and 3·81 d (J = 8 Hz, $C_{(28)}H_2$), 4·65 bs and 5·01 bs (=CH_2). For $C_{30}H_{48}O_3$ (456·7) calculated: 78·89% C, 10·59% H; found: 79·13% C, 10·81% H.

Methyl 19β,28-Epoxy-1,10-seco-18α-olean-10(25)-en-1-oate (*XV*)

A solution of lactone *XIII* (120 mg) in a mixture of methanol (10 ml) and sulfuric acid (0·1 ml) was allowed to stand at room temperature for 2 h, then diluted with ether and worked up according to procedure *A*. Yield, 105 mg (85%) of methyl ester *XV* which crystallizes from chloroform–methanol either in platelets with m.p. 99–102°C or needles with m.p. 117–119°C, $[\alpha]_D$ −24°. IR spectrum: 1 728, 1 439, 1 174 ($COOCH_3$), 1 642 and 893 (C=CH_2), 1 030 cm^{-1}

(COC). ^{1}H NMR spectrum: 0·79, 0·82, 0·94, 0·97, 0·99, and 1·04 (6 × CH_3), 3·54 s ($C_{(19)}H$) 3·42 d and 3·77 d (J = 8 Hz, $C_{(28)}H_2$), 3·64 s (OCH_3), 4·64 bs and 5·00 bs (=CH_2). For C_{31}·$H_{50}O_3$ (470·7) calculated: 79·10% C, 10·71% H; found: 78·92% C, 10·85% H. The same methyl ester was also obtained on reaction of acid *XIV* with diazomethane.

1α,2α;19β,28-Diepoxy-18α-oleanan-3-one (*XVII*)

A solution of ketone *XVI* (130 mg) and 3-chloroperbenzoic acid (500 mg) in dichloromethane (6 ml) was allowed to stand at room temperature for 12 days, then diluted with chloroform and worked up according to procedure *B*. Preparative chromatography on a silica gel thin-layer (12 g) gave 100 mg (74%) of epoxy ketone *XVII* and 22 mg of a mixture of epoxy ketone *XVII* with an unidentified by-product which decomposed during attempts at purification. Epoxy ketone *XVII* was identical with a preparation from literature[10]. M.p. 264–266°C (chloroform–ethanol), $[\alpha]_D$ +121°. Ref.[10] gives m.p. 269°C, $[\alpha]_D$ +124°. IR spectrum: 1 708 (C=O), 1 039 cm^{-1} (COC). ^{1}H NMR spectrum: 0·81, 0·91, and 0·94 (3 × CH_3), 0·98 (2 × CH_3), 1·02 and 1·09 (2 × CH_3), 3·35 d and 3·59 d (J = 4·7 Hz, $C_{(1)}H$ and $C_{(2)}H$), 3·54 s ($C_{(19)}H$), 3·45 d and 3·78 d (J = 7·9 Hz, $C_{(28)}H$).

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