SALT LAKES OF THE AFRICAN RIFT SYSTEM: A VALUABLE RESEARCH OPPORTUNITY FOR INSIGHT INTO NATURE'S CONCENRTATED MULTI-ELECTROLYTE SCIENCE

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ABSTRACT

The Tanzanian rift system salt lakes present significant cultural, ecological, recreational and economical values. Beyond the wealth of minerals, resources of these rift salt lakes include unique wildlife, for instance Lesser Flamingo (Phoenicopterus minor) and Cichlids (Alcolapia spp), which adapted chemically aggressive environments like that of Lake Natron. The general hydrochemical equilibria of these salt lakes is governed by Na⁺ > K⁺ >> Ca²⁺ ~ Mg²⁺ : (CO₃⁻² + HCO₃⁻) > Cl > $SO_4^{2-} \sim F$ ionic concentration pattern. Salt accumulation mechanism in these endorheic lakes is considered to base on thermo-mechanical disintegrative stress of the geochemistry sourcing. The rains chemically interact with rift terrains, which are dominated with natrocarbonatitic volcanic materials, thus transporting dissolved salts to the lakes. In addition, hot springs around the shores of these lakes carry significant salinity loads into the lakes. The salt reserves magnifies through recurring evaporative concentration in seasonal cycles. Despite this huge wealth of mineral resource potential of the Tanzanian rift salt lakes, they are still underutilized, thus contributing less into the national economy. It is envisaged that studies on phase rule on separation of pure salts from lake brines as demonstrated herein have a direct industrial value addition to Tanzanian rift salt lakes.

INTRODUCTION

Economically significant reserves of the multi-electrolyte concentrates/evaporites that typify the lakes of the African rift system are to be found in Tanzania in host rift lake locations fitting the description "endorheic" (closed) basins. Most prominent among these are lakes Natron, Manyara and Eyasi plus smaller ones of lesser expanse and prominence such as Kitangiri, Balangida, Singidani, Balangidalelu, Basotu and Basoda.

Whereas the chemistry and aquatic science of salt lakes globally elsewhere have benefited from substantial scientific research; critical knowledge gaps, scanty and outdated scientific literature and diminished prioritization for the Tanzanian resources have been responsible for the stagnation in research productivity. As a result there is meager input of this salt lake electrolyte resource into the Tanzanian economy. This is indeed in conflict with the pressing country-wide demand for soda ash and allied minerals as key industrial feedstock, which currently wholly relies on importation. This realization has prompted a re-launch of local investigative chemical scientific effort by local research groups to provide quantity and quality evaluation and updating, as well as harness the scholarship value availed by the challenging domain of natural hyper-saline multi-electrolyte science.

Important scientific contrast and departures from dilute aqueous text book chemistry have been shown to characterize and apply to hyper-concentrated multi-electrolyte chemical environments. The chemistry of extremely concentrated electrolytes is known to approach that of salt melts, a frontier scientific domain that demands the development of newer models for a clear understanding of the governing equilibria. Chemical speciation, the new equilibria and processes such as pH, pKa's buffer capacities, the extreme aggressive chemical environment and adaptive mechanisms evolved by resident bioforms, the phase rule tool for extraction and separation science for pure salts may be cited among interesting rewarding key research themes thus catalyzed and pursued in the project. Many of the Tanzanian and regional rift valley salt lakes fall under the category of "soda lakes" of high pH \geq 10 (from anionic dominance of carbonates). The Na⁺ > K⁺ >> Ca²⁺ ~ Mg²⁺ : (CO₃²⁻ + HCO₃⁻) > Cl⁻ > SO₄²⁻~ F⁻ ionic concentration pattern is typical (Table 1) save for few cases such as Lake Singidani where [CO3²⁻ + HCO₃⁻] is minor and closely mirrors the non-soda Dead Sea brine chemistry.

 Table 1:
 Trend of brine ionic concentrations in Tanzanian rift valley lakes (Kameka 2006).

	Cations	Anions
Dead Sea	$Mg^{2+} > Na^+ > Ca^{2+} > K^+$	$Cl^{-} > Br^{-} >> SO_4^{2^-} \sim CO_3^{2^-}$
L. Natron	$Na^+ > K^+ >> Ca^{2+} \sim Mg^{2+}$	$(CO_3^{2-} + HCO_3) > Cl^{-} > SO_4^{2-} \sim F^{-}$
Ethiopian Lakes	$Na^+ > K^+ > Ca^{2+} \sim Mg^{2+}$	$(CO_3^{2-} + HCO_3^{-}) > Cl^{-} > F^{-} > PO_4^{3-}$
Australian Lakes	$Na^+ > K^+ > Ca^{2+}$	$Cl^{-} > SO_4^{2^-} > (CO_3^{2^-} + HCO_3^{-})$
L. Eyasi	$Na^+ > K^+ >> Ca^{2+} \sim Mg^{2+}$	$Cl^{-} > (CO_{3}^{2-} + HCO_{3}) > SO_{4}^{2-} > F^{-}$
L. Singidani	$Na^+ > Ca^{2+} \sim Mg^{2+} > K^+$	$Cl^{-} > SO_4^{2^-} > (CO_3^{2^-} + HCO_3^{-}) > F^{-}$
L. Balangida	$Na^+ > K^+ >> Ca^{2+} \sim Mg^{2+}$	$Cl^{-} > (CO_{3}^{2-} + HCO_{3}^{-}) > SO_{4}^{2-} > PO_{4}^{3-}$
L. Kitangiri	$Na^+ > Ca^{2+} > K^+ > Mg^{2+}$	$Cl > (CO_3^{2-} + HCO_3) > SO_4^{2-} > F > PO_4^{3-}$
L. Manyara	$Na^+ > K^+ > Ca^{2+} \sim Mg^{2+}$	$(CO_3^{2-} + HCO_3^{-}) > Cl^{-} > SO_4^{2-} > PO_4^{3-} > F^{-}$
L. Magadi	$Na^+ > K^+ >> Ca^{2+} \sim Mg^{2+}$	$(CO_3^{2-} + HCO_3) > Cl^{-} > SO_4^{2-} > F^{-}$
Sea Water	$Na^+ >> Mg^{2+} > Ca^{2+} \sim K^+$	$Cl^{-} >> SO_4^{-2-} > (CO_3^{-2-} + HCO_3^{}) > Br^{-} > F^{}$

Ionic concentrations are location and season dependent, peaking downstream in open stabilized lake brines and swinging with the dry and wet seasons where they become concentrated during dry spells and vice versa during the wet season. The salt accumulation mechanism hinges in salt lake basin dischargelessness of the endorheic basin which guarantees solute retention, while the negative hydrological balance, high desiccating temperatures and aridity of rift localities is responsible for hyped levels of the evaporative processes to yield the electrolyte concentrates. Selective salt precipitation into bedded sediments is another modifier of the chemical constitution patterns. The mineral salt resource potential of local salt lakes as sought out by our local effort as well as literature sources is presented here in terms of tonnage estimates for the major economically significant salts for the individual salt lakes. These lakes are proving to host competitively ample resources locally, regionally and globally, and in some of the instances such as Lake Natron, the tonnages excel any single source elsewhere worldwide in terms of soda ash reserve volumes.

RESOURCES OF THE RIFT SALT LAKES

East African Rift lakes (Fig. 1) and Tanzanian lakes in particular, support a flourishing tourism industry centred on a unique wildlife endowment particularly the cross border migratory Lesser Flamingo (*Phoenicopterus minor*) populations and a rich bird life associated with lake ecosystems. Apart from the resident fish varieties, for example Cichlids such as *Alcolapia spp* of lake Natron, that can adapt to these extreme chemically aggressive environments, fishing is not an economically significant industry on the soda lakes.

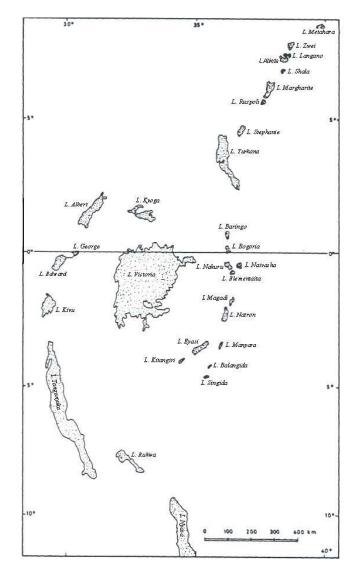


Figure 1: Closed lake basins of the East African Rift System.

The mineral potential of Tanzanian salt lakes remain largely unexploited to-date except the *Minjingu* phosphate deposits (Orr and Grantham 1931, Qulwi 1995) which is an ancient dried-up lake bed extension of Lake Manyara and the largest known such deposit in the Southern Africa Development Cooperation (SADC) region. Unlike Lake Magadi (Kenya) where commercial exploitation commenced in 1914, commercial salt production has remained virtually insignificant for the abundantly rich Tanzanian salt and soda save for peripheral lakes. domestic utilization such as medicinal, snuff additive, laundry soda and food cookability improver. Soda ash is the principal industrial feedstock chemical now wholly globally sourced from nature's soda lake reserves as the cheaper alternative, after the more expensive Solvay and Le Blanc Na₂CO₃ industrial production process options were abandoned after proving unattractive cost-wise compared to

mining of nature's cheaper supplies. In the African region, Lake Magadi continues to dominate the production while Sua Pan (Botswana) and Abiata (Ethiopia) are significant producers. An estimate of the breakdown of the reserves in Tanzanian salt lakes (Kameka 2006, Mwakoba *et al.* 1992), in terms of tonnage, is presented in Table 2. Lake Natron is singularly impressive and surpasses most others in the rift zone and in the region beyond (globally), and the economic development of its key industrial mineral reserves promises a new era of meaningful resource prosperity.

Table 2: The mineral	potential of T	Fanzanian rift	lakes in to	onnage estimates	(Kameka 2006).

	L. Natron	L. Balangida	L. Eyasi
NaCl	20,000,00	240,000	2,471,000
Na - $(CO_3^{2-} + HCO_3^{-})$	26,000,000	208,000	2,118,000
Na_2SO_4	900,000	152,000	n.d.
KCl	n.d.	8700	n.d.
Na ₃ PO ₄	30,000	n.d.	n.d.

n.d = not determined

Hydrochemical equilibria

For *soda* lakes, carbonates $(CO_3^{2-} + HCO_3^{-})$ are the dominant brine electrolytes concentration-wise in the majority of cases. The governing brine chemical equilibria, such as pH, carbonate, fluoride, phosphate and silicate, modified by high brine ionic strength conditions, has been explored (Guest and Stevens 1951, Kameka 2006, Mosha *et al.* 1996, Mwakoba *et al.* 1992, Ngambeki 1996, Qulwi 1995).

The equilibria are fundamentally important and have a governing determining role on solubility, formation, sedimentation, ionsolvent behaviour, toxicity of various forms, ecological mobilization and bio-availability pathways which are critical for food webs in the basin ecosystem. In highly concentrated multi-electrolytes, these processes are subject to major alteration (Horne 1969, Mosha *et al.* 1996) and defy conventional dilute chemical methods of interpretation.

FACTORS AFFECTING THE COMPOSITION OF LAKE BRINES pH

This is strictly a measure of a_{H+} where a =activity. It is known that $a_{H+} \neq$ effective [H⁺] if the activity coefficient $f \neq$ unity, due to restrictions on ion mobility and reactivity. In our work, we have adopted that a two pH unit downward adjustment is required (Mwakoba et al., 1992) for a direct conventional pH meter readout (which bases on the manufacturer's calibrants) to cater for this fundamental complication. Extent of reaction in these hyper saline multi-ion media is known to increase with concentration of non-participating (indifferent) electrolyte, which maximizes charge separation. It is known that pH measurement is sensitive to both $[H^+]$ and inter-ionic forces. Changes occur in the liquid junction potential at the salt bridge test solution interface. An asymmetric potential arises in the glass electrode during transfers (from dilute to concentrated solutions).

Carbonate equilibria

Carbonate $(CO_3^{2-} + HCO_3^{-})$ dominates as the governing equilibrium in soda lakes on the abundance criterion as shown in Table 1 and

2. It is among the most important equilibria in the soda lakes. An excellent treatment of the carbonate equilibrium and its consequences in soda lakes is given elsewhere (Eugester, 1970). The results of our work point generally to discernible suppression of the carbonate dissociation constants (pKa_1 and Pka_2) with ionic strength as shown in Table 3.

 pKa_2

Table 3:Carbonate pKa d	lata for selected rift soda lakes.
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L. Balangida (30 °C)	10.29	443.4	5.26 (5.18)	8.36 (8.41)
	5.09	230.4	5.38 (5.43)	8.83 (8.77)
L. Manyara (30 °C)	1.389	55.7	6.09 (6.14)	9.23 (9.30)
	1.024	43.1	6.00 (6.06)	9.49 (9.38)
Thermodynamic (25 °C)	0	0	6.37	10.25

TDS g/L

С

Phosphate equilibria and buffer capacities

Apart from its importance in the lake basin food webs, phosphate and the three-step phosphate dissociative equilibrium is an important determinant of the buffer capacity, so vital for survival of bio-forms in the chemically aggressive and challenging brine environment. Limitations of our procedure (potentiometry) permits only evaluation of pKa_1 and pKa_2 while pKa_3 eludes the method due to indiscernibility, based on inadequacy of separation of equivalence points (> six log units minimum separation requirement for adjacent equivalent points to be discernible). In common with findings by other workers (Bates 1948) who examined sea water, the acid dissociation constants of phosphoric acid increase with ionic strength (salinity). A higher pKa denotes a weaker acid. Buffer capacity based on the phosphate equilibrium was shown to increase with ionic strength and attains impressive magnitudes ranging from 36 to 1000 times that of pure water which has the magnitude of 4.6 x 10^{-7} . These magnitudes maximize tolerance of aquatic bio-forms, mitigating against large sudden potentially fatal disruptive pH swings. These chemistry tolerance mechanisms complement the bioadaptations that have evolved in response to the extreme aggressive environments, permitting species, albeit few, to survive therein.

Silicate equilibria

 pKa_1

Silicate equilibria have an important role in the salt lake ecosystem. It mobilizes silicon for bio-uptake as the basis for the various food webs in the aquatic environment, starting at the lowest trophic level(s). The element is also bio-mobilized for incorporation into skeletal structures. For aforementioned reasons regarding indiscernability of some of the equivalent points for the PO_4^{3-} case, the method enabled evaluation of pKa_1 and pKa_2 using silicic acid in artificially re-constituted "brines" of varying ionic strength to reproduce the target natural brines. The values obtained were in agreement to those of other investigators (Bates 1948) and to the thermodynamic value (after effecting appropriate extrapolations).

Salt accumulation mechanisms

Climate-driven and geochemistry sourcing categories have been advanced to account for the current levels of accumulated electrolytes. In the former version, rain chemically interacts with rocks, presubjected thermo-mechanical to disintegrative stress by day versus night temperature differences, typically over 50 °C versus below 10 °C, respectively. Such rock weathering is typically depicted and exemplified in rock formations in rift terrain such as the weathered, disintegrated rock face seen in Fig. 2 in the Lake Natron precinct. The short rains work and leach the

terrain, with the dilute rain leacheate inflow accumulating in the endorheic basin, eventually summing up to present day reserves via repetitive evaporative cycles seasonal over concentration millennia. Hot. windy desiccating conditions. and negative hydrological (evaporation exceeding balance precipitation) and aridity, are fingerprint features of rift basin locations. Salt lake brines are richer in Na⁺ than K⁺ despite almost equal natural crustal abundance (quoted at 2.9 and 2.6% respectively), as the latter bonds more efficiently and is more strongly retained by silicate rocks, thus selectively resisting leach processes into aquatic bodies.



Figure 2: Progress and effects of weathering of Lake Natron Basin rock formations; a hill in the lake basin (left) and close up details of the vertical rock face revealing the extent of the disintegrative thermo-mechanical stress influence (right).

The geochemistry of the locations of these salt lakes has a defining influence on the constitution of the accumulated salts. With the natrocarbonatitic volcanicity that dominates the Tanzanian rift system, the same will be reflected in salt lake electrolyte constitution in which soda ash type minerals will expectedly command dominance. The singularity is highlighted by the world's best known example of a natrocarbonatite lava type volcano that spews out this mineral type during eruptions; the Oldoinyo Lengai (Fig. 3) in the Lake Natron basin, this being among the only two known world-wide, the other being Homa Mountain in the Winam Gulf.



Figure 3: Natrocarbonititic volcanicity: Lake Natron's Oldoinyo Lengai Volcano (seen with the 2007 eruptive lava flows) which contributes into the lake basin's soda ash dominance as is manifest in the vast soda ash evaporites reserves in the lake (Fig. 4).



Figure 4: The vast soda ash evaporite reserve expanses of Lake Natron. (a) Saturated brine in equilibrium with the crystallizing soda, (b) Dry soda ash on the surface.

The geochemistry of a salt lake location may exert its influence on the lake's electrolyte accumulation mechanism via the geothermal springs which feed into the lake brine and whose total contribution can sum up to a dominating bulk for the target salt lake. Some of the well quoted striking examples are the respective contributions supported by daily salt load inputs of 1000 and 500 tons into Lakes Natron and Magadi respectively via the thermal spring sources that dot the periphery of the lake.

Atmospheric carbonic acid sourcing is another contribution to the overall mineral

chemistry and regeneration in the lake basin. Rain with its carbonic acid (H₂CO₃) load derived from dissolved carbon dioxide is a key player and source in the basin mineral regeneration/replenishment mechanisms. The acid contents of the atmospheric precipitation (brief rains) flow lake ward rock terrain, through leaching and chemically interacting with rock mineral content en-route that has been pre-released by the disintegrative thermo-mechanical stress mechanisms described earlier. All the enumerated mechanisms operating in a lake basin therefore sum up or partially account for the current observed levels and continual

regeneration/replenishment of the mineral reserves. This may explain why e.g. the soda ash mineral reserves such as Lake Magadi may appear as defying depletion despite a lengthy (over 100-year) industrial exploitation history.

Another modifier of the salt accumulation mechanism and hydrochemical setting for salt lakes is via early precipitation of Ca^{2+} and Mg^{2+} carbonates and gypsum as well as fluoride to bedded sediments. This implies alterations in terms of enrichment of bedded bottom sediments in terms of the less soluble types, correspondingly depleted from the aqueous electrolyte content above.

Natrocarbonatite type minerals: Stability and the governing temperature and pressure conditions

An investigation of the stability of various soda ash minerals as a function of carbon dioxide partial pressure (P_{CO_2}) has been undertaken, and this has revealed a variety of soda ash minerals as in Fig. 5 (Eugester 1970). Enumerated in the list for soda ash allied minerals typically encountered in Lake Natron are: Halite [NaCl], Nitratine [NaNO₃], Kogarkoite [Na₃FSO₄], Niter $[KNO_3],$ Thermonatrite [Na₂CO₃.H₂O], $[Na_{3}H(CO_{3})_{2}.2H_{2}O],$ Trona Pirsossonite $[Na_2Ca(CO_3)_2.2H_2O]$ and Analcime [Na(Si₂Al)O₆.H₂O], for which examples of the typifying identity and quantification, X-Ray Diffractommetry spectra. which featured and informed this work are shown in Fig. 6 (a) and (b).

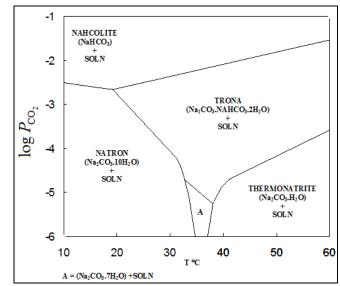


Figure 5: The stability of various soda ash minerals as a function of carbon dioxide partial pressure (P_{CO_2}) .

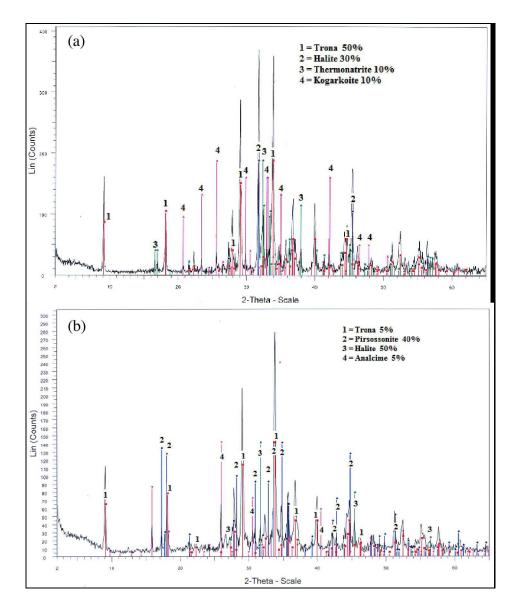


Figure 6: X-Ray diffractograms of natural Tanzanian salt lake (L. Natron) soda ash evaporites yielding identity and quantification of the soda ash allied minerals. (a) A sample of soda evaporites sourced at Eng'onguremit – Gelai flats [36 M 0172227, UTM 9724083]. (b) Sample of embedded crystalline material below crystallizing brine sourced at Takano [36 M 0822186, UTM 9717446].

Seasonal and locational dependence of electrolyte concentrations

Lake volume shrinkage during dry months interprets into brine electrolyte concentration increase. A representative example is Lake Natron, where in January 2011, the authors observed an estimate of over 60% dry-up of the lake (Fig. 4b). In this season, the brine concentrations registered Total Dissolved Solids (TDS) up to 24,900 ppm (Fig. 7). However, the situation

reverses in rainy seasons, leading to dilution. As observed in Fig. 7, the highest brine concentration during wet season was 314.5 ppm.

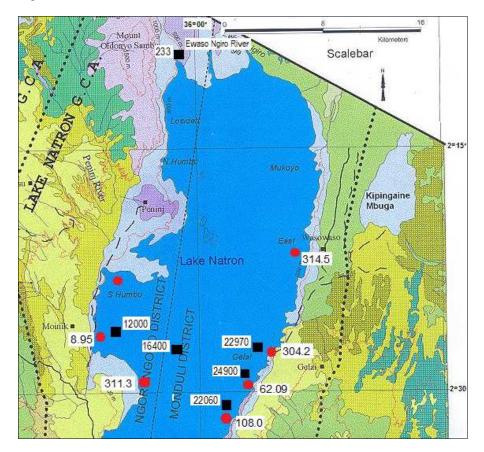


Figure 7. A map of part of the Lake Natron Basin showing sampling sites and respective brine concentrations (TDS in ppm). Wet season () and dry season ().

Open lake brines register the maximum electrolyte concentrations, while upstream sources (wells, streams, springs) show progressive dilution in that order. Depth constitution variance dependence is unimportant since these lakes have rather nominal depths, often below the one-to-two metre depth mark. The salt concentration peaks maximizes and stabilizes in the 'open lake' away from any diluting influence and/or inflow (streams, creeks, lagoons). Presence of aquatic life forms is an indicator pointing to dilution influence for brines in the locational vicinity. Very rarely, the compass axis direction can prove a concentration gradient variables well, as testified by the Lake Natron case, where a north-south concentration trending is occasioned by a major southward inflowing freshwater Ewasong'iro river whose lake diluting influence is maximum in the north, but diminishes southward, with maximum brine concentrations attained towards the south in the Wosiwosi/Tokano salt flat locations (Fig. 7).

THE PHASE RULE TOOL IN SALT SEPARATION SCIENCE

The phase rule serves the basics on which to anchor the separation science for "useful" pure salts such as NaCl, Na₂CO₃, Na₂SO₄, K_2CO_3 and K_2SO_4 from the natural aqueous multi-electrolytes. Manifestation of nature's solubility equilirbia and the applicable principle(s), can, for example, include the "Bull's eye pattern" which attends the drying-up progress for a salt lake. Here crystallizations start with the least soluble on the periphery, systematically progressing inward under those governing principles (Gale 1938, Robertson 1929, Teeple 1929).

separation Indigenous "pure salt" technologies have evolved around soda lakeside populations, for example for the Katwe volcanic crater lakes of western Uganda (Arad and Morton 1969). The same is seen in the L. Eyasi (Orr and Grantham 1931) and Lake Kitangiri (Tulia Division 1999) cases in Tanzania where end-products of exceptional purity were encountered. For the chemical investigative work, it is needed to carry out solubility equilibria studies in reconstituted laboratory test multi-electrolyte establish the crystallization brines to pathways.

Quaternary systems replicating the brine ionic strength conditions and electrolyte compositions in individual lakes were studied at 30 °C, 40 °C, 50 °C and 70 °C so as to construct isotherms and derive phase diagrams using both the Teeple and Jänecké approaches for discerning the crystallization pathways starting with a replicated constitutionally specified salt lake mother liquor (Sheshadri 1976). The Jänecké version, (Fig. 8 (b)), lends itself to proven ease of interpretation in phase rule separation science, and was used to design successful crystallization paths revealing the following mother liquor-to-product schemes:

- (ii) System NaCl +Na₂CO₃+ Na₂SO₄+ H₂O yields burkeite, Na₂CO₃.nH₂O, sodium sulphate and halite.
- (iii) System $Na_2CO_3 + Na_2SO_4 + NaHCO_3 + H_2O$ yields burkeite, trona, nahcolite and sodium sulphate.

The notation Na_2Cl_2 is a phase diagram stoichiometry convenience (improvisation) to cater for NaCl *vis-à-vis* Na_2CO_3 and Na_2SO_4 . Based on our work, recovery efficiency for a process design intended for extraction of pure salts from a soda brine, example Lake Eyasi, has demonstrated the encouragingly excellent recovery profile (Table 4). Values (except for % recovery – last column) are in kilograms.

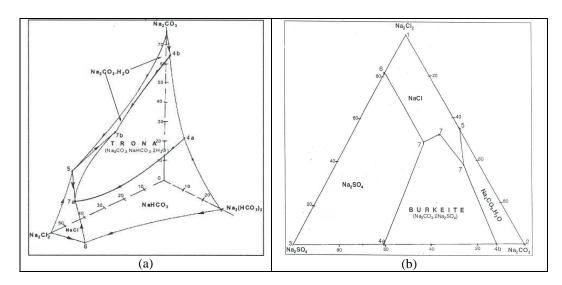


Figure 8: The quaternary system: NaCl + Na₂CO₃ + NaHCO₃ + H₂O at 50 °C: (a) Teeple; (b) Jänecké version (Sheshadri 1976).

Table 4: Pure salt recovery profile for artificially re-constituted , Eyasi brine.

Salt	Initial (mother liquor)	Extracted	Residue (bitterns)	% Recovery
NaCl	1362	1216	146	89.3
Na ₂ CO ₃	188	136	52	72.4
Na_2SO_4	111	92	18	83.8

CONCLUSION

The promising mineral resource potential of the rift salt lakes in Tanzania has remained largely unexploited with no significant contribution into the national economy, which currently relies on importation for key salts. mainly industrial carbonates. bicarbonates, sulphates, chlorides and phosphates. These industrial salts are no longer chemically manufactured, but solely mined from salt lakes elsewhere globally. Detailed scientific chemical knowledge of these lake systems, beyond the scanty and outdated available in the literature, is a key interventional pre-requisite towards harnessing and prioritizing these resources. Research targeting these knowledge gaps is among the basics for this to happen. Apart

from addressing quantity and quality, hydrochemistry of natural hyper-saline multi-electrolytes is scientifically rewarding, exciting and challenging, due to fundamental departures from dilute aqueous systems, in terms of chemical processes and equilibria, that govern such systems. Critical in such environments are the key alterations in solubility, formation, sedimentation, ionsolvent behaviour, toxicity of various forms, ecological mobilization and bioavailability pathways for food webs in the ecosystem. Additionally, phase rule studies on separation of pure salts from lake brine concentrates as demonstrated in this work, have a direct industrial value, and hitherto, only those indigenous extraction technologies independently evolved by lakeside populations are in evidence. The salt minerals of these lakes, together with other resources of these rift systems, ought to register their rightful and meaningful contribution to the national economy through those strategies, apart from catalyzing quality academic research.

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