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Supramole ular hemistry is defined as " hemistry beyond the mole ule", bearing on the organized entities of higher omplexity that result from the asso iation of two or more hemi al spe ies held together by intermole ular for es. Now, the appli ations of supramole ular hemistry in the realm of radio hemistry have attra ted mu h attention. In this arti le, the separation of important metal ions by two kinds of supramole ular systems and ioni imprinted polymers (IIP) is dis ussed, whi h demonstrates the signifi ant roles of supramole ular systems in the field of radio hemistry. Meanwhile, the first type is supermole ules resulted from the intermole ular asso iation of a host mole ule and its substrates, where the host mole ules are mainly ma ro y li ompounds (su h as rown ethers, alixarene, alix rown ethers and so on). The se ond type is supramole ular assemblies indu ed by the spontaneous asso iation of a large undefined number of omponents into a spe ifi phase, in luding reversed mi elles, mi roemulsions and liquid membranes. IIPs an re ognize and separate metal ions sele tively, whi h realizes the re ognition fun tion of supramole ular hemistry. The transport fun tion of supramole ular hemistry is also well interpreted in the separation of metal ions by liquid membrane. Besides, the perspe tives of supramole ular hemistry in the field of radio hemistry are also presented.

supramole ular hemistry; radio hemistry; separation of metal ions; ma ro y li ompounds;

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ioni imprinting

ǖ ɔ (supramole ular hemistry) ζ Ξ Ξ w ɔ ɔ Ã ɵ ǖ Ə Ώ ´ ئۆm Ẻ ᛀm ' ɔ [1]。 ǖ ɔ ɔ طm ש , Ά 、 、 、 ᛀ Ặ , Њ Џ ɔ ῴ 。 ǖ ɔ ىن (1987 ɔ)、 ɑ Louis Pasteur Jean-Marie Lehn ε "ǖ ɔ ", "ǖ Ж "。 Lehn ǖ , ǖ ɔ Ƀ ǖ ש Ξ ط[1]: (1) ǖ (supermole ules), m ǖ (m ɔ שm) ئۆ ǖ ἇ Ɑ ىن ɔ , ǖ ´ Ẻ ' ; (2) ǖ (supramole ular assemblies), ǖ ה ´ У ( ) Ẻ m ' ǖ , 、 、ش 、 、 。 ǖ ẻ ي ζ ǖ : 、 、 ᛀ , ǖ ىن ' Ƀ ἇ ᛀ 。 ɔ Џ ɔ ǖ , Ϛ ǖ 。 ɑ Cameron Њ 1910 Ό ɔ (radio hemistry) τ , ◗: ɔ ש ẹ ᑲ У ɔ ᛀ m [2]。 m ה , ɔ ط ἇ , ẹ ىن

ᛀ m щ ɔ (hemistry of radioelements), Ƀ ᛀ ɔ 。 ῴ (ᛀ) ɔ , w ש Ặ Ά ש ɔ ; ˜ (ᛀ ζ Sr、Cs) ɔ ᛀ , w ש ǖ 、 ɔ ᛀ Ά ש ɔ [2]。 , ɔ ɔ Ά Δ , ẹ Δ " " 。 ζ Ξ ǖ ᛀ ◌ ´ ǖ , ǖ ɔ ئۆ ɔ ط 。 m ǖ Ƀ ζ ǖ 、 、 ɔ ´ 。 J ǖ ζ א 、 ᛀ 。 ◌ ´ Ά ǖ , ئ Њ ɔ Ξ ǖ 。 ǖ ἇ ' ǖ 。 ئۆ ǖ , ǖ ' ἇ 。 ǖ ɔ ئۆ ɔ ط ῴ Ə ǖ 。

1967 Pedersen ´ w , ɔ ´ (、 、 、) ה 。 Њ ǖ Ɑ O、S、N, ' Ά ´ , Ύ Ẻ Ḩ , غ ' w 、 ئ ǖ 。 ᵷm , Ḩ ىن غ , ъ ' w ئ ǖ ἇ ἇ 。

2. 1

ɔ ´ ئۆ ǖ ɔ ἇ Ặ [3—7]。 ה , ἇ Ά ´ Ÿ 。 Binnemans [8] 、 w ש Ά UO_2^{2+} ´ Ÿ : UO_2^{2+} ' 6 Ɑ Ƀ ´ ; ئۆ Ά UO_2^{2+} Ḩ ; ئۆ Ά UO_2^{2+} Ḩ ; Њ ἇ Ḩ ה 。 ᑲ ǖ ẹ , ' w ζ ǖ Ά ´ Ÿ[9,10]。 Grüner [9] ´ m Ώ , ה

, , Na^+ ; , Cs^+ 。

Sr^{2+} 。 -18--6(DCH18C6)- Sr^{2+}, , Na^+、K^+、Cs^+、Ni^{2+}、Fe^{3+}、Al^{3+}、Cr^{3+}、Mo^{6+} 。

DCH18C6 Sr^{2+} , K^+ Mo^{6+} , DCH18C6 , Sr^{2+} K^+ [11—13]。 SREX(strontium extra tion) 0.1 M 4,4′-(′)- -18- -6(DtBuCH18C6, 1) 1. M (TBP) ISOPAR® L [14,1],

J ﺨ 21- -7(DCH21C7)[40] ɛ ˥ Ḣ, ‖ (PFOA⁻) ɛ א ʼ w ǚ ᾆ Sr^{2+}、UO_2^{2+}、Cs^+ ˥。 ǚ ה [41—43], ئ CO_2 ˥ , Ά ˥ Ḣ ئ - ζ- ˊ , Ά א Ў ә ˜ CO_2 , ˥ 。 ˥ ζ ˥ Ỷ Ҍ ˥ Ḣ ש̇ Ḩ ئ CO_2 。

2. 2

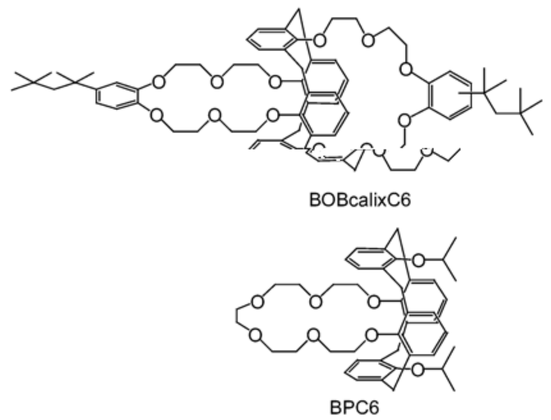
Ά ˊ א ᾆ m Ḩ , Ҍ ẹ Ė ʼ , Ύ Ά 、 w ש̇ ǚ ζ ฿ˊ , ɛ ǚ ɔ ⅄ ˜ ɕ ɥ ζ ɔˊ [44]。 ئ ǚ ς Ꞷ , ئ Δ Ặ [49], ʼ ә ɛ ﺨ ╥ ʼ ǚ ئ ئ ῎Ω, Ύ ʼ 、 [4 ,46] w ש̇ [47,48] ǚ ש̇ , ъ ǚ 、 w ש̇ ẹ ђ 。

ئ ɔ m J ﺨ ﺨ ﺨ ɔ [0, 1]、 ﺨ ɔ [2] ⅄ ء [3— 6] Ҍ Ln/An ǚ 。 [6] w ש̇ [8] 6 8 ﺨ ﺨ- , -J r ﺨ ﺨ ﺨ ɔ (CMPO) ╥ , CMPO ,Eu^{3+} ⅄ Am^{3+} ˥ ⅄ [0]。 Ҍ ء [6] , ء ɔ N ᗡ ל ˥ w ש̇ ῗ [46]。 Le ouvey [9]ˊ m , ה ǚ Ά UO_2^{2+} ˊ ῎Ω。 Ramírez [60] Ặ Ж ɔ -ʊr ﺨ [6] Ḩ ῎Ω, ה ǚ UO_2^{2+} ⅄ Th^{4+} ǚ ῎Ω ђ Ҍ La^{3+}、Eu^{3+}、Y^{3+}。 Ҍ ẹ ђ ǚ , [4] 4 ﺨ ɔˊ ʼ w ئ Na^+、K^+、Cs^+、NH_4^+ ئ ϴ ; ئ ˊ Li^+。 ǚ ῎Ω ה , ﺨ w ש̇ ﺨ O ᗡ ˊ ˊ Li^+, ῎Ω ئ ˊ [7, 8]。 w ʼ ﻍ ﺨ ъ ǚ , NO_3^- Ã Ҝ ; Ã ɵ ᾆ , ʼ w NO_3^- Ã Ҝ

。 ˊ ฿ˊ ˥ ς Ặ [61]。 w ɔˊ ǚ , Ḩ ⅄ Ҍ ɘ (;) ˊ ﻍ ﺨ ╠ ә 。

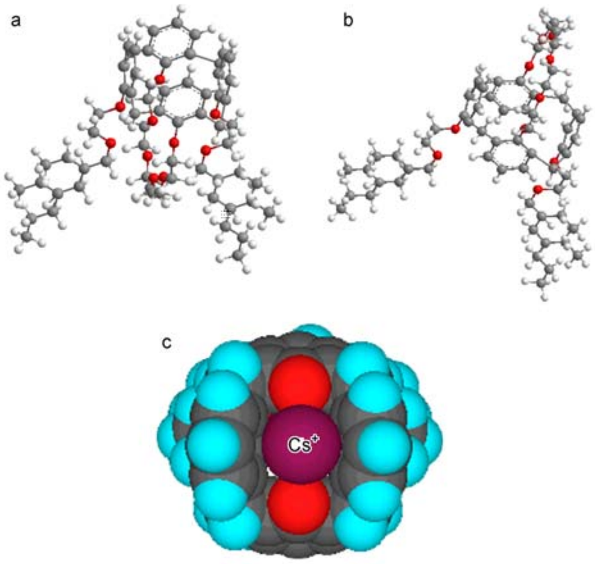
2. 3

Ė m , ǚ ʼ ⅄ Ξ ζ ǚ Ж ╥ , ╠ ә ئ Ό ᾆ ῎Ω。 ئ ǚ , Cs^+ ˥ Ό ˥ ῎Ω w ש̇ [62]。 ╞ ᾀ ɔˊ Г ﺨ [4] -6(BPC6, ه 2)/ ъ ˥ ^{137}Cs ˥ [63—66]。 1,3-[(2,4- J ф ﺨ ﺨ ф ﺨ)]-2,4- -6- [4] (alix[4]arene-R14, ه 3) m ي ǚ ᾆ Ḣ。 , HNO_3 ; , Ά - alix[4]arene-R14 , - alix[4]arene-R14 Ό Cs^+ ᾆ ῎Ω。 ز ئ SiO_2 ɔ ˊ ʼ w ǚ [24,67—70]。 Ҍ alix[4]arene-R14 ⅄ ˊ ز ئ SiO_2 ɔ ˊ , ئˊ HNO_3 ; , Ό ђ Ч , Ύ Ҍ ه 3 ǚ , ẹ ђ 、 ﻪ 、 ﻪ , Cs^+ ᾆ ῎Ω w ש̇ 。 Sessler ˊ m ฿ ᾆ Ḩ [4] ⅄ w ש̇ ᾆ Ḩ -6- [4] [71], ה ʼ w Ά CsF ئ ز 1∶1 ˊ 。 ئ ϴ ; , ʼ w Ặ Ά CsF ˊ 。



Ҍ Cs^+ ˥ ǚ [72,73]

Mole ule stru tures of alix rown ethers in the extra tion of Cs^+ [72,73]



(a) -, (b) - alix[4]arene-R14 ῦ
ẇ ẅ( ) Ά Cs(Ⅰ) ῦ [68]

Mole ular stru ture of (a) - and (b) - alix[4]arene-R14 and ( ) effe tive mat hing of size between the alixarene avity and Cs(Ⅰ) ion as well as π-bonding intera tions with the arene groups[68]

ૡ ᄀ , ᄀ
ς Ă 。Dai Ά Rogers
´ Ə[72], [4] Ј (ʊ - -6)
(BOB alixC6, 2) ૡ Cs^+ ᄀ,
K^+、Na^+、Sr^{2+} Cs^+ ᄀ ἵ。
, Na^+ ᅦ Sr^{2+} ᄂ Ά S, Cs^+
ἵ ʾ ẇ 。 Cs^+ K^+ ૡ
Ḥ ᄀ ʜ, ʾ ع ɛ
。
ૡ ᄀ ῦ
, Ό Cs^+ - ᄀ
[73]。ૡ [C mim][NTf_2] , ά
BPC6(2) ʾ ẇ ъ˘ Cs^+
ᄀ Cs^+; ૡ Ə; , ẹ ᄀ Cs^+ ῦ
ʾ 10^3 ẇ ɔ, Њ ẇ Ə ɛ Ḥ
BPC6 ʾ Cs^+ ᄀ ῦ (10),
ς Ђ Њ ẇ BOB alix6 ɛ ᄀ Ḥ ૡ
[C mim][NTf_2] Cs^+ ᄀ 。
Ə Ḥ, ʾ Cs^+ ᄀ Ὼ。
, BPC6-[C mim][NTf_2] ᄀ Cs^+ ζ
Ξ , BPC6 ᅦ Cs^+
´ Ά C mim^+ , ẇ ẅ Cs^+ Ά
C mim^+ ה 。

3.1 א

Ḥ、 Ḥ、
ᅦ ╙ ˘ ̉ Ὼ
[74]。 ῦ Њ ,
, ع Ə ɛᵐ ῦ ч Ẻ
ῦ Ὼ[7]。
Ҝ ᄀ ɔ ᄀ Ḥ ૡ ᄀ
ẇ ╥ ῦ ૡ, ᄀ ẇ ẅ Ά
Ə ɛ , Ξ
ᄀ Ḥ ᅦ ´ ῦ [76]。20 ἵ 80 ɥ
ἐ, Ὑ ẇ [77,78] ה
、
ᄀ Ḥ ૡ ᄀ ع ˜ ɔ˜
ᄀ。ђ ᄀ Ḥ ɔ ɔ
Ꞵ (W/O) ی ,
ẇ ´ Њ ,
´ Ẻ ה ,
ع ᾇ , ૡ ,
ᄀ。
ૡ / ᄀ Ҝ Ḥ(
)、 Ḥ W/O ی , Ő
ᄀ Ḥ, ᄀ 。Њ ᄀ Ḥ Ẻ
, ς ᄂ Ά 。, Osseo-Asare [79] ૡ ૡ ; , ά , 8-Ј ɸ
-7- ‖ Ј -6- (LIX 63) ᄀ Ni(Ⅱ); Yang [80] ά , -Ј Ə ɛ Ḥ, ẇ є
(TRPO) UO_2^{2+} ᄀ。ᵐ З
ᄀ , Ẻ ẻ ی א ῦ , ૡ
( ) ૡ, ૡ / ᄀ
ς ʾ ẇ , Ḥ,
ɔ˜ 、 ᄀ Ḥ ʾ
, ᄀ; ç N1923
ૡ Ə; ʾ א غ, ᄀ U(Ⅵ)
, ᄀ Ḥ , ᄀ ʾ [81]。
[82—86]。
ъ , ᄀ ῦ
, ʾ ẇ´ ᄀ , ᾇ
、ɔ ᄀ ῦ 、 є [7]。Ὑ
ẇ[87—91]、Neuman[92,93] ᅦ Osseo-Asare[94—96]
Ḥ ᄀ ,
ה א ᄀ ʾ ẇ
ᄀ ᅦ ɔ ᄀ 。ᵐ Њ
א ૡ, ʾ ẇ ˘

[79,97]

[98—101]

J (2-ϕ)

(HDEHP) (TOA)

[102,103]

[104—107]

HDEHP、2-ϕ 2-ϕ

(P 07)、

[76]

Winsor Ⅱ (Winsor Ⅱ

W/O

[7]

Neuman

[93]

[108] Ni、Co

[108—111]

[112] TBP

IR、^{1}H NMR、

TBP P═O H^+

W/O Chiarizia

[113—11] SANS TBP- J -HNO_3

U(Ⅵ)、Zr(Ⅳ)、Pu(Ⅳ)、Th(Ⅳ)

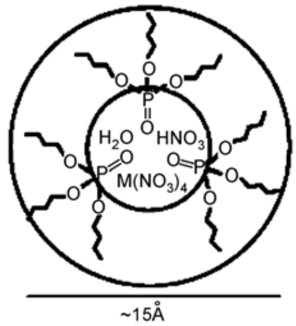
Winsor Ⅱ

4

[99,100]

Sun [116,117] N1923

Fe(Ⅲ)、Th(Ⅳ) N1923



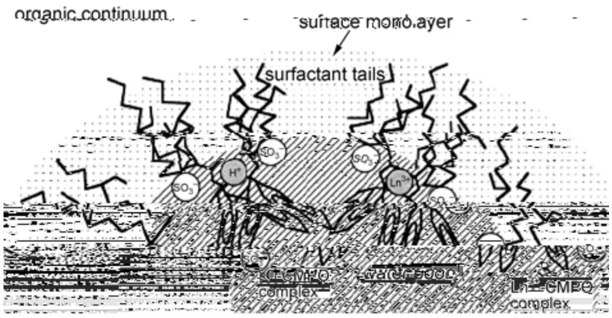
TBP 4 [114]

Idealized two-dimensional pi ture of organi phase parti les for the extra tion of the nitrate salt of a tetravalent ation from nitri a id aqueous solutions into a TBP solution in -o tane: small reverse mi elles[114]

Naganawa [118] J (2-ϕ) (AOT) J (CMPO) AOT--HNO_3

H^+-CMPO AOT

CMPO M^+-CMPO AOT



AOT Ln^{3+}-CMPO H^+-CMPO [118]

A model for the interfa ial retention of Ln^{3+}-CMPO and H^+-CMPO omplexes in the AOT reversed mi roemulsion[118]

Osseo-Asare[96,119] ε є Winsor Ⅲ (Winsor Ⅲ , Ξ ( )), є Ό Winsor Ⅱ ε Winsor Ⅲ 。Nave[120] Baxter , є , ε є ε 。

TBP Th(Ⅳ)、U(Ⅵ) є [121—124]。

, CO_2 , CO_2 。1987 Smith AOT [12], Johnston CO_2 "water-in-CO_2" [126], CO_2 "water-in-CO_2" , [127—132], [128] Sr^{2+}[133] 。 , CO_2 , CO_2 ε , , , 。 。

3.2

, є , , ε , 。 , , , , , , "　" 。6 ——(support liquid membrane, SLM) 。 , , , , ε [61,134—142]。 ε Cs^+、Sr^{2+} 。 , Kumar[143] -

pH

[138,1 0—1 2] Izatt [1 0] [ ]

(=4,6,8) Cs^+

Cs^+

Cs^+

Cs^+ Chang [1 2]

[4] Cs^+

(ioni imprinted polymer, IIP)

M^+

M^+

M^+

M^+

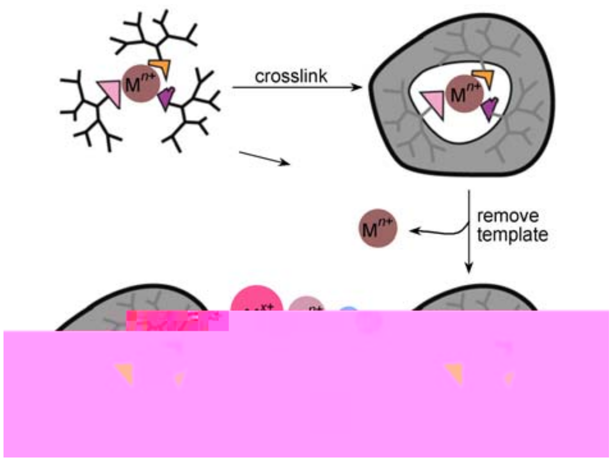
M_1^+, M_2^+

(7)

IIP

(EGDMA) (DVB)

Singh [1 3]



S hemati illustration of ioni imprinting te hnology

(SALO), 4- (4-VP),
(UO_2^{2+})
(MAA), EGDMA, UO_2^{2+}
IIP 9 μmol/g,
Th^{4+}, Cu^{2+}, Co^{2+}, Ni^{2+}, Zn^{2+} CrO_4^{2-}
UO_2^{2+}
IIP, IIP
Dai
UO_2^{2+} [1 4,1] , IIP
Th-IIP[1 6—1 8], Th^{4+}
, 7- -8-
(DCQ), 4-VP
(Sty), DVB
Nd-IIP[1 9], Sm-IIP[160], Dy-IIP[161], Er-IIP[162]
Y-IIP[163]

IIP

Shamsipur [164] MAA, EGDMA
UO_2^{2+} UO_2^{2+}

IIP 2.9 μmol/g, Fe^{3+}、Th^{4+} UO_2^{2+}

UO_2^{2+}。

La-IIP[16]、Dy-IIP[16]、Ce-IIP[16,166]、Nd-IIP[167]、Gd-IIP[168]、Sr-IIP[169,170]、Cs-IIP[171]

。

IIP

[162,172—174]。 γ-

[162,17]。 IIP

;

。

IIP

IIP

IIP

IIP

。

。

(1)

。

[176]。

[177,178],

。

Sr

Sr, Sr

[179,180]。

。

(2)

(cyclodextrins)

[44,181,182]。

[183—18]。

。

(3)

;

[82]。

(cloud point extraction, CPE)。

[84]。

U(Ⅵ)、Th(Ⅳ)

[186—189]。

。

30

。

。

Jean-Marie Lehn

[1]:

( )

( )

;

。

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。

。

18C6 18- -6
18- rown-6

4-VP 4- ϕ
4-vinylpyridine

AOT J (2- ϕ)
bis(2-ethylhexyl) sulfosu inate sodium

BF_4^-
tetrafluoroborate anion

BOB alixC6 [4] J (- -6)
alix[4]arene-bis(tert-o tylbenzo- rown-6)

BPC6 [4] -6
bis(2-propyloxy) alix[4] rown-6

alix[4]arene-R14 1,3-[(2,4- J ϕ ϕ)]-2,4--6- [4]
1,3-[(2,4-diethyl-heptylethoxy)oxy]-2,4-rown-6- alix[4]arene

CMPO - , - J
o tyl (phenyl)- , -diisobutyl arbamoylmethylphosphine oxide

C mim^+ 1- -3-
1-alkyl-3-methylimidazolium ation

CPE
loud point extra tion

DCH18C6 J -18- -6
di y lohexano-18- rown-6

DCH21C7 J 21- -7
di y lohexano-21- rown-7

DCQ ,7- J -8-
,7-di hloroquinoline-8-phenol

DtBuCH18C6 4,4′-( )- J -18- -6
4,4′-( )-di-(tert-butyl y lohexano)-18-rown-6

DVB J ϕ
divinylbenzene

EGDMA ϕ J J
ethylenegly ol dimetha rylate

HDEHP J (2- ϕ)
bis(2-ethylhexyl)phosphori a id

IIP ○
ioni imprinted polymer

LIX 63 ,8- J ϕ -7- ‖ J -6-
,8-diethyl-7-hydroxy-6-dode anone oxime

MAA
metha ryli a id

NTf_2^-
bis(trifluoromethylsulfonyl)imide anion

P 07 2- ϕ 2- ϕ
2-ethylhexyl 2-ethylhexylphosphonate

PF_6^-
hexafluorophosphate anion

$PFOA^-$ ‖
perfluoroo tanoate anion

PIM
polymer in lusion membrane

SALO
sali ylaldoxime

SLM
support liquid membrane

SPEC strontium/ esium partitioning from HLW by extra tion hromatography

SREX strontium extra tion

Sty ϕ
styrene

TBP
tributyl phosphate

TOA
tri- -o tylamine

TRPO
trialkylphosphine oxide

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