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陆相硅质岩成因类型及构造—气候指示意义

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摘要 硅质岩形成于特定的地球化学条件, 具有重要的构造—气候—成岩指示意义。我国硅质岩的研究主要集中于海相地层, 陆相硅质岩虽分布广泛但研究却很薄弱, 成因解释由于借鉴海相燧石经验, 以地球化学分析为首要手段, 结论存在片面性和单一性, 可能会引起古老大陆重要气候—构造—环境信息的遗漏缺失。国内外陆相硅质岩全面调研表明, 陆相燧石主要存在三种成因类型, 包括准同生燧石、埋藏成岩燧石和表生成岩硅结岩, 各类燧石岩石矿物学特征不同, 具有独特的古气候—古地理指示意义, 因此精细的岩石—矿物学分析是辨别陆相燧石的首要手段。蒸发背景和pH变化是三类陆相燧石硅质溶解、沉降和交代的主要原因, 且陆相燧石可以当作一类特殊蒸发岩。

关键词 燧石; 硅结岩; 蒸发岩; pH变化; 碱湖

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0 引言

硅质岩形成于特定的地球化学环境, 具有重要的气候、构造、环境、成岩等指示意义^[1], 在国内外引起广泛关注。我国燧石的研究主要集中于中元古代到二叠纪海相地层, 包括华北、华南、塔里木板块的大陆边缘型燧石和昆仑—秦岭造山带、三江特提斯造山带的大洋型燧石^[2]。海相燧石普遍被认为是热水成因^[3-9]和生物成因^[10-11], 少数为次生交代成因^[12-13]。海相燧石的硅质一般认为来源于生物^[14-15], 与放射虫、海绵虫、硅藻等硅质生物活动有关^[16-17]。我国陆相盆地同样发育较多燧石, 但针对性的研究明显偏少, 且依据地球化学特征普遍被解释为热水沉积, 如新疆三塘湖盆地中二叠统芦草沟组中的硅质喷爆岩、藻纹层燧石、条带状和串珠状燧石^[18], 准噶尔盆地西北缘风城组结核状燧石^[19], 以及云南临沧陆相含煤碎屑岩盆地锆矿床中新统层状燧石^[20]。仅少数陆相燧石被解释为生物^[21]、交代^[22]或蒸发成岩成因^[23]。

显而易见, 我国陆相燧石的研究借鉴海相燧石研究经验, 重视热液成因。然而, 湖泊受局部物源、气候、构造抬升等外界因素的影响, 水体化学性质如

温度、pH等季节性变化频繁, 导致水体中的硅质溶解度远不如海洋环境稳定, 若直接借鉴海相燧石研究经验, 可能会使结果引发较大偏差。湖泊可在地表环境发育独特的原始沉降硅质, 也可在表生作用下改造先前存在的沉积岩而形成硅结岩, 这些类型的燧石在海相沉积中不存在, 这几类陆相硅质岩在国际上被广泛报道, 而在我国陆相沉积中却并未引起重视。此外, 我国陆相燧石研究, 重视地球化学手段, 忽视岩石学—矿物学差异, 易导致陆相燧石成因片面、单一。为此, 本文从SiO₂矿物类型着手, 阐明各类硅质矿物成因的指示意义; 从物理化学角度论述陆相硅质沉降及富集的主要原因; 概括陆相燧石的主要类型及特征; 针对大陆燧石的成因提出主要研究方法; 最后综述大陆燧石的气候—构造意义, 弥补国内陆相燧石研究的薄弱, 避免燧石重要而宝贵的地质信息缺漏。

1 硅质矿物类型

SiO₂矿物主要包含非晶质蛋白石, 晶质石英、玉髓和斜硅石, 这些硅质矿物在陆相硅质岩中均有发

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育。自从 Folk *et al.*^[24]报道正玉髓和水玉髓是蒸发岩曾经存在的重要指标,硅质矿物类型在指示沉积环境和成岩条件方面引起了广泛重视^[25-27]。

蛋白石类矿物呈亚稳定性,含有不同程度的结晶度、晶体结构和含水量。Jones *et al.*^[28]根据XRD非晶质硅质分布模式,将蛋白石矿物分成三组:Opal-A, Opal-C, 和 Opal-CT。Opal-A 主要在硅质微生物体中出现,但也可以是非生物成因。Opal-C 在沉积物中很少见,Opal-CT 是蛋白石中分布最普遍的矿物,其结构随含水量和晶体无序度而异^[29]。在埋藏成岩过程中,蛋白石将经历连续溶解—沉淀—重结晶过程(亦称为老化,aging),转变为石英^[30-31]。蛋白石转化程度是时间和温度的函数,可在淡水成岩过程中加速^[32-33]。

颗粒状晶质硅质矿物,根据晶体大小分为微晶石英(microcrystalline quartz <20 μm)和亮晶石英(megaquartz >20 μm)。Maliva *et al.*^[34]认为亮晶石英并不是由微晶石英随时间推移重结晶产生,而是在交代或胶结过程中直接产生。玉髓主要指纤维状晶质硅质,根据c-轴与晶体长轴方向的关系可分为四种类型。“chalcedony”一词专指负(延性)玉髓,正玉髓(quartzine)主要指正延性玉髓,水玉髓(lutecite)主要指纤维轴与晶体c-轴相交30°的玉髓,Z型玉髓(zebraic chalcedony)主要是指呈螺旋状或Z字阶梯状的玉髓。玉髓的不同结构与形成的水化学环境存在一定联系,酸性或无硫酸盐环境发育的玉髓主要是负玉髓(chalcedony),碱性或富硫酸盐或Mg的环境主要发育正玉髓和Z字型玉髓^[24]。此外,还存在一类晶质硅质矿物称为微火焰石英“microflamboyant quartz”,介于颗粒石英和纤维状石英之间^[35]。

斜硅石(moganite)是石英的亚稳定单斜结构的同质异相^[36]。斜硅石与石英在镜下很难区分,可以通过Rietveld改进的XRD分析,或者拉曼和核磁共振分析区分。斜硅石与石英在燧石中往往可以共存,尤其是在蒸发环境中^[27]。斜硅石常与Magadi-type 燧石相伴生,Heaney^[27]认为斜硅石是含水麦羟硅钠石的成岩转化物,Bustillo^[37]则认为斜硅石可以通过含镁的蒙脱石硅化而成。

2 硅质沉淀的物理化学条件

一般自然水体中二氧化硅浓度很低,溪水和地下水二氧化硅浓度介于10~60 mg/kg,海水在1~2 mg/kg之间^[38],这些浓度远低于非晶质硅质的平衡溶解度。生物活动,如硅藻和放射虫类的新陈代谢,会进一步加重水体中硅的不饱和状态^[38]。因此,自然水体,如河水、淡水湖泊、海水等,很难达到非晶质硅质的饱和度,硅质沉淀往往需要不寻常的外加条件去改变水体中硅质溶解度^[116]。

水体中硅质溶解度是pH、温度、压力的函数^[39],浅湖区或一般深度的湖泊,其压力变化对硅质溶解度的影响几乎可忽略不计。pH是影响水体中硅质溶解—沉降的最重要因素^[39],当pH在1~9范围内变化时,硅质溶解度几乎与pH变化无关,当pH>9时,硅质溶解度迅速增加^[40-41](图1a)。因此,在pH较高的湖水中,泥级或粉砂级碎屑石英和火山灰中的硅质将会溶解,而当pH降低,硅质则会重新发生沉淀^[42]。温度对硅质溶解度亦影响较大,硅溶解度随水温增高呈倍数增加^[43](图1b),当富硅热水遇到冷水时,硅的溶解度可降低10~20倍,呈过饱和状态,导致硅质直接沉淀。此外,硅质生物的新陈代谢作用吸收水

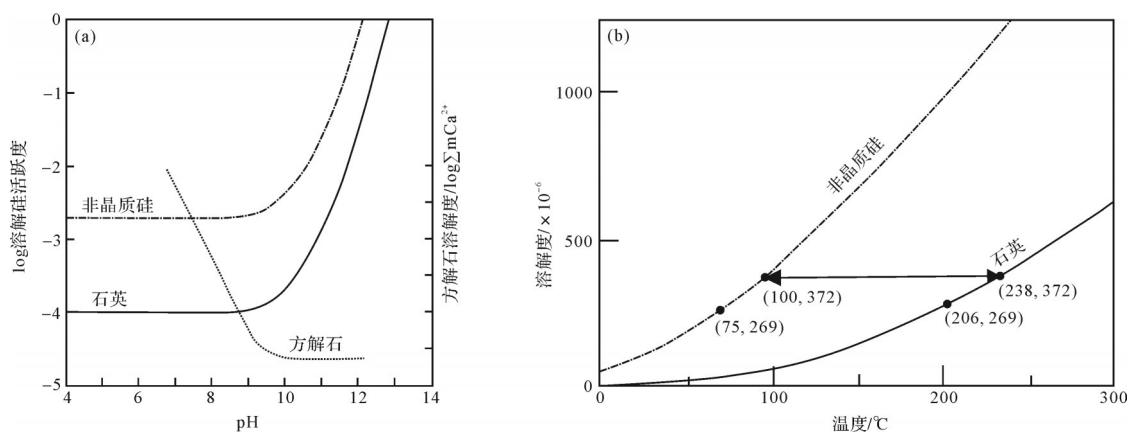


图1 非晶质硅和晶质硅在水体中的溶解度随pH^[41](a)和温度^[43](b)的变化趋势

Fig.1 Solubility curves for the silica phases with respect to pH^[41] and temperature^[43]

体中的硅质,包括少量放射虫、硅鞭藻、硅质鞭毛虫和海绵虫,这是海洋中硅质沉降的主要方式^[44-45],但在湖泊中生物沉降的硅含量视硅质生物种类和数量而定。对于前中生代海洋或湖泊,硅藻不繁盛,微生物介入可造成硅质沉淀^[26]。Laschet^[46]总结了水体中硅质直接沉淀的情况:1)富硅流体的蒸发浓缩;2)高温富硅流体的冷却;3)碱性富硅流体的中和;4)动荡的富硅流体变得稳定;5)CO₂产生,存在或者浓度增加;6)高压富硅流体的压力释放;7)不同盐度、碱度、硅质浓度的流体混合。硅质间接沉降条件包括:1)其他矿物相或有机质对硅质的吸收;2)硅分泌生物对溶解硅的利用。

3 陆相燧石的主要成因类型

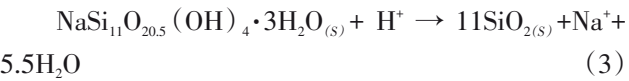
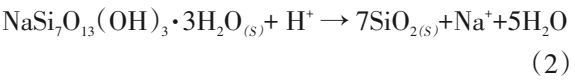
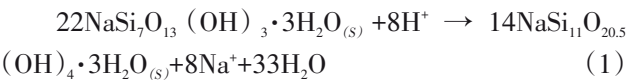
陆相水体环境的pH、温度及生物数量变化频繁,发育燧石的类型较海相沉积复杂,根据形成机制分为:准同生、早期埋藏成岩和表生成岩。

3.1 准同生燧石

准同生燧石指在低温地表或近地表环境下,由早期含水富硅矿物(Opal或含水硅酸盐)在沉淀后迅速脱水或失钠转变而成(表1)。

3.1.1 Magadi-type 燧石

自Eugster^[47-48]在肯尼亚东非裂谷Magadi湖的更新世沉积物中发现非生物湖相燧石以来,多个古老湖盆地层中陆续报道该类型燧石,如西澳大利亚Yerrida盆地古元古界Killara组的Bartle段^[49],南澳大利亚寒武纪Officer盆地^[50],苏格兰北部中泥盆统Old Red砂岩^[51-52],意大利北部二叠纪Bolzano河湖相层序^[53],美国西部侏罗纪—更新世地层^[54],以及世界著名的始新世绿河组^[55]。这一类型燧石是世界著名的Magadi-type燧石(表1)。



Magadi-type 燧石发育于半干旱碱湖地区,燧石由前期化学沉淀的麦羟硅钠石(magadiite, NaSi₇O₁₃(OH)₃·3H₂O)转化而成^[47-48,56-57]。如前所述,高碱性水体中硅质溶解度较高(图1a),不能直接沉淀出硅质,但湖水在贫铝富钠的情况下,可直接沉淀出含水含钠的麦羟硅钠石。麦羟硅钠石不稳定,在沉淀后很快通过失水、失钠形成燧石(公式2),也可先形成中间产物水羟硅钠石(kenyaite, NaSi₁₁O_{20.5}(OH)₄·3H₂O)(公式1),再转化成燧石(公式3)。这种转化过程主要发生在雨水或地表水流经的近地表部位,pH或Na⁺活性降低有助于转化。尽管这一过程涉及交代作用,但转化过程往往直接发生在麦羟硅钠石沉积之后,且只需几百年即可完成转化,因此Magadi-type燧石可近似代表原始沉积产物。

麦羟硅钠石直接从湖水中沉淀可全盆沉积,因此Magadi-type燧石单层分布广阔,厚度在5~65 cm之间,与粉砂岩或黏土互层(图2a~c),少数在黏土中呈结核状或透镜状(图2d)。由于麦羟硅钠石是直接沉积产物,通常会发育同沉积构造,包括软沉积变形和滑塌构造(图2a~c)^[50-52]。麦羟硅钠石或水羟硅钠石在脱水失钠转化成燧石的过程中,体积减小,发育V字型的收缩缝(图2e)^[50]。此外,Magadi-type燧石中可能分散含量不等的杂质,包括方解石、方沸石、钠长石、菱铁矿、伊利石、钾长石等,这些杂质含量呈现向心式规律变化,比如在燧石中心富集,在边缘接近缺失。Magadi-type燧石主要在碱湖蒸发环境中形

表1 现代湖泊中燧石特征对比:Magadi湖(肯尼亚),Coorong湖(澳大利亚)^[16]

Table 1 Chert formation in modern lakes: Lake Magadi (Kenya) and Lake Coorong (Australia)^[16]

	Magadi-type 燧石	Coorong-type 燧石
水化学	富钠,碱性,碳酸盐湖泊 干旱季节:高pH>9,硅质溶解 潮湿季节:pH波动(8~11),硅质沉降	富镁,碱性,碳酸盐湖泊 干旱季节:低pH(6.5~7),硅质沉降 潮湿季节:高pH(9.5~10.2),硅质溶解
pH控制	淡水输入和蒸发	季节性生物活动变化
燧石形成过程	Magadiite(含水含钠硅酸盐)沉降,随后脱水、脱钠转化成纯燧石	Opal-CT直接沉降,随后转化成燧石
燧石沉积构造	收缩缝 软沉积变形 角砾化	软沉积变形 角砾化 滑塌
共生矿物	天然碱	菱镁矿,镁方解石,白云石

成,常常伴生碱性矿物(如苏打石,碳钠钙石)的形成,后期经过淋滤、溶解,被方解石、白云石或硅质充填,形成蒸发岩假晶(图2f)。

3.1.2 Coorong-type 燧石

Peterson *et al.*^[40]首次在南澳大利亚 Coorong 潟湖中发现直接沉淀的非生物硅质(Opal-A),该区域也因报道世界上首例原生白云石而著名^[58-60]。在气候潮湿的季节,Coorong 湖水营养物质来源充足,微生物光合作用活跃,吸收大量 CO₂,促使水体 pH 升高,最高可达 10.2。高碱性水(pH 介于 9.5~10.2)使碎屑硅质发生溶解,释放大量溶解硅在湖水中。在气候干旱的季节,早期生长的植物体死亡,发生腐烂、降解、产生有机酸,致使湖水 pH 降低至 6.5,水体中硅质溶解度降低,非晶质硅质大量沉淀出来。与 Magadi-type 燧石不同,Coorong-type 燧石由早期 Opal-A 脱水

转化而成(表 1)。Colinvaus *et al.*^[61]在 Galapagos 岛屿的一个火山湖岩心柱中,发现了相似的带状硅质胶状物(silica gel, Opal-A),硅质发育于水深 30 m 的还原环境环境中,硫化物较多,水体盐度 52‰,有机质生产率高。

Coorong-type 燧石主要指微生物诱导 pH 变化,导致非晶质硅质(Opal-A)沉降、再脱水转化成的一类燧石。Wheeler *et al.*^[62]用 Coorong-type 燧石模式解释了加利福利亚州北部的 Deep River 盆地 Newark 群中的层状燧石。Wells^[63]用该模式解释在犹他州中部的古新一始新世 Flagstaff 湖泊发育一类较为特殊的结核状燧石,这类燧石呈层状分布,但彼此不连续,似乎置于剥蚀表面或无沉积表面,虽然呈椭球形,但并没有搬运痕迹,附近也没有细碎屑物包围。一些燧石层分布在并没有破坏的白云石夹层中,因此也不

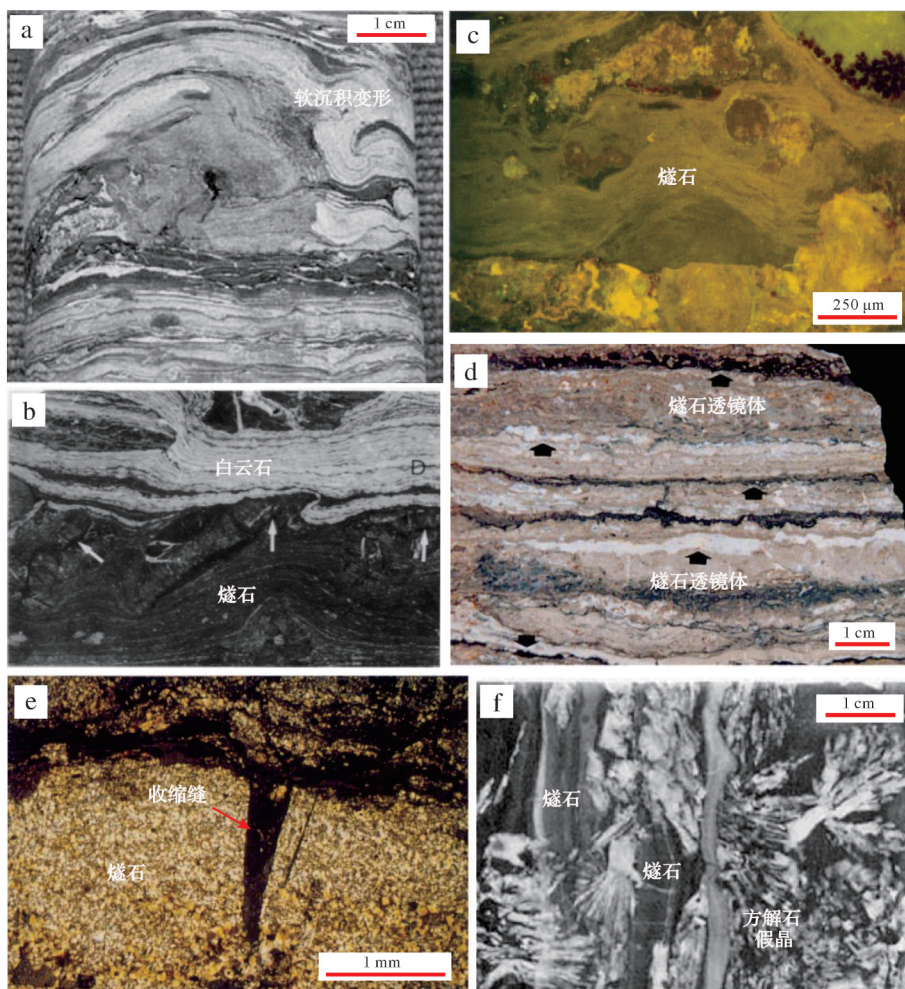


图2 发表的古老 Magadi-type 燧石岩心特征

(a)燧石上下岩层发生软沉积变形^[50]; (b)燧石层发生软沉积变形^[51]; (c)荧光照片,燧石塑性结构^[53]; (d)不规则透镜状^[53]; (e)燧石中的 V 字形收缩缝^[53]; (f)燧石伴生的方解石假晶,方解石交代菊花状苏打石^[52]

Fig.2 Published typical features of Magadi-type cherts

可能是沉积后的产物。Wells^[63]认为结核状燧石这些成层分布、彼此不连续、挤压上下层的奇特特征,是机械压实的结果。湖泊的一次突发pH下降事件,如暴雨,导致硅质大规模沉降,原始含水硅质层在湖浪的作用下来回翻滚,形成圆球状燧石,通过失水、变形和埋藏,沉降在湖底碳酸盐泥中。

3.1.3 Bogoria-type 燧石

肯尼亚 Bogoria 湖与 Magadi 湖同属东非裂谷系碱性盐湖群, Renaut *et al.*^[64]在 Bogoria 湖发现一种区别于 Magadi-type 燧石、而与热液有关的原始硅质沉积,我们称之为 Bogoria-type 燧石。Bogoria 湖岸线附近分布有 220 多个热泉口,在部分热泉口(>80 °C)附近的低洼处出现硅质沉积,硅质以胶结砾石和富集成硅壳为主,硅壳厚度可达 15 cm。这些硅质沉积主要成分是 Opal,富硅藻。Bogoria-type 燧石局限于断裂附近,与高温泉水遇到冷湖水、温度降低导致硅质溶解度降低有关。Renaut *et al.*^[65]在 Bogoria 湖泉口 Loburu 附近发现了生物主导、热液辅助形成的另一种硅质沉积,主要以微生物硅质壳、针状微型叠层石和硅壳为主。热液中的硅质沉降主要与微生物和生物薄膜有关,因为这些成分含有有利于硅质成核的活跃组分(OH⁻和羧基)。这类硅质虽与微生物密切相关,但其沉降过程属于被动沉积,不需要活微生物的代谢作用,主要依靠有机质的负电性质,吸引附近液体中的金属离子。

3.2 埋藏成岩燧石(burial diagenesis)

准同生燧石在湖相沉积物中较为特殊,其形成和富集往往与特殊沉积环境或地质事件联系在一起,如高碱性水体、火山喷发、热液输入等。实际上,除热泉地区和少数碱性湖泊外,世界上很少有湖泊正在沉积硅质。古老陆相沉积岩中的燧石,大多数被解释为交代成因^[66-70]。埋藏成岩燧石主要是在埋藏过程中,湖相碳酸盐沉积物发生强烈硅化形成的燧石。

3.2.1 硅化发生时间

碳酸盐地层的硅化可发生在埋藏成岩阶段的任何时期^[71],但要在地层中富集一定规模大小的燧石,一般在地层尚未固结的早成岩阶段。早成岩阶段,沉积物埋藏较浅,但与湖水不直接接触,温度压力没有显著变化。此阶段含硅流体流动较为通畅,可发生大规模硅化。目前,尚未有文章报道中成岩和晚成岩阶段陆相碳酸盐硅化,形成丰富燧石的事件^[41]。

早成岩过程中燧石形成的具体时间,一般通过与碳酸盐矿物胶结或成岩时间相对比而得出^[41]。Bustillo^[41]重点研究了 10 个早期硅化成岩的典型例子,早期硅化的证据包括:1)代表原始湖相或沼泽相的沉积特征,如有机质、根茎、洞穴、纹层等,被硅化破坏严重^[67];2)硅质结核形成于湖泊干涸、泥裂形成之前^[63];3)空隙充填的硅质沉淀发生在早期碳酸盐胶结形成之前或期间^[72];4)硅化发生在文石转化成方解石之前^[25,73];5)硅化发生在早期白云岩化之前;6)硅化与早期去白云岩化有关,均源于湖水中大气水的输入^[66];7)燧石碎屑混入年轻的沉积物中^[74];8)燧石的层间脱水产生裂缝、卷曲、尖灭、褶皱及蘑菇型结构^[75];9)燧石与周围碳酸盐存在差异压实^[76];10)未固结碳酸盐岩内的燧石完全硬化会产生脆弱的块体,在未固结碳酸盐岩中开裂并移位^[77]。

总体而言,陆相碳酸盐岩的硅化时间一般与湖泊从浅湖演化到沼泽环境有关,是湖退序列的最后—部分。在此阶段,碳酸盐沉积物经历了水位的频繁升降^[67,78]。碳酸盐岩的硅化主要与沼泽环境相关,该环境有机质含量较高,比如根茎、木质植物,在降解时会产生有机酸,降低pH,促使硅质沉降。

3.2.2 硅化过程

碳酸盐岩硅化的主要物质来源为硅质微生物。生物硅质(Opal A)的稳定性远远不如其他形式硅质,一旦硅质微生物包含于碳酸盐沉积物中,硅质就会发生重排列。异常高生物硅质的沉积环境,如湖泊(淡水至超咸水)和沼泽环境,最有利于发生早期硅化。在火山盆地中,火山灰和火山岩的蚀变是硅化的重要物质来源。在其他情况下,碎屑硅质的溶解、黏土矿物转化释放的硅质以及泉水输入的硅质,均可以是早期硅化的物质来源。

早期成岩过程中造成碳酸盐矿物溶解、硅质沉降的因素主要是pH变化,pH大于9,方解石沉降而硅质溶解,pH小于9,硅质沉降而方解石溶解(图1a)。埋藏环境下有三种机制可降低孔隙水中的pH:1)有机质分解产生CO₂,或者生物活动产生CO₂^[14,79];2)含水硫化物氧化,在氧化还原界面降低pH;3)海水与大陆水的混合造成方解石溶解和硅质沉降^[15]。

成岩阶段中硅化作用以充填空隙、交代碳酸盐矿物最为常见。早期成岩阶段,空隙主要有根铸模孔,白云化岩石中的孔洞、内碎屑之间的孔隙、收缩缝,生物壳铸模孔等。早期成岩阶段形成的燧石,其

形态受空隙类型和碳酸盐形态影响,以结核状(图3a,b)和不规则状(图3c)为主,部分充填生物壳体(图3d)和蒸发岩溶解形成的铸体(图3e)。与湖相硅结岩中硅质直接充填蒸发岩铸模不同(见3.3.3),埋藏成岩过程中盐类矿物铸模往往先被方解石或白云石充填,后期再被硅质交代。硅质交代碳酸盐岩具有高度选择性,母岩中方解石被硅化,白云石不受影响(图3f)。碳酸盐选择性硅化过程受pH、溶解硅活性、以及前期岩化程度(如孔隙度和渗透率)的影响。

3.3 表生成岩硅结岩(meteoric diagenesis)

表生成岩作用形成的燧石称为硅结岩(silcrete),是由 Lamplugh^[80]首次提出,主要是指近地表环境硅质聚集或交代土壤、沉积物、岩石或风化物质而形成的坚硬块体。硅结岩在澳大利亚、南非和西欧地区分布最广,在其他局部地区亦有分布^[81],时代贯穿前寒武纪到全新世。Summerfield^[82]将硅结岩(silcrete)分

为土壤硅结岩和地下水硅结岩,Nash^[81]在此基础上又增加了湖相硅结岩类型^[81](表2)。

3.3.1 土壤硅结岩(Pedogenic silcretes)

土壤硅结岩是在渗流环境下通过交替的蒸发、淋滤、渗透、沉淀作用形成,发生在碳酸盐沉积中断、地表环境暴露时期。土壤硅结岩的形成时间较长(>10⁶年),主要形成于稳定基底或盆地边缘的土壤层中^[83-84],指示沉积间断或不整合面,横向上可延伸较远^[84-85]。土壤硅结岩主要出现在热带—亚热带具有交替干湿变化特征的环境,赋存于泥质母岩中^[84,86-87]。土壤硅结岩的硅质主要来源于土壤层^[84,88]。

土壤硅结岩具有特定的剖面结构,一个典型的硅结岩剖面可划分为两部分^[81]。上部分具有柱状的硅质胶结,存在溶解和渗滤的证据,可能形成假角砾岩,更罕见的情况下形成结核状丘体。下部分以淀积为特征,被有序度不高的硅质轻微胶结,经常具有结核状面貌。

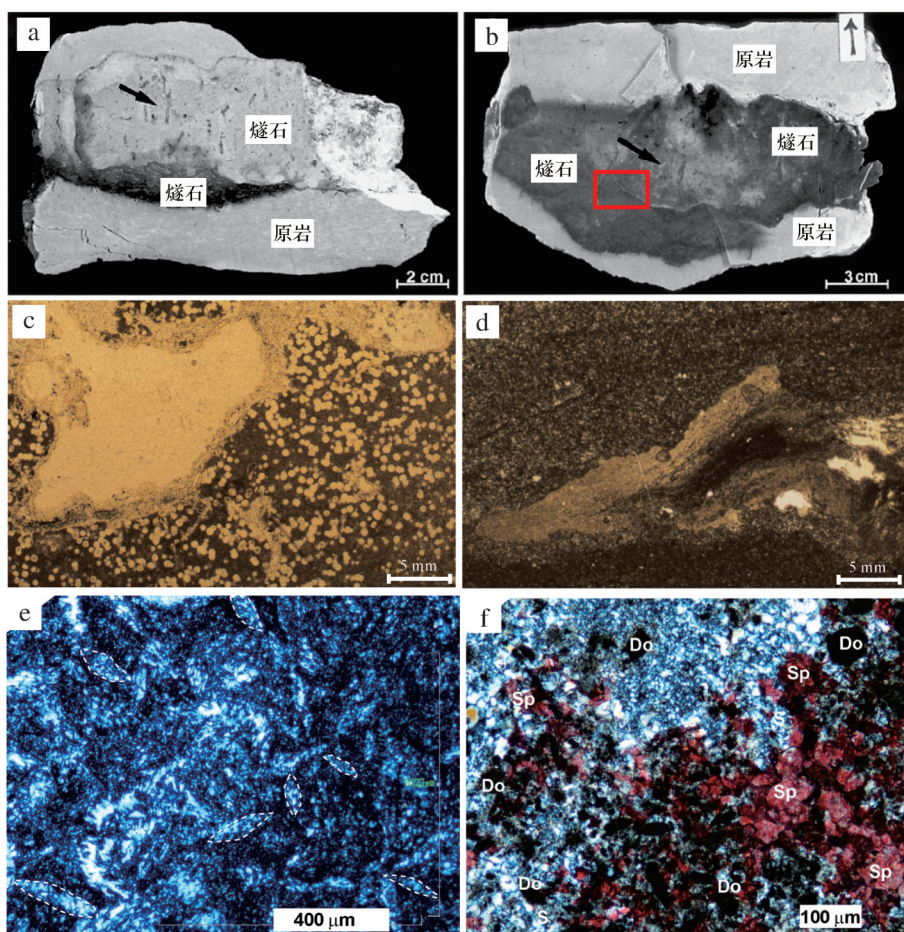


图3 埋藏成岩燧石的岩石—矿物学特征

(a,b)结核状燧石^[67]; (c)湖相泥晶灰岩中玉髓以推进的方式交代方解石^[41]; (d)湖相泥晶灰岩中生物壳体被选择性硅化^[41]; (e)燧石中显示硬石膏晶型(白色虚线框)^[68]; (f)选择性硅化作用^[68](Sp-亮晶方解石, Do-白云石)

Fig.3 Petrology and mineralogy of burial diagenetic cherts

表 2 主要硅结岩类型及其特征、形成环境对比^[81]
Table 2 Comparison of different silcretes and their morphology, structure, and environments^[81]

类型	宏观面貌	宿主	结构、化学性质	古环境
土壤 硅结岩	柱状	高岭石化沉积物	原始岩层结构破坏	土壤剖面, 热带或亚热带干湿交替, 土壤剖面硅化
			微晶石英为主	
			富 Ti	
			低 Al 和 Fe	
	连接		淀积特征	土壤剖面, 热带或亚热带干湿交替, 蒸发
			原始岩层结构破坏	
			Opal 为主	
			低 Ti	
纵向剖面分布	富黏土和 Fe			
	淀积特征			
地下水 硅结岩	块状	砂	原始岩层结构保存	地下水位与 峡谷相当
	透镜状、扁平状	砂砾	微观面貌简单	
			无淀积特征	温带或热带
	不规则状	碳酸盐岩	原始岩层结构保存	喀斯特循环
	结核状	富黏土沉积物	微观面貌简单	地下水水位起伏
			无淀积特征	温带或热带
湖相 硅结岩	结核状	湖相	简单	边缘相至蒸发凹陷
		干盐湖	燧石形状	干旱或半干旱

3.3.2 地下水硅结岩(groundwater silcretes)

地下水硅结岩是地下水水位变化或地下水发生侧向流动,造成原始地层发生硅化而形成的岩石。地下水硅结岩缺乏像土壤硅结岩那样特征鲜明的剖面组成和复杂面貌,其结构相对简单^[84]。与土壤硅结岩不同,地下水硅结岩的形成并不破坏原始碳酸盐岩的结构和构造(表 2)。地下水硅结岩的硅化程度与孔隙度具有一定关联,硅质主要以空隙充填和交代碳酸盐矿物为主^[89]。硅化作用可发生于风化或未风化地层中,在近地表或地下深度 100 m 以内的地层中均可发生,形成不连续硅质透镜体或空间重叠的燧石席。

3.3.3 湖相硅结岩

湖相硅结岩普遍发育于内流盆地中的季节性湖泊、暂时性积水或干盐湖内部或附近。在现代蒸发湖泊环境,硅质的沉淀主要受 pH 和盐度变化控制。例如,一次雨水或洪水的侵入就会使得季节性湖泊盐度降低,造成硅质溶解度降低,继而发生沉淀。湖泊中大规模水体混合区域主要位于边缘,高于地下水水位,硅化主要发生于湖泊边缘,与湖退相关^[90-95]。

湖相硅结岩的硅质趋于不规则形状,透镜状或结核状,形似花椰菜^[96](图 4a,b),以交代早期蒸发岩为主,硅质主要是正玉髓和水玉髓^[97-101]。Milliken^[35]总结了硅质交代蒸发岩矿物的岩相学特征:硅质一般呈结核状,最常见的是正玉髓,也包括水玉髓,波状微型石英等。硅质中可能夹杂各种各样的基质矿物(图 4c,d),矿物类型取决于先前湖泊沉积物组成。例如,非洲南部卡拉哈里沙漠绿色湖相硅结岩中含有海绿石—伊利石^[91]。

4 古老陆相燧石成因分析

4.1 准同生,埋藏成岩,还是表生成岩?

岩石矿物学是研究古老陆相燧石成因最为有效和直接的手段。根据燧石宏观上的沉积结构和岩石组成,微观上的硅质矿物类型的识别以及与其他矿物的成岩关系,基本可以判断燧石成因。

(1) 层状或纹层状燧石:大部分层状或纹层状陆相燧石是原始硅质沉积物脱水产物,仅有少部分是岩层完全被硅质交代的结果。根据层状燧石中特定

的沉积结构,如软沉积变形(图2a~c)、滑脱结构(图2b)、角砾、V字形收缩缝(图2e)等,可区别准同生燧石与交代成因燧石。准同生燧石在沉淀过程中,往往含有水体中的杂质矿物,这在交代矿物中并不常见。Magadi-type 燧石和 Coorong-type 燧石同属准同生燧石,均发育于碱水蒸发环境中,以发育软沉积构造变形和收缩缝为特征(表1)。Magadi-type 燧石发育于富钠碱水环境中,Coorong-type 燧石发育于富镁碱水环境中(表1)。此外,Magadi-type 燧石具有以下独有特征:1)向下的收缩缝和窗棂构造,源于magadiite向石英转化过程中的失水失钠、体积减小(图5a,b);2)较大的蒸发岩晶体铸模,源于伴生的盐类矿物(如苏打石)溶解后被硅质充填(图5c);3)石英具直线或网格状晶体取向,源于前矿物magadiite由碟状晶体堆积形成的球状集合体组成,具有优势结晶取向(图5d)^[102]。

(2) 结核状或不规则状燧石:结核状、透镜状、扁平状或不规则状燧石一般是次生成因,以空隙充填或交代碳酸盐—蒸发岩矿物为主。该类燧石研究可依据:1)硅质规则外形,如盐类矿物铸模(图3e)、生物壳体(图3d)等,可判断硅质的前驱物;2)硅质特殊外形,如花椰菜燧石(图4a,b),为湖相硅结岩;3)硅

质中交代残留矿物,如花椰菜中的硬石膏包体(图4c,d);4)根据燧石中残留的矿物成分,可建立成岩序列,燧石在交代碳酸盐矿物时,往往交代不完全,残留有原始矿物的包体(图3d,f、图4b~d),且硅质交代碳酸盐矿物具有选择性交代现象(图3f),相对于自形白云石,方解石和文石更易被硅质交代。

(3) 土壤硅结岩和地下水硅结岩的硅质主要以胶结物形式出现,硅质充填空隙或交代基质和骨架颗粒。根据骨架颗粒的密度及接触形式,硅结岩分为颗粒支撑结构、基底支撑结构、基质结构以及砾石型结构。Summerfield^[82]总结了硅结岩微观结构可能形成的一些方式,原始物质在经过淋滤、溶解、充填、交代后,原始结构可能完全改变。如颗粒支撑的砂岩在发生表生硅化作用后,形成基底式胶结结构,基底式胶结砂岩在基质和骨架颗粒被完全交代的情况下,变成基质型硅结岩。

4.2 硅质来源研究

目前,我国大部分陆相燧石研究主要利用地球化学手段研究硅质来源。实际上,陆相沉积环境不缺乏潜在的硅质来源,普遍具有多源性。陆源碎屑中的石英、长石、黏土矿物等,以及火山碎屑中的凝灰质,在湖水或孔隙水pH大于9的情况下,均有可能

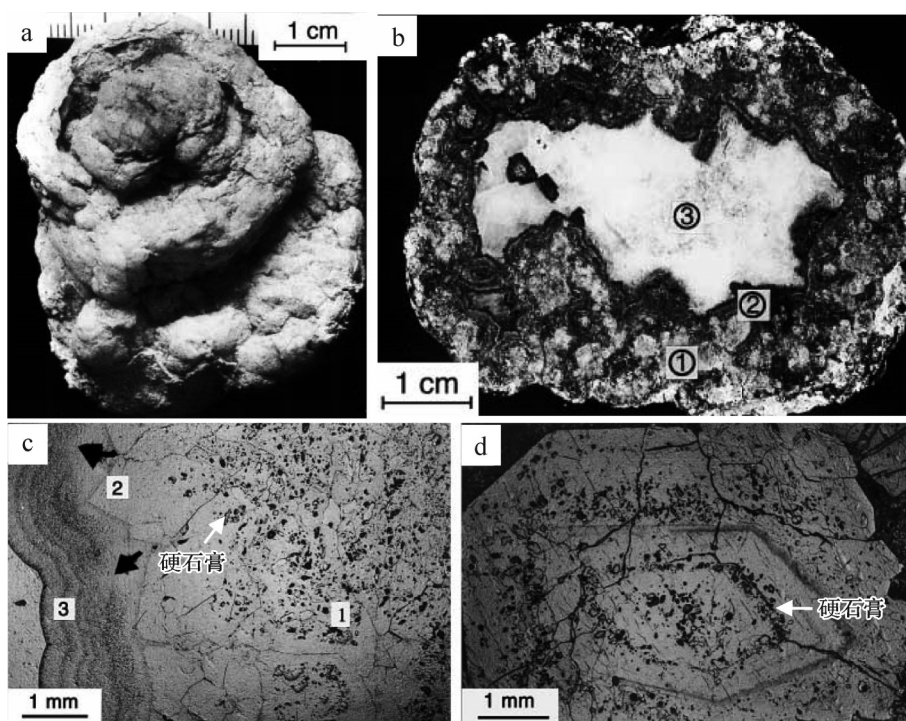


图4 大气水成岩改造湖相沉积物形成的特征性菜花状燧石^[96]

(a)花椰菜燧石结核外部形态;(b)花椰菜燧石结核内部组成:①亮晶石英,②带状白云石,③方解石胶结;(c)燧石结核中不同石英相的排列:1.亮晶石英中含有丰富的硬石膏包裹体;2.亮晶石英中不含硬石膏包裹体;3.正玉髓环带;(d)燧石结核中自形石英展示出不同期次生长带

Fig.4 Typical cauliflower cherts in meteoric diagenetically modified lacustrine sediments^[96]

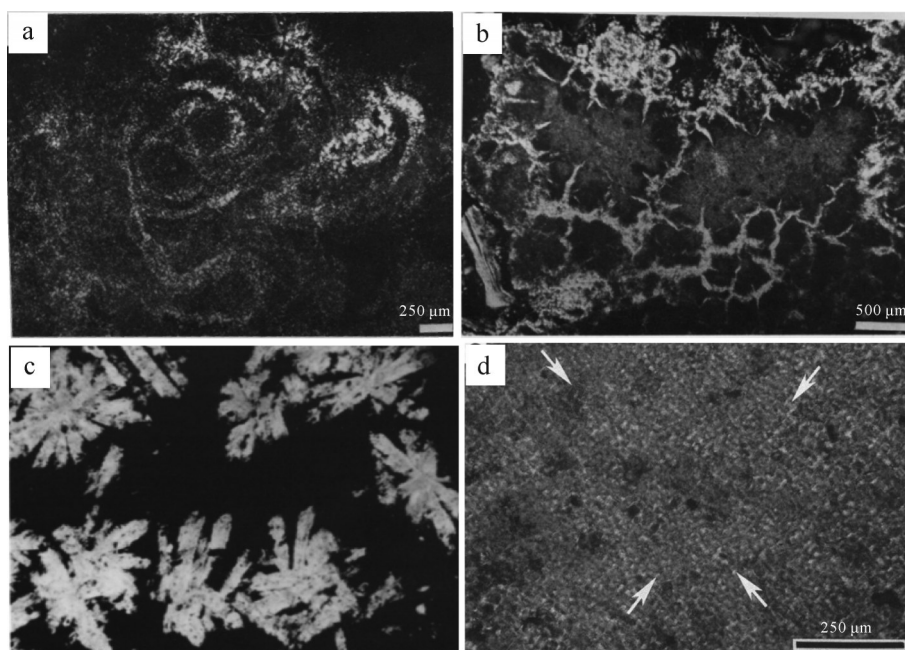


图5 Magadi-type 燧石微观矿物特征

(a)同心网状裂缝充填正玉髓^[102]; (b)燧石中的网状裂缝^[102]; (c)石英交代蒸发岩矿物(苏打石)后形成的假晶^[52]; (d)石英晶体格子状分布^[102]

Fig.5 Microscopic features of Magadi-type cherts

提供溶解硅质,在蒸发条件下溶解硅质即可发生富集。因此,蒸发碱湖是最易形成燧石的大陆环境,报道的三类准同生燧石均发生于该环境中;其次是湖泊边缘相,该相带受地下水影响,在干旱条件下水体蒸发程度远大于湖泊中心。

热泉对湖泊中溶解硅的贡献有限。一般来说,湖盆中热泉的温度不可能太高($<100\text{ }^{\circ}\text{C}$),如Bogoria湖岸边的220个热泉,温度介于 $36\text{ }^{\circ}\text{C}\sim 100\text{ }^{\circ}\text{C}$ ^[64]。水温在 $100\text{ }^{\circ}\text{C}$ 以下,石英溶解度随温度增加的幅度较小,温度增加对碎屑硅质溶解的影响有限(图1b),远小于因pH增加至9导致硅质溶解度增加的幅度。即使是热液型Bogoria-type 燧石,亦发育在碱湖中。碱性环境和蒸发条件是绝大多数陆相燧石发育的共同点。Renaut *et al.*^[65]指出,若热泉水中的硅质含量并没有达到非晶质硅质的饱和度,蒸发作用比温度降低更能促进硅质沉降,此时微生物物质可为硅质提供初始结核场所。因此,对于受地表环境控制的准同生燧石和硅结岩来说,研究pH变化和蒸发条件比研究具体硅质来源更有意义。

对于受地表条件较小的埋藏成岩燧石,硅质并非来源于原始湖泊或地下水中的溶解硅,而是原始湖泊中已沉积硅质的溶解。此时,可利用高分辨率扫描电镜可观察是否存在大量硅质微生物,从而判断生物贡献。或利用XRD和主量元素判断燧石中 SiO_2 含量、

矿物类型、和杂质矿物,因为不同成因的燧石 SiO_2 含量不同,副矿物的种类亦有差别。Yamamoto^[103]将燧石硅质来源分为四个端元(热液、生物、火山岩和陆源),利用 $\text{Al}/(\text{Al}+\text{Fe}+\text{Mn})$ 和 $\text{Al}_2\text{O}_3/\text{TiO}_2$ 比值计算不同来源的端元对燧石的贡献。

5 古老陆相燧石的构造—气候意义

不同成因的陆相燧石代表不同的成岩环境。准同生燧石的存在,本身就代表了特定成因的湖泊环境,近地表硅结岩和早成岩燧石的氢氧同位素则记录了地表水和地下水同位素比值变化,从而可以与气候或者早期地貌变化联系起来。

(1) 准同生燧石反映特殊的构造—气候环境。Magadi-type 燧石可指示古老的碱湖蒸发环境。目前,较多陆相燧石的研究致力于Magadi-type 燧石的识别,尤其针对原始沉积结构严重变形、岩石组合已发生改变的前寒武纪变质地层, Magadi-type 燧石的出现可代表在前寒武纪时期存在陆相碱性湖相蒸发环境,这对前寒武纪大陆的古地理重建具有重要意义。Coogrong-type 燧石同样发育于强蒸发盐湖环境,但主要发现于有机质丰富的富镁贫钠的碱性水体环境(表1)。Bogoria-type 燧石可直接指示断裂—热液活动和古热液喷口所在地。

(2) 次生成岩燧石(包括埋藏成岩燧石和土壤硅结岩)可指示地表环境和气候变化。目前除构造成因的燧石外,大陆岩层的硅化过程主要发生在近地表环境,例如土壤硅化作用,直接受近地表环境的影响,主要发育于干湿变化频繁的气候条件下,潮湿气候有利于硅质淋滤,干燥气候促进硅质沉降。燧石中氢氧同位素比值的变化反映了在强烈蒸发条件下地表水的氢氧同位素比值变化,从而可以与气候或者地貌变化联系起来。燧石的氧同位素测试对象是结构氧(O-Si-O),氢同位素测试对象是微晶燧石中原位OH,原位OH主要指相邻羧基团在脱水反应时留下的羟基氢和微型流体包裹体中的水分子。不同成因的燧石能很好地记录古水文变化,如果燧石是直接通过交代碳酸盐矿物而形成,那么氧同位素反映的是硅化过程中水体条件,如果燧石是早期Opal脱水而成,则氧同位素反映的是Opal重结晶过程中的信息。埋藏成岩燧石可作为一个重要工具去研究碳酸盐岩发生的化学和物理转化,例如燧石中的 $\delta^{18}\text{O}$ 就记录了碳酸盐岩的热演化史。大陆碳酸盐岩中燧石的氧同位素变化较大,大的燧石结核可反映多期结晶过程。

(3) 地下水硅结岩反映古地形变化。地下水硅结岩的形成与气候的变化联系较少,主要受古地形变化的影响。该类硅化作用一般与地下水水位变化或者沉积物中地下水的流动有关,或者与河流输入硅质有关。

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Genesis of Continental Siliceous Rocks and Their Tectonic-climatic Significance

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Abstract: Many siliceous rocks are located at key stratigraphic intervals and can provide critical information regarding the formation of sedimentary basins, tectonic orogens, and climatic settings. Previous studies have mainly focused on the siliceous rocks that developed within marine strata in China. However, in continental strata of China, siliceous rocks also have a wide distribution. Affected by the rich experience in studying marine siliceous rocks, most previous work of continental siliceous rocks was mainly based on geochemical analysis and has attributed continental siliceous rocks to biological and hydrothermal origins. Since the occurrences of continental siliceous rocks and their host rocks are much different from those hosted in marine deposits, the application of marine chert formation models to continental siliceous rocks will lead to the bias of some important conclusions on climate, tectonics, and environments. We review the genesis of continental siliceous rocks, and three types of continental siliceous rocks are introduced: penecontemporaneous cherts, burial diagenetic cherts, and meteoric diagenetic silcretes. Based on silica precursors and formation settings, three penecontemporaneous cherts are identified: Magadi-type cherts, Coorong-type cherts, and Bogoria-type cherts. Magadi-type cherts are commonly associated with saline, alkaline lakes and may be bedded lake-wide or localized in nodular deposits, forming from a sodium silicate precursor such as magadiite ($\text{Na-Si}_7\text{O}_{13}[\text{OH}]_3 \cdot 3\text{H}_2\text{O}$). Coorong-type cherts are deposited by direct Opal-crystobalite precipitation due to seasonal water pH variation and are also associated with saline, alkaline lakes. Bogoria-type cherts form from a siliceous, probably gelatinous precursor that precipitated around submerged hot springs during high lake level. Burial diagenetic cherts are formed during burial by replacing other minerals, mostly carbonates. Most of the silicification processes encountered in continental carbonates are interpreted as having occurred during the early burial diagenesis of lacustrine or palustrine sediments. Meteoric diagenetic silcretes or siliceous duricrusts are defined as the indurated products of surficial and penesurficial silica accumulation and are formed by cementation and/or low-temperature replacements of all types of rocks and sediments. Silcretes are further divided into pedogenic and groundwater silcretes. When studying continental silica rocks, petrological and mineralogical analysis are the first and foremost means for determining type. Bedded (>1 mm) and laminated (<1 mm) cherts are mostly formed during penecontemporaneous periods and are

transformed from hydrous precursors, but the possibility of the total replacement of bedded carbonated by silica cannot be excluded. Slumping, soft-sediment deformation, V-shaped shrinkage, and brecciation can further verify penecontemporaneous cherts. Characteristics of Magadi-type cherts that differ from Coorong-type cherts are (1) a ground-mass mosaic of fine quartz crystals that vary in orientation from random to rectilinear; (2) finely disseminated inclusions of silicate clays, zeolites, and/or carbonates (predominantly calcite); (3) large crystal molds (probably after trona) concentrated near sample margins; and (4) inward-directed shrinkage cracks and/or more irregular internal voids filled with chalcedony, silicate clays, zeolites, and/or carbonates (predominantly calcite). Nodular cherts are mainly diagenetic products. Based on the mold shapes and mineral remains in cherts, the replacement precursors of silica can be identified and the paragenesis between silica and other minerals can be rebuilt. Cauliflower cherts are generally lacustrine silcretes. Pedogenetic and groundwater silcretes mainly appear in the form of cements, filling voids and replacing the matrix between skeleton particles. The origins of continental cherts are quite varied. If water pH increases higher than 9, many materials in lakes can release dissolved silica into the water, like detrital quartz, silicates, and tuff materials. A pH of 9 is a key point for silica precipitation, above which silica solubility can increase exponentially. Temperature variation is not an important condition for increasing silica concentrations in continental settings, as water temperatures are generally lower than 100 °C in continental settings and not sufficient to greatly enhance silica solubility. In this regard, continental cherts are mainly associated with evaporative alkaline waters. Evaporation and pH variations are the main drivers inducing the dissolution, precipitation, and replacement of silica in continental conditions. In most cases, continental silica rocks can be regarded as a special evaporite.

Key words: cherts; silcretes; evaporites; pH fluctuations; alkaline lakes